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# PROCEEDINGS
OF THE
# INTERNATIONAL CONFERENCE ON THE PEACEFUL USES OF ATOMIC ENERGY

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Volume 13
## LEGAL, ADMINISTRATIVE, HEALTH AND SAFETY ASPECTS OF LARGE-SCALE USE OF NUCLEAR ENERGY

# Proceedings of the International Conference on the Peaceful Uses of Atomic Energy

Held in Geneva
8 August - 20 August 1955

## Volume 13
## Legal, Administrative, Health and Safety Aspects of Large-Scale Use of Nuclear Energy



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# PREFACE

The Proceedings of the International Conference on the Peaceful Uses of Atomic Energy are published in a series of 16 volumes, as follows:

| Volume Number | Title | Sessions Included |
|---|---|---|
| 1 | The World's Requirements for Energy; The Role of Nuclear Power | 2, 3.2, 4.1, 4.2, 5, 24.2. |
| 2 | Physics; Research Reactors | 6A, 7A, 8A, 9A, 10A.1. |
| 3 | Power Reactors | 10A.2, 3.1, 11A, 12A, 13A, 14A. |
| 4 | Cross Sections Important to Reactor Design | 15A, 16A, 17A, 18A. |
| 5 | Physics of Reactor Design | 19A, 20A, 21A, 22A, 23A. |
| 6 | Geology of Uranium and Thorium | 6B, 7B. |
| 7 | Nuclear Chemistry and the Effects of Irradiation | 8B, 9B, 10B, 11B, 12B, 13B. |
| 8 | Production Technology of the Materials Used for Nuclear Energy | 14B, 15B, 16B, 17B. |
| 9 | Reactor Technology and Chemical Processing | 7.3, 18B, 19B, 20B, 21B, 22B, 23B. |
| 10 | Radioactive Isotopes and Nuclear Radiations in Medicine | 7.2 (Med.), 8C, 9C, 10C. |
| 11 | Biological Effects of Radiation | 6.1, 11C, 12C, 13C.1. |
| 12 | Radioactive Isotopes and Ionizing Radiations in Agriculture, Physiology and Biochemistry | 7.2 (Agric.), 13C.2, 14C, 15C, 16C. |
| 13 | Legal, Administrative, Health and Safety Aspects of Large-Scale Use of Nuclear Energy | 4.3, 6.2, 17C, 18C. |
| 14 | General Aspects of the Use of Radioactive Isotopes; Dosimetry | 7.1, 19C, 20C. |
| 15 | Applications of Radioactive Isotopes and Fission Products in Research and Industry | 21C, 22C, 23C. |
| 16 | Record of the Conference | 1, 24.1, 24.3. |

These volumes include all the papers submitted to the Geneva Conference, as edited by the Scientific Secretaries. The efforts of the Scientific Secretaries have been directed primarily towards scientific accuracy. Editing for style has been minimal in the interests of early publication. This may be noted especially in the English translations of certain papers submitted in French, Russian and Spanish. In a few instances, the titles of papers have been edited to reflect more accurately the content of those papers.

The editors principally responsible for the preparation of these volumes were: Robert A. Charpie, Donald J. Dewar, André Finkelstein, John Gaunt, Jacob A. Goedkoop, Elwyn O. Hughes, Leonard F. Lamerton, Aleksandar Milojević, Clifford Mosbacher, César A. Sastre, and Brian E. Urquhart.

The verbatim records of the Conference are included in the pertinent volumes. These verbatim records contain the author's corrections and, where necessary for scientific accuracy, the editing changes of the Scientific Secretaries, who have also been responsible for inserting slides, diagrams and sketches at appropriate points. In the record of each session, slides are numbered in numerical order through all presentations. Where the slide duplicates an illustration in the submitted paper, appropriate reference is made and the illustration does not appear in the record of the session.

Volume 16, "The Record of the Conference," includes the complete programme of the Conference, a numerical index of papers and an author's index, the list of delegates, the records of the opening and closing sessions and the complete texts of the evening lectures.

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# Session 4.3

## LEGAL AND ADMINISTRATIVE PROBLEMS

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# Administrative and Legal Problems on the Use of Radioisotopes in Japan

By K. Suzue,* Japan

## 1. TRAINING

In order to train users of radioisotopes in the safe handling of radioactive materials and to prevent potential radiation hazards among the users, a training course is held regularly once or twice a year. The course lasts a week and consists of a series of lectures and laboratory experiments. To date, the training course was held at Tokyo University in 1951; at Kyoto University in 1952; at both Kyushu University and the National Institute for Agriculture in 1953; and at both Hokkaido and Keio Universities in 1954. Each course was held under the sponsorship of the Japan Radioisotope Association and with the support of the Scientific and Technical Administration Committee (STAC) of the Prime Minister's Office and of the Ministries concerned. The course given is generally in conformity with that of the Oak Ridge Institute of Nuclear Studies. Participants per course amounted to as many as 500 in number, and many of them were permitted to participate in laboratory experiments.

In connection with training and health safety in the use of radioisotopes, the following students were ordered to advanced countries: (1) H. Yoshikawa (Prof. of Tokyo University) to the Oak Ridge Institute in 1951; (2) F. Yamazaki (Staff member of Scientific Research Institute Ltd.) to the Oak Ridge Institute in 1951; (3) T. Inoue (Technical Officer of the National Personnel Authority) to University of London in 1953; (4) S. Nishigaki (Technical Officer of the National Institute for Agriculture) to the Oak Ridge Institute in 1954; (5) M. Izawa (Technical Officer of the National Institute of Health) to the Oak Ridge Institute in 1955; (6) Y. Hirata (Member of Scientific Research Institute Ltd.) to the Oak Ridge Institute in 1955; and (7) N. Ikeda (Assist. Prof. of Tokyo Kyoiku University) to the Oak Ridge Institute in 1955.

## 2. REGULATIONS

The STAC is taking a leading part in the preparation of "Radioactive Substances Control Act" which has been under consideration since 1952. The object of the Act is to prevent the occurrence of radiation hazards as well as to ensure public safety by controlling the production, delivery, possession, use, measurement and other aspects of the handling of radioactive substances and radiation-emitting apparatus, and by calling for the survey and disposal of radioactive contaminated materials.

The points now under discussion are as follows: (1) criteria for permitting the production, delivery, use, and so on; (2) disposal; (3) export and import; (4) transportation; (5) measurement and measuring instruments; (6) education of radiation workers; (7) qualifications of supervisors; (8) action in accidents; (9) safety inspection; (10) restrictions on the engagement of minors and inexperienced persons; (11) health examination; and (12) handling of radiation-affected persons.

The organization of the National Research Institute for Radiation, which the Science Council of Japan has recommended the Government to set up, is now under consideration in the STAC in expectation of its establishment in 1956. University courses on radiation therapy have already been set up in various universities, but the Institute is intended to be the sole consolidated basic radiation research organ in Japan.

The organization of the Institute will be approximately as shown in Table I.

## 3. WORKERS' COMPENSATION†

### General

The problem of compensation for the workman injured by harmful radiation and radioactive materials is of rather recent origin in Japan. The problem of compensation for marine workers has been discussed in the Diet but this subject has so many social, political, and even diplomatic implications that it would be out of place to discuss it here. We shall describe here the present situation of compensation for common workmen relating to injury caused by radiation, which has been in force for several years since the war.

### Compensation for Workmen by Labor Standard Law

Historically, the workmen compensation system established by law in Japan goes back for many years. In 1916 we had already a system of compensation for the worker with a considerable wide coverage as provided by the Factory Law. The Labor Standard

* Chief, Office of the Scientific and Technical Administration Committee, Prime Minister's Office.

† By the Labor Standards Bureau, Ministry of Labor.

**Table I. The Programme of the Organization of the National Research Institute for Radiation**

| *Name of section* | *Name of laboratory* | *Contents of research* |
|---|---|---|
| Physical section | Measurement laboratory | Research on measuring instruments and methods of radiation measurement |
| | First instrument laboratory | Research on diagnostic instruments |
| | Second instrument laboratory | Research on therapeutic instruments |
| | Sensitive materials laboratory | Research on X-ray films, radioautograph, and other sensitive materials |
| Chemical section | First chemical laboratory | Biochemical research on radiation |
| | Second chemical laboratory | Research on separating and processing of radioisotopes, and preparation of isotope-labeled compounds |
| | Third chemical laboratory | Research on chemical disposal of contamination |
| Biological section | First biological laboratory | Research on influence of radiation on human bodies |
| | Second biological laboratory | Research on radiobiology |
| Genetics section | Genetics laboratory | Research on genetic effects of radiation |
| First medical section | Radiation laboratory | Research on radiation to be used as a means of diagnosis |
| | Radioisotope laboratory | Research on radioisotopes to be used as a means of diagnosis |
| Second medical section | Radiation laboratory | Research on radiation to be used as a means of therapy |
| | Radioisotope laboratory | Research on radioisotopes to be used as a means of therapy |
| Third medical section | First medical laboratory | Research on maximum permissible dose of radiation in human beings |
| | Second medical laboratory | Research on diagnosis and therapy of radiation hazards |
| Work section | | Metallic, electric, and glass works, photographing (including reproduction of literature), breeding and cultivation, and management of apparatus and nondurable goods, for the purpose of promoting researches in each section |
| Office | | General Affairs Division; Accounting Division; Equipment Division; Library |

Law is its successor in a developed and improved form, under which compensation for workmen injured by harmful radiation is ensured.

In Chapter VIII of this Law we have several articles governing workmen's compensation. Article 75 stipulates the compensation for the worker who falls ill consequent upon the performance of duty, and Article 76 assures the worker of the income equivalent to 60% of his average wage as the nonduty compensation. When a worker dies in the performance of his duty, the employer must, under Article 79 of the Law, pay compensation equivalent to 1000 days average wages to the bereaved family.

In case the worker who receives compensation under Article 75 fails to recover from the injury or illness in three years from the date of his first medical treatment, the employer may discontinue the compensation prescribed in this Law after paying an expiry compensation equivalent to 1200 days' average wages of the worker. This is stipulated by Article 81 of the Law.

### Compensation for Injury or Illness Caused by Harmful Radiation

These above-mentioned Articles relate to the compensation of workers. Under the terms of the Enforcement Ordinance of the Labor Standards Law a wide range of occupational sickness is covered by Article 75 of the Law. This consists of 38 items of sickness or poisoning. The fourth of these items covers diseases caused by radium rays, ultraviolet rays, X-ray and other injurious radiations. Under Article 35 of the ordinance we compensate the worker who is suffering from diseases caused by harmful radiations.

Moreover, the scope of medical treatment is set out under the stipulations of Article 75 of the Law, including the following items—(1) medical examination; (2) supply of medicines or materials for medical treatment; (3) operations and other medical treatments; (4) hospitalization; (5) nursing; and (6) transference of the patient—which are included in Article 36 of the Ordinance. Article 37 also stipulates, in case the worker should be injured, fall ill or die while on duty in the working place or in buildings connected with the enterprise for which he works, the employer must arrange for a medical examination by a physician without delay.

However, these Articles do not indicate the seriousness of the disease for which workers shall be compensated. Since the condition of disorder caused by harmful radiations is continuous and normally be-

comes worse very gradually, it is very difficult to distinguish the disordered from the normal. Therefore, we had to determine the degree of disorder caused by harmful radiations by the following regulation.

In a case where a worker, who is usually engaged in jobs in which he is likely to be exposed to harmful radiations emitted by radium, X-ray tubes, or other ionising radiations emitted suffers from radiation sickness, this shall be considered as the disease regulated in the fourth item of Article 35 of the Enforcement Ordinance in the following circumstances:

1. When there is any cancer, ulcer, etc., of the skin.
2. When the number of red corpuscles in 1 $cm^3$ of capillary blood becomes regularly less than 4 million in case of male, or less than 3.5 million in case of female.
3. When the number of white corpuscles in 1 $cm^3$ of capillary blood becomes regularly less than 4000.

The meaning of "regularly" in these cases is as follows: it indicates that blood has been taken from the damaged workman twice in two consecutive days without any difference being shown in the average blood corpuscle count.

In addition, Article 85 of the Law prescribes that persons who have any objections to the recognition of the occupational illness or death, the method of medical treatment, the amount of compensation awarded or other matters pertaining to the compensation, may demand an investigation or arbitration, and the employing office is also authorized to demand a medical examination or autopsy when the office deems it necessary.

### Workmen Compensation Insurance

In order to fulfill his duty as prescribed by the Law, the employer sometimes has to shoulder very big burdens.

Therefore, we have a system of Workmen Compensation Insurance, which is one of the most important social insurance systems in Japan. It lessens the burden on the employer and ensures compensation for the worker.

The employer pays a premium to the government every year and the government gives the benefit to the worker who has to be compensated.

For the enforcement of the system we have the Workmen Compensation Insurance Law which has the same coverage for occupational diseases as the Labor Standard Law.

Every employer who employs a certain number of workers in hazardous conditions has to enter the system, and the worker who has been engaged in hazardous work, even if he is now retired from that work, never loses the right to be compensated, if the necessity arises.

There is a small gap in this system. If the number of workers who are engaged in hazardous work is very small in proportion to the number of other workers in the same enterprise, the employer need not enter the insurance system. Thus X-ray technicians or doctors exposed to hazardous radiations in a big hospital may sometimes lose some of the advantages of insurance, but this does not mean they lose the right to be compensated under the Labor Standard Law.

The Workmen Compensation Insurance has been in force since 1948 and costs more than 10,000 million yen every year.

## PRESENT SITUATION

From 1950 to the present, we have statistics of occupational diseases which were compensated under the above described regulations every year.

The following numbers are those which have been approved as compensated cases under the fourth item of Article 35 of the Ordinance: 1950, 8; 1951, 29; 1952, 24; 1953, 11; 1954, 36.

However, we cannot be so optimistic about the radiation hazards to which more than 20,000 workers, including medical doctors, X-ray technicians, nurses, X-ray operators of heavy industries and other workers are exposed, because there seem to be numerous unreported cases of radiation disease.

In a limited survey carried out very recently, the following surprising findings were made:

We examined 65 doctors, 75 X-ray technicians, 42 other workers at medical installations, 32 industrial workers operating X-ray machines or working with radiosotopes. Among them were found 9 doctors, 22 X-ray technicians, 9 medical workers, and 4 industrial workers who required medical treatment or rest. The proportion of affected persons was almost 20% of the total number examined, which was a surprisingly large proportion. However, as the criteria used were very strict, it is hoped that the real level of radiation sickness is a little lower than these statistics indicate. But it must be presumed, at present, that there may be a considerable number of unreported, or not yet discovered, cases of radiation disease.

We are therefore now making great efforts to establish the real extent of the hazards, to learn how to prevent it and to protect workers from hazardous conditions.

# Administrative and Legal Problems of Widespread Use of High-Level Radiation Sources—Industrial Health and Safety. Radiological Health-Safety Codes

By W. Binks,* UK

Radiation hygiene involves the study of the effects of ionizing radiation and the adoption of measures to prevent radiation injury. As regards the effects of radiation, the knowledge gained from earlier experiences with X-rays and radium has been supplemented in recent years through biological studies with a number of artificially produced radioactive isotopes. On the basis of such knowledge, recommendations have periodically been drawn up by various national and international bodies on the practical measures to be adopted to avoid injury to radiological workers. The production and widespread use of vastly increased quantities of radioactive materials do not present any radically new problems regarding radiation effects. They do, however, create not only a more difficult problem in protecting those engaged occupationally in the use of radioactive materials but also a growing public health problem.

## RESPONSIBILITY FOR PROTECTIVE MEASURES

Those responsible for framing protective measures have, in the past, tended to rely on the method of "self-regulation", that is, one in which guidance in the form of recommendations has been placed at the disposal of radiological workers and their employers whom, it has been hoped, would be sufficiently knowledgeable and thoughtful to exercise reasonable measures of control to prevent injury either to themselves or to others. While, in the main, this policy has worked, the actions of some defaulters have led to the view that stronger measures of control are required, particularly now that the consequences of such defaulting may also seriously affect the health of the public.

The primary responsibility for radiation hygiene now rests with the State. It is, indeed, becoming increasingly difficult to find a Ministry which has no part to play in radiation protection. In shouldering this responsibility, the State has to decide what measures it will adopt to control the position. It is no easy decision to make.

Since it is impossible to prohibit all use of ionizing radiations, and since any significant radiation level above the natural level of radiation from cosmic and terrestrial sources to which man has always been subjected is believed to have deleterious consequences, it is clear that, whatever control measures are sought, some degree of damage, however small, must be accepted. A calculated risk has then to be faced, both as regards injury to radiological workers and injury to the community, in a genetic sense, which may result from the exposure of "large populations" to very low-level radiation from radioactive waste products. In this connection the International Commission on Radiological Protection[1] (ICRP) has recommended that, for those occupationally exposed, the basic maximum permissible weekly dose to the critical body organs should be 0.3 rem in one week, while, for prolonged exposure of a large population, this maximum permissible level should be reduced by a factor of 10. Each country will, however, have to decide how close it will work to these maxima.

## LEGISLATIVE CONTROL VERSUS OTHER FORMS OF CONTROL

In devising control measures, several aspects must be considered:

An important difference between radiation hazards and many other occupational hazards is that the appearance of radiation effects is delayed, sometimes for 15 to 20 years. Accordingly, the problem must be viewed not in terms of injuries or absence of injuries at the present moment but in terms of what might be anticipated some years hence if inadequate measures are advocated from the start.

There are several disadvantages if complete legislative control is introduced. The doses permitted under all circumstances and the techniques to be followed must be defined very closely. This will greatly impede developments in the peaceful applications of atomic energy. In any case, it seems impossible to ensure that all the statutory rules are obeyed, however large an inspectorial staff is engaged.

Unfortunately in no country has the state been reached when recommendations plus self-regulation are adequate.

Pressure of economic forces is leading to demands for as much relaxation as possible in the permissible levels of radiation.

* Radiological Protection Service (Ministry of Health and Medical Research Council) Sutton, Surrey, England.

It is clear that the legal and administrative measures which are adopted will have a great bearing on the rate of expansion of industrial and other applications of radioisotopes and on the future health status of individual radiological workers and of the community at large. Though some measure of legislative control appears to be necessary, the statutes should be as simple and restricted as possible. The State could lay down the maximum permissible levels which must not be surpassed, but the method of achieving this statutory demand could be through codes of good practice, through guidance and supervision from health-physics groups and through the appropriate education and training of all concerned. The codes would not aim at being detailed inflexible specifications of technical procedures but would be in the nature of performance codes, giving guidance to the users of radioisotopes on the design of laboratories, shielding, general working procedures, disposal of radioactive waste, transportation of radioactive isotopes and tests for monitoring the radiation received by personnel and for checking any clinical effects. The heads of radiological departments† would thus be granted a reasonable measure of freedom in designing laboratories and in devising the techniques and protective measures to be adopted. At the same time, some of the responsibilities would devolve upon them.

This is basically the policy being pursued in the United Kingdom. Clearly there is a need for uniformity amongst the codes intended for the various specialised groups. It is hoped to effect such uniformity through liaison between the various Ministries involved, acting through an administrative and scientific advisory committee.

## BASIC FEATURES OF CODES OF PRACTICE

In preparing its recommendations on radiation safety standards, the ICRP has adopted the policy of dealing with the basic principles of radiation protection and of leaving to the various national protection committees the task of introducing such detailed technical statutory regulations or codes of practice as are considered best suited to the needs of their individual countries. The latest report of the Commission deals with the maximum permissible weekly doses for external and internal radiation sources (including the maximum permissible body burdens and maximum permissible concentrations in air and water of a large number of radioisotopes now in use); also with the general principles regarding working conditions in radiological departments and with some useful experimental data on the absorption of direct and scattered X-radiation. Unfortunately, the new report of the Commission does not deal with the handling and disposal of radioactive materials, although a subcommittee is actively engaged in preparing recommendations on these aspects.

† The ICRP defines a "radiological department" as a department or area in a medical, industrial or research organization where there is a potential radiation hazard. This same meaning will be adopted in the present contribution.

Pending the completion and publication of this additional ICRP document, it might be of interest to Delegates to the Conference to learn something about the general trend of thought in the United Kingdom as to the basic features of a code of practice to cover the use of radioactive isotopes.

In the first place, there is general acceptance of the values for the maximum permissible weekly doses and for the isotope levels in the body, in air and in water recommended by the ICRP for radiation workers. As these values are only a rough guide to the relative harmfulness of the isotopes which might be deposited in the body during operations involving their manipulation in unsealed form, the next step has been to prepare a broad classification of the isotopes according to their relative radiotoxicity, taking into account, where possible, other factors such as the types of compound in which the isotopes appear, the specific activity (depending on the degree of dilution or, in the case of naturally-occurring radioactive isotopes, on the long life), the volatility and the relative radiation doses to critical organs and tissues (including the gastro-intestinal tract) when accidental ingestion or inhalation occurs. In turn these factors have to be considered in relation to the radiochemical laboratory facilities to be provided for the quantities of isotopes to be manipulated and to the complexity of the procedures involved. The isotopes are regarded as falling into four main classes of relative toxicity, shown in Table I.

It is felt that three grades of laboratory should suffice for the handling of the various quantities of isotopes given in Table II. The lowest grade of laboratory (Grade *C*) would be any modern conventional chemical laboratory, fitted with at least one good fume hood and having floors covered with linoleum and work benches provided with non-absorbent tops or disposable covers. A Grade *B* laboratory would be a high-grade chemical laboratory which may have to be provided with glove boxes as well as fume hoods. As regards the top grade of laboratory,

**Table I**

| *Relative radiotoxicity of isotope* | *Classification* | *Grade of laboratory required for isotope at rough levels of activity specified below* | | |
|---|---|---|---|---|
| | | *C* | *B* | *A* |
| Very high | 1 | < 10 μc | 100 μc – 1 mc | > 10 mc |
| High | 2 | <100 μc | 1 mc – 10 mc | >100 mc |
| Moderate | 3 | < 1 mc | 10 mc – 100 mc | > 1 c |
| Slight | 4 | < 10 mc | 100 mc – 1 c | > 10 c |

Modifying factors to be applied to the above quantities, according to the complexity of the procedures to be followed.

| *Procedure* | *Modifying factor* |
|---|---|
| Storage (stock solutions) | × 100 |
| Very simple wet operations | × 10 |
| Normal chemical operations | × 1 |
| Complex wet operations with risk of spills<br>Simple dry operations | × 0.1 |
| Dry and dusty operations | × 0.01 |

**Table II.**
**Classification of Isotopes according to Relative Radiotoxicity per Unit Amount**

| | The isotopes in each class are listed in order of increasing atomic number. |
|---|---|
| Class 1 (very high toxicity) | $Sr^{90}$ + $Y^{90}$, *$Pb^{210}$ + $Bi^{210}$ (Ra D + E), $Po^{210}$, $At^{211}$, $Ra^{226}$ + 55% *daughter products, $Ac^{227}$, *$U^{238}$, $Pu^{239}$, *$Am^{241}$, $Cm^{242}$ |
| Class 2 (high toxicity) | $Ca^{45}$, *$Fe^{59}$, $Sr^{89}$, $Y^{91}$, $Ru^{106}$ + *$Rh^{106}$, $I^{131}$, *$Ba^{140}$ + $La^{140}$, $Ce^{144}$ + *$Pr^{144}$, $Sm^{151}$, *$Eu^{154}$, *$Tm^{170}$, *$Th^{234}$ + *$Pa^{234}$, *natural thorium, *natural uranium |
| Class 3 (moderate toxicity) | *$Na^{24}$, $P^{32}$, $S^{35}$, $Cl^{36}$, *$K^{42}$, *$Sc^{46}$, $Sc^{47}$, *$Sc^{48}$, *$V^{48}$, *$Mn^{56}$, $Fe^{55}$, *$Co^{60}$, $Ni^{59}$, *$Cu^{64}$, *$Zn^{65}$, *$Ga^{72}$, *$As^{76}$, *$Rb^{86}$, *$Zr^{95}$ + *$Nb^{95}$, *$Nb^{95}$, *$Mo^{99}$, $Tc^{99}$, *$Rh^{105}$, $Pd^{109}$ + $Rh^{109}$, *$Ag^{105}$, $Ag^{111}$, $Cd^{109}$ + *$Ag^{109}$, *$Sn^{113}$, *$Te^{127}$, *$Te^{129}$, $Cs^{137}$ + *$Ba^{137}$, *$La^{140}$, $Pr^{143}$, $Pm^{147}$, *$Ho^{166}$, *$Lu^{177}$, *$Ta^{182}$, *$W^{181}$, *$Re^{183}$, *$Ir^{190}$, $Ir^{192}$, *$Pt^{191}$, *$Pt^{193}$, *$Au^{190}$, *$Au^{196}$, *$Au^{199}$, *$Tl^{200}$, $Tl^{202}$, $Tl^{204}$, *$Pb^{203}$ |
| Class 4 (slight toxicity) | $H^3$, *$Be^7$, $C^{14}$, $F^{18}$, *$Cr^{51}$, $Ge^{71}$, *$Tl^{201}$. |

* $\gamma$-emitters.

it is most likely that the specified quantities of unsealed isotopes will be confined to work inside atomic energy research and production organizations, for which specially designed laboratories are required. From these two classifications, coupled with general information about laboratory facilities, techniques and protective clothing, it is possible to deal with one of the most frequent requests from potential users of radioisotopes, namely, as to the type of laboratory and facilities required to deal with certain levels of activity of specified unsealed isotopes.

For those who work with sealed sources of radioactive isotopes, the main problem —barring accidental fracture of the source containers—is to protect, partly by distance and partly by shielding, against the effects of penetrating $\gamma$-rays on the internal body tissues and of $\beta$-rays on the more exposed parts of the body. A reasonable amount of technical data has already been accumulated about absorption of $\beta$- and $\gamma$-rays in various materials.

It is appreciated that there is a need for radiation surveys of new or modified radiological departments, and for "personnel tests" of a clinical and physical nature. The physical tests have to deal with both external and internal radiation hazards. For external radiations, the amount received by personnel should be continuously or periodically checked by means of suitable ionization chambers or film badges carried on the body. Where there is a potential internal radiation hazard associated with work with unsealed radioactive isotopes, it may be necessary to assess the body burdens of workers from time to time from measurements of the radioactivity of the excreta (particularly of the urine) and, in certain cases, from measurement of the $\gamma$-rays or bremsstrahlung emitted by the body.

Storage and transportation of isotopes are further matters calling for guidance. The problems here, in so far as personnel are concerned, are relatively simple to cope with, since in the main they are confined to external radiation hazards. But there is a difficult administrative problem, both at the national and international level, where transportation of isotopes by public transport authorities necessitates a certain degree of segregation from other merchandise, such as sensitive unprocessed photographic film, which might be damaged by the radiation. It is the difficulty in securing the required degree of segregation, and not any difficulty in protecting transport personnel, which has delayed transport agreements.

Of all the problems created by atomic energy developments, none seems to have given rise to greater difficulties, or caused more public alarm, than that of the disposal of radioactive waste. Radioactive waste may leave an establishment in the form of a gaseous effluent (from an atomic energy pile or from the flue of a hospital incinerator in which is burned any highly contaminated clothing or bedding of patients undergoing treatment with internally administered radioisotopes); in the form of liquid effluents (including excreta from radioactive patients) which may enter drinking water supplies by direct or indirect routes or may flow into river estuaries; or in the form of solid waste which, at low levels of activity, can be removed as ordinary waste by the sanitary authorities or, at higher levels of activity, be placed in disposal pits or buried at sea. In the United Kingdom, most of the waste disposal problems arising up to the present time have been dealt with satisfactorily, though a workable scheme has yet to be devised for centralised solid waste disposal.

Guidance has also to be given on decontamination procedures and on the permissible levels of contamination for personnel, for personal and protective clothing, for "active laboratories" and "inactive areas", for equipment, and for patients' clothing and bedding before it is considered safe to release to public laundries. The recommended levels for parts of the body, for personal clothing, hospital bedding and inactive areas are $10^{-5}$ $\mu c/cm^2$ of surface for $\alpha$-emitters of very high toxicity (see Table I) and $10^{-4}$ $\mu c/cm^2$ for $\beta$-emitters of all classes. These levels are increased by a factor of 10 for protective clothing, active laboratories and equipment.

## ADMINISTRATIVE PROBLEMS ASSOCIATED WITH THE IMPLEMENTATION OF CODES OF PRACTICE

Many codes of practice refer to the duties of appropriate protection organizations or health physicists or other qualified experts in giving advice on the plans of new radiological departments, and in carrying out personnel tests and radiation surveys of departments. In the United Kingdom, the major radiotherapeutic departments which are using up to 2000 curies of sealed $\gamma$-ray emitters for teletherapy treatment or up to a few curies per year of isotopes

such as $P^{32}$ and $I^{131}$ for internal administration to patients have health physicists on the staff who are responsible for the necessary safety measures and monitoring procedures. Again, the departments within the Atomic Energy Authority for the United Kingdom have health-physics sections which perform the duty laid upon the Authority in the Atomic Energy Authority Act, 1954, "to secure that no ionizing radiations from anything on any premises occupied by them, or from any waste discharged (in whatever form) or from any premises occupied by them, cause any hurt to any person or damage to any property, whether he or it is on any such premises or elsewhere." But outside these specialist groups there are several thousands of workers in smaller hospitals and in industrial, University and Government departments who look to others to provide these surveying and monitoring services. This work, at present undertaken by the National Physical Laboratory, is to be transferred to, and expanded under, the new Radiological Protection Service, which is a centralised unit set up by the Ministry of Health and Medical Research Council. The aim of the Service is to give as much help as possible to radiation workers in dealing with protection problems and to further the general policy of the country to maintain a satisfactory state of affairs as regards radiation hazards through guidance, supervision and education rather than wholly through statutory regulations.

In attempting to operate protection services, it appears that some departure from ICRP principles is necessary. The Commission expresses permissible weekly dose in terms of the quantity of ionizing radiation received *in one week* and not *per week*. Limitations in available staff have however, in general, resulted in radiation monitoring tests being conducted over a period of not less than 2 weeks. Furthermore, difficulties arise as to the procedure to be followed after a person has been over-exposed, either accidentally, or in a planned emergency which may arise, with high-level activities. If he has received, say, a dose of 1.2 rems in one week (that is, four times the permissible dose), should he be removed completely from duties involving radiation exposure for the next three weeks or should he be allowed to continue working under conditions involving much less radiation exposure for a compensatory period of, say, 12 weeks until the average for the total 13-week period is below the permissible weekly dose? If the former course is adopted, work may be impeded; if the latter—and it does not seem unreasonable—permissible weekly dose is, in effect, averaged over a period of 13 weeks.

## INTERNATIONAL COOPERATION

There are, of course, several administrative features of an international character. There is need for the collection and dissemination of basic information about radiation effects and protective measures. Again, transportation of radioactive materials, disposal of radioactive waste, and agreement on a symbol for radiation hazards are matters of international concern, whilst a serious accident to a nuclear power reactor in any country is likely to have world-wide repercussions.

It would seem that there is likely to be a race between nations in developing nuclear power reactors and in attempting to capture world atomic markets. It is not impossible that, in such a race, competitive considerations will lead to drastic economies on radiation protection being urged. There is thus need for international agreement on the minimum standards to be allowed. In these respects, international bodies such as the International Commission on Radiological Protection, the International Labour Office and the World Health Organization have a very important function to fulfil.

## REFERENCES

1. Recommendations of the International Commission on Radiological Protection, British Journal of Radiology, Supplement No. 6 (1955).

# The General Problems of Protection Against Radiations from the Public Health Point of View

**World Health Organization**

The appearance of radiation hazards dates from the discovery of X-rays and radium at the turn of the century. The adverse biological effects of excess exposure to radiation* were soon recognized, but very little was done to protect against them until the first world war, when the diagnostic use of X-rays became widespread. After the war, the need for radiological protection was recognized in many countries, first among enlightened groups in hospitals, and later on a national basis. In 1925, the first international effort was made to broaden the scope of radiological protection when the preliminary steps were taken for the formation of the International Commission on Radiological Protection.†

The first recommendations of the Commission were aimed principally at the medical profession and to some extent at the luminous dial painters and the industrial radiographers. The recommendations were largely empirical and based on the experience of those groups whose operational methods were considered desirable. The discovery of uranium fission brought radiological protection to the status of an important industrial health consideration. This was immediately recognized by the scientists who developed the first nuclear energy plants in the USA, and every effort was expended to ensure the safety of their workers. Similar efforts have now spread to every known atomic energy centre.

Fundamental research in radiobiology aimed at putting protection measures on a sound scientific basis is now well established in many countries, and at least three nations which have atomic energy establishments have agreed to pool their knowledge of radiological protection. Since specialists of these nations are also active in the ICRP, the recommendations of this body benefit from the findings of the conferences of the three nations, and also from valuable contributions from other countries which have long radiological experience but which are not yet working on atomic energy. The recommendations of the Commission have no legal standing, yet they are adopted by many nations because they represent the most comprehensive knowledge in the field at the present time.

The world is now facing a change in the problem of radiological protection, and this has been recognized for some time. With the widespread use of nuclear power, the hazards due to radiation are no longer confined to radiologists and atomic-energy workers. The problem has spread beyond the confines of industrial medicine to become an aspect of public health.

## THE SCOPE OF PUBLIC HEALTH PROBLEMS

Man has always been exposed to radiation from cosmic and other natural sources. Present developments in nuclear energy are such that, with the passage of time, the radiation background, in other words, the ambient radiation level, will be raised significantly by radiation sources of man's own making, if these are not controlled. From what is already known about the biological effects of radiation, an intensification of the radiation background is likely to lead to somatic and genetic effects in man. The former will occur in the population exposed while the latter will accumulate and affect future generations. It is the responsibility of public health to do as much as possible to prevent the radiation background from rising unnecessarily fast and to too high a value. This problem exists now, on a small scale, as a result of the operation of nuclear energy plants, and the protective aspect is now being dealt with at the industrial level. As the use of nuclear power for industrial purposes extends, this will no longer be possible. Although the broad aspects of the public health problem are well understood and are simply stated above, there is much to be learned about the operational procedures.

* Throughout this paper the term "radiation" is used in the sense of ionizing radiation.

† The establishment of the International Commission on Radiological Protection (ICRP) was envisaged at the First Congress of Radiology in 1925 and it was formally established in 1928 under the auspices of the International Congress of Radiology with which it has since been affiliated. It is a non-governmental organization composed of individuals chosen on the basis of their recognized activity in the fields of radiology, radiation protection, physics, biology, genetics, biochemistry and biophysics without regard to nationality. In addition to the main Commission there are five International Sub-Committees which deal with various aspects of radiation protection. The ICRP deals with the basic principles of radiation protection and leaves individuals the right and responsibility of introducing the detailed technical regulations, recommendations or codes of practice best suited to their needs. It meets usually every three years and issues recommendations on radiation protection which are revised from time to time. The last recommendations were prepared at the 1953 meeting and are now being published.

The experience of the past ten years has brought to light methods which might be used successfully to prevent the radiation background from rising, but the precise numerical data relating radiation background to effects on the world population are lacking. Such numerical data are all-important if public health is to function efficiently in its relations with the development of nuclear power. The correct numerical answer to this problem is not at hand and will not be forthcoming for some time. Therefore, the way is open to the two extreme courses of overcaution or undue leniency.

The public health problem at the moment is not an easy one. It is further complicated by the fact that public health depends on the degree of organization of a community, and since the types of communities affected by the development of nuclear energy will be diverse, it will be most essential that operating standards be clearly defined.

The present codes of practice for dealing with radioactive effluent from atomic energy plants are compiled on the supposition that an insignificant proportion of the world population is involved. When the exposed population becomes significant, these codes may have to be made more exacting. The limiting factor in compiling standards is, and is likely to remain for some time, knowledge of human genetics, a subject on which there are few relevant quantitative data. The only hope for a quick answer of a qualitative nature applicable to human genetics is the work now going on in the genetics laboratories of the world. The fate of an irradiated population of certain organisms could give information which could be applied qualitatively to the human race. But time is short, and public health must fulfill its obligation by intelligent control so that general exposure to radiation background will not soon reach levels from which there is no return.

With this problem before it, public health must profit from the experience of the industry during the last ten years; it must not be accused of hindering the development of nuclear power and thus depriving the world of its benefits. It must cooperate now with those responsible for the technical development of nuclear power. The industrial experience of the past has shown that the public health problems which have been foreseen and attended to ahead of time have proved less costly than those discovered too late, after a period of operation.

## THE BROAD PUBLIC HEALTH ASPECTS

Since public health activities must be integrated with the other disciplines involved in the development of nuclear energy, it must first have representation of the highest order capable of appreciating the problems as they arise in other scientific fields. Such personnel does not exist in the required numbers at present, and the first task of public health is therefore to embark on a comprehensive training programme. It is necessary, on the other hand, for the engineer and physicist to appreciate the public health problem. Since this is a rapidly expanding field, public health personnel will have to be kept informed of the latest developments. These developments will be taking place all over the globe and it will be a task in itself to sort out the important findings from the vast body of literature which will grow up. This is already a problem even at this early stage.

The task of public health will be lightened considerably by the provision of adequate protection in new installations, and for this reason it must collaborate at the early design stage. Present nuclear energy plants suffer much from the fact that not enough protection was provided for in their design, and they therefore need to be patrolled by large radiological-safety crews. Although safety crews will never be completely abolished from nuclear power installations, it is desirable for economic reasons that they be kept as small as possible by providing for maximal protection in the design of plants. Much experience in providing protection has already been accumulated, and this has an additional appeal on the basis of lower operating costs. It is assumed that as experience is gained, accidents in nuclear plants will become as rare as in other types of industrial plants. The troubles still encountered in present nuclear energy plants were generated when operations were started and knowledge was inadequate. This is a warning to remind us that a correct start be made, for difficulties arising from a false start in this field will make their presence felt for a long time. Since the development of nuclear power is now a world concern, it is also necessary that the public health programme be co-ordinated at the highest national level so that there can be full international co-operation.

Another important aspect of public health interest will be the siting of nuclear energy plants. In general, these will be of two kinds: reactors producing power and plants to process the spent fuel. The reactors may have to be close to large centres of population. In general these will not present much of a health problem unless there is an accident or the effluent systems are not properly designed. Reactors must therefore be built, at least at this stage, on the assumption that there is always a possibility of an accident, and thus the proper safeguards must be incorporated.

The more difficult installations will be the fuel reprocessing plants though these, with improved technology, could be quite safe. At the present stage we must expect the usual amounts of low-activity wastes which arise from one reason or another and which will have to be disposed of locally. The siting of such a plant will therefore be determined by transportation facilities and proximity to a suitable site for disposal into the ground.

Working practices in nuclear energy installations are now well established; the records for radiation safety in such plants are probably the best in the world. However, it will be necessary in the interest of public health to institute broader precautions. At

the moment the problem of protection of the communities is looked after by the staff of the nuclear energy plants themselves. When such plants become numerous these public health aspects will have to be the responsibility of public health workers. Codes of practice will have to be worked out with the plant personnel and must be of an eminently practical nature if they are to be effective.

## THE PARTICULAR PROBLEMS

Nuclear energy plants will impinge on public health by means of their waste products, and via the widespread distribution of radioactive isotopes. The practical public health problem will then be to assess and control the radioactive wastes and to ensure that the radioactive isotopes distributed are safely transported and disposed of. The wastes are of three kinds, gaseous, liquid and solid.

The air may be polluted from stacks which emit either radioactive particles or gases. Particulate waste can be filtered, and there is already much experience in this field. The filters now used are of the highest quality in order to trap the smallest particles. Their introduction into a ventilation system places additional requirements upon the air flow and pressure heads which are reflected in the cost of the installation. However, it is cheaper to design the air system properly at the beginning than to attempt to modify it later when radioactive fallout in the vicinity of the plant becomes intolerable. Some of the fission products are noble gases and can pass through any filter, but on the whole these are short-lived and can be stored, trapped and allowed to decay. This cannot be done with krypton-85, which has a ten-year half-life and which might become hazardous under adverse meteorological conditions in crowded industrial areas. The use of filters and gas traps changes the air pollution problem to one of the disposal of solid contaminated waste. In this regard public health services must be equipped to carry out air monitoring to be assured that conditions are safe in populated areas. The present safe standards for this form of disposal are contained in the recommendations of the ICRP (1953).

The use of waterways for the disposal of radioactive materials is obviously not an acceptable solution. It is almost impossible, even with the best control, to prevent small quantities of such waste from reaching water which may be used for drinking or which may contain edible organisms. The standards for such pollution are again contained in ICRP (1953) and public health authorities must be in a position to verify them. This problem is now being dealt with by industry. It differs from the usual pollution problem in that the actual amount of harmful waste is very small and its toxicity is measured in terms of its radioactivity. It must be remembered, however, that some biological systems have the power to concentrate certain radioactive elements so that a water monitoring programme must include the measuring of radioactivity present in water, edible organisms, as well as some of the lower forms of life and sediments.

At present much of these dilute wastes is dealt with by running them into the ground where the radioactive elements which are in small bulk are taken up and retained, mostly by the clay. The practice of getting rid of wastes in this manner is economically desirable but care must be exercised in its execution, especially as it may become widespread. The presence of large quantities of chemicals in the waste solution may destroy the soil's property of retaining the radioactive elements. Also the movements of these wastes are governed by the properties of the soil itself and the movements of the ground water. These two factors must be carefully studied in the area before such disposals are undertaken. A more conservative approach to ground disposal is the use of specially prepared beds, so that the active soil is retained under control and the effluent from the beds monitored and disposed of to the ground if sufficiently decontaminated. The disposal of wastes of low radioactive content in the ground is so important economically that it may well be a deciding factor in siting reactor fuel processing plants until more advanced chemical processes eliminate the problem entirely.

The burial of contaminated solid material raises much the same problems as the release into the ground of contaminated solutions. The site must be chosen, therefore, with much the same considerations in mind.

A further problem arises in this connection in that many of these solid wastes such as scrap steel have a commercial value and there is a tendency to return them for further industrial use. This will lead to a low level of radioactive contamination in supposedly new material. It is evident from this that public health measures must include radiation monitoring at a low level.

A much bigger public health problem is the disposal of highly radioactive wastes. To date there is no good solution for this problem, and many methods are being studied. In the meantime the wastes are stored in tanks awaiting permanent disposal. From the standpoint of public health these wastes will have to be confined so that they will not come in contact with humans in harmful concentrations. For countries which are crowded together and which lack space it may be desirable to have a common waste disposal site. The use of many scattered disposal sites for highly radioactive wastes whether on land or at sea might lead to the spread of radioactive material, for there is a high probability this material will escape if the sites and methods used are not properly chosen. A more satisfactory alternative would be to have a few recognized sites throughout the world which would be considered safe for the deposit of high-activity waste, and that such sites would be controlled on an international basis.

## A SUGGESTED PUBLIC HEALTH PROGRAMME

For public health to play its part in the development of nuclear power it must be sure that its demands are consistent with sound principles of radiological protection. Already in many quarters there is the suspicion that protection against radiation is being overdone. To allay these suspicions a public health programme must be designed to seek the knowledge, which is now lacking, on the quantitative effects of chronic low-level irradiation on humans.

It seems now possible to suggest a programme of work which might be logically undertaken by public health along with the development of nuclear power. This programme is not intended to be all-inclusive and would certainly be subject to modification in the light of review by technically competent bodies such as the ICRP. The intention is to create a starting point for discussion in the hope of stimulating action in the right direction. The following items among others might be usefully included in such a programme.

### The Training of Public Health Personnel

This is one of the immediate important requirements. The field of nuclear technology is expanding rapidly. It is necessary that public health authorities join in this expansion as soon as possible. The number of public health personnel trained in this specialized field is at present inadequate, especially in countries which as yet have no nuclear energy programmes. Public health personnel must be trained to such a level as to command the respect of their colleagues in physics, chemistry and engineering if their advice is to be heeded. The detailed requirements of such a training programme and the method in which it will be effected are dealt with in another paper prepared by WHO.

### The Dissemination of Pertinent Public Health Information

There is at present a vast body of knowledge concerning the radiological health problems associated with nuclear energy. Some consideration has also been given to the general problems of public health. For various reasons this knowledge is not available in a cordinated and condensed form. This undesirable state of affairs is one which can be effectively corrected if the present information were co-ordinated and made available to those requiring it. The effort necessary to do this would be considerable and a way to attack the problem would be for an international organization such as WHO to select important public health subjects and to have them discussed amongst groups of experts working in the respective fields. At the same time due attention should be paid to the necessity of dealing with new information as it becomes available. Such an information service will be invaluable to public health administrations, especially in countries which have had no previous connection with radiation problems.

### A World-Wide Scientific Study of the Somatic Effects of Low-Level Radiation on Humans in Relation to the Intensification of the Radiation Background

Such a study must undoubtedly be supported by work on animals which would on the whole yield qualitative data. This subject will be dealt with in other papers presented at this Conference. Quantitative observations on the effect of radiation on humans have been sadly lacking. For this reason there is a great effort taking place to try to extrapolate the results of animal experiments to get quantitative information which would be of use to public health. A first step in this programme would be to reconsider the present system of collection of vital statistics and to add to it or modify it in the light of the health problems of the nuclear age. The standardization of statistical information to include radiation effects could well be aided by advice from the ICRP.

The incidence of the radiation-associated diseases would have to be studied in relation to the intensification of the radiation background. For this purpose specific radioactive isotopes which have been shown to be damaging to animals, such as strontium-90, might be selected for initial scrutiny. The assays at low radiation levels which enter into this work require special technical skills and the use of advanced measuring equipment. This technical knowledge must be acquired by health personnel. A difficult technical study of this kind would be much more effective on a world-wide basis if those undertaking it have at their disposal uniform methods and common standards so that all results could be ultimately compared. It must be appreciated that such a research would be a long-term one, but there seems to be no other way of obtaining the required information.

A particular aspect of this work would be the study of human beings irradiated as a consequence of occupational or other conditions. These are to be found among the following: (1) patients submitted to radiation therapy; (2) workers in medical radiology; (3) workers in the nuclear energy industry; and (4) workers in the uranium mining industry.

International standardization and inter-comparison of experiences would again be of great value in this field. It must be realized that although radiation is easily measured it is very difficult to get reasonable agreement when these measurements are made in diverse ways and places.

## A STUDY OF THE RADIATION-INDUCED GENETIC DEFECTS IN THE HUMAN RACE

This is an extremely difficult problem and a clear method of attack is not evident. The genetic condition of the human race is in a dynamic stage and it would be almost impossible to distinguish small shifts due to radiation, yet it is this factor that is presently influencing the limit which is considered acceptable as the world-wide radiation background. An experiment on mammals such as rats would perhaps give

qualitative information on the fate of the human population when exposed to additional low-level radiation. These experiments are of value in general because they will tell whether an irradiated population tends to get stronger, retain the status quo or deteriorate. This is the public health value of such work.

Some information on humans could be obtained from a genetic study of selected groups such as the offspring of persons exposed to radiation by their work and of communities living at high altitudes.

In any case genetic information on humans will take a long time to collect and a comprehensive programme on genetics should be undertaken as soon as possible. Here again uniformly recorded statistical information would be essential.

Only if a comprehensive public health programme of the type outlined above is initiated will world-wide radiation protection be efficient. Although work on radiations has proceeded for fifty years, knowledge of biological effects is still deficient. And yet, this generation has an obligation to posterity to hand down the legacy of nuclear technology not only with all its useful potentialities but also with corresponding knowledge of how to deal with its inherent hazards.

# The Achievement of Radiation Protection by Legislative and Other Means

**By Lauriston S. Taylor,* USA**

## I. VOLUNTARY PROTECTION MEASURES

The potential hazards of ionizing radiation were recognized within a few years of its discovery. Minor precautions in its use took place beginning at that time, but it was not until about 1920 that the seriousness of radiation hazards was first generally recognized. The increased hazard was brought about in the main by the advent of the hot-cathode Coolidge tube that produced quantities of radiation never before known. In one sense the situation was comparable to that of the last 10 years when again we have been confronted with radiation problems, that while not essentially new in nature, were of a considerably greater magnitude because of the much wider variety and size of sources. This, of course, was brought about by the use of artificially-produced radioactive isotopes in medicine, industry and research. As in the 1920's, we are again facing an increase in the problem of protection and because the magnitudes of today are so much greater, we find ourselves cautiously looking at more stringent means of controlling radiation.

From the outset, various means and methods have been employed by countries, states, cities and individuals to control radiation. In this report, a number of such examples will be given, but it should not be implied that these represent the only cases wherein radiation protection efforts have been carried out. In addition to those cases of which the author is aware, there are undoubtedly many other cases of which he is unaware.

For many years, the efforts toward radiation control and radiation safety were mainly on a voluntary basis. In some cases these efforts were materially assisted by some central laboratory or other organization. For example, in 1925 Sievert in Sweden offered the services of his Institute for the provision of radiation safety inspection and evaluation. The use of these services was up to the individual but they were available upon request. At the outset of this program, there were only a few dozen radiation installations in Sweden. Of course, as in the rest of the world, the number increased rapidly until in the 30's it began to appear that some more formalized inspection procedures would be desirable. Accordingly, in 1941 a State act was passed which required the licensing and inspection of all radiation installations. The responsibility for this was delegated to the Radiophysics Institute in Stockholm. Both plans of radiation control in Sweden have worked well and they have an enviable reputation for radiation safety in that country.

Similarly in England, a voluntary radiation inspection service was started in the early 20's. In this case, the services were performed, upon request, by the National Physical Laboratory in Teddington. At the present time no compulsory radiation legislation exists in England although it is understood that plans for legislation are now being worked upon and may go into effect in the near future.

In New Zealand, radiation protection regulations were established in 1951 under their Radioactive Substances Act of 1949. Their regulations call for licensing and inspection of all radiation sources and the program apparently seems to be working out satisfactorily. The field and laboratory services are performed by the Dominion X-ray and Radium Laboratory of the Department of Health in Christchurch. It is not known whether or not they had similar services available on a voluntary basis prior to 1951.

Germany has for many years provided voluntary radiation inspection services through its Physikalisch-Technische Bundesanstalt. Likewise, other countries, such as France, Denmark, Italy, and Canada, have had voluntary radiation-control programs for many years.

The control of radiation in the United States has been on a less centralized basis. This is occasioned in the main by the very large distances involved over the country as a whole, and the very large number of radiation sources which would be subject to control. The problem is further complicated by the fact that any control measures would normally fall to each of the 48 States. The National Bureau of Standards, from which stems much of our radiation protection information, does not regard the provision of radiation inspection and control functions as falling within its purview. While the U. S. radiation safety record is possibly not as good as in those countries where control services were available (there is no good basis for comparison), it has nevertheless

* National Bureau of Standards.

not been a serious problem in comparison with other industrial hazards and accidents. Within recent years, there appears to be a growing desire to set up radiation legislation and regulations in the individual States. While at the moment there are only one or two states having laws dealing with radiation protection, at least a dozen states are actively working on the problem, and it is anticipated that within a few years a substantial number of the more highly industrialized States, at least, will have some degree of radiation regulation.

## II. GUIDANCE PROVIDED BY NATIONAL AND INTERNATIONAL ORGANIZATIONS

Guidance in the matter of radiation control in different countries has been provided, to a major extent, by the recommendations of the International Commission on Radiological Protection. This Commission was established in 1928 under the sponsorship of the International Congresses of Radiology. It has remained in active operation ever since and has provided a substantial degree of world leadership in the field. Prior to 1928, several countries, notably Sweden, England, and Germany, had some limited recommendations or codes of practice relating to radiation safety. These were developed by committees of their national radiological organizations and formed the basis for inspection under their voluntary protection programs.

The first international recommendations on radiation protection were based, to a large extent, upon the British recommendations that were in existence in 1928. Their recommendations were relatively simple and dealt with the basic aspects of protection without going into a great deal of detail. They have served admirably as a basis for many of our later developments.

In the United States, the Advisory Committee on X-ray and Radium Protection was established in 1929, under the sponsorship of the National Bureau of Standards. This was made up initially of representatives of the several medical and radiological organizations and the National Equipment Manufacturers Association. In 1946, because of the tremendous growth in radiation usage during the preceding few years, this committee was substantially enlarged in membership and the scope of the activities were, at the same time, greatly broadened. Its name was also changed to the National Committee on Radiation Protection. Recommendations of this committee have been published as handbooks of the National Bureau of Standards. Prior to the war, the committee produced handbooks containing codes of practice for protection against X-rays, gamma rays from radium, radioactive luminous compounds, etc. Since 1946, the earlier handbooks have been revised and, in addition, various other fields of activity have been covered. These have included the disposal of radioactive waste, safe handling of radioactive isotopes, maximum permissible exposures to internal and external radiation, monitoring methods, protection against radiation from radium, betatrons, and so on. In all, the NCRP has produced since 1946, 13 such handbooks and are presently working on 5 more.

In addition to the work of the National Committee on Radiation Protection, whose main purpose is to establish the basic principles and philosophy of radiation protection, the American Standards Association has developed an extensive code for industrial protection in the use of X-rays and radium. At the present time, the ASA is engaged in a substantial extension of its industrial protection codes into many other areas.

As in the United States, England, Germany and some of the other countries have also substantially extended the scope of their radiation protection recommendations, so that at the present time a number of countries are in the position of having fairly comprehensive radiation protection rules available for use.

In the formulation of recommendations by the International Commission on Radiological Protection, an essential guiding principle for the first twenty years was that its recommendations be kept as simple and concise as possible. An endeavor was made to set forth the fundamentals and the factors upon which international agreement could readily be obtained. Some deviation from this principle took place at the 1953 meetings of the Commission. At that time, very much more elaborate recommendations were prepared. In fact, the three chapters dealing with X-ray protection, the permissible amounts of radioactive isotopes in body, air, and water, and the permissible exposures to radiation from external sources were, to a considerable extent, based upon the corresponding handbooks prepared by the National Committee on Radiation Protection.

In the author's opinion this practice may present some serious difficulties. After all, the recommendations on X-ray protection that served as a model for the 1953 ICRP recommendations were developed around the needs and practices as they exist in the United States. While there are not vast differences between different countries, there are nevertheless sufficient differences to make international agreement on such detailed recommendations a somewhat difficult procedure. The matter of obtaining acceptance of all of this detail turned out to be a very formidable task and it is believed that much of this difficulty could be removed in the future by developing the whole chapter on a less detailed basis.

## III. SPECIFICITY VS OBJECTIVE REGULATIONS

In the development of radiation protection recommendations or regulations, there are two classes of regulations that may be developed. These would be termed "specificity" or "objective" regulations. In the specificity regulations, all details are spelled out specifically and in detail so that one is told not only what to do but virtually how to do it. This has cer-

tain obvious advantages, particularly for groups or organizations that do not have the necessary background or do not care to think through the detailed problems for themselves. On the other hand, a specificity regulation lacks flexibility and probably requires more frequent changes than the objective regulation. With an art that is developing as rapidly as radiation protection, our knowledge of protection needs is increasing steadily and we find ourselves constantly confronted with the task of revising a specificity regulation in order to keep abreast of modern technology.

Compared with this, the objective regulation spells out the main objectives to be accomplished by any of several available means. Such regulations would include information on permissible exposure, degrees of responsibility, certain factors relating to the disposal of radioactive materials, and so on. However, such a regulation would not include any detail showing how these objectives may be obtained. For information leading to the achievement of the goals, the user is referred to various sources of detailed information, such as the detailed recommendations in the handbooks of the National Committee on Radiation Protection or the American Standards Association. Since the basic principles of radiation protection are not expected to change very rapidly, the use of an objective regulation allows greater flexibility and encourages greater ingenuity in achieving the desired goals. It also necessitates less frequent changes as our knowledge and skill in the radiation field increases. Most of the handbooks of the NCRP are of the objective type. Notable exceptions to this are Handbook 41, Medical X-ray Protection up to Two Million Volts (now being revised), and Handbook 54, Protection Against Radiations from Radium, Cobalt-60, and Cesium-137. This latter is an outgrowth of the original handbook on radium protection.

## IV. THE TREND TOWARD LEGISLATIVE REGULATIONS

Because of the greatly increased and more widespread use of all kinds and varieties of radiation sources within the past few years, there is growing concern in all countries over the control of radiation hazards. At the same time, there appears to be a growing belief that the problem can no longer be met on a purely voluntary basis, and there is an expanding world pattern of thought leading to greater and greater control of radiation through legislative means. This trend has also been observable in the United States and much of the remainder of this report will relate to its experience in this field over the last few years.

With the increasing likelihood that radioactive wastes may no longer be readily confined to the point of use and may even escape from one country to another, it is highly likely that there will be increased international concern over the control of this form of radiation hazard. The problem may well cut across many countries where practices and procedures differ substantially, and it will be increasingly important to be able to arrive at the fundamental aspects and fundamental requirements of radiation protection in a manner that will be acceptable to all countries. In this regard, some of the experience in the United States may possibly be helpful.

Since there is individual state control in the US and this, then, involves 48 different states, it might be said that we have a localized proving ground which in some limited measure would compare with the international relationships between many countries. At the same time, the US has certain federal controls relating to radiation protection and these may be likened to international control exercised over a number of countries. It is not claimed that the practices and experiences in the United States are necessarily the best, but it is believed that they should be useful as a study model for international consideration.

## V. VOLUNTARY COMPLIANCE

As already noted above, the US has had for some 25 years, various codes relating to radiation protection. The use of these has been strictly on a voluntary compliance basis and as such they have served an extremely useful purpose. Accompanying these a positive effort has been made in the direction of education in radiation protection. Improvements in our practices have been noticeable as a result of these educational efforts and it is my own belief that education alone would provide a satisfactory control of radiation except under rather unusual circumstances. It is, of course, the unusual circumstances that brings about the pressure for more direct legislative regulation of radiation hazards.

With the exception of a few municipalities, there were no legal radiation regulations in this country until about 1950. On the other hand, any radiation damage case that has reached the courts has for many years been settled on the basis of the recommendations contained in the NCRP handbooks. Thus, while in themselves having no legal status whatever, they have served as a solid foundation for many legal decisions on radiation protection in the US. In some cases the recommendations of the NCRP have been adopted officially by some of our radiological organizations and, through these organizations, have been passed on to their members.

Generally speaking, no compulsion has been involved in bringing about their use but there are cases where hospitals, for examples, have been required to comply with the NCRP recommendations in order that they be certified by organizational authorities. In the industrial field, pressure exerted through the welfare organizations of labor unions has been instrumental in bringing about the adoption of the safety recommendations of either the American Standards Association or the NCRP. In these situations the unions have demanded compliance with the recommendations as a part of the specified working conditions of the employees.

## VI. FEDERAL REGULATIONS

Action by the federal government in the field of radiation regulation has been very limited until quite recently. The main federal requirement of any long standing has been one promulgated by the Food and Drug Administration. This organization while not in a position to promulgate rules and regulations relating to the achievement of radiation protection, called attention to the necessity of protection by requiring certain labelling on all X-ray equipment. This labelling points out that the equipment was dangerous and that its use should be limited to qualified individuals.

The Atomic Energy Act of 1946 made some indirect references to the needs for the establishment of radiation regulations for radioactive isotopes, but the interpretation of this act was never such that the Atomic Energy Commission could decide that the establishment of radiation regulations was clearly within its prerogatives. This situation seems to have been rectified in the Atomic Energy Act of 1954, as a result of which, the Atomic Energy Commission is now preparing fairly detailed regulations relating to matters of protection against radiations from Commission-produced isotopes. It is not within their power to exert similar control over X-rays or the radiations produced by natural radioactive sources such as radium. It should be pointed out that the proposed AEC regulations will, in the main, be of the objective type and that reference will have to be made to other standard sources of information to accomplish the desired objectives.

## VII. STATE REGULATIONS

Brief mention will be made of some current State actions in the field of radiation protection and mention will also be made of regulations that either exist or are being proposed for early adoption. As of this date (1 May 1955) there is only one state that has a fairly comprehensive set of radiation control regulations in force. These regulations have been promulgated by the Department of Labor of the State of California and cover all sources of radiation including artificial and natural radioactivity, X-rays, etc. The regulations are of a mixed type, being in the main of an objective type but including some specific details. In this state, there is no established procedure for the registration or licensing of radiation sources. They have a limited inspection service available and are authorized to inspect according to their wishes and capacity to do so.

In New York State, the Department of Health has promulgated a set of regulations which are expected to go into effect this year. These are based fairly closely upon a pattern developed by the NCRP and are strictly of the objective type. They call for the registration of all sources of radiation exceeding certain specified levels. It might be pointed out that for radioactive isotopes these levels vary from one isotope to another. In addition, the New York State Department of Labor is expecting to promulgate a second set of regulations. These will be quite different in format from those promulgated by the Health Department but thus far, they appear to be in substantially good agreement insofar as the basic principles are concerned. They also require registration with the Department of Labor. In these regulations there has been established a rather elaborate pattern of classification of radiation sources according to the potential hazard produced by each. This classification of sources provides certain administrative simplifications, in that the regulatory body can devise a given set of inspection procedures for each source classification. On the other hand, this will undoubtedly lead to increased complication in registration and there will undoubtedly be various classifications of sources within the bounds of a given institution.

Superposed on both of these regulations will be the federal regulations relating to the licensing of the uses of artificial radioactive isotopes. Here again there is fairly good agreement as to the basic principles between the federal and the state regulations. The good agreement between these several sets of regulations has to a considerable extent been brought about through the efforts of the NCRP which has served in an advisory capacity to both the state and federal governments in the matter of basic principles.

It might be mentioned, in passing, that the City of New York also has some limited regulations relating to certain types of X-ray apparatus. Thus in New York City a radiation user may find that he has to comply with four different sets of regulations, each differing in detail even though the basic principles are somewhat closely related.

Other states, while working actively on the problem, have not yet reached the definite regulation stage. The State of Pennsylvania, Department of Labor, has been working for some time on a general regulation to cover all sources of radiation. This will call for registration of sources. The Pennsylvania code is also of the mixed type containing regulations of both a specific and objective type. Other states, such as West Virginia, New Jersey, and Michigan, are also giving consideration to the problems.

Probably the simplest radiation legislation is that proposed for the State of Massachusetts. This Act, which consists of only one paragraph, authorizes the Department of Health to carry out such protective measures as it deems necessary and which are in accordance with the recommendations of the National Committee on Radiation Protection. In many respects, this may appear to be the most ideal and most flexible solution to the problem. It is pointed out by some, however, that such legislation will probably prove, on the basis of experience in other fields, to be inadequate for the problem. One reason for this is that such legislation does not make it readily possible to set up the necessary organization and control procedures. With this left uncertain, it is feared that

proper regulatory systems will not be readily established. This is something yet to be proven, at least insofar as the radiation protection field is concerned. On the other hand, it appears to many that this type of code would be preferable in its brevity, to one that was so replete with detail as to hamper its feasibility of adequate enforcement.

It will be noted, in reference to some of the state codes above, that registration has been called for. In no cases have any of these states established a requirement for licensing. It is believed and hoped that registration alone will serve to provide the control agency with the necessary information as to existing and proposed sources and that licensing, together with all of the encumbrances that will accompany it, will not be necessary. Generally speaking, registration will consist in simply notifying the control agency of the existence of a radiation source together with some relatively simple specified information describing its general nature. Use of the source can proceed with or without inspection. This differs materially from licensure where it is presumed that before a license would be granted, all of the details regarding the use would have to be studied and improved. Of course the Atomic Energy Commission will require its own licensing, regardless of state actions.

Admittedly, licensing and advance approval may stop, at the origin, some abuses of radiation usage. On the other hand, it is obvious that such strict control will also materially hamper the development of some radiation usages and in many cases seriously delay putting a radiation source into operation. This is to be weighed against the possibility of some radiation harm resulting from improper use. It is one of the risks which accompanies radiation use, but on the basis of experience in the past, it is believed that the risk is not large and should be acceptable in any case.

## VIII. NCRP RECOMMENDATIONS ON RADIATION REGULATION

To provide some degree of guidance for the states engaged in developing radiation protection regulations, the National Committee on Radiation Protection established a subcommittee for the purposes of considering the problem. In a sense, this work was undertaken in spite of the rather strong feeling that the Committee had held for many years that regulations were not desirable. However, it was also recognized that, regardless of how the Committee felt, states were obviously going ahead with developments on their own. As long as this appeared to be the situation, it was agreed that its many years of experience should be applied to the problem and that the Committee should be of whatever assistance possible to those states. It is the attitude of the NCRP today that it does not care to recommend or oppose adoption of radiation control regulations by states.

Regulations should be in a form as flexible as possible. In other words, a simple enabling act authorizing the establishment of radiation control measures would on its face appear to be adequate. The control authorities could then refer to NCRP or ASA handbooks according to their needs. However, it has been found that most states tend to avoid any situation which appears to "adopt by reference" any codes or recommendations prepared by other bodies. In fact this is unconstitutional in many states. It is true that this procedure has been followed successfully in such cases as the National Electrical Safety Code. However, the resistance against this procedure appears to be so strong that efforts to overcome it do not seem to be profitable. To the layman, it would appear that the states have become used to dealing with what appears to be cumbersome methods of control in other areas, and that they would rather extend these same methods than to try to work out something new which on its face appears simpler. Admittedly, the problems are not simple once one studies them closely. However, it is believed that adoption by reference in the radiation field could save the states a great deal of difficulty and at the same time provide a high degree of uniformity between the control measures or regulations in the many states.

There appear to be two main obstacles to the principle of adoption by reference. An understandable objection is that by simply indicating that the recommendations of a particular organization would be adopted by the state, commits the state to future actions of this particular organization. While the state may be completely content with past actions, they have no assurance that future actions would be equally acceptable. It is understandable that they might balk at such a blanket adoption by reference, but it is done often. A second problem lies in the fact that outside groups have prepared very few codes that are general enough in nature to warrant adoption *in toto* by more than a few of the states. Generally speaking most recommendations or codes contain too much detail, since they are designed primarily to provide broad principles of guidance. Furthermore, unless a set of recommendations is adopted *in toto*, the state may be faced with the necessity of making exceptions, and this can be even more cumbersome than selecting from the recommendations portions which they wish to use.

The National Committee on Radiation Protection has recently completed a pattern for a radiation control Act and a sample set of regulations for use under that or other Acts. Accompanying this is a detailed discussion of the problems that may be involved in the adoption of radiation protection legislation or regulations. While some several hundred copies of these recommendations have been circulated about the country, they have not yet been published. It is expected that they will be at an early date.

In preparing the regulations, one of the basic guiding principles has been to keep them as simple

as possible, free of details, and at the same time to set forth the main fundamental objectives that it is desired to accomplish. Where, of necessity, some detail and numerical quantities have had to be included, these have been limited to the factors which it is believed should be kept uniform over all the states. If this degree of uniformity is maintained over the states, they may add such detail as they wish and still not involve the country as a whole in conflicting basic requirements for radiation control.

In dealing with the possible use of the regulations, the NCRP strongly recommends that the states adopt only those portions of the regulations that apply to the problems that may exist within the state. Furthermore the state should not adopt any more of the regulations than can be adequately enforced at the time of adoption. As experience in radiation regulation grows, the degree of coverage by the regulations can be broadened and extended. It is felt strongly that regulations that are unenforceable may do more harm than no regulations at all.

## IX. ESSENTIAL FEATURES OF RADIATION REGULATIONS

Given below is a very brief outline of the general areas that are covered by the proposed uniform regulations.

At the start, the scope and application of the regulations is set forth so that the public will know the nature of the problem with which the state is attempting to deal.

This is followed by a section giving certain definitions. Only those definitions are included where an exact understanding of the terms is essential to an understanding of the regulations themselves. Technical definitions of scientific terms in common use are not included since they encumber the regulations.

Certain exemptions from the regulations are essential. A regulation covering all forms of radiation from large sources down to a radioactive wristwatch would of course be impossible of enforcement. Therefore, it is necessary to set certain levels of radiation below which control is not required. Not subject to control would be quantities of radioactive material so small that if the entire amount were taken internally, continuously or at one time by a person, no serious harm would be likely to result to that person. Appropriate levels in line with this principle have been assigned to all of the radioactive materials that are normally used in this country.

The standards against which radiation control will be exercised are based on the permissible exposure levels that have been published by the NCRP and the ICRP. For the sake of convenience, these exposure levels and quantities of radioactive materials are listed in the regulations.

To provide the control agency with a knowledge of where radiation sources exist, registration is called for. The process of registration is simple and may consist either of writing a letter to the control agency or completing a form supplied by the agency.

One section deals with the problem of personnel monitoring and area radiation surveys. This does not tell how to perform such operations but indicates the type of measurement that should be carried out to insure safety of workers. In this section, it is indicated that surveys may be omitted or discontinued where past experience or measurements have indicated that the operating conditions are likely always to remain safe.

Certain exposure records and reports are required. These will be generally maintained by the radiation user, but may be inspected by the control agency if it so wishes. The only time that any personnel exposures need to be reported directly to the control agency is when they have exceeded by a factor of 5 the permissible amounts specified in the regulations.

Another section deals with the delegation of responsibility for safety in a radiation installation. By the provisions in this section, it is hoped that the state can effectively shift the direct responsibility for safety to the user, by describing what the user must do and for what he must be responsible. By this means it is believed that much less rigorous examination of installations by state authorities will be required. Of course from time to time they will want to spot-check operations, but if the responsibility provisions are followed, there should be reasonable assurance of continued safe working conditions.

A few essential details regarding the storage of radioactive materials are described. Generally speaking, these rules specify storage under such conditions that any escaping radiation will be below the specified permissible levels.

Similarly in the requirements with regard to radioactive contamination control, the objective is to keep individual exposures below the permissible amounts. For this particular problem, many different practices and procedures are available to the individual laboratories and it is felt unwise to try to describe these factors in detail. A section on labelling provides some degree of uniformity in the marking of radiation work areas, sources and containers of radioactive materials.

The problem of the disposal of radioactive wastes is a much more difficult one and there are many gaps in our knowledge of how this may be accomplished. Here again, the regulations require essentially that no radioactive materials be disposed of in such a way that they can concentrate at levels and in places that may be harmful to human beings. Any radioactive waste disposals must be done with the approval of the state authorities, and adequate records of the nature and place of disposal must be maintained. Where there is any likelihood of radioactive wastes, either in the air or water, exposing large population groups to radiation, the exposure levels for such groups are set at 1/10th of those recommended for

occupational exposure. This is to provide some safety factor, since control of radiations under some circumstances will be very difficult. It also tends to lower the average population exposure and hence minimize possible future genetic effects. Individual, non-occupational exposures may be averaged over one year.

The last section of the regulations gives the technical standards, guides, and general information to be used in achieving the requirements of the regulations. At the outset, it is indicated that the numerical quantities included in this section should be subject to change as more current and up-to-date information becomes available on the subject. The agency should be empowered to make changes in the technical standards whenever the situation so warrants.

The technical data in this section includes the following items:

1. The minimum quantities of each individual isotope that is subject to regulation or control.

2. Maximum permissible dose from external sources of radiation. This consists of the seven rules taken from Handbook 59, and are similar to the external exposure limits used by the ICRP.

3. Maximum permissible neutron fluxes. These are taken from Handbook 55 and the recommendations of the ICRP.

4. A table of the relative biological efficiency of different types of radiation including neutrons and alpha particles. This is taken from the recommendations of the ICRP and NCRP.

5. Maximum permissible amount of radioisotopes in the total body and maximum permissible concentrations of radioactive isotopes in air and water. This material is taken from Handbook 52 of the NCRP and is in general agreement with the recommendations of the ICRP.

It will be noted that all of the technical contents of this section are selected from existing and available handbooks or other sources of information. For guidance, and an indication of the ways and means by which protection can be achieved, the user is referred to these sources for detailed information. This material is not included as a part of the regulations since it can be expected that improvements in protection techniques will be made from time to time.

For further information regarding the work of the NCRP and for information regarding the development of regulations for the control of radiation, the reader is referred to the bibliography at the end.

The following is a listing of the handbooks on radiation protection that have been prepared by the NCRP or are presently in the course of preparation.

Handbook 27 Safe Handling of Radioactive Luminous Compounds

Handbook 41 (Under Revision) Medical X-ray Protection Up to Two Million Volts

Handbook 42 (Under Revision) Safe Handling of Radioactive Isotopes

Handbook 48 Control and Removal of Radioactive Contamination in Laboratories

Handbook 49 Recommendations for Waste Disposal of Phosphorus-32 and Iodine-131 for Medical Users

Handbook 51 Radiological Monitoring Methods and Instruments

Handbook 52 Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water

Handbook 54 Recommendations for the Disposal of Carbon-14 Wastes

Handbook 55 Protection Against Radiations from Radium, Cobalt-60, and Cesium-137

Handbook 55 Protection Against Betatron-Synchrotron Radiations up to 100 Million Electron Volts

Handbook 56 Safe Handling of Cadavers Containing Radioactive Isotopes

Handbook 58 Radioactive-Waste Disposal in the Ocean

Handbook 59 Permissible Dose from External Sources of Ionizing Radiation

In Preparation:

Protection Against X-rays up to 2 Million Volts (Revision of Handbook 41)

Disposal of Radioactive Waste by Burial in Soil

Protection Against Neutrons

Protection Against High Energy Electrons from Accelerators

Regulation of Radiation Exposure by Legislative Means

Disposal of Radioactive Wastes by Incineration

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3. Tabershaw, Irving R. and Harris, Saul J., *Administrative Problems in Radiation Protection*, Nucleonics *Vol. 12*, pgs. 8–13.

4. Hutton, G. L., *Public Control of Radiation Emitters*, Public Health Reports, December 1954.

# An Account of Atomic Energy Developments in Australia

The commencement of the production of uranium from Australian ores, and the construction of a large atomic energy research establishment, by the Australian Atomic Energy Commission, have been outward signs of the great interest which is growing in the Commonwealth in the peaceful application of nuclear energy.

In fact the interest goes back a long way, and it is the purpose of this paper to record the steps which have been taken, by the Commonwealth and State Governments, by Universities, by Industries and by Scientists and Engineers, to bring the development to its present state.

Credit for the first Australian interest in this matter must go to Mr. Oliphant, who, during the war, instigated a search for uranium in South Australia, where minerals containing this metal were known to occur. No actual development took place at that time, but after the war the Government of South Australia, through its Department of Mines, continued the search which led ultimately to the development of the Radium Hill uranium mine.

In 1949 the Commonwealth Government established the "Industrial Atomic Energy Policy Committee" to advise the Government on possible industrial applications of this new source of energy. The Chairman of the Committee was Mr. Oliphant, and the members were Mr. White as Deputy Chairman, Mr. Martin, Mr. Baxter, Mr. Raggatt, Mr. Goodes and Mr. Breen.

This committee collected much valuable information and defined the problems which would have to be faced in any development of atomic energy in Australia. The committee ultimately came to the conclusion that its terms of reference were too narrow to enable it to discharge its responsibilities in the way it felt it should, and it recommended its own dissolution and the creation of an alternative form of organisation.

In April, 1952, the Government established an Atomic Energy Policy Committee under the Minister for Supply. It consisted of Mr. Stevens, Chairman, representing the Department of Supply, Mr. Martin representing the Department of Defence, Mr. Wheeler or Mr. Hibberd as alternates representing the Secretary to the Treasury, Mr. Cochrane representing the Department of Defence Production, Mr. Raggatt representing the Department of National Development and Mr. White representing the Commonwealth Scientific and Industrial Research Organisation.

This Committee made important recommendations regarding the development of uranium resources in Australia, and after seeking advice from Sir John Cockcroft, who visited Australia at that time, recommended to the Minister for Supply, Mr. Beale, the establishment of an Australian Atomic Energy Commission with statutory powers.

The Commonwealth Government accepted this recommendation and the Commission was duly established, its members being selected in November, 1952, and appointed by the Governor-General, on 17th April, 1953.

The composition of the Commission was: Mr. Stevens, Chairman and whole time member; Mr. Baxter, Deputy Chairman and part time member; and Mr. Murray, part time member.

The Commission is directly responsible to the Minister for Supply. The Atomic Energy Act, 1953, gives the Commission the statutory powers under which it operates.

Broadly, the Commission has two main fields of activity, that related to the discovery and production of uranium, and that related to the development of the uses of atomic energy for industrial and other purposes.

The powers of the Commission operate only within the limits of the constitutional authority of the Commonwealth Government.

Except as may be necessary to the defence of the Commonwealth, the States are free to develop their own uranium resources. The Commission's direct responsibilities here lie within the Territories of the Commonwealth.

In relation to the use of atomic energy, the Commission is given authority in comprehensive terms to develop the practical uses of atomic energy, to construct and operate plant for this purpose and to undertake, organise and generally foster scientific research with a view to the advancement of technology in this field.

In surveying what the Commission has done since its appointment, it will be convenient to divide the activities relating to the procurement of uranium, from those relating to its utilisation, and to consider them separately.

By 1953, at the inception of the Commission's work, considerable progress had been made in Australia towards the discovery and development of uranium supplies. In 1949, a local prospector had discovered indications of uranium ore at Rum Jungle in the Northern Territory, and the Bureau of Mineral

Resources had carried out extensive investigations in this difficult country and by 1952 had proved the presence of an ore body of commercial importance. These investigations also revealed several other promising deposits in the same area.

During 1952, prior to the appointment of the Commission, agreements had been made with Combined Development Agency of the United States and Great Britain, in relation to the production of uranium at Rum Jungle. An agreement had also been made with Consolidated Zinc Pty. Ltd. who subsequently formed a subsidiary company, Territory Enterprises Pty. Ltd., to construct and operate a mine and ore treatment plant at this site.

These agreements provided a means for the development of these new resources in the far north, and for a large capital investment in the work. Special clauses protected Australian interests and ensured uranium supplies for internal use as required.

The Commission became responsible for these arrangements and for their development and extension. The plant was finished in record time and was set in motion by the Prime Minister, Mr. Menzies, on 17th September, 1954.

In parallel with this work, the Bureau of Mineral Resources, directed by Mr. Nye, had, as agents for the Commission, embarked upon a most extensive programme of airborne scintillometer surveys designed to locate radioactive deposits in other parts of the Northern Territory. These surveys have since been extended into some of the States by arrangements with State Mines Departments. The results of surveys have been published as a guide to private prospectors and mining companies, many of whom are now engaged in exploring and developing discoveries made in this way.

The policy followed by the Commission has been to encourage private enterprise to play a major part in the production of uranium, and to place as few restrictions as possible in their path. This policy has been backed by a declared ore buying programme with fixed prices to enable companies to plan ahead with security.

While not directly the concern of the Atomic Energy Commission, the activities of the Government of South Australia must be mentioned here, or else a very incomplete picture of uranium development in Australia would be presented. The South Australian Department of Mines has pursued an energetic and successful search for uranium minerals in that State, and has also done much work on methods of extraction and purification. The Radium Hill mine and concentrator was opened in 1954 and a modern treatment plant at Port Pirie is expected to be complete in 1955. This will be the second uranium producer in Australia.

It is difficult at the moment to form a considered judgment on the uranium position in Australia. Two producers, Rum Jungle and Radium Hill, are established. A great many other prospects have been discovered and some are being explored to establish the quantity and quality of ore they contain. Some will almost certainly prove to be workable on a commercial basis, but how many will do so, and what will be their capacity is not at present known. It seems likely that Australia will have adequate uranium for her internal requirements, but whether she will have a substantial exportable surplus cannot yet be determined.

In order to assist the Commission in the discharge of its responsibilities in the mining fields, an advisory committee on uranium mining was established in June, 1953. The committee consists of Mr. Murray, General Manager of the Mount Lyell Mining and Railway Co. Ltd., and a member of the Commission; Mr. Anderson, a director of Consolidated Zinc Pty. Ltd.; Mr. Kruttschnitt, a director of Mount Isa Mines Ltd.; and Mr. Raggatt, Secretary of the Department of National Development. Mr. Murray is chairman of the committee. This committee, comprised of men with great experience in the problems of mining and geology in Australia, has given invaluable help to the Commission.

In considering its responsibilities in relation to the development of uses for atomic energy, the Commission found it had inherited much less than had been the case with uranium production.

The greater part of the knowledge in the world on the production and use of atomic energy had come from the great wartime effort of Great Britain, the United States and Canada. This work had been carried out in secret.

Australia, as a country, had played no part in the work, though a number of Australian scientists, notably Mr. Oliphant, in their private capacities had played important roles.

At the end of the war the security arrangements between the three participating countries were continued, together with an arrangement for the exchange of atomic energy information, and it appeared that Australia might be permanently excluded from access to the important data necessary for operating in the atomic energy field.

There were a number of scientists in Australia with practical and inside knowledge of atomic energy work, but these men were in general not free to impart this information as they were bound by security undertakings.

There were also several university departments interested in atomic energy matters from whom the Commission could expect to obtain help and advice. Notable among these were the Schools of Physics at Melbourne University and the National University, where Mr. Martin, Mr. Oliphant and Mr. Titterton and their staffs had established international reputations by their research in nuclear physics. At Sydney University, Mr. Messel was commencing work on similar lines. In the N.S.W. University of Technology Mr. Baxter and Mr. Myers, who had recently returned from Harwell, were developing chemical engineering and metallurgical work related to atomic

energy problems, and in Melbourne Mr. Anderson, then about to return from Harwell, and Mr. Duncan were about to develop a strong School of Radio Chemistry.

In addition to this, the Commonwealth Scientific and Industrial Research Organization and the Department of Supply, in anticipation of developments in Australia, had made an arrangement some years before whereby Australian scientists on the staffs of these organisations could be placed at Harwell to work in the British Atomic Energy Organisation and gain experience in atomic energy work. This had been possible only on the understanding that these men would not report to Australia the work they were doing, and would treat their knowledge as secret until released from this requirement by the British authorities. Nevertheless, this group of about a dozen men represented an important potential for Australia development.

It was clear that the Commission had to make a choice between two ways of proceeding: firstly, to try to obtain access to the large amount of secret information possessed by the Atomic Energy Powers, and then to work in partnership with them; or to start at the beginning and laboriously work out the fundamental information and know-how, remaking the mistakes that others had made years before at great expense of money and time.

It was realised immediately that the first of these alternatives was the only practical one, the second probably being quite outside Australia's capacity in money or manpower.

In 1953, Mr. White and Mr. Baxter paid an extended visit to Britain, America and Canada to explore the possibility of some cooperative arrangement. The prospects were found to be promising. Further discussions were continued between Australia and Britain on a Government to Government level, during which Lord Cherwell paid a visit to Australia. In 1954, Mr. Stevens, Mr. Martin and Mr. Baxter visited Britain to continue the discussions and a little later the Minister for Supply, Mr. Beale, announced that an arrangement had been made whereby the British Government had offered to make available to Australia knowledge and know-how arising from Britain's industrial atomic energy programme. Further, Britain would enter into a cooperative research programme with Australia for furthering the development and application of atomic energy within the Commonwealth for industrial purposes. The arrangement provided for the Australian Atomic Energy Commission to create a research and development organisation, while Britain would provide facilities for training Australian scientists and engineers in that country.

Shortly after this agreement had been announced, the Minister for Supply stated that the Government had approved a programme submitted by the Commission for the creation of a research and development organisation in Australia.

This programme provides for the construction, on a site about twenty miles south of Sydney, of a comprehensive group of laboratories for research in problems of chemistry, metallurgy, mechanical and chemical engineering, physics, and other branches of science and engineering, relevant to reactor technology and atomic power production. This installation is also to include a large research reactor of a very modern type, designed to give a very high neutron flux, primarily for materials testing purposes but also for general research.

The estimated cost of the reactor and laboratories has been given at £A5,500,000.

To operate these laboratories, the Commission is creating a scientific staff, planned at the moment to reach fifty senior scientists, with a total personnel of about 400.

The senior members of this group are Chief of Research and Development, Mr. Watson-Munro; Chief Engineer and Deputy Chief of Research, Mr. Dalton; Head of the Chemistry Department, Mr. Miles; Head of the Metallurgy Department, Mr. Alder.

The Commission has made it clear that its programme, while part of an overall Commonwealth effort, will also be a self-contained attempt to develop economic industrial power from uranium.

The main problems which must be overcome before the realisation of industrial atomic power at an economic price are problems in engineering, metallurgy and chemistry, and could largely be summarised as problems in materials.

The Commission has indicated that its main effort will be directed to the development of high temperature, fluid fuel reactors suitable for power production, and the fact that its laboratories provide for extensive work in hot metallurgy and chemistry and in engineering is an indication of the kind of problems, both fundamental and applied, which the Commission expects to have to tackle. The choice of an ultra high neutron flux reactor for materials testing also indicates the kind of work to be undertaken.

To assist it in its work in the scientific and technological fields, the Commission has appointed a Scientific Advisory Committee. This committee consists of Mr. Oliphant, Mr. Martin, Mr. White, Mr. Myers, Mr. Hunter, Mr. Anderson, Mr. Brain, and Mr. Baxter (Chairman). This committee has guided the Commission throughout in its approach to scientific and technical problems.

The Act under which the Commission operates, not only empowers it to conduct research and development work, but to initiate and support such work in universities and other institutions. It may also take steps to support the training of scientists and engineers in those fields in which it is interested.

So far, the Commission has taken certain steps in this direction and has declared its intention in future to widen the scope of these activities within the limits of the funds which are available to it for the purpose.

Undergraduate scholarships in universities have been created in geology, geophysics and in metallurgy in order to increase the output of graduates in these subjects from Australian universities.

A system of post-graduate scholarships in science and engineering has been introduced. Here universities have been invited to nominate candidates and topics for research, and to those selected by the Commission, scholarships have been awarded, through the university, and grants for equipment have been made.

The Commission has also placed research contracts with some university schools for work on projects of direct interest to the Commission.

The Commission has announced its intention of providing special facilities for research and advanced training for engineers and scientists from universities and from industry in its laboratories at Sydney. No detailed scheme has been put forward yet but it would seem likely that the pattern already established in America at Oak Ridge and the Argonne, in Canada at Chalk River, and in Britain at Harwell, might be followed in Australia. This would involve post-doctorate, or even pre-doctorate research fellowships, under the auspices of Australian universities, but carried out in the Commission's laboratories, and periodical post-graduate schools, for training engineers and scientists on a shorter term basis in reactor technology, hot chemistry and other special features of this work, which cannot be done effectively anywhere but in a large and extensive establishment such as the Commission will control.

It is already clear that the development of industrial atomic energy, as it takes place, will make demands upon Australian industry for new facilities and techniques, and may in some cases alter the existing industrial pattern in a significant manner.

These changes may not be very rapid, but the Commission has felt that right from the start it should be in close communication with leaders of industry so that they may learn of trends which the Commission sees arising from its own and other people's research work. In turn, industrial leaders will be able to guide and assist the Commission when it is faced with problems of this sort.

The Minister for Supply recently announced that a Business Advisory Group had been formed, and that twenty leaders of Australian industry representing most of the larger industrial groups in the country had agreed to serve upon it. This Committee will be of great assistance to the Commission in the development of industrial atomic energy in Australia.

It will be seen from the foregoing that conditions have now been created in which the development of the peaceful uses of atomic energy can proceed rapidly in Australia.

The Commonwealth Government has, through its Atomic Energy Commission, set up an organisation which will bring together all the resources of Governments, of Industry and of the Universities and use them so that Australia may benefit as rapidly as possible from the new knowledge which is flowing from her own efforts and from those of her friends overseas. As the exchange of information on an international basis develops, through the efforts of the United Nations, Australia will be able to benefit by what she can learn from others, and will have something to contribute in return.

# Swedish Atomic Energy Company—A Cooperation between Government and Private Industry

**By Harry Brynielsson,* Sweden**

As early as December, 1945, the Swedish Government appointed Atomkommittén, the Atomic Energy Commission, to make proposals for the steps which should be taken towards the utilization of nuclear energy. In its first report in the beginning of 1946 the commission recommended an improvement of the resources of scientific institutes working in the field of nuclear physics and nuclear chemistry. For this purpose the commission received a grant of Swedish kronor 2,000,000. The commission also financially supported investigations on the uranium-containing alum shales and the possibilities of extracting uranium from them. Research on these problems was also initiated by a group of industries.

In its second report of April, 1947, the commission proposed the formation of a special organization, a company, to take care of the applied research and the industrial problems affecting the atomic energy programme. At the end of 1947 the Swedish Atomic Energy Company (Aktiebolaget Atomenergi) was founded.

Since the formation of the company, the main task of the commission, besides being an advisor to the Government on questions regarding atomic energy, is to stimulate and support fundamental research in nuclear physics and nuclear chemistry. Sweden is a member of the European organization for nuclear research, CERN, and is represented in this organization by the commission. The commission is under the Ministry of Education and has a yearly budget of about Swedish kronor 4,000,000 (= about $800,000). At present there are 13 permanent members; most of them are scientists, but there are also representatives from State authorities and industry. The Chairman is Governor Jacobsson and the Secretary General is Mr. Funke.

There are many and difficult problems connected with the peaceful application of atomic energy, and in order to solve them as effectively as possible, a cooperation between State and industry is important. In order to facilitate a collaboration, the Atomic Energy Company was formed as a company jointly owned by the State and some private enterprises, which were already showing real interest in the new energy source. The State owns 4/7 and private industrial interests 3/7 of the share capital. This amounts at the moment to 3½ million Swedish kronor (= about $700,000), and with regard to the company's extensive development programme an increase is being planned, with the above-mentioned proportions retained. The Atomic Energy Company is under the Ministry of Commerce. The private shareholders are at present about 20 large enterprises, mainly important consumers and producers of electricity and manufacturers of electrical and mechanical equipment. In connection with the above-mentioned increase of the share capital a greater number of industrial participants is anticipated.

Four of the Board's members, including the Chairman, are appointed by the Government and three, including the Vice-Chairman, by the private shareholders. There is close cooperation between the Atomic Energy Commission and the Atomic Energy Company, and some of the members of the commission are also members of the Board. Chairman of the Board is Governor Jacobsson, Vice-Chairman is Mr. Bengtson, and Managing Director of the company, Mr. Brynielsson.

The form of a company (aktiebolag) was chosen, as this type of enterprise by tradition has proved to be efficient in the realization of industrial operation. It was, however, obvious from the beginning that the company would not have any income of importance for many years. The current costs are met by annual Government funds. These funds from the start of the company up to June 30th, 1956, amount to approximately 61 million Swedish kronor (= approximately $12,000,000). Private industry contributes to the work in progress by supplying goods at favourable prices and with short delivery time, and by helping with premises and technical advice. The private contribution to the programme is expected to increase when more industrial problems arise. The share capital has been invested in buildings and other permanent installations.

According to the articles of association, the scope of operations of the company is "to prospect for and produce fundamental materials necessary for the use of atomic energy, to build experimental reactors and later, on a larger scale, reactors for exploiting atomic energy for research, commerce and industry, and to take care of the research as well as the industrial and

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commercial activity connected with the before-mentioned programme."

The company is at present organized in two departments, the Department of Chemistry under Mr. Svenke, and the Department of Physics under Mr. Eklund. Each department is divided into several sections.

The main task for the Department of Chemistry has been to develop methods for the extraction of uranium from alum shales. These shales occur in large quantities but have a uranium content of only about 200 grams per metric ton. In 1953, extraction on an industrial scale was started at Kvarntorp, producing a concentrate containing about 20% uranium. This concentrate is purified in a chemical plant in Stockholm, where also uranium metal and fuel elements will be produced.

The sections of the Department of Chemistry are: general chemistry; analytical chemistry; chemical technology; physical metallurgy; metal production; extraction plant (Kvarntorp); chemical plant (Stockholm); prospecting; and purchase and general services.

After preparatory work, the Department of Physics has built the first natural uranium, heavy water experimental reactor of 300 kw, which started in the summer of 1954. Plans are now under way for a second development reactor of some ten thousand kw, which is scheduled to be in operation in 3-4 years. Different types of power reactors are also being studied.

The sections of the Department of Physics are: theoretical physics; neutron physics; general and health physics; electronics; mechanical construction and heat transfer; reactor operation; nuclear chemistry; and purchase and general services.

In the administration a detailed bookkeeping system has been built up in order to keep a check on the costs of the different projects. A civil and constructional engineering section also belongs to the administration and has as one of its main tasks to plan and build the new research centre, Studsvik, on the coast of the Baltic, about 80 kilometres south of Stockholm, where the company has acquired an area of about 150 hectares for the next reactor and future experimental work in the reactor field.

For the planning of the work of the company the Board draws up a general programme of development for a period for example of 5 years. This has to be approved by the Government. Each autumn the management of the company makes a working programme on broad lines for the next budget year and calculates the necessary amount of money to carry out this programme. After this plan has been approved by the Board, the corresponding demand for money is given in to the State authorities.

Of course, such programmes cannot be binding in details. The rapid development in this domain often causes changes of the plans drawn up. Such modifications have to be decided by the Board of Directors, and if they are of major consequence, especially from the economic point of view, also approved by the State authorities. By having the majority of the shares, the State has secured control of the general policy of the company. For the execution of work, the management has the same independence as in other private companies.

In carrying out the programme, the company cooperates with many Government authorities and private enterprises.

As mentioned, there is close cooperation with the Atomic Energy Commission. Some of the fundamental research supported by the commission is of immediate interest to the company, and the research reactors of the company will also be used for fundamental research. Foreign scientists and lecturers are often invited jointly by the commission and the company. The present set-up consisting of a purely Government organization and a company where the Government is principal shareholder, but where also private enterprises actively take part, has proved to function well.

Prospecting for minerals of interest in atomic energy work is carried out in close cooperation with the Geological Survey of Sweden which also is undertaking several investigations for the company. Exchange of information also takes place with some big mining companies, which are prospecting for other minerals. The company is at present working out a programme to stimulate private prospecting for minerals of interest.

In Sweden the protection against radiation is supervised by the Institute of Radiophysics. The work of the section of health physics of the company is carried out in close contact with this organization. The authority which will eventually handle reactor safety questions, will certainly cooperate closely with the company and have access to all experience obtained there.

In June, 1954, the Atomic Energy Company took part as a founder member in the establishment of the European Atomic Energy Society (EAES) which is concerned with the practical applications of atomic energy, chiefly the construction of reactors and connected problems. This organization aims to achieve an increased exchange of experience between countries having a practical atomic energy programme.

The future power producing reactors in Sweden will probably be built by private industries and owned and run by the Swedish State Power Board and private power producers, each of which to-day contributes about 50% of the electricity produced in the country. For the development and design of these reactors, the supply of nuclear fuel and handling of burnt out fuel elements the Atomic Energy Company will form a central organization. It is to be hoped that the cooperation between State and industry, already begun, will continue and promote a rapid development of atomic energy in Sweden.

# Some Administrative and Legal Problems Related to the Widespread Use of High-Level Radiation Sources

**By William Mitchell,* USA**

The development of an atomic energy industry resulting in the widespread use of high-level radiation sources will create many problems requiring administrative or legal solutions which at first glance seem unusual or unique and of considerable magnitude. This is a common experience attending the growth of any new industry which widely affects the activities and conduct of the general public and which is dependent upon continuing technological advances. The automobile, the airplane, and the radio, afford recent examples. The important thing is to view these problems in their proper perspective, to fit their solutions to the technical developments, and to minimize so far as possible any impeding influence which they might have upon the progress of technology.

The potential danger inherent in the many processes in the atomic energy industry and the many unknown factors which still exist in the field have caused considerable speculation concerning the magnitude of the problems created and a tendency to exaggerate. Constant reference to a catastrophic incident and the prevalent but mistaken notion of an "atomic explosion" being capable of occurrence in a reactor have contributed greatly to the fears and doubts of the public and of potential members of the industry. These fears and doubts have a deterring effect on continuing development and progress and the prompt application of the results. Thus, a first step for any nation undertaking an atomic energy program should be an educative process designed to put the problems in proper perspective.

Complacency would be foolhardy, for grave danger certainly exists, but atomic energy hazards can be brought under complete control with proper safeguards in the design and operation of reactors and in the distribution and use of radioactive materials. It is with this understanding, rather than with emphasis on pessimistic or abnormal assumptions, that the administrative and legal problems in the field should be approached.

Against the background of operating experience, this paper will summarize the major administrative and legal problems discussed in detail in other papers submitted under this agenda item, sketch the scheme of control and regulation employed in the United States to protect health and safety, outline some of the issues of legal liability which might arise in the event of an accident, and present some observations with respect to the important problem of insurance.

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## BACKGROUND

The operating history of 25 reactors in the United States for the years 1943 through 1954 shows no accidents involving radiation injury sufficiently serious to cause lost time of personnel during 606,686 operating hours and 17,799,000 man-hours. With respect to critical facilities† during the same period, a *single* accident resulted in injury to 4 persons and 768 lost man-hours. There were no fatalities. This one incident occurred during 36,196 operating hours and 1,440,000 man-hours. Two laboratory accidents connected with critical assemblies of fissionable material occurred during the period surveyed. In each of these incidents, one man died and a few were injured by radioactive effects. These are the only fatalities known to be attributable to work in atomic energy installations since 1942. This remarkable safety record is far superior to the record of industry in general.

It is known that a similar record has been compiled in the United Kingdom. There, according to available statistics, with a cumulative experience of some 50,000 man-years, no deaths attributable to radioactivity have occurred, not a single case of permanent or temporary injury from external radiation has arisen, and the safety rate with respect to internal hazards is indicated as being a very small percentage of the best industrial rate, with no deaths and only one case of potential injury noted during a survey encompassing 10,000 man-years in a plant where the risk is deemed greater than most others in the industry.

The Canadian experience with a major reactor breakdown is of special interest and provides some first-hand knowledge concerning public safety and reactor operations. The breakdown of the Canadian

† A "critical facility" may be defined as a device designed to test at low power the critical mass neutron flux distribution, and other characteristics of a flexible arrangement of nuclear fuel, materials of construction, coolants, and other reactor components.

NRX reactor in 1953 due to the failure of the control system resulted in considerable physical damage to the reactor structure and the release of large amounts of radioactive materials. But there were no fatalities and none of the plant personnel was significantly affected by the radioactivity during the accident or in the subsequent decontamination, dismantling, and general clean-up operation. There was no mechanical damage outside the reactor itself, and after a precautionary evacuation, plant personnel were able to return on the next following work day. According to Canadian authority not the least of the lessons learned from the experience "is that large amounts of radioactivity can be handled safely even though they are spread over large areas and throughout a complicated industrial-type plant."

None of this means that reactor hazards should be taken lightly or the problems they create over-simplified. But the record does show what can be accomplished through an understanding of the dangers and the technology involved. It lends convincing support to the body of informed (but by no means unanimous) opinion which is strongly convinced that with proper attention to reactor design and the application of proven protective and control measures, the accidents that may occur in connection with the use and operation of nuclear facilities will be no more frequent and no more dangerous than the accidents which occur in many other industries.

## THE ADMINISTRATIVE AND LEGAL PROBLEMS IN SUMMARY

The control of radiation hazards is neither easy nor cheap, and the problem influences almost every aspect of atomic energy work. It is an important factor in the development of industrial processes, in designing plants and equipment, in the location of facilities, and in planning and scheduling work. It influences employee medical programs, selection and training of personnel, establishment of work routines, the clothing that workers wear, and even the way they wash their hands and where they can eat and smoke. It is estimated that in the United States approximately $100 per worker a year is spent in radiation protection. This represents about 1% of operating expenses, and based upon a 5% net return on investment would be equivalent to 20% of total profits in a private industrial operation.

Other United States papers discuss in specific detail many of the administrative and legal problems which have developed in connection with the control of radiation hazards and which will assume prominence as the use of radiation sources is broadened. The following are indicated as among the most important matters which must be considered:

1. The control of reactor design and containment through the establishment of specific codes (similar to those which have been developed in connection with boilers and pressure vessels) with adequate engineering design standards to insure safety in reactor performance.

2. The development of site requirements for the location of reactors which balance the hazards of operation with such factors as capital and labor costs, the availability of a water supply, the proximity to consumer load and the costs of transmission, the distance from fuel reprocessing centers, and the disposal of waste.

3. The training of industry personnel to develop competency in the handling of radioactive materials and in operating and manipulating the controls of nuclear facilities.

4. The development of a system of inspection and inspection procedures to assure that principles of safe operation are followed and that necessary relations between important operational features exist.

5. The development of a system of control to regulate the ownership and use of nuclear materials, the construction and operation of nuclear facilities, the distribution and use of by-product materials, and the health-safety standards for protection against radiation hazards.

6. The establishment of the rules of legal liability for personal injury or property damage, and the availability of insurance adequate to the needs of the atomic energy industry under those rules.

The main concern of this paper is with the last two problems.

## REGULATORY CONTROL IN THE UNITED STATES

It is, of course, recognized that the system of regulatory control designed to protect health and safety in the United States may not be appropriate under other forms of government, yet an outline of that system should be helpful to other countries now faced with the problems of organization and regulation.

The basis for all control in the United States is the Atomic Energy Act of 1954, a complex piece of legislation which throughout its many sections shows a predominant concern with the protection of public health and safety and provides the broad outlines of a plan for the control of materials and facilities essential to the industry. In general, the control scheme adopted has two main features: (1) a system of licensing which permits a review of proposed activities and the imposition of such conditions as may be deemed necessary, and (2) continuing supervision of a licensee's activities through the conditions and terms included in the license, through rules and regulations, through inspections, and through reports

### Materials Control

Three types of materials are subject to control special nuclear material (consisting of such materials as uranium-233, uranium enriched in the isotope $U^{235}$ and plutonium), source material (including natural uranium and thorium), and by-product materia (or radioactive isotopes produced in a reactor). By

statute, title to all special nuclear material, wherever produced in the United States, vests in the Government. However, the Atomic Energy Commission may issue licenses for the possession of this material and may distribute it to qualified applicants for purposes of research and development, medical therapy, and for the operation of licensed facilities. The private ownership of source material is permitted under the law, but a license is required before any person may transfer or use important quantities of source material after removal from its place of deposit in nature. The licensing controls cover all phases of commercial processing operations, including the milling of the ore, the refining of the ore to metal, and the fabrication of the metal into reactor fuel elements. With respect to by-product material, or radioisotopes, the Commission is authorized to issue licenses to applicants seeking to use such material for research and development purposes, for medical therapy, industrial uses, agricultural uses, or such other useful applications as may be developed.

As to each of the three groups of materials, the Act authorizes the Commission to "establish by rule, regulation, or order, such standards and instructions to govern the possession and use of" the material "as the Commission may deem necessary or desirable," among other things, "to minimize danger to life or property." By this provision the legislative branch of the Government has, in effect, delegated to an executive agency the authority to make the laws to protect health and safety which will govern all persons who possess or use special nuclear, source, or by-product materials. This technique of delegating authority, subject only to broad legislative standards, is familiar to students of constitutional and administrative law and is utilized generally when the field of regulation requires special knowledge or expertise and when flexibility is necessary or desirable.

### Facilities Control

The statute makes it unlawful to own or possess a reactor or other facilities which produce or use special nuclear material without a license. The system imposed is a two-step procedure. An applicant must first obtain a "construction permit" under which the reactor is built, and, after construction is completed, a license must be obtained for the facility. The construction permit is a recognition of the fact that safe operation is related to design and method of construction, and gives an opportunity to determine by inspection before potentially dangerous operations are begun whether safety standards have been met and whether the reactor will operate safely. Upon completion of the project a license will issue subject to "such conditions as the Commission may by rule or regulation establish." A reactor license may not issue if in the opinion of the Commission it would be "inimical . . . to the health and safety of the public," and all applicants must demonstrate and agree that they are equipped to and will observe "such safety standards to protect health and to minimize danger to life or property as the Commission may by rule establish." Further, prospective licensees must exhibit appropriate technical and financial qualifications, character, and such other qualifications as the Commission "may deem appropriate." Finally, the operators of facilities must be licensed under prescribed conditions after individual qualifications have been determined.

Under the broad statutory power which it exercises over materials and facilities, the Atomic Energy Commission is in the process of issuing or revising a series of regulations dealing with special nuclear material, source material, by-product material, facilities, facility operators, health and safety standards, and the rules of practice and procedure before the Commission for those who seek Commission action or who are subject to Commission orders. These regulations are necessarily lengthy and complicated, and it would serve little purpose to analyze them here in detail. There are, however, some common features and techniques employed in them to protect health and safety which should be of general interest.

First, it should be emphasized that the health and safety standards to be established for the atomic energy industry are not directed to the usual hazards associated with normal industrial activity but are addressed essentially to those risks which are peculiar or unique—the risk of a reactor breakdown, the risk of a release of fission products, the risk of exposure to excessive radiation. They are directed to the protection of both workers in the industry and the general public.

Basically, all of the regulations will require that:

1. A licensee must be a reliable person qualified through training and experience to use material which comes into his possession safely and for the purposes for which it is requested.
2. A licensee's equipment and facilities must be adequate to protect health and minimize danger to life and property.
3. The location of the proposed use must be suitable for the purpose.
4. The only use to which material may be put must be authorized by law and stated in the license.
5. A licensee must not transfer material or a facility except to a person authorized to receive it under a license.

The licensing regulations will not prescribe detailed accounting procedures with respect to materials received nor the type of physical protection to be given the material. They will not prescribe the kind of monitoring equipment licensees must have, the kind of shielding for various types of reactors, the procedures to be employed in the case of an emergency, or the times, places, and manner of performing surveys and monitoring. Rather, general health-safety standards are established and licensees will be required to utilize such equipment and procedures as are necessary for compliance. Finally, and perhaps

above all, the licensing regulations will have flexibility to cover circumstances where knowledge is imperfect, experience is insufficient to establish binding requirements of general applicability, or where the conditions of use or the nature of the activity are likely to vary infinitely.

The regulations establishing the standards for protection against radiation hazards arising out of licensed activities prescribe the operating limitations, conditions, and procedures which must be met under penalty of license revocation, injunction, or, in certain cases, criminal punishment. These regulations are concerned with the following major items:

1. Limitations on the permissible doses of radiation which may be absorbed by an individual due to exposure in both controlled areas where access is restricted and uncontrolled areas, limitations on the amounts or concentration of radioactive materials permitted to remain on exposed surfaces, and limitations on the amounts of radioactive effluent which may be released, discharged, or disposed of into air or water.

2. Hazard control, including surveys (or evaluations of radiation hazards under specific sets of conditions), personnel monitoring, respiratory protection, caution signs, signals and labels in radiation and concentration areas and for containers of radioactive materials, storage of material, and the training of personnel in the safe handling of material, the proper use of radiation-measuring devices, monitoring instruments, protective equipment and other devices furnished for their protection, and the procedures to be observed in the case of accident.

3. Control over the disposal of radioactive waste by burial in the soil, burial in the ocean, and by discharge into public sewers, through the imposition of conditions relating to disposal locations and the manner by which disposal may be accomplished.

4. The maintenance of records by licensees showing individual radiation exposures and the results of surveys, and reporting requirements in instances of over-exposure.

Here, also, the regulations are flexible, the limitations may be extended upon the showing of an operational need, and, in any case, it is provided that the Commission may grant such exemptions from the requirements of the regulations as may be authorized by law and which will not endanger life or property.

In drafting all the regulations certain basic considerations have prevailed. Regulations have the force of law; thus, they must to the greatest extent feasible be simple, concise, and unambiguous. They should not be written for the health physicist but for members of the industry, the general public, and the courts which may be called upon to enforce or interpret them. It is essential that they be understandable to people untrained in the field. Further, each requirement imposed must be justified by genuine and substantial considerations of health and safety and not by notions of desirable practice or good housekeeping. Material which is in the nature of a suggestion or advice, while it may be published elsewhere, should not be included in regulations, and matters which cannot be treated with precision and definiteness should be left for individual solution in specific cases by appropriate provisions in licenses and the issuance of appropriate *ad hoc* orders.

As stated before, this scheme for controlling radiation hazards through a system of licensing and regulation may not be acceptable or practicable in other countries. Further, it is recognized that knowledge of the problems which will be encountered in the expanding use of radiation sources is far from complete and that further understanding may require constant modification and revision. But in the United States there is a considerable experience in the handling of materials and in the operation of nuclear facilities. The control scheme outlined above is based on that experience, and indicates one way to meet the problems which will be encountered. Certainly, here is an area in the field of atomic energy which offers great opportunity for an international exchange of ideas, information, and accumulated experience.

## PUBLIC LIABILITY

There has been considerable discussion, particularly among lawyers, concerning the matter of civil liability for radiation damage and much speculation concerning the liability rules which will apply in the event of an atomic accident. It is reasonable to assume that the utilization of atomic energy will raise unique problems but it is difficult to conceive of any which cannot be resolved within the framework of existing legal systems. It is equally difficult to assume the answers, and the principles of legal responsibility which will prevail must await the facts and practicalities of particular cases. There are, however, certain precedents in the law which by way of analogy indicate future issues which may arise.

### Liability of Owners and Operators of Facilities

An accident causing public damage will raise the issue of strict liability, or liability without fault, under which proof of negligence is unnecessary. In 1866, the English Court of Exchequer first announced the doctrine that one "who for his own purposes brings on his lands and collects and keeps there anything likely to do mischief if it escapes, must keep it at his peril, and, if he does not do so, is *prima facie* answerable for all the damage which is the natural consequence of its escape."

In affirming, the House of Lords limited use of the rule to situations involving a "non-natural" use of the land [*Rylands v. Fletcher*, L.R. 3 H.L. 330 (1868), affirming L.R. 1 Ex. 265 (1866)]. The doctrine is incorporated in the American Restatement of Torts, which recognizes the general rule

that there is no liability for "unintentional and non-negligent" conduct even where harm results, but announces a single class of exceptions for so-called "ultrahazardous activities." Section 159 states that: ". . . one who carries on an ultrahazardous activity is liable to another whose person, land or chattels the actor should recognize as likely to be harmed by the unpreventable miscarriage of the activity for harm resulting thereto from that which makes the activity ultrahazardous, although the utmost care is exercised to prevent the harm."

This concept of strict liability has been applied to the storage of explosives, to blasting, and to ground damage from aviation. Its extension to damage from radiation caused by escaping fission products, in those countries which accept the doctrine, would seem to be consistent with the generalized rule of ultrahazardous activities.

It is, however, far from clear that one could support a general statement that strict liability will be applied in all cases of atomic accidents. Much will depend on technological developments, on the availability of insurance permitting the risk to be spread, and on prevailing social values, particularly where the operation involved is for the benefit of the public generally and is essential to the good of the State as a community. There are, furthermore, certain legal defenses which might succeed—the fault of the plaintiff, intervention by a third-party, acts of God, normal or ordinary use of the land, and statutory authority. The latter two may well prevail in the typical fact situation which can be hypothesized. The English Courts themselves have excluded absolute liability where the activity in question was "merely . . . the ordinary use of the land or such a use as is proper for the general benefit of the community," *Richards v. Lothian,* [1913] A.C. 263 (P.C.), and it has been indicated that the manufacturer of explosives in wartime may be an "ordinary user," see *Read v. Lyons,* [1945] K.B. 216, 240 (C.A. 1944). Legislative permission to conduct an activity has the same effect as "natural user." In *Northwestern Utilities, Ltd. v. London Guarantee & Accident Co.,* 154 L.T.R. 89 (P.C. 1936), the rule of strict liability was held inapplicable to a utility company whose gas escaped into a basement and exploded, on the ground that the company located and used its pipes in accordance with statutory permission. *A fortiori* if, in addition to statutory authority, a defendant could show that his activities in all respects were conducted in accordance with official regulations and standards.

The presence of the State as a party in any litigation due to the ownership of the reactor fuel will raise additional questions relating to (*a*) the scope of the State's liability—compare Section 2 (1)(c) of the British Crown Proceedings Act (10 & II Geo. 6, c.44) which imposes governmental liability absolutely by reason of the ownership or control of an ex-hazardous instrumentality with Section 410 (a) of the United States Federal Torts Claims Act which apparently requires a "negligent or wrongful act or omission" of a government employee; and (*b*) liability for discretionary acts—see *Dalehite v. United States,* 346 U.S. 15 (1953), relieving the United States Government from liability in connection with the Texas City disaster by reason of the discretion and policy decisions involved in the Government's ammonium nitrate fertilizer program.

If it should be required that proof of negligence is a condition to the imposition of liability, there is a further principle in tort law which will benefit a plaintiff and ease the problems of proof; namely, the principle of *res ipsa loquitur.* Basically, this doctrine, which applies when the cause of the injury or damage is under the sole control of a defendant and experience indicates that the accident causing the harm will not happen if due care is exercised, permits the drawing of inferences of negligence from a mere recitation of the occurrence. It has been applied in a variety of circumstances—an unexplained explosion in a powder factory, boiler explosions, unexplained airplane accidents, bursting bottles, falling ceilings—and it is quite likely that an argument will be made for application in a case involving a reactor accident. The following language from an opinion of one of our state courts in a boiler case indicates the approach which may be taken:

"Boilers sometimes explode. Comparing the number of explosions with the extent of the use of boilers, explosions are not frequent. If they are kept in proper condition and repair, and if they are operated properly, explosions are unusual. Whether the *res ipsa* doctrine, which permits an inference of negligence from the fact of an explosion, should apply is largely a question of how justice in such cases is most practically and fairly administered. There is nothing illegally illogical in permitting the inference to be drawn. Usually the party injured is without information upon which he may with certainty allege the exact cause, and is without direct proof. Perhaps the exact cause is incapable of ascertainment. The actual proof, if any, is with the party having the management of the instrumentality. These are practical considerations. We think the jury should have been permitted to draw an inference of negligence . . . from the occurrence of the explosion . . ." *Kleinman v. Banner Laundry Co., 150 Minn. 515 (1921).*

This somewhat lengthy identification of a problem of civil liability has been presented only to indicate that the questions which arise are substantial and the answers not easily perceived. The facts in any case will predominate and shape the results. Special care should be taken to avoid hasty generalizations concerning the applicable rules, in order to avoid the mistakes that were made, for example, when the automobile first presented novel questions of negligence and liability.

### Liability of Manufacturers and Suppliers

A defect in a component part of a reactor, faulty construction of reactor facilities, and the mishandling or misuse of radioactive products may cause widespread damage and present the problem of the liability of the manufacturer, constructor, or supplier to injured third persons (*i.e.*, persons other than immediate contractors or buyers).

The general rule in the United States is that:

"A manufacturer who fails to exercise reasonable care in the manufacture of a chattel which, unless carefully made, he should recognize as involving an unreasonable risk of causing substantial bodily harm to those who lawfully use it for a purpose for which it was manufactured and to those whom the supplier should expect to be in the vicinity of its probable use, is subject to liability for bodily harm caused to them by its lawful use in a manner and for a purpose for which it was manufactured." — *Restatement of Torts, § 395.*

This principle was originally discussed in terms of "dangerous substances," but has been so extended by recent decisions as to render the concept of "dangerous" practically meaningless. Now, if substantial harm can be foreseen and if the chattel is defective the rule applies.

An extension of the doctrine in *Moran v. Pittsburgh-Des Moines Steel Co.*, 166 F. 2d 908 (3rd Circ. 1948) is of special interest in the atomic energy industry. Defendant, under contract with a public utility company, designed, furnished materials for, and constructed a tank on the utility's land for the storage of liquified natural gas. Thirteen months after completion and acceptance of the tank, it ruptured, releasing large quantities of gas and causing fires and explosions in which more than 100 lives were lost. An employee of the utility company engaged in work unconnected with the storage of gas was killed, and an action for wrongful death was brought against the builder of the tank. The court held the defendant liable for negligent defects in manufacture to one who might reasonably be expected to be in the vicinity of the chattel's use and, also, that the principle applied even though the tank when installed technically became part of the structure and land of the utility company.

The decision is important in that it includes within the rule not only manufacturers of equipment but building contractors as well, and presumably defective design and engineering.

In any case, however, according to prevailing authority there must be proof of negligence. Attempts to extend the doctrine of strict liability to manufacturers of articles or equipment which prove to have a defect that causes injury have not as yet met with much success; but it can be expected that this new principle of liability will be advanced in cases of injury or damage due to reactor break-down. Acceptance will depend upon the courts and circumstances.

The commercial distribution of radioactive products will also present liability problems. Modern case law holds the manufacturer liable for injury due to inherently dangerous articles marketed without the necessary cautionary statements. A danger is inherent when it derives from the nature of the article itself, as opposed to dangers resulting from a defectively made article that is ordinarily harmless. Negligence attaches not to the manufacturing, but to the distributing and marketing process and is founded on the failure to give proper instructions and warning.

The very good chance that the defenses of contributory negligence and assumption of risk will present recovery in most cases arising out of the distribution of radioactive products may inspire the argument that strict liability should attach in order to stimulate standards of conduct needed to protect the public. Using as analogy the statutory liability imposed with respect to foods, drugs and cosmetics, it may be advanced that when the distribution of radioactive products is subject to control through licensing and regulation proof of a violation of the regulations and the conditions of the license will constitute conclusive evidence of negligence. Here, again, we merely identify the nature of the problem and do not presume to supply the answers.‡

## INSURANCE

Questions of liability lead to the problem of insurance, about which there has been considerable discussion and study. The matter has become of immediate interest in the United States only recently, for while the atomic energy program was financed wholly from public funds the Government assumed directly all risk of loss. Private industry is now being asked to assume the risks and private insurors to insure them. The principal question concerns the availability of private insurance at reasonable rates adequate to the needs of the industry. The problem is complicated by the magnitude of possible loss, the limits to the size of risk which an insurance underwriter can assume, the lack of the type of actuarial data needed to appraise the probabilities involved, and the absence of any definite conclusions by the industry itself as to the amount of insurance it needs.

In the United States, an Insurance Study Group, composed of executives of leading companies in the insurance industry, was appointed to review the insurance problems created by expanded industrial participation in atomic energy and to develop infor-

‡ There are other legal problems which might be mentioned: Since radiation injury may not become apparent for some time, statutes of limitations may have to be changed or special treatment afforded such injuries. International transportation of materials and a catastrophic incident causing widespread damage over a large geographical area may raise questions of jurisdiction and the choice of law. A "mass" tort may present procedural questions. For problems related to workmen's compensation, see Greene, "Workmen's Compensation Aspects of the Peaceful Uses of Atomic Energy", P/323, Session 4.3, Vol. 13, these Proceedings.

mation and criteria with respect to the insurability of atomic energy installations and undertakings. The group's interim conclusions contained in a preliminary report recently filed are informative and helpful and seem to present a more optimistic picture than was originally expected. They may be summarized as follows:

1. The catastrophe potential is more serious than anything now known in the insurance industry, but the possibility of a serious catastrophe seems very remote.

2. The insurance capacity now applicable to the more hazardous types of chemical operations appears to be adequate to cover *physical damage to reactor plants*. However, if a reactor is located in proximity to large existing industrial plants, the increased exposure of these plants may be beyond the capacity of the insurance industry and further study of the problem is indicated.

3. *Physical damage losses to reactors and machinery* incidental to atomic power plant installations can probably be handled in the same way that boiler and machinery coverage on extra-hazardous machinery is now handled in other industries. Radioactive contamination of machinery and equipment and the containing buildings resulting from reactor failure presents new hazards requiring further investigation by the insurance industry to determine the nature and scope of coverage that can be afforded.

4. Insurance against *loss of use* presents a special and difficult problem reflecting both the expenses of decontamination and the long delays involved in the process. Such insurance is, in effect, a guarantee of successful operation, and it is believed that if any such protection is available it will be somewhat limited in amount.

5. *Workmen's compensation* for employees engaged in the operation of reactors and auxiliary equipment can be handled by existing insurance facilities. The major catastrophe problem here is with respect to employees of other plants in which the air and water supply might become contaminated. Whether such employees would come within the terms of a workmen's compensation policy remains a question.

6. The *public liability hazards* resulting from injury to persons or damage to property and arising from the manufacture, construction, ownership or operation of power reactors can be insured by existing insurance facilities up to the limits of liability normally available to more hazardous types of industrial enterprises.

The most serious problem as to the amount of insurance available lies in the field of what is termed "third party liability insurance" and arises from the very high aggregate dollar amount of claims which might arise in the event of a possible, though not necessarily probable, catastrophic accident. Such claims might be made directly by the person suffering injury or damage, or they might arise as subrogation actions on the part of insurers called upon to pay the loss in the first instance. In any event, the impact upon casualty underwriters may be very great and there might flow into the liability insurance market from a single accident unprecedented claims aggregating the total monetary damage assessed for loss of life, personal injury, and damage to property.

Notwithstanding the complications involved, the report of the Study Group concludes with the belief "that the insurance industry can work out an aggregate limit of liability for all parties at interest substantially equivalent to those normally required by other major industrial enterprises."

Essentially, the questions which remain, after the general conclusions of the group are translated into *amounts* of coverage, must be answered by the atomic energy industry: How much coverage does the industry want? Is that amount greater than is available in the commercial market? If so, is the desire for excess coverage reasonable and warranted?

If it is concluded that excess coverage is required and can be justified on the basis of need and if it is determined that the lack of such coverage is proven to be a deterrent to participation and progress in the field, some measure of Government assistance may be indicated. Several suggestions have been made concerning the form which such assistance should take: the development of a scheme similar to that which prevailed with respect to war damage, government reinsurance, direct government excess coverage insurance, and general indemnity legislation which would obligate the government to pay for losses exceeding those commercially insured. No decision has been made in the United States in this regard and no such determination can be made until the limits of desired coverage are defined and justified.

# Administrative Problems in the Industrial Utilization of Atomic Energy

**By George G. Manov,* USA**

Time telescopes today's living—the coal industry is some 2000 years old; the steam age approximately 100, and atomic energy only ten short years. Yet in this space of time, most of the still unsolved technical problems involved in the commercial application of atomic energy are well on their way toward solution. The administrative problems, however, are just beginning to be recognized.

In this paper attention is focused away from the purely scientific and engineering aspects of the utilization of atomic energy and is directed instead toward an examination of some of the nontechnical problems involved. Before this new force can take its place within the normal framework of industrial economy, questions regarding the financing of atomic energy facilities, the location of A-power stations nearer centers of population, the control of radiation hazards, and the insurance aspects are among those that must be answered. Last, but not least, one of the very important remaining tasks is the education of the executive.

## FINANCING ATOMIC ENERGY DEVELOPMENTS

For the purpose of this discussion, atomic energy facilities are defined to include radioisotope laboratories, gamma irradiation installations, research reactors, power reactors, and fuel processing establishments.

The United States government has spent a total \$12.3 billion ($\$12.3 \times 10^9$) in the atomic energy program since 1941, of which \$4.8 billion may be said to represent the present physical value of the industrial plant. By comparison, the contribution of private capital to the atomic energy program has been rather small. From 1951 through 1954, private industry, chiefly through the power-study groups, spent \$8 million. With the passage of the Atomic Energy Act of 1954, the relative proportion of expenditures may change as capital enters the field.

Several banking and investment houses have expressed interest in seeking to increase their knowledge of atomic energy not only on a technical basis but also to identify the need for risk capital and the probable financial gain therefrom. There seems to be a clear realization, however, that atomic energy offers no quick return on invested capital.

If one assumes that in 1960 one new electric power plant in every ten constructed may be powered by atomic energy, it would appear that funds of approximately \$1 billion per year might be required, about one-half of which would be for the nuclear portion of the plants. From an administrative point of view, such risk capital might require, and be qualified for, incentives that are available to similar industries such as ordnance and aviation. Reviewing the past history of American enterprise, one does not doubt that such risk capital will be forthcoming.

## LOCATION OF ATOMIC ENERGY FACILITIES

Surprisingly enough, only six short years ago there was considerable apprehension expressed concerning the locating of radioisotope laboratories in highly populated areas. Today, there are well over 3000 laboratories using radioisotopes, many of them situated in the downtown areas of large cities. It is to be expected that as more experience is gained in the operation of research and power reactors, as well as other atomic energy installations, nuclear hazards will find their proper place in relation to other industrial hazards with which we are more familiar.

The first large-scale nuclear reactor was located in the sparsely populated hills at what is now Oak Ridge, Tennessee, and following conventional practice in designing explosive plants (for want of a better guide), each group of processing facilities was placed in a separate valley to minimize the over-all effect of any possible explosion. The production plant at Hanford, Washington, and the National Reactor Test Station at Arco, Idaho, were located with the thought that the most practicable form of protection was a large exclusion area. This reasoning is still valid for installations which are for the most part experimental in nature and must of necessity possess sufficient flexibility to permit a whole spectrum of testing and research operations.

### Reduction in Size of Exclusion Area

However, if atomic power is to be put to practical use, it is imperative that the size of the exclusion area be held within reasonable limits, if only because of (1) the amount of capital tied up in the exclusion area, and (2) the absence of sufficiently large areas near metropolitan and industrial centers. For example, a nuclear reactor of 500,000 kw capacity serving

* US Atomic Energy Commission, Washington, D.C.

a city of 500,000 population, would require an exclusion area of several hundred square miles.

It is evident that atomic power will not be able to compete on an economic basis with conventional power if such large areas are required. It would be possible to reduce the capital investment in real estate by moving the plants farther away, but it is generally conceded that locating generating facilities 200 miles or more from large consuming centers is impracticable because the line losses and the cost of installing transmission towers fail to be compensated for by the savings in the cost of land.

One major step forward has been the placing of a prototype reactor (for the submarine "Sea Wolf") inside a steel sphere 225 feet in diameter. This sphere with a cross-sectional area of nearly one acre was constructed at a cost of about $2 million. If one assumes that 6000 horsepower are required by conventional methods to propel a submarine of the same length as the Sea Wolf at conventional speeds, the cost of the "Land Area" is approximately 10 mills/kwh. In considering this example, it must be remembered that no attempt is being made here to produce competitive electric power.

A second step in reducing the cost of "real estate" has been taken with the construction of 60,000 kw Pressurized Water Reactor at Shippingport, Pennsylvania. This reactor is enclosed in a steel shell. The approximate cost of constructing this shell was about $2 million, indicating an exclusion area cost of 0.8 mills/kwh. The land cost of a conventional power plant is negligible in terms of mills/kwh. Improvements in the design of containment areas, the use of reactors that are self-regulating and a more realistic approach to the problem of reactor hazards will permit the locating of A-power reactors in the same geographical location as conventional power plants.

It is not a corollary that a chemical reprocessing plant must be located next to each atomic power plant. Indeed, some thoughts have been expressed with respect to concentrating fuel reprocessing plants in several locations throughout the United States, each plant to reprocess the spent fuel rods from a group of power plants.

## COST OF ROUTINE RADIOLOGICAL SAFETY PROTECTION

What are the costs of routine radiological safety protection in an atomic energy installation?†

From the administrative point of view, however, it is interesting to note that the Atomic Energy Commission spends approximately $100 per year per man to insure radiation protection, and that the costs of other safety devices (conventional type) are quite small by comparison. It is estimated that the cost of radiation protection is about one per cent of the total operating expenses, and that if it were desired to protect against radiation by an additional factor of 10, the cost would be two per cent of the total operating costs. This would appear to be quite small, until one realizes that this additional one per cent would correspond to 20 per cent of the total profits of a commercial enterprise, based on 5 per cent net profit. To an administrator, it therefore becomes of considerable importance to assay the dollar cost of radiation protection.

### Approaches to Cost Reduction

What approach should be used in arriving at a proper level of radiation protection? The traditional manner has been to introduce a new process method into industry and to determine by case histories to what extent the hazards actually appear. This is the actuarial or the statistical approach. If time were to permit, this might be one sound way of ascertaining the true extent of radiological hazards.

A second approach might be to guard against all possibilities and to provide for protective devices and for radiation safety codes complete in such detail as to preclude the possibility of accidents happening. In this day and age such an approach would be comparable to the old-time requirement that each automobile must be preceded by a man with a red flag, and that if two automobiles met at an intersection neither should proceed until the other had passed. This would indeed insure a perfect safety record in the industry if that industry could survive at all.

But there is a third, or preventive, approach. The work of the Atomic Energy Commission and its contractors during the past fourteen years has indicated that enormous quantities of radioactivity can be handled safely. Accident rates in the Commission's operations involving some 134,000 contractor personnel are noticeably lower than in other conventional industries, and in the peaceful applications of atomic energy there have been no fatalities caused by atomic radiation. This is an amazing record when compared to conventional hazards in other industries.

Lost-time injuries per million man-hours from data supplied by the National Safety Council and the Atomic Energy Commission are as follows: electrical utilities, 11; non-ferrous metals and products, 10; *average for all industries,* 8.2; miscellaneous manufacturing, 6.1; chemical, 5.5; AEC, 2.3; and communications, 1.8. It should be remembered that the statistics for the atomic energy program include accidents not related to atomic radiation itself, such as in normal construction work. From these figures it would appear that workers in atomic energy plants are more safety-conscious than persons in conventional industrial plants. It is axiomatic that a vigorous employee safety program strongly supported by top management pays dividends.

In this discussion, there is no intent to over-simplify the problem of employee safety, but it should be stressed that adequate protection against radiation can be obtained and that the know-how is available. With film badges, radiation-detection devices, contamination-detection equipment, etc., on hand, there

† There is a great deal of technical information available on the subject of radiation protection, and the details will not be discussed here.

is no reason why the safe utilization of atomic energy on an industrial scale cannot be achieved.

But, there is room for belief in the argument that perhaps safety is being over-done. There are as many as fifty different ways of shutting down some reactors, and frequently shut-downs occur because of failure of a safety device. To draw an analogy, it might be helpful in some cases if we had an attachment for an automobile that would automatically sound the horn, turn off the ignition and apply the brakes whenever the pressure in one of the tires, for example, fell below a certain predetermined value. Yet one could conceive of many situations in which it would be both unwise and even dangerous to have such a shut-down occur. As our knowledge progresses in the identification of the key points of reactor safety, so will the instrumentation become simplified and our confidence in the operating reliability of reactors increased.

In short, considerable thought is being given, and direct action is being taken, to insure so far as is humanly possible that the atomic age becomes a part of the normal American industrial economy with maximum regard to the health and safety of the workers.

## LEGISLATIVE CONTROL OF RADIATION HAZARDS

### Control at the Federal Level

Radioactive "byproduct" materials in the United States are obtainable only from the Atomic Energy Commission or its authorized secondary suppliers and distributors. The Atomic Energy Acts of 1946 and 1954 lay down certain criteria for the possession of radioactive material. Section 81 of the 1954 Act contains the statement:

". . . The Commission shall not permit the distribution of any byproduct material to any licensee, and shall recall or order the recall of any distributed material from any licensee, who is not equipped to observe or who fails to observe such safety standards to protect health as may be established by the Commission or who uses such material in violation of law or regulation of the Commission or in a manner other than as disclosed in the application therefor or approved by the Commission. . . ."

Section 81 also states that:

"No person may transfer or receive in interstate commerce, manufacture, produce, transfer, acquire, own, possess, import, or export any byproduct material, except to the extent authorized by this section. . . ."

In anticipation of the increase of industrial participation in the atomic energy program, the Atomic Energy Commission has taken a number of steps to maintain adequate administrative control at the Federal level. The Advisory Committee on Reactor Safeguards, established five years ago, continues to report to the Commission. A Reactor Hazards Evaluation Staff has been established whose duties are to develop standards, guides and codes for the design, operation, supervision, containment and location of reactors in order to protect the public health and safety, and to evaluate proposals for new reactors and of significant changes in existing reactors.

The Commission has also established a Licensing Division which will be responsible for issuing licenses to various firms and individuals desirous of engaging in the applications of atomic energy either on a research or industrial basis. In the case of radioisotopes, these procedures have been in force since 1946 and provide for control at the source of supply (allocation procedures) and for control at the point of use (inspection of facilities).

An Inspection Division has been established whose duties will include the careful supervision of the Commission's and contractor's staff to insure that health and safety standards are maintained in the operations of the atomic energy program. Additional contemplated regulations involve licenses for operators of reactors and other utilization and production facilities, together with Federal health and safety standards.

### Control at the State and Other Levels

Various states have become interested in considering legislation to control the potential hazards in the atomic energy program. Among these are New York, Pennsylvania, California and Connecticut.

The National Committee on Radiation Protection under the sponsorship of the National Bureau of Standards has published a number of handbooks which, while lacking in legal authority, are nevertheless looked upon as recommended manuals of "good practice."‡ In addition, the NCRP recently formed a subcommittee to study the feasibility of state regulation for the control of radiation exposure. A draft interim report has been circulated within the Subcommittee containing suggestions as to the form that such legislation might take.

The Boiler and Pressure Vessel Committee of the American Society of Mechanical Engineers has established a sub-group on Nuclear Power. In addition, other industrial organizations such as the American Standards Association, American Society for Testing Materials, and others, have likewise evidenced interest in the preparation of safety codes. The subject will not be dealt with in detail except to point out that a number of qualified groups are earnestly at work on the problem and that from this array of talent, both technical and administrative, there should evolve suggestions for a workable system of legislative and technical control of radiation hazards.

‡ Handbooks recently issued include NBS-42: Safe Handling of Radioactive Isotopes; NBS-47: Recommendations of the International Commission on Radiological Protection; NBS-48: Control and Removal of Radioactive Contamination in Laboratories; NBS-49: Recommendations for Waste Disposal of Phosphorus-32 and Iodine-131 for Medical Users; NBS-52: Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water; and NBS-54: Protection Against Radiations From Radium, Cobalt-60, and Cesium-137.

It is to be hoped, however, that in the enthusiasm to formulate such controls, the various suggestions will not have been transformed into legislation which might be unduly restrictive or which might work at cross purposes from one state to the next. One recalls that there is a legend—in Africa—that the Giraffe was designed by a Committee.

This comment applies also to the question of international regulations and codes for health and safety. American as well as European industrial firms may likewise find it advantageous to work toward international safety codes to permit normal commerce in the field of applied atomic energy. A start has been made with the several meetings of the National Committee on Radiation Protection, the most recent of which took place in Stockholm in August, 1953, and which considered maximum permissible limits of radiation exposure to individuals. In Paris, on June 23, 1955, at a meeting sponsored by UNESCO (United Nations Educational, Scientific and Cultural Organization), there was discussed the question of international standards for the shipment of radioactive materials.

It is hoped that many more such conferences can be held at the various national and international levels, and agreements reached on the over-all approach and general technical content on such proposed regulations, before individual and differing regulations are adopted by each nation.

## INSURANCE PROBLEMS

Under the new Atomic Energy Act, the insurance picture may change from one in which little insurance is purchased as such (self-insurance) toward a situation where more normal conditions are expected to apply. In this respect the new Atomic Energy Act says in part ". . . the licensee will hold the United States and the Commission harmless from any damages resulting from the use or possession of special nuclear materials by the licensee."

The average insurance executive has not had the time to devote himself to a study of nuclear physics or radiological safety. In attempting to assess the impact of atomic energy upon the insurance field, he has had perforce to rely on fragmentary information gained in part from semi-technical articles, and colored by the recurrent misidentification of a nuclear reactor with a nuclear weapon.

In 1950, when the radioisotopes program was well underway, there arose the question of rates for personal life insurance for atomic energy workers, public liability, property damage and workmen's compensation rates. Under the sponsorship of the Atomic Energy Commission and the Brookhaven National Laboratory a series of seminars was held February 6–10, 1950, with the representatives of the major insurance companies. These seminars did much to clear the atmosphere and were mutually helpful to the isotope user and to the insurer. As a result of in-service training programs, insurance companies now possess trained personnel sufficient to answer inquiries sent to them by other insurance companies. The over-all result is that the use of radioisotopes has been placed in its true perspective within the spectrum of other industrial operations.

We are now faced with the analogous problem of insurance in the field of the nuclear reactors. Understandably, insurance executives are troubled with the thought of a run-away reactor contaminating tens of thousands of homes, automobiles and other valuable property, and the possibility of one company being faced with a multitude of individual personal damage suits totalling perhaps ten to one-hundred million dollars.

Purely from an administrative point of view, it would be extremely helpful to have had one or more major reactor failures, if only to have some frame of reference with respect to the frequency of accidents and extent of the damage involved. Where would our insurance rates and our safety codes for conventional power plants be if we had yet to experience our first boiler failure?

Differentiation should be made between the *probability* of loss which largely determines the insurance rate and the *possibility* of loss which is a maximum catastrophic loss for which an insurance company might be held liable. In neither of these categories is there actuarial or statistical experience, and rates must be set largely on the basis of the maximum amount of liability that might be involved. In many respects the situation parallels closely that of writing insurance on the first commercial airliners.

In this valley of uncertainty, it is helpful to point out the record of the several dozen operating reactors and critical assemblies in this country. The Hanford reactors have been in operation almost twelve years with no more maintenance than would be expected for a pioneering-type reactor. The Canadian NRX reactor failure is probably the severest to date, and yet even here the reactor has since been put back into operation at increased power. One other case involved the planned failure of the experimental boiling-water reactor experiment. Here, in the course of a scientific investigation, it was necessary to determine the parameters of a reactor involving extremely high rates of control-rod withdrawal and tremendous surges in power. The experimental data obtained should be of considerable assistance in determining the boundary conditions in the design of this particular type reactor. It is to be hoped that additional experiments will be carried out so that more of these planned failures will be made to take place, and that from these data the rudiments of a nuclear reactor safety-code can be set down on paper. Eventually there may be established for reactor safety, an organization which might be the counterpart of the National Underwriters Laboratory.

These operating experiences, some of them with reactors working at full capacity, yield at least two points on the curve in terms of possible failure per million reactor-hours. Because "two" is a very small

number statistically, it is difficult to place much faith in any extended extrapolation of this record and yet it must, of necessity, form the baseline in our consideration of reactor hazards.

The Engineering Test Reactor, as well as the Materials Test Reactor, will continue to furnish much valuable information regarding metallurgical properties under the influence of high intensities of radiation. Facilities now under construction at the National Reactor Test Station at Arco, Idaho, include provision for transient-testing and safety determinations for various types of reactors. Other research elsewhere includes development of safety "fuses" for reactor cores. Meanwhile, within the next five years there may be from ten to twenty university-type research reactors located in various parts of the United States. The safety records in their operation should do much to influence the administrative thinking in terms of power reactors.

Undoubtedly, improvements will be made. For example, the Swimming Pool Reactor displayed at the Geneva Conference possesses a completely automatic start-up and power-level control system. By contrast, it may be recalled rather humorously that the first commercial electrical distribution system consisted of bare copper wires laid in a wooden trench.

The Atomic Energy Commission has recently granted security clearances to a committee of top insurance executives to study the general problem of insuring industrial power reactors. As a result of detailed technical conferences with Commission and other personnel and of visits to operating installations, this Committee may be in a position to make some specific recommendations.§

During the next few years, perhaps some form of government assistance may be necessary to help carry the risk of any catastrophic damages. As a precedent, an organization similar to the War Damage Corporation, as established by the US Government during World War II, or assignment of such responsibility to a present organization, for example the AEC, might be among the solutions considered.

It is believed that in time the insurance industry will solve the problems of insuring reactor operations against all types of hazards in the same manner that it solved analogous problems of air and railroad transportation.

## SUMMARY

The industrial application of atomic energy offers many new and challenging administrative problems. None of these is insurmountable; they can be and must be put into their proper perspective in order that atomic energy may rightfully take its place in the world economy.

§ The report of this Committee will be published shortly.

# Problems in the Legal and Administrative Control of a Program for Distribution of Radioisotopes

**By Stephen P. Cobb, Jr.,* USA**

Radioactive isotopes can be of immeasurable benefit to science and technology. Their potentialities were known early in the 20th century through the use of naturally-occurring radioactive materials. The scope of their usefulness broadened in the 1930's when it was learned that elements throughout the periodic table could be made radioactive through the use of high energy accelerators. Then after World War II and the development of nuclear reactors, the availability of radiosotopes increased tremendously, with a corresponding decrease in cost. Since then it has been possible to explore more completely the full scope of radioisotope usefulness.

Although radiation can be extremely useful, it is also potentially harmful if not controlled. During the early years following the isolation of radium few controls were maintained. There were abuses in the handling of radium which led to deaths in subsequent years, with symptoms strongly pointing to radium poisoning. Many curative powers were attributed to radium by unscrupulous persons anxious only to exploit the material. Some efforts were undertaken in the early 1920's to curb unrestricted medical use of radium through standardization of such uses. The efforts to acquaint people with the potential hazards of radiation and to limit the use of radiation emitters were worthwhile. However, the groups studying the hazards from radiation could only make recommendations for the protection of public health and safety; they had no authority to enforce their decisions. Nevertheless, their work was important and has continued; data assembled by committees of experts in the field of radiation protection currently is forming the basis for various types of health and safety regulations.

It was believed necessary in the United States at the time the Atomic Energy Act of 1946 was drafted to provide certain legal controls over the distribution of radioisotopes, byproducts of uranium reactors. The fact that these isotopes would, in all probability, be available in much larger quantities and at lower costs than heretofore made some type of control all the more necessary.

The type of controls set up and exercised by the United States may not be applicable to radioisotope distribution programs in every country. The kind and extent of programs will vary considerably depending upon such factors as, to name only a few: (1) the legal basis for control of an atomic energy program; (2) the scope of isotope utilization, for example, whether the materials will be limited to medicine and research or whether they will be used industrially as well; and (3) the quantities of materials to be used. The following discussion of the program carried out by the US will serve to outline some of the problems and a method of handling them.

## LEGAL BASIS FOR CONTROL

The basis under which the Atomic Energy Commission controls the distribution of radioisotopes is the Atomic Energy Act. The original Act of 1946 and the 1954 amendment both contain express provisions concerning distribution of "byproduct materials," as the radioisotopes are called. The 1954 Act states that byproduct materials may be furnished " . . . for research or development purposes, for medical therapy, industrial uses, agricultural uses, or such other useful applications as may be developed."[1] At the same time, the Act recognizes the dangers involved in handling radiation emitters, for it also states that the Commission "shall not permit distribution of any byproduct material . . . and shall recall or order the recall of any distributed material from any licensee who is not equipped to observe or fails to observe such safety standards to protect health as may be established by the Commission. . . ."[1]

Thus, a specific act was passed to set up controls over nuclear energy applications and a specific Commission established to carry them out. This might not be necessary in all countries; in fact, some countries have delegated the authority for radiation protection and inspection to existing groups, both within and outside of the Government. During the early days of radiation utilization, much of this inspection was done on a voluntary basis. However, as the amount of radioactive substances has increased in various countries, either through import or through construction and operation of reactors, voluntary radiation inspection is being replaced by regulatory controls.

Although it would be possible to establish voluntary standards, it is unlikely that any enterprise

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involving potentially hazardous materials, such as radiation emitters, can continue to operate successfully on a voluntary basis. In fact, the experience of US Government agencies, such as the Federal Food and Drug Administration and Federal Trade Commission, has indicated that an agency can not expect 100 per cent voluntary compliance with minimum safety standards.

One means of compulsory control is through issuance of regulations which implement basic control laws, and which may be broad or specific as the situation requires. The US AEC has issued certain regulations governing the possession, use, transfer, and disposal of radioisotopes. Other regulations outlining minimum health and safety standards will be published in the future. The currently published regulations outline procedures to be followed when obtaining a license to use the materials; criteria for necessary training and experience before a license can be issued; the type of records to be kept, etc. Although this type of regulation may not be necessary for limited programs, US experience has indicated that it is desirable to publish all criteria of training, experience, equipment, facilities necessary for obtaining radioisotopes. In this way all persons can determine what the requirements are; furthermore, these requirements will be applicable to all.

A very important type of regulation, and at the same time a difficult one to formulate, is a set of health and safety standards. The AEC has been delegated this authority under the Act and is in the process of drafting such standards. In compiling these regulations, great reliance has been placed on the recommendations of the National Committee on Radiation Protection, whose members in turn are associated with the International Commission on Radiological Protection. Thus, advantage is taken of the recommendations of experts in the field from all over the world, who have been concerned with radiation protection standards for many years. The International Commission on Radiological Protection has in general, endeavored to base its recommendations on fundamentals in simple and concise terms, which can be translated into regulatory language.

As in the case of licensing regulations, health and safety standards which are published can serve to indicate to all the standards which must be met. If later experience in the field of radiological safety warrants amending the regulations, this can be done without undue difficulty. It must not be assumed that a given set of standards will exist without modification for a long period of time especially in the rapidly expanding field of radioisotope utilization and protection.

Even though the emphasis has been placed on regulations, it is not intended to discount the real value of recommendations of the National Committee on Radiation Protection and the International Commission on Radiological Protection. As indicated previously, these recommendations form the basis for regulations. Also for those interested in more detailed information as to the basis for the recommendations, it may be found in handbooks prepared by the NCRP and distributed by the National Bureau of Standards. Any country contemplating a set of regulations or codes would find these handbooks useful as starting points in drafting such codes.

## ADMINISTRATIVE CONTROLS

The US AEC established an organizational group within the Commission to handle the radioisotope distribution program. This group, the Isotopes Division, has had several functions: (1) production control—coordination of isotope production to insure that the desired types and quantities of materials are available from the various reactors throughout the country; (2) distribution control—administration of a licensing program, with the establishment of basic criteria of training and experience sufficient to permit wide, yet safe, utilization in scientific research, medicine, industry and other fields; (3) inspection and visitation—evaluation of users' facilities plus consultation on radiological safety; (4) promotion—education in safe radiological practices, assistance in establishment of training courses in radioisotope utilization, and preparation and delivery of articles, talks, etc., on isotope utilization.

A group such as the Isotopes Division may be a more formal organization than all countries would need. The functions of the division might be spread among individual groups. However, in all probability these functions will need to be undertaken in varying degrees in any radioisotope program. In the US, the Isotopes Division has served a useful function as a central group for coordination of licensing and regulatory activities and as a center for spreading of information relative to radioisotope utilization. Such centralization has merit, particularly at the beginning of a program. As the program expands, however, and trained users increase in number the need for continued centralization tends to decrease.

## COOPERATION WITH OTHER GOVERNMENT AGENCIES

Although the Atomic Energy Commission is the principal group controlling radioisotope distribution, other Government agencies are concerned with different phases of the program. For example, transport of radioisotopes across state lines must conform to regulations of the Interstate Commerce Commission or the Postal Department; inclusion of radioisotopes in foods or drugs, even for experimental purposes, comes within the scope of the Federal Food, Drug and Cosmetic Act; a function of the US Public Health Service for a number of years has been radiological health activities. The AEC has cooperated with these other agencies to work out mutually satisfactory arrangements in connection with radioisotope distribution and utilization.

### Transportation Groups

The wide variety of types of radioactive isotopes, from the short-lived, strong gamma-ray emitters to the long-lived weak beta-ray emitters present an equally wide variety of packaging and shipping problems. The unique features of radiation bring about transportation problems not ordinarily encountered; for example, a sturdy, heavy box suitable for ordinary materials may still present radiation hazards when it contains a strong gamma-ray emitter. Ionizing radiation may have an effect on photographic materials and navigation instruments.

A Subcommittee of the National Research Council studied the problem as it affected transportation within the United States. This Subcommittee received comments among others from representatives of railroads, Air Transport Association, Atomic Energy Commission, photographic industry, radium and radon industry and developed certain standards for shipment of radioisotopes. Packaging and shipping regulations were based on three principles: (1) there must be a workable compromise between no shielding and gross overshielding, so that reasonable protection would be afforded to undeveloped films in transit, to employees of the carrier, passengers, and livestock and plants in transit; (2) the magnitude of the hazard from a radioactive parcel depends upon the kind and nature of the radiation emerging from the parcel, not on the name of the radioisotope involved; (3) the amount and chemical nature of the radioactive substance in the parcel is of interest only in case of an accident in which a spill or leakage may occur, resulting in possibility of ingestion, inhalation or absorption of the radioactive material. Thus those radioisotopes which are concentrated in the body, such as bone seekers, would present a greater hazard than those eliminated rapidly.[2]

For purposes of classification for transportation within the United States, packages containing radiation emitters have been placed into three groups: Group I, packages containing radioactive material from which gamma radiation at the surface may exceed 10 milliroentgens per 24 hours; Group II, packages containing neutron-emitting sources; Group III, packages containing radioactive material emitting electrically-charged corpuscular rays only (beta rays or alpha particles) and the surface radiation will not exceed 10 milliroentgens per 24 hours. Most short-lived isotopes and those materials particularly useful in medicine fall into Group I, while many pure beta-ray emitters fall into Group III.

The shipping regulations further state that external shipping containers must be so constructed that the gamma radiation at any point of readily available surface will not exceed 200 milliroentgens per hour. This applies both to rail or air transport. The regulations also specify the type of inner containers for liquid radioactive materials to minimize danger of leakage in case of breakage. Special precautions must be taken in case of transport of physiologically hazardous materials, such as radium, strontium-90, polonium-210, barium-140.

At the time these regulations were adopted they met the needs of isotope users. However, the utilization of these materials now has grown to such a point that there is some question that the regulations are presently too restrictive. This is particularly true for air shipment from a reactor site producing large quantities of isotopes. If it is determined that the regulations present real hardships to manufacturers and suppliers of radioisotopes, the situation can be reexamined and possibly the rules altered.

It is true that some countries transport radioactive isotopes in wing tips of airplanes. This is not done in the US, but experience has shown this to be workable, and no ill effects result either to navigation instruments, or airline personnel. Such a procedure, of course, lessens the weight of container necessary and offers savings in transportation costs. If such arrangements appear feasible in a specific case they should be explored. Nevertheless, the basic problems of health protection and shielding from radiation remain the same. Cooperation between the distributors of the radio-activity and the shippers is essential, and this can best be obtained through mutual education and discussion.

### Public Health Agencies

In the US there are regulations administered by the Food and Drug Administration which are designed to assure the public of pure foods and drugs. As is well known, one of the wide uses of radioisotopes is in the field of medicine and medical research. The AEC, therefore, has cooperated with the Food and Drug Administration, to assist radioisotope users to comply with the regulations of this group. For example, radioisotopes intended for use as "new drugs" (which term applies to medical and physiological research programs) must be labeled in accordance with provisions of the Federal Food, Drug and Cosmetic Act. The means for meeting these labeling requirements have been worked out by a special committee, formed for the purpose, consisting of members of the Food and Drug Administration and the AEC.

Other problems which have been considered by this committee are the possible sale of beef from animals which previously had received external radiation doses and the extent, if any, to which food materials may contain radioisotopes before the materials are considered adulterated. A potentially large scale use of radiation is in the sterilization or pasteurization of foods and the Food and Drug Administration is concerned with this problem, particularly as to whether harmful side effects may take place during irradiation or whether foods are damaged during the process. Such matters will need to be worked out among the groups performing the scientific studies on such processes, the Food and Drug Administration, Public Health Service officials. the Atomic Energy Commission.

Other groups in the US are obligated by law to protect the health of the public. For example, the US Public Health Service has been concerned for a number of years with protection of people from the harmful effects of radiation. Even after the formation of the Atomic Energy Commission, the Public Health Service retained this interest. It has cooperated with the AEC in conducting studies on decontamination of waters contaminated with radioactivity, problems associated with the disposal of radioactive wastes, evaluating hazards associated with the wide variety of radioisotopes available from nuclear reactors, surveying the dangers associated with other types of radiation-emitters, such as X-ray machines and particle accelerators. The Public Health Service is also vitally interested in the development of sound radiological safety programs in the various states in the United States. In this latter regard, it has set up programs under which state and local health officials can acquire basic knowledge of radiation and radiation protection. The AEC cooperates and assists in these programs by providing personnel for lectures and demonstrations.

A public health problem which becomes more important as the amount of radioactivity in use becomes larger is that of waste disposal. In general, there are two methods of controlling wastes: (1) concentration and storage and (2) dilution and dispersal. Disposal may be accomplished as follows: (1) short-lived radioactive liquids may be controlled by storing them until they have decayed to a safe level and/or by dilution with water; (2) long-lived radioactive liquids may be controlled by concentration or evaporation to the solid state after which they may be stored or buried; (3) gaseous and airborne materials may be controlled by filtration and dilution with air; (4) solid radioactive and contaminated materials may be disposed of by burial in the soil or disposal in the ocean under controlled conditions.

The extent to which special facilities need to be set up will depend upon the size of the isotope program. In a medical program, for example, which is primarily a diagnostic one, waste-disposal problems may be insignificant, since the amounts of material used will be very small and usually of short-life. Ordinary waste disposal facilities may be satisfactory in such cases. For a program involving therapy and larger number of patients, special holding tanks for liquid wastes may be necessary.

The waste-disposal problem is such an important one that the National Committee on Radiation Protection has compiled four handbooks on the subject and is currently preparing two additional ones. The matter of disposal of radioactive wastes should be carefully considered by any group initiating an isotopes program.

As in other instances discussed throughout this paper, exactly similar problems probably will not be faced by all countries engaging in radioisotope programs. However, many countries have found it desirable, at least in the beginning to delegate authority for review and/or control of radioisotope programs to public health and sanitation groups. These groups doubtless would face general problems similar to those under review by the AEC and the Food and Drug Administration, especially if the isotope programs are devoted primarily to the field of medicine. The US has found it practical to work out such problems through a committee organization; other countries may or may not wish to adopt the same procedure. Scientific personnel trained in radiation problems could assist Public Health groups or Public Health officials could receive training themselves; either procedure would appear to be sound practice regardless of the size of a radioisotope program.

## TRAINING AND EDUCATION

Before any radioisotope program can expand, it is essential that there be at least a small group of well-trained persons available. Such a group can serve as a nucleus for spreading basic knowledge of isotope techniques, thus permitting an increasing expansion in utilization.

When the program for distributing reactor-produced radioisotopes began in the United States, there were relatively few people in the country who had experience in handling radiation emitters. One of the important tasks of the AEC was to encourage the establishment of training courses to increase the number of persons qualified to use isotopes.

There was set up in Oak Ridge, Tennessee, a special training course specifically devoted to radioisotope techniques. It is a laboratory lecture course designed to cover fundamental concepts and techniques involved in handling radioactive materials. Completion of this course or others like it would generally qualify a person to handle tracer quantities of isotopes for research and development activities. Such a course does not, nor is it intended to, qualify someone for a particular, specialized use of isotopes. Rather it is just the first step in acquainting people with the problems involved in handling radioisotopes, and therein lies its value.

A next step in training for particular uses of isotopes usually involves more specialized training, obtained through "on-the-job" experience with a group using isotopes in the particular manner of interest. Little, if any, formalized training courses are available for specialization, although some medical courses, for example, are being started in some locations. More and more basic training courses may well become part of established curricula in colleges and universities as interest in isotopes continues to grow.

The need for education and training has been recognized everywhere that isotopes are used. Some countries aside from the US, with advanced nuclear energy programs, have set up similar training courses for their own nationals and for nationals of other countries as well. For those countries just beginning

to enter into the field, it has appeared desirable to send a few people to the US, or to other places to receive the basic training courses, plus perhaps additional specialized training. These people in turn have been able in some instances to establish in their own countries periodic training programs for teaching isotope techniques on a regional basis. This is a highly worthwhile procedure and is to be encouraged wherever possible.

Although actual training is the most desirable method of learning isotope techniques, procurement of technical information also plays an important role. For example, information on radiation protection, monitoring and measuring equipment, laboratory design, laboratory equipment for handling various levels of radioactivity, handling of wastes and their disposal are all important to people beginning isotopes programs. Isotope bibliographies also may be of assistance to persons initiating plans for utilizing isotopes, as well as to those who already have programs underway. The AEC has in the past made available technical information in the nuclear energy field, including isotope utilization, and it is continuing this program.

The problems associated with training and dissemination of information are ones which any country using, or planning to use, radioactive materials should deal with from the very beginning. The manner in which these problems are faced will, to a large extent, determine the speed and scope of growth of the entire program.

## CONCLUSION

Because of the fact that, until recently, the sources of supply of the main quantities of radioisotopes have been Government-owned, the radioisotope distribution program in the US has been a Government monopoly. This will not necessarily continue when privately-owned reactors get into operation. However, there has been a background of several years' experience in the administration of the radioisotope distribution program, so that certain controls can be relaxed.

In all probability, any country just entering into a distribution program would find it desirable to establish rather strict rules regarding utilization. This was true in the US in 1946; for example, authorizations (or licenses) were issued for periods not exceeding six months for a single isotope for a single purpose. Now, on the other hand, a variety of licenses can be issued with varying degrees of breadth of scope, depending upon the particular situation and for so long as two years' duration. Other countries may find it practical to relax certain rigid controls as the program becomes more firmly established.

Any moderate, or large scale, isotope utilization program will doubtless have to deal with insurance problems and workmen's compensation problems. However, these are problems which are encountered in other forms of business enterprises and, hence, have not been discussed in this paper.

This paper has endeavored to point out certain of the administrative and legal problems which arise in a radioisotope distribution program. Although the US experience was used as an example, exactly the same problems may or may not arise in every case and, of course, they may not be handled in the same fashion. Nevertheless, many of the problems discussed are basic in nature and must be solved to some degree depending upon individual situations.

## REFERENCES

1. Atomic Energy Act of 1954, Chapter 8, Section 81.
2. Nuclear Science Series, Preliminary Report No. 11, *Physical, Biological, Administrative Problems Associated with the Transportation of Radioactive Substances.* National Research Council (Washington, D. C.).

# Administration Problems in Radiation Protection in New York State

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The development of control techniques for chemical, physical or infectious hazards in industry follows certain fundamental medical and engineering principles which are modified in accord with the particular properties of the agent. The control of the hazard from ionizing radiation is no exception. For several years, various agencies in New York State have worked with radiation control and while no definitive program has been established, progress has been made in defining the special technical and administrative factors. The purpose of this paper is two-fold: (1) to develop the basis for the decisions which are currently being taken and (2) to describe the administrative problems. Because of New York State's size, its industrial development and its well organized health and safety agencies, most of the factors which have arisen can be considered representative of other jurisdictions.

The following principles have guided us in formulating our administrative controls:

## CONTROL MUST BE FLEXIBLE

The use of radiation is growing and its boundaries have as yet not been defined. Preventive methods must be elastic and allow for expansion both in number and in type of all kinds of encapsulated or loose radioactive materials and radiation equipment and machines. To date, the principal uses and hazards are still in the medical and allied professions (physicians, dentists, veterinarians, etc.) but the industrial applications are extremely widespread and expanding. Illustrative of this growth are the number and type of radiation sources in New York State where there are more than 1500 industrial establishments with a potential radiation hazard.

There are about 750 shoe fluoroscopes, some 125 industrial radiographic installations and at least 25 X-ray diffraction units. We have on record 400 radium-bearing static eliminators installed in various plants. The polonium-type static eliminator is also widely used, but the number manufactured and distributed is not available to us. The number of plants using cobalt-60 or sealed radium for radiography is unknown, but a minimum estimate would be 125. In many installations, the original sources are being replaced by those of larger size. Miscellaneous ionizing emissions from high voltage instruments such as electron microscopes and accelerators total at least 15. There are two radium-processing plants in the state and 20 more are associated with dial painting but no definite figures can be established as to the intermediate and ultimate users of these radium dials. The hazard from the use of isotopes in industry such as beta-ray thickness gauges or as tracers in processing, research, etc., are estimated in the several hundreds. There are still apparatuses which may produce stray radiation but these have not yet been adequately defined, such as theatre or projection TV tubes. This enumeration excludes the potential hazard from power reactors and the associated operations of such installations.

Such variations in energy sources, sizes, and establishments demands code rules which are flexible and responsive to change. The concept of flexibility is best expressed by placing the emphasis on "performance" rather than on "specification" in the codes controlling the radiation hazard. Since radiation is easily detected and measured if the potential sources are known, specific control techniques need not be itemized as they can be individually tailored and applied. It is recognized that a "performance" code is more difficult to enforce, but the chief aim at this time is education, not enforcement.

## ALL IONIZING RADIATION IS POTENTIALLY HARMFUL

While certain maximum allowable standards are needed, no amount of radiation should be permitted if methods are available to eliminate it. The standards as determined by the National Committee on Radiation Protection and other scientific bodies suffice as bench marks for engineering control. Generally, the most restrictive criteria should be used provided it is possible to achieve them from an engineering and economic standpoint. This principle seems extreme if taken literally, as everybody cannot be fully protected. We cannot create an absolutely safe environment any more than we can a biologically sterile one. Radiation, however, is peculiar in that there is a cumulative effect. Chronic radiation is no different than acute radiation except that the end point is delayed. There is increasing

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evidence that each ray produces some damage and that while the effect may not be immediately detectable or perhaps even meaningful in an average lifetime, the cumulative effect contributes to a number of adverse biological changes. Blood disturbances, carcinogenesis, cataract formation, genetic effects and degenerative changes associated with aging are some of the known sequalae.

Maximum permissible levels should not be considered final or rigid and provision for the accumulation of data to allow for re-evaluation must be included in any control program. Proper records and provisions for periodic review are essential. The accumulation of lifetime data on exposed individuals will be meaningful only if all possible radioactive sources of exposure are recorded. The daily bombardment of cosmic rays is uniform at various sea levels and the trace amounts from radioactive materials in air, food and water can be approximated. The amount of fall-out is apparently very small but has to be taken into account. The greatest variation at this time, however, occurs from individual exposure to diagnostic and therapeutic X-ray and radioactive material willfully applied to the human being. The amount of damage produced in this way cannot be assessed accurately but the high incidence of leukemia and skin lesions in radiologists are evidence of its occurrence.

Industrial injury from ionizing radiation also does not permit accurate evaluation. This is more substantial than is apparent from a review of the number of workmen's compensation cases due to ionizing radiation during the years 1945 to 1952. In New York State during this period there were only 6 closed cases of accidental injury or disease due to radiation but it is reasonable to assume that more injuries occurred in this period which were not reported. Because of administrative regulations, this low incidence of industrial occurrence does not reflect the hazard, the effectiveness of control methods or the number of people involved in the use of ionizing radiation. The tragedies of the radium dial painters of the 1920's must not be repeated to convince us of the danger of the unrestricted use of these materials.

## RADIATION DAMAGE IS PREVENTABLE

One of the brightest chapters in the history of preventive medicine is the prevention of damage from stray ionizing radiation. This has been magnificently demonstrated in the government installations concerned with atomic energy where workers have been and are far safer than in the average radiologist's office. Radiation is easily detected and measured by appropriate instruments and suitable techniques such as distance and shielding with lead, concrete, water and other materials are available to control any hazard.

The problem is primarily one of finding the potential hazard; that is, of knowing where radiation is being used. Man himself cannot be relied upon to act as an indicator of radiation. Its presence is insensible and the clinical effects too slow and too vague in the preventable stages to be recognized. It is, therefore, imperative that each regulating agency be appraised of its use. If it is reported, appropriate measurements may be taken and control techniques enforced. There is more instrumentation and technical knowledge available for the control of ionizing emissions than for any other physical, chemical or infectious agent.

## RADIATION CONTROL IS A FUNCTION OF INDUSTRIAL HYGIENE

With the development of atomic energy, a new profession, health physics, appeared. This was natural in the atomic installations where intimate understanding of radiation physics was necessary so that potential biologic effects could be evaluated and protective methods devised. It is logical that the chief safety officer, then and now, in such an establishment would be an individual trained primarily in radiation physics. The problem is quite different in industry where there is widespread use of relatively small amounts of radiation. In a shoe store, in a printing plant or a foundry, the problem of radiation is a minute one compared to the over-all health and safety hazards which exist and there is no need for a specialist such as a health physicist.

Industrial hygiene, the professional field concerned with the control of our industrial health hazards, considers radiation as another type of hazard. In some ways it is easier to handle than others, since instrumentation and engineering methods are so highly advanced. Industrial hygienists moreover are experienced in the attitudes, organization, operational problems, finances and goals of industry and have in the past successfully enlarged their scope of interest to include all hazards as they appeared. In some industries, industrial hygienists have special qualifications; for example, a chemical engineering background is most useful in the petroleum industry and in large atomic energy plants, the safety director may be trained primarily in health physics. Widespread utilization of this wonderful new tool of modern technology will be delayed until many people become familiar with its presence. At this time there is need for people who are moderately acquainted with radiation, but able to recognize its potentialities for harm and understanding enough to call upon the health physicist when a problem arises which is beyond their technical capacity.

## EXISTING GOVERNMENT AGENCIES ARE CAPABLE OF CONTROLLING RADIATION

Health physicists and other radiation specialists suggest that a separate department be established in each state to cover all aspects of radiation safety in industry, hospitals, air and water supply, power reactors, etc. This is contrary to our present set-up

where control of various hazards is allocated to the different state agencies depending upon their jurisdiction, staff and function.

There is no sound reason for considering ionizing radiation as particularly different. A separate agency would create duplication, confusion and loss of time. It is the common complaint of industry that there is an endless parade of governmental inspectors representing different and even the same state agency, each interested in a minute aspect of their work. Dealing with each agency is expensive, time consuming and tedious. Safety inspectors looking for only radiation hazards would add to this burden without increasing efficiency. A commission of experts in an advisory or consultative capacity may serve a useful function but the prime responsibility should be given to the functioning state agencies. Peace-time progress can be achieved only as the general public becomes familiar with radiation. This maxim also applies to the various branches of the state government which have to learn about radiation and incorporate its handling into their present functions. A specialized training program for inspection personnel in the regulatory agencies will dispel many of the misconceptions concerning radiation hazards. Furthermore, they offer a realistic means for effective control methods.

The administrative problems encountered by the various state agencies concerned with radiation safety are currently being evaluated and discussed. There is expectation that there will be more extensive use of atomic energy as a result of technological advances and the stimulus provided by the US Atomic Energy Act of 1954. Safety regulations must be adopted by the states to achieve the high standards set by the Federal atomic establishments. Radiation safety is expensive and if it is not subsidized by government, we can expect industrial managements to use all devices that save money. Rules, therefore, must be promulgated so that the ignorant or unscrupulous do not benefit at the expense of the public and the worker. Education should be the main approach, but some regulatory discipline is also necessary.

In New York State, the major burden for radiation safety falls upon the Health and Labor Departments and both have taken steps toward the adoption of safety regulations. Regulation 1G of Chapter 16 of the New York State Sanitary Code concerns itself entirely with radiation. This code was scheduled to go into effect July 1, 1955 but its effective date has been indefinitely postponed. Public hearings have been held on radiation code rule No. 38 of the Board of Standards and Appeals of the New York State Labor Department and modifications are currently being made. No date has as yet been set for further hearings but it is expected that the code will be adopted some time during 1955.

It is worth pointing out that there is a basic difference in the approach between the two Departments. The Labor Department is more of a regulatory agency than the Health Department. In this instance, however, the Labor Department recognizes that in the case of radiation, education is more important than specific control measures. Its code also stresses "performance", i.e., the attainment of established criteria rather than rigid design techniques for each individual radiation source or for each industrial use. There is no basic difference between the two codes, except for the definition of jurisdiction currently being negotiated. Decisions affecting these matters will depend upon legal interpretation as well as on the ability of each Department to carry out the work in terms of time, staff, equipment and geography.

Jurisdiction and responsibility of each department are legally established. The Labor Department is responsible for all harmful exposures to factory workers. All environmental radiological problems and all willful exposure of humans to radiation are the prime concern of the Health Department. There are, however, a number of "gray" areas in which both departments have a common interest which are worth describing:

### I. Veterinarian Clinics

Veterinarian clinics may be considered allied to the medical profession but the workers and technicians are employees and should be afforded protection under the Labor Law. The extent of this protection has not been clarified in occupations of this kind. The Labor Department is not concerned with the self-employed professional such as the veterinarians themselves.

### II. Educational Institutions

Educational institutions hire "mechanics", working men" and "laborers" who are defined as covered employees by the Labor Law. On the other hand, educational institutions ordinarily do not come under the "places" covered by this law. In an engineering institution, the nature of the problems are such that the experiences of the Labor Department could be more effectively applied than those of the Health Department. The converse may be true in an institution which carries out medical studies.

### III. Research Laboratories

Research laboratories are either private, commercial or attached to a large industry. Jurisdiction by either Department would have to be evaluated on the basis of the type of work and the ownership.

### IV. Transportation

Employees engaged in the transportation industry are normally not covered by the health and safety provisions of either the Labor Law or Public Health Law and both departments might have a legitimate interest in various phases of this activity. Jurisdiction might well belong to the Division of Safety of the executive branch or the Interstate Commerce Commission if the carrier operates in more than one state.

### V. Shoe Stores

Shoe stores are mercantile establishments with employees, but the radiation hazard is primarily to the public. Hence, both departments have a legitimate interest in this operation and both have cooperated in a state-wide survey and control program which is currently being completed.

### VI. Waste Disposal

Pollution of air and water by radioactive wastes is a responsibility of the Health Department. The most effective means of control is at the source and it would seem logical that the Labor Department could carry out this function best. The Health Department has responsibility for non-radioactive air pollutants but the Labor Department has lent its technical support to the establishment of controls in this important area.

### VII. Power Reactors

The issue of jurisdiction over nuclear reactors which produce electrical energy is somewhat ambiguous. Ordinarily, the Public Service Commission is in charge of all service matters relating to the production of power. Discussions about this phase of power reactors are still in the preliminary stages.

These indeterminate areas illustrate the problems of field service and final determination of responsibility may well be based upon the personnel and equipment available. Both the Health and Labor Departments have health physicists on their staffs who can act as experts and consultants to the field force. Health Departments are administered by local units whereas the Labor Department has a corps of inspectors operating under central direction. Although the training and education of many of the Labor Department inspectors are not considered adequate for more than the most routine kind of technical problems, under competent direction they can serve for case finding, routine inspections and compliance. The Health Department has a number of trained sanitary engineers in local units but these are few in number and not sufficient for total coverage. All these people in both departments are capable of contributing to the effective administration of radiation controls. Which approach will prove to be the most effective still remains to be determined in the future.

It is anticipated that there will be no serious conflict with any regulations of the Atomic Energy Commission which may be adopted in the future, provided the states carry out their responsibility for health and safety. New York State, through its Health and Labor Departments, is doing so by adopting specific codes for radiation control. A code rule is only as good as it is enforceable and many details of the present Labor Department's code are still being examined for their workability. The areas which need the greatest amount of consideration because of divergent viewpoints or lack of experience are:

### I. Registration

Knowing where radiation is being used is considered the greatest single instrument toward effective control; hence, registration of those employing radiation is absolutely essential. Registration must be distinguished from licensing in that it entails only informing the regulating agency of the use of radiation sources or equipment or of significant changes in amount or process. Data on the number of people exposed, the approximate degree of hazard by stating the type and energy of radioactive material, should be sufficient initially.

### II. Standards

A maximum permissible limit must be set. This exposure limit in past years has been consistently revised downward on the advice of the radiation experts. Adoption of any criteria by a state agency must have scientific validation and it has been suggested that the standards of a committee of experts such as the National Committee on Radiation Protection be made mandatory. No state agency, however, willingly relinquishes its autonomous status and therefore, qualified staff experts in each agency are necessary. Advisory Committees may be appointed or a group such as the National Committee for Radiation Protection may be designated as the advisory body.

The need for competent and qualified experts in each regulatory agency goes beyond the setting of standards. The agency must be in a position to verify and weigh the opinions of qualified experts who appear before it. It must determine the instrumentation used and must classify the hazard from each of the radioactive sources. This classification of source, according to degree of hazards, governs the number and type of preventive measures and follow-up visits. This is an important function for which the agency must accept responsibility.

### III. Records

Many types of records are needed for administrative purposes. Registration forms, reports of environmental conditions, medical records on individuals, etc., are some of the records which provide the administrative tools for control and regulation. The handling of these records requires both intelligence and experience. Radiation control is a living and hence a changing problem, and revision will be necessary as experience and time dictate.

### IV. Education

The worker must be informed where radiation is used, particularly where distance is one of the control factors. Management is reluctant to inform workers of the potential hazards of their job, particularly when under ordinary conditions control methods are efficient and no hazard exists. Unjustified complaints and perhaps even claims for damages are occasionally made by workers in such circumstances. Nevertheless, it is extremely im-

portant that all be informed by proper means; labels, signals, etc., where radiation is present. Since radiation can be measured and controlled, no damage to humans is anticipated and unjustified complaints can be denied through scientific evidence.

There are many other administrative details which have not been satisfactorily defined. Experience, however, will be the only teacher and in a year following the adoption of a code, definitive answers should be available for many of these questions.

# Regulations Applicable to the Use of Radioisotopes in France

**By L. Bugnard* and J. Vergne,† France**

From the moment of the discovery of the artificial radioactive isotopes by F. and I. Joliot-Curie, and following the work of Hevesy and Urey, the use of these nuclides for research work began in France. It developed a great deal, beginning in 1947, when it became possible to obtain, at reasonable prices, and practically without any limitations as to quantity and quality, certain radioisotopes produced in the United States, in Great Britain, or by the C.E.A., either in nuclear reactors or with large particle accelerators.

Progress, in this field, was conditioned by:

1. The steady increase in the number of competent workers capable of making proper use of the new potentialities in research laboratories, hospitals, and industry. Theoretical courses were developed, in the relevant Departments of the Universities, and this was helped, from this year on, by the creation of a third cycle of Higher Education. Specialized practical courses, given twice a year, have been organized since 1950, for the training of radioactivity technicians, through the Commissariat à l'Energie Atomique, the Centre National de la Recherche Scientifique, and the Institut National d'Hygiène.
2. National control of the distribution and uses of radioisotopes.

Along with the development of scientific and technical progress, the use of radioisotopes became ever more widespread, and laws and various regulations came into being one by one, to lay down the rules applicable to the import, export, transfer, transport and utilization of the radioisotopes.

## A. PREPARATION, IMPORT, EXPORT, POSSESSION, SALE, DISTRIBUTION AND USE OF THE ARTIFICIAL RADIOELEMENTS

As early as 1934 (decree of 12-5-1934 and regulations of 12-26-1934, Official Gazette of 1-1-1935) the dangers offered by the radioactive substances, as well as the precautions to be taken in order to avoid them, were brought to the knowledge of the personnel working in all the installations where these substances are being prepared and used. The regulations underscored the precautions to be taken according to the various circumstances encountered in practical use, in order to establish protection against the noxious effects of radioactive products; they also provided for measures of protection and organization, as well as for medical checkups on the personnel. However, it was only when the manufacture of artificial radioelements in large amounts and at reduced prices became significant, that the regulations could become more specific.

On September 13, 1947 (Official Gazette 10–4–47) a Committee presided over by the Minister of Public Health and Population was set up, to make a study of the therapeutic and biological applications of the radioisotopes.

In addition, due to the dangers which they may offer in the fields of human and veterinary medicine, the artificial radioelements and the preparations of all kinds made radioactive by the incorporation of radioelements, or by any and all other methods were added to the list of poisonous substances, for which legal provisions are set up in the decree of November 19, 1948.

Parallel to this, a decree of July 30, 1949 set up an interministerial commission for the import of artificial radioelements from abroad. In order to check on the reality of the guarantees required by the Government of the exporting state, to ensure the best possible utilization of the artificial radioelements, to permit the control of their use, and to ensure publication of the results obtained with them, all applications for the allocation or purchase of artificial radioelements were made subject, by this decree, to the approval, either of the Minister of Public Health, if they were destined for research in the field of human biology or therapeutic applications, or of the Minister of National Education, if they were meant for research falling within the fields of physics, chemistry, animal or vegetal biology. Each Minister worked with the help of a specialized committee, the members of which had been designated by a decision of December 6, 1949. The composition of the committee assigned to the Minister of National Education was modified by a decision of June 28, 1952.

Representatives of the Commissariat à l'Energie Atomique sat on these two committees, presided over, respectively, by Mr. Aujaleu (Public Health) and Mr. Dupouy (National Education).

However, provisions which were more explicit still were later spelled out by the law of July 19, 1952 (embodied in the Pharmaceutical Code by the

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Original language: French.

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† Head of the Legal Department of the C.E.A.

decree of August 25, 1952, then in the Public Health Code by the decree of October 5, 1953, chapter 2, title 3, book 5, articles 631 *et al.*).

This law, as well as the decree of May 3, 1954, which defined the conditions of its application, constitutes, at the present time, the main basis of the regulations applicable to the artificial radioelements in France. These texts, which repeal the decree of July 30, 1949, set up regulations applicable to the preparation, import, export, utilization, sale, possession and distribution of artificial radioelements. They also set up an interministerial committee in charge of submitting opinions on the matters relating to these radioelements.

This Committee consists of: a representative of each one of the six ministries interested; a representative of the Institute National d'Hygiène; a representative of the Centre National de la Recherche Scientifique; and two representatives of the Commissariat à l'Energie Atomique.

This Committee is presided over by Mr. de Lavit, Conseiller d'Etat; the vice president is Mr. Goldschmidt, C.E.A. Director of Foreign Affairs, and a Department head of the C.E.A., Mr. Fisher, who has been appointed its permanent secretary, as a nonvoting member.

The Interministerial Commission, at its plenary sessions, formulates its opinions or proposals pertaining to all questions of a general order having to do with the preparation or application of regulations applicable to the artificial radioelements, and particularly: the preparation, import and processing of artificial radioelements, in any and all forms; the general conditions applicable to the calibration, possession, transportation, sale, distribution of, and trade in, these products; the general conditions of utilization of the artificial radioelements, and the measures of protection against the effects of their radiation; the general rules according to which advertisements can be done, it being specified that any and all publicity bearing on the use of artificial radioelements or products which contain them in human or veterinary medicine is prohibited, except as meant for physicians, veterinarians or pharmacists, with due regard for the provisions in force.

This Committee is made up of two sections which must give an opinion on the preparation, import, export, transfer and conditions of utilization specifically applicable to the artificial radioelements; the first section reviews the applications bearing on human biology or therapeutic applications, the second those bearing on the other applications.

Accordingly, the preparation, import and export by any physical person or legal entity other than the C.E.A., and the transfer by anybody of artificial radioelements or products which contain them, for applications in human biology or therapeutics, are subject to the previous authorization of the Minister of Public Health and Population, after hearing the first section.

For all applications other than those described above, the preparation, import, and export of the artificial radioelements or products which contain them by any physical person or legal entity other than the C.E.A. are subject to the authorization of the Minister of National Education when the installation which prepares or uses them is placed under his jurisdiction; in all other cases, this authorization is given by the Prime Minister.

The decisions pertaining to authorizations are taken after hearing the second section of the commission.

The C.E.A. may transfer, for purposes other than those bearing on human biology or on therapeutic applications, the artificial radioelements which it holds, following a favorable opinion by the second section of the Committee.

Transfer by all other holders will be authorized by the Prime Minister or by the National Education Minister, as the case may be, following an opinion of the second section of the Committee.

In each and every case, whether the artificial radioelements are destined for human biology and for therapeutic applications or not, the specific conditions under which they must be used have to conform with the general regulations pertaining to artificial radioelements; but, in the case of inadequacies or insufficiency in those regulations, these conditions of utilization are set at the time of each individual authorization by the authority having jurisdiction to authorize the transfer, after hearing the first or the second section of the Committee, according to the case.

In order to make the procedure simpler in the numerous cases in which there is no particular difficulty, the Interministerial Committee and its sections may delegate their authority to two of their members, who will jointly examine the ordinary requests.

The authorizations are personal, but the holders may be allowed to transfer the products which they hold to other users. Penal sanctions are provided (fines and imprisonment) for all the infractions bearing on the preparation, import, export and utilization of the artificial radioelements. In addition, the beneficiaries of individual authorizations are subject to permanent control bearing on the processing, keeping and transporting of the artificial radioelements, as well as on the supervision of their use and verification of current inventories.

Finally, the adding of artificial radioelements, or products containing them, to foodstuffs, articles of hygiene and the so-called beauty products, is prohibited.

### B. TRANSPORTATION OF ARTIFICIAL RADIOELEMENTS

The regulations applicable to the transportation of artificial radioelements are in constant development, and various projects are under way in order

to harmonize the present provisions with international recommendations.

At the present time, we must distinguish between:

### I. The Provisions Having to Do with Transportation

#### a. Railroad and Road Transportation, River Transportation

The radioactive substances belong to group 41-420 of the fourth category of toxic materials, which are among the dangerous and foul materials. Transportation of such materials is regulated by decisions taken by the Minister of Public Works, Transport and Tourism, after hearing the Committee on Transportation of Dangerous and Foul Materials, created by the decree of February 27, 1941.

A decision of April 15, 1945 approved the regulation applicable to transportation of the dangerous and foul materials by railroad, road and river. This text was later modified by a certain number of decisions which appeared in the official gazette.

The packages containing radioactive substances must include lead shielding and a double wall for the purpose of creating an insulated area around the dangerous radiation zone.

The container for the radium or radioactive product shall be kept in lead shielding, the thickness of which shall be no less than: 10 millimeters when there are no more than 100 milligrams of radium or a quantity of another radioelement of equivalent activity; 20 millimeters when there are between 100 and 300 milligrams of radium; 30 millimeters, when there are between 300 milligrams and 1 gm of radium; and 50 millimeters when there is over 1 gm and up to 3 gm of radium.

This lead shielding must be placed inside a double walled wooden case, so built that the two walls are assembled. Both must be of sufficient thickness for the unit to be perfectly resistant, the thickness of the walls cannot be under 10 millimeters, and the space between them is to be filled with some suitable material (sawdust, excelsior), the thickness of this filling being at least: 10 centimeters when there are no more than 100 milligrams of radium; 20 centimeters when there are from 100 to 300 milligrams of radium; 25 centimeters when there are 300 milligrams to 1 gm of radium; and 35 centimeters when there is more than 1 gm of radium.‡

The parcels which contain 3 gm of radium, or more of a quantity of another radioelement having an activity equivalent to that of 3 gm of radium, are acceptable only if the shipping manifest is accompanied with an authorization delivered by the Minister after hearing the opinion of the Committee on the Transportation of Dangerous and Foul Materials. This authorization is to specify the packaging requirements.

‡ In the case of cubic shaped parcels, the above rules mean outside dimensions of at least: 26 cm for less than 100 milligrams; 48 cm between 100 and 300 milligrams; 60 cm between 300 milligrams and 1 gm; and 84 cm for more than 1 gm.

The parcels which contain artificial radioelements must carry, in very plain sight, some red labels with a death's head on a white background, with the inscription: "Radioactive material, do not stand near this parcel".

#### b. Transportation by the Post Office Administration

According to decree No. 2337 of August 27, 1943 and the current regulations, the maximum quantity of radioactive materials which can be shipped, with a statement of its value, is 100 milligrams of radium or 100 millicuries of another radioelement.

The source must be housed in a lead shield having a minimum thickness of 2 cm, placed at the center of a cubic parcel having sides 30 cm long. A white label on a blue background showing "radioactive materials" must be affixed to the case.

#### c. Airplane Transportation

There are no special regulations for transportation by airplane, but the international rules (essentially

**Table I**

| *Ailments caused by X-rays, natural or artificial radioactive substances, or any other corpuscle emitters* | *Duration of Compensation* |
|---|---|
| Serious progressive anemia of the hypoplastic or aplastic type | 3 years |
| Slight progressive anemia of the hypoplastic or aplastic type | 1 year |
| Leukopenia attended by neutropenia | 1 year |
| Leukoses or leukemoid conditions | 3 years |
| Hemorrhagic syndrome | 1 year |
| Blepharitis or conjunctivitis | 7 days |
| Keratitis | 1 year |
| Cataract | 5 years |
| Acute or chronic lesions of the skin and mucosae | 10 years |
| Radionecrosis of the bone | 5 years |
| Bone sarcoma | 15 years |
| Broncho-pulmonary cancer due to inhalation | 10 years |

*Work which may cause the ailments*

All work exposing the subject to the action of X-rays, natural or artificial radioactive substances, or to any other corpuscle emitters, in particular:

Extraction and processing of radioactive materials

Processing of radioactive substances

Preparation of radioactive chemicals and pharmaceuticals

Preparation and application of radium-bearing luminescent products

Research or measurements on radioactive substances and X-rays in the laboratories—manufacture of equipment for radium therapy and X-ray equipment

Work which exposes the workers to radiation in the hospitals, clinics, dispensaries, medical offices, dental and radiological clinics, private hospitals and anticancer centers. Work in all the industries or businesses which use X-rays, radioactive substances, and/or substances or devices which emit the radiations listed above

those of the Interstate Commerce Commission) are customarily followed.

1. The maximum quantity transported is 2000 millicuries of radium, or its equivalent of another radioelement.

2. The intensity of the doses of gamma rays at one meter of the parcel must not exceed 10 milliroentgens per hour, or a neutron flux having an equivalent biological effect.

3. Intensity of the radiation must be less than 200 milliroentgens per hour at the surface of the parcel.

4. The inside package is a metal container, lead sealed; for liquid substances, a water and gas tight container made of thick glass must be used and a material that can absorb everything in the event of a leak must be placed inside the shielding.

5. The stiff and resistant outer parcel must be at least four inches along its smallest dimension.

6. A label "Radioactive material" must be showing on the box.

### II. Special Rules Applicable to the Customs Duties and Administrative Formalities for the Import of Artificial Radioelements

1. The National Center of Scientific Research obtained, in March 1949, of the General Direction of the Customs and Excise Taxes, permission to withdraw shipments of scientific equipment and chemical products, provisionally without any import license, provided this document be produced within two months of taking it out of the customs. This method also was valid for the isotopes.

2. By a decision of the Minister of Finance and Economical Affairs, taken on May 23, 1951, the import customs duties were waived for natural and artificial radioelements appearing under numbers 360A and 360 B of the customs schedules.

3. In addition, in a notice to the importers of products originating in and coming from countries which are members of the European Economic Cooperation Organization, dated January 7, 1955, all quantitative restrictions were waived for the import of the products designated in a table appended to the present paper, in which the artificial radioelements under 360 B, the origin of which is from the countries of the O.E.C. or their overseas territories.

### III. Occupational Diseases

Since the issuance of the decree of December 31, 1946, modified on December 9, 1950, the affections brought about by X-rays, naturally radioactive substances or artificially radioactive substances, or any other corpuscle emitters, are classified among occupational diseases.

Table I sums up the present regulations.

# Insurance for Reactors

**By C. R. Williams,* USA**

Since the announcement of the self-sustaining nuclear chain reaction at the University of Chicago, December 2, 1942, it has been obvious that a source of tremendous amounts of energy had been made available to mankind. Developments in the field of useful, beneficial applications of this kind of energy have been carried on under government control in several countries.

There are, however, many potential hazards to both people and property in this program. They include not only normal industrial accident and occupational disease exposures, but added problems related to radioactivity. Indemnification for injury or death of those employed in the operations; for damage to equipment and property directly involved; for injury or death to persons not connected with the projects and for damage to property outside the operations are problems whether the nuclear energy program is operated by governments or by private corporations.

In the case of government operations, such indemnification, whether for damaged plant facilities or injury of employees or the public, can be made from available public funds. Private industry, on the other hand, must either be prepared to pay such losses from its own resources or to purchase insurance protection.

In the United States, the Atomic Energy Act of 1946 created the Atomic Energy Commission and vested in this Commission full responsibility for all developments in the nuclear energy field. By its control over fissionable material, the Commission exercised control over all nuclear developments in the United States. Private industry's participation in this program was only through contracts with the Atomic Energy Commission.

During this period, the Federal Government assumed responsibility for all liability for injury or damage resulting from any operations within the scope of the program.

Under this arrangement an excellent safety record has been compiled, particularly in the reactor program. It has been the result of giving safety high priority in both design and operation of reactors.

The Atomic Energy Act of 1954 has opened to American private enterprise the opportunity to obtain fuels for nuclear reactors to be used for research, testing or power production, under a lease arrangement. This program is to be carried on under certain Government controls exercised through licensing. At the same time full responsibility for all liability for damage resulting from reactor operations must be assumed by private industry. This is made eminently clear in Chapter 6, Section 53 of the Atomic Energy Act of 1954: "(8) The licensee will hold the United States and the Commission harmless from any damages resulting from the use or possession of special nuclear materials by the licensee."

This clause has led industry to examine its potential liabilities and to assess the kinds of insurance protection which would be needed. Since reactor failure might, under certain circumstances, cause severe losses, particularly in areas surrounding the reactor plant, many industrial groups interested in participating in the reactor program have felt that their potential liability may in some cases exceed their resources. Thus, when such liabilities are removed from governments and assumed by private industry, a new problem—that of financial protection—arises.

As a result of this, segments of American industry began to explore the possibilities of obtaining from insurance companies the financial protection against liability, which it considered necessary.

This search revealed a reticence on the part of insurance carriers to become involved in risks of such magnitude. This reticence was based on the fact that the liability limits requested were much higher than the industry had ever before been asked to provide; that legal limitations imposed on insurance carriers relative to liability limits on a single risk precluded certain coverages for such high limits; that the insurance industry had little knowledge of what it was being asked to cover; that the entire private reactor program was, in fact, experimental; that no assurance had been provided as to the development of standards of construction and operation of reactors; that no specific approval and inspection program had been set up. Considerable progress is being made to overcome many of these objections.

While the insurance industry appreciates industry's reluctance to risk its entire corporate resources on a single project it is no more anxious to risk its very existence on a single experimental industrial venture. All concerned realize and accept the possibility, however remote, that extensive losses could be caused by failure of the reactor unit of a nuclear

* Liberty Mutual Insurance Company, Boston, Massachusetts.

power plant, for example. The degree of hazard is related to reactor type, design and construction; the location of the reactor in relation to people and property; and the caliber and attitude of both management and operating personnel.

During normal operation of a reactor there are few problems that seriously bother informed insurance underwriters. The impasse is reached in considering a major reactor failure and its effects not only on the reactor and operating personnel but also on areas surrounding the plant. Such an accident may impose on the management of the company operating the plant a liability of extensive proportions. The serious problems arise from possible large and unpredictable losses under the general heading of public liability. A serious accident could involve injury and death to people living and working in the area around the reactor operation as well as contamination of structures in the vicinity. Industry appears to feel that such losses could be extremely large. Insurance against this type of loss is provided under policies which specify limits of liability which the insurer is willing to accept. These limits are related to many factors, but in any event are, at the present time, considerably less than those which industry appears to want.

## THE REACTOR

There have been many reactors operated during the past few years by several different countries. They have compiled excellent safety records. There has been only one reported reactor failure and the principal damage in this case was to the installation itself. The entire cost of this accident was borne by the government operating the reactor. Under private operation, it would have been the responsibility of the operating company.

During the first few years of the period when many groups are involved in reactor development, the possibility of accidents will be greatest. This is because much of the development will be experimental and there are many unknowns in several phases of the reactor field. As knowledge and experience are gained, the risk will decrease.

Under private operation, then, many factors must be considered in evaluating the possibility of loss and potential cost of indemnification for damage. These criteria must be applied to each reactor unit separately. They include: use to which the reactor is to be put; reactor type and design; location; amount of containment; quality of supervision and operating personnel; safety program and inspection.

### Reactor Use

At this stage of the program, privately built reactors in the United States will probably be used for either research, testing of materials or production of power. The degree of risk will depend in part upon the use to be made of a given reactor.

Most *research reactors* contemplated at the present time are to be built by universities and other research organizations for the joint purposes of undertaking nuclear research and training personnel in reactor design and operation. Such reactors will probably be relatively small. Because of the nature of their use they will, of course, be designed for extreme flexibility. In addition, they will be used by large numbers of people, many of whom will have had no previous reactor experience. The safety of such reactors will depend more upon the quality and integrity of supervision than it will on design. Therefore it is imperative that reactors of this type should either be located far away from populated areas or they should be completely contained.

It is fairly obvious that remote location is a deterrent to effective use of the equipment. In a university, for example, it is essential that the installation be located in a place which is reasonably convenient for students' use. It thus becomes obvious that for a reactor of this type, effective containment is necessary.

In addition to universities it is also contemplated that some hospitals will be interested in having small reactors located on their premises for medical research and, possibly, therapy. As in the case of university research reactors, supervision of operation of these units must be of the highest quality. In addition, because of their location, a medical unit should also be designed for complete containment.

Because of the small size of research reactors, the amount of damage and contamination resulting from a reactor failure would be limited. The nature of the operations and the relatively large numbers of people who might be working with them, however, increases the possibility of incidents occurring. Thus from an insurance standpoint reactors of this type will probably be scrutinized with great care.

It is entirely probable that under the program of reactor development by private industry in the United States, some companies will be interested in the production and operation of *materials testing reactors* to assist industry in its reactor development program. There is at the present time no available information on private reactors of this type. If they should be proposed, they will have to be examined individually in the light of the various factors which will be discussed below.

By far the greatest amount of interest today has been in the development of *nuclear reactors for power purposes*. Several designs have been proposed and several companies or groups of companies will design and build such plants. In spite of the fact that these are to be full-scale reactors operated for the purpose of producing power, the entire program can be considered as being experimental in nature. This is because many different kinds of reactors are to be built and operated with a view to determining the kinds of reactors which are most practical and most economical for power production.

Such reactors will be considerably less flexible than research reactors and will probably be operated at more uniform power levels. In addition, the supervision and staff will be more stable and will be limited

in number. These factors will decrease the possibility of accidents. The severity of any incident which might occur could be much greater than in the case of research reactors.

### Reactor Type

Since the development of nuclear reactors is in its infancy, there have been many experimental types designed. There are unquestionably many other approaches which have not yet been explored. One of the purposes of private participation in the United States reactor program is to bring the viewpoint and ingenuity of industry into the program.

The objectives in reactor design are to increase efficiency and decrease cost (both initial and operating). By far the best protection against injury or damage is an inherently safe design. None of the types of reactors being proposed by industrial groups today can be said to be inherently safe. Some are safer than others and there is no question but that reactor design will be an important factor in eliminating the possibility of reactor failure. In cases where industry is requesting insurance, design will be an important consideration in determining whether a particular reactor is insurable.

### Reactor Location

In addition to considering the reactor, it is important to know in detail the potential exposure in terms of people and structures. The total indemnification cost will be related to the reactor location in terms of population, adjacent industry and the value of surrounding property.

One means of reducing losses through death, injury, physical damage or radioactive contamination is by the use of so-called exclusion areas. Obviously, if a reactor is placed in an isolated location, the possibility of catastrophe is reduced to a minimum. During the early phases of reactor development, large exclusion areas were utilized. With the development of nuclear power and building of reactors for teaching, research and medical applications, the usefulness of the installations will depend on their being located close to population centers. As has been stated, this means that greater reliance must be placed on design, containment and personnel.

### Containment

By containment is meant the enclosure of either the reactor core or the reactor and working area in a vessel or building capable of withstanding any possible pressure or missiles which may be produced as a result of a reactor failure. The purpose of such containment is to prevent the escape of radioactive contamination to surrounding areas. If the enclosure is the building housing the entire reactor unit, provision must be made for rapid evacuation of operating personnel from the building.

### Quality of Supervision and Operating Personnel

The safety of a nuclear reactor depends to a large degree on the skill and experience of both supervisors and operators. As in the case of industrial accidents today, human failure will probably play a significant role in any reactor accidents which may occur in the future. Thus a combination of intelligent supervision and well-trained operators will be a potent means of accident prevention. This must, of course, be combined with a safety conscious management and an efficient safety organization. Constant vigilance will be even more important in the future than it has been in the past.

### Inspection

During the first few years when private industrial research groups become involved in nuclear reactor design, construction and operation, it is essential that activities of such groups as the Advisory Committee on Reactor Safety and a safety inspection group be continued. It is particularly important that details of design be scrutinized carefully to assure that maximum safety is being built into any reactor. Furthermore, careful evaluation must be made of the proposed locations of these devices. This must be correlated with design and any plans for containment. It is also necessary during this early period of transition that inspection be made from time to time by competent people to assure continuous safe operation of any reactor.

Since the consequences of a reactor failure involving the dissemination of radioactive materials are so serious, the reactor operators must never be permitted to forget their responsibility not only to their fellow employees but also to the general public to prevent any incidents which may endanger either.

## PROBLEMS OF INDEMNIFICATION

There are many kinds of indemnification problems which can result from the operation of reactors. These may range from minor incidents involving damage to the reactor itself to major accidents which could involve dissemination of radioactive materials over wide areas around the plant. There are questions related to injury of employees directly concerned with reactor operations; damage to the facility itself; the effects of fire in any of the equipment; damages resulting from shut-down and a consequent failure to deliver power; and many compounded problems related to the responsibilities of part manufacturers if the failure of any particular component can be said to be responsible for any reactor accident. In addition to this there is the problem of indemnification of persons working in adjacent industrial plants but not employed by the owner of the reactor and also payment of loss to the individuals living in areas around the reactor operations who suffer either injury or damage to their property.

The problems of indemnification are present regardless of whether a reactor is owned and operated by a governmental agency or by private corporations. There is only a difference in methods of handling payments for such losses. As has been stated pre-

viously, in the case of government operations any payments for such losses will come from public funds. Private industry, particularly in the United States, is desirous of having complete insurance coverage to protect itself against all financial loss which can be related in any way to reactor operations.

### Injury to Employees

Injury to employees working on reactors or contingent equipment is paid for in the United States under a system known as Workmen's Compensation. The basic concept of compensation for injury while at work is almost universal. It varies only in mechanism of administration.

As far as reactor operations are concerned it seems to be the general consensus of opinion that Workmen's Compensation insurance imposes no problems that cannot be solved. It is possible to estimate maximum potential loss in that a specific number of employees will generally be involved. Limits of liability are established by law in the United States and many other countries.

Charges for this kind of insurance coverage are usually related to the amount of payroll and are based on accident experience. Since there is no experience from private operation of nuclear reactors it will be necessary to establish rates on the basis of experience within the Atomic Energy Commission together with an estimate of the added risk of widely expanded operations among a relatively inexperienced group.

In the normal operation of a reactor there are few hazards which cannot be evaluated with some accuracy. It should be possible to establish a reasonable base insurance charge for the reactor operation related to its basic use. This might mean one rate for power plants, another for materials testing reactors and a third for research reactors. In addition, there will probably be an added cost related to potential radiation exposures during normal operation. Consideration will also be given in rating to a possible catastrophe in the event of reactor failure.

### Damage to Facilities

A second important area of indemnification is related to any damage which can occur to the reactor unit and its ancillary equipment, the building in which it is housed and associated structures. Such damage could occur as a result of fire, equipment failure or of breakdown of reactor components. As in the case of injury to employees there is no serious problem about limits of liability. The exact value of all of this equipment and buildings will be known and their cost of replacement will be subject to accurate estimates. This, then, will determine the amount of insurance protection which would be required to cover damage or loss of any or all of the reactor facilities.

The question of rates will be related to reactor design, to materials and construction of the reactor unit, types of construction of the buildings involved as well as fire protection facilities. The cost of insurance of this type will, of necessity, be higher than normal rates for damage to equipment and facilities because of the added possibility of radioactive contamination. This is because the cost of decontamination can be extremely high.

### Injury or Damage to the Public

The most serious problem and one which is going to be most difficult to solve is that related to indemnification for injury or damage which may be done outside the reactor facility. If one assumes the worst possible kind of an accident it is conceivable that under proper conditions large amounts of radioactive material could be spread over a wide area outside the reactor. Radiation levels could reach intensities sufficient to produce injury and death to the exposed people and could produce serious contamination of the surrounding structures. It is fairly obvious, then, that in a congested region, the cost of such damage could be extremely high. It is difficult to predict, in the light of our present knowledge, exactly how high this cost would be. This is why industries in the United States desire to have limits of liability insurance of this nature high enough to protect their entire corporate resources.

From the standpoint of the private insurer there are definite limitations on the amount of risk which can be assumed on any individual enterprise. Even by pooling a world-wide insurance capacity it may not be possible at this time to provide general liability insurance protection in the amounts now being asked by industry. It is possible only to make an educated guess as to how much liability protection is actually necessary. It now appears as though the insurance industry is reaching the point of being willing to provide all types of insurance for most private reactor operation within the range of limits now available for industrial operations of all types. Beyond this, other arrangements will be necessary.

### Business Interruptions

In the case of a reactor for the production of power another possible contingency is that of a liability for failure to produce power which is essential for other operations. In other words, if a reactor breakdown resulted in a loss of power production which caused customers utilizing such power to shut down their operations, they might be entitled to some type of indemnification for their loss of business. In the case of a major accident with attendant long-term shut-down such losses could also be extremely high, particularly if several industrial customers were involved.

This kind of situation, however, should not develop to very serious proportions during the next few years since most projected power plants will merely supply power to a grid system and their shut-down consequently would not result in a complete power loss for any particular area. In any areas where a nuclear power plant is the sole source of power this could be a difficult problem.

### Other Problems

A matter of major concern to the insurance industry is related to the fact that under certain circumstances the indemnification of losses could be compounded. This could result from an incident in which, for example, a component part of a reactor is found to be the cause of the accident and the reactor plant itself is damaged; that employees in a neighboring industrial plant are injured; that considerable property in the area is contaminated by radioactive materials. In such a situation it can be pointed out that the operator of the reactor is primarily responsible for its failure and, therefore, the injury to the public; damage to the reactor itself and damage to surrounding property are all his responsibility. The employees of the adjacent industrial plant, however, are covered by law usually under insurance policies carried by their employer. The case could be further complicated if the company operating the reactor claims that the manufacturer of a defective part was in fact responsible for the entire incident. The possibilities of long and costly litigation in this case are almost unlimited. The costs would have to be added to the total costs of the actual damage itself.

There are some segments of the insurance industry which believe that it would be desirable to find methods of providing financial protection on more or less a package basis in order to avoid the kind of situation described above.

## A POSSIBLE PROGRAM

It seems reasonably obvious from this discussion that because of the unusual nature of the reactor program, some means of providing indemnification in the event of damage resulting from a serious reactor incident must be found. As has already been pointed out in the case of governmental operation in such installations the money, if any, simply comes from public funds. In the case of private industry some other method must be found to accomplish this purpose. In view of the lack of valid information upon which to base sound judgment it would seem reasonable to suggest the possibility of a joint effort involving the insurance industry and the Federal Government providing all necessary financial protection to private operators of nuclear reactors. Under such a program the insurance industry through pooling or a similar device would provide all coverages necessary to the maximum capacity of the industry. During the first few years when these projects are experimental and industry is gaining experience in design and operation of nuclear reactors protection above the capacity limit of the insurance industry could be provided by the Federal Government as excess coverage. This would mean that with the exception of an extreme catastrophe the indemnification of loss in all private nuclear energy operations would be handled by insurance carriers on a routine basis. At the end of a five-year period of private operation the problem should be re-examined. Sufficient data should be available by that time to permit an intelligent evaluation of the insurance protection actually required with such operations and of the ability of insurance carriers to meet this requirement.

During the period of early development industry must play its part in doing everything possible to prevent loss. This can be done through design and construction giving safety high priority. Losses can be kept down by use of maximum practical exclusion areas and containment. Further protection can be obtained by the employment of adequate health and safety staffs and by compliance with all standards which may be promulgated. Cooperation with all inspection agencies will also help in reducing the possibility of accidents. By wholeheartedly supporting all accident prevention programs industrial management can help considerably, not only in reducing the possibility of reactor catastrophe but in reassuring the insurance industry.

Insurance companies will provide an added incentive to safe design and operation of reactors by risking their money. If insurance companies work as a group, they should be able to exercise a powerful influence for safety. It is greatly in the interest of everyone concerned that every reasonable safeguard be used to prevent a serious accident.

One of the important contributions of an insurance program is that insurers will have an active interest in controlling all hazards. They will also be able to assist in developing safety and safe practices through their technical staffs.

# Workmen's Compensation Aspects of the Peaceful Uses of Atomic Energy

**By Bruce A. Greene,* USA**

The growing use of atomic energy for peaceful purposes has intensified the need for adequate provisions in workmen's compensation laws to afford the workers full protection in the event of an injury due to exposure to the hazards created by atomic energy. Many of the problems which are anticipated under the workmen's compensation laws in the field of atomic energy are similar to those which already exist in other industries such as chemical manufacturing. However, the lack of any adequate experience to date with atomic energy cases makes it difficult to determine whether the same standards recommended for occupational diseases in other industries will apply to industries utilizing atomic energy. The purpose of this paper, therefore, is to raise some basic questions regarding workmen's compensation problems which it is anticipated may arise in the field of atomic energy and to suggest the answers, wherever possible.

## FULL COVERAGE OF OCCUPATIONAL DISEASES SHOULD BE RECOMMENDED TO PROTECT SUCH WORKERS

In general, two methods of providing coverage for occupational diseases are followed under the workmen's compensation laws in this country. One method is called the schedule type of coverage whereby only those diseases listed under the schedule in the law are covered. The other is the full coverage type of provision under which all diseases are covered, usually by making the definition of the term "injury" broad enough to include any known diseases and any further unknown diseases resulting from employment conditions. The definition recommended by the United States Department of Labor for this purpose reads as follows:

"Injury means mental or physical harm to an employee arising out of employment, including such disease or infection as arises out of such employment."

More than half of the laws in this country now provide full coverage. This includes those States where the schedule type of coverage method was followed originally but full coverage has been obtained subsequently by adding to the list of diseases covered a provision such as "any and all occupational diseases" or "all other occupational diseases." Protection to workers who are injured on the job due to a radiation hazard is afforded also by a number of the States with schedule coverage which specifically include diseases due to exposure to radioactive substances and materials.

The experience in the United States indicates that, in order to give the worker adequate protection, full coverage of occupational diseases should be provided either through the definition of the term "injury" or by adding a catch-all provision to the schedule of diseases. A specific list, no matter how complete when compiled, can never keep pace with the new diseases that emerge, because of the lag between recognition of the disease and requisite legislative action.

## UNLIMITED MEDICAL AID SHOULD BE GRANTED TO THE WORKER WITHOUT REGARD TO THE PERIOD OF TIME REQUIRED OR COST INVOLVED

One of the most important provisions of workmen's compensation laws relates to the extent to which the worker is entitled to medical and hospital care in event of an injury while on the job. Most of the workmen's compensation laws in the United States provide for unlimited medical aid. Examples have been cited where the medical cost has been over $100,000 in a single case which the employer or his insurance carrier have paid. In some of the States, the unlimited medical benefits are granted specifically by statute. In other States, the statutes may prescribe medical aid subject to a certain maximum cost or period of time but give the workmen's compensation agency authority to extend the benefits for such time or amount as it may consider necessary. It is assumed that in such States injured workers would generally receive the benefit of full protection against the cost of medical care.

About one-third of our laws still limit the cost of medical aid or the period of time in which it may be given or include both types of limitations. In such States, however, it is not uncommon for the employer or insurance carrier to provide medical care over and beyond the legal requirements. Most employers recognize that the best medical care reduces costs by lessening the period during which such care is needed, and in many cases, lessening the degree of permanent disability suffered by the worker.

* US Department of Labor, Bureau of Labor Standards.

The International Labor Organization, the United States Department of Labor, and the International Association of Industrial Accident Boards and Commissions, composed of representatives of the various State and Canadian Provincial workmen's compensation agencies, have always favored unlimited medical benefits as the desirable standard for workmen's compensation laws. Radiation diseases are usually slow in developing and may extend over long periods resulting in a heavy medical cost. Laws should be designed to give full medical protection to the worker who suffers such an injury.

### WHAT TYPE OF PROVISION REGARDING SELECTION OF DOCTOR AND SUPERVISION OF MEDICAL CARE UNDER WORKMEN'S COMPENSATION LAW SHOULD BE ADOPTED TO ASSURE THAT SPECIALISTS IN RADIATION DISEASES WILL BE AVAILABLE TO TREAT WORKERS SUFFERING WITH SUCH DISEASES?

The medical-aid provisions of workmen's compensation laws involve the problem of the method in selecting the physician or surgeon to attend the injured worker. Various methods are provided for under the laws. A survey of the provisions for selection of attending physicians made by the statistical committee of the IAIABC in 1949 showed that, in most states, the law provides for the choice to be made directly by the employer or insurance carrier. In a few states, the selection is made by the worker from a panel made up by the employer or carrier. In about one-fourth of the states, the worker has some form of "free choice" but only a few of these authorize unlimited "free choice." In actual practice, it is quite common for employers or insurance carriers to forego their legal rights and to allow the worker his choice of a physician.

The key to the provision for selection of the doctor is the extent to which supervision over medical care is exercised by the workmen's compensation agency. In supervising medical care, compensation officials state that one of the main points to control is that the injured worker is treated by a physician, surgeon, or specialist whose competence to treat the type of injury sustained has been determined by recognized medical organizations. Inexpert medical care often proves expensive and may have a very harmful effect on the rehabilitation of the injured.

Since relatively few doctors are experienced with and qualified to handle radiation diseases, it is important that the worker who is suffering from such disease has the opportunity to consult a specialist, whenever this is possible.

Regardless of the method of selection of doctor being followed, it is extremely important that the workmen's compensation agency be given the necessary authority and medical staff to supervise and control the quality of the medical treatment being given to the injured worker. This should include the authority to request a change in the doctor, hospital, or other medical facility whenever in the judgment of the agency such change is desirable.

### SHOULD THE SAME TYPE OF MEDICAL EVIDENCE BE REQUIRED TO PROVE THE ORIGIN OF THE DISEASE IN RADIATION CASES AS IN OTHER DISEASES OR SHOULD SPECIAL PROVISION BE MADE FOR SUCH EVIDENCE?

One of the most difficult problems in many workmen's compensation cases involving occupational disease is the difficulty of proving causal relationship between disability and employment. This may be particularly true in radiation diseases since so little is known about these diseases and how much exposure is required to cause an injury. To facilitate presentation of evidence by an injured worker in such cases some method or special provision should be designed for workmen's compensation laws. The workmen's compensation law might include, for example, a specific presumption provision stating that for the enforcement of a claim under the workmen's compensation law, it shall be presumed, in the absence of substantial evidence to the contrary, that disability or death which is found attributable to the effects of radiation, either directly or by aggravation of underlying pathology, arise out of and in the course of employment where radioactive substances were used. Other possible methods should be studied to meet this problem.

### SHOULD THE USUAL TIME LIMITS FOR FILING CLAIMS IN WORKMEN'S COMPENSATION CASES BE AMENDED TO MEET THE SPECIAL PROBLEMS WHERE SUCH DISEASE MAY BE SLOW IN DEVELOPING AND DISABILITY MAY NOT OCCUR UNTIL 5, 10, OR MORE YEARS AFTER LAST EXPOSURE?

Time limitations for filing claims in workmen's compensation laws based on date of injury or date of last exposure may deny a worker with a radiation disease his right to compensation. Where a disease is slow in developing, such time limitations may expire before the injured worker suffers a disability or before he knows that he is suffering from such disease.

One method of meeting this problem is to provide that the time limitation shall not begin to run until the employee knows or ought to have known the nature of his disability and its relation to his employment. Further study should be given to this whole problem of the extent to which time limitations should be relaxed in radiation cases including the possibility of a time limitation provision which would not begin to run until the injured employee suffered a loss of wage-earning capacity.

### WHAT METHOD OF INSURANCE IS CONSIDERED BEST, GOVERNMENT OR PRIVATE, IN VIEW OF THE UNCERTAINTY OF THE HAZARDS INVOLVED AND THE LARGE SCALE DESTRUCTION WHICH MIGHT RESULT FROM ONE ACCIDENT?

At the present time, workers employed in plants producing atomic energy in the United States are protected by the State workmen's compensation law

of the State where the plant is located. Under an arrangement between the Federal government and the private contractor who operates the plant, the private contractor is reimbursed by the Federal government for the full cost of workmen's compensation protection. The usual type of workmen's compensation insurance system has not been followed, therefore, in such plants as a means of protecting the employer against any claims which may arise due to radiation diseases.

With the extension of the use of atomic energy for peaceful uses, and particularly the installation of atomic reactors, a study is needed to determine whether private insurance carriers, state funds, or self-insurers will be willing and able to handle radiation hazards or whether some other system such as a special government reinsurance fund, or other type of fund can be designed to meet this problem. The employer should have available to him at reasonable rates some method of insuring himself against the uncertainty of the radiation hazards and the large scale destruction which might result from one accident.

One of the most difficult insurance problems which will require careful study and may involve special insurance arrangements is in regard to radiation injuries which may occur to employees as a result of their employment in a plant located in the vicinity of a plant utilizing atomic energy. The application of third party liability procedures under workmen's compensation laws in such cases will require study as to the extent to which a special government insurance plan or other system may be needed to cover such contingencies.

### SHOULD ANY SPECIAL METHODS OR TECHNIQUES FOR ADMINISTRATION OF CASES INVOLVING ATOMIC ENERGY DISEASES BE RECOMMENDED, SUCH AS SPECIAL BOARDS?

The Wisconsin compensation authorities, who have had the longest experience in this country with the operation of full coverage for occupational disease, state that the settlement of occupational disease claims is no more difficult than adjudication of accidental injury cases and that no special administrative machinery is needed. Nevertheless, in some states the administration of the occupational disease provisions has been handicapped by the establishment of elaborate procedures and arrangements, such as medical boards, for settling occupational disease claims. The primary purpose of such machinery was to safeguard against any abuses of the coverage of occupational diseases.

Whether radiation diseases would require any special administrative methods or techniques is a question which should be studied. A Subcommittee on Industrial Relations of the American College of Surgeons did recommend in 1954 that "panels of impartial medically qualified experts should be established within the compensation systems" to assist the compensation agencies in the performance of their duties. The decision of these medical panels would be advisory only and the final decision would be made by the head of the workmen's compensation agency. A panel, for example, on radiation diseases might be very useful in determining the relationship of the disease and employment in contested cases. It might also be of great assistance to the medical director of the workmen's compensation agency in supervising the adequacy of medical care and the need for medical rehabilitation of workers injured due to exposure to radiation hazards.

### THE AMOUNT OF COMPENSATION BENEFITS SHOULD BE THE SAME FOR RADIATION DISEASES AS FOR OTHER INDUSTRIAL INJURIES

Under the workmen's compensation laws in the United States, the benefits for disability in case of occupational diseases are usually the same as for accidental injuries except with respect to silicosis, asbestosis, or other dust diseases. Because of the fear of a heavy compensation cost for dust diseases, some state workmen's compensation laws have placed special limitations on the benefits payable in such cases. However, the trend in recent years has been to remove such limitations and to treat all occupational diseases in the same manner as other injuries. The experience of a number of States has indicated that there is no valid reason why compensation for occupational diseases should be different than for other industrial injuries.

### SHOULD SECOND-INJURY FUND PROVISIONS BE EXPANDED TO COVER WORKERS WHO ARE SUFFERING FROM RADIATION DISEASES TO FACILITATE THEIR RE-EMPLOYMENT?

Workers who are suffering from radiation diseases but who may not be totally disabled may find difficulty in obtaining a job. Employers may hesitate to employ such disabled workers for fear of being liable for a heavy workmen's compensation cost if they should suffer further injury. To meet such problems, second-injury funds have been established under most of our state workmen's compensation laws so that when a second-injury occurs the employer has to pay only for the last injury, the remainder of the award being paid from the fund. The disabled worker receives full benefits for the disability resulting from the combined injuries.

Most of our second-injury fund provisions are limited at the present time to the loss of an arm, leg, eye, or other member of the body. Consideration should be given to broadening the coverage of such funds to include any type of permanent disability. This type of provision would facilitate the employment of workers disabled because of radiation disease and would help to prevent pressure on such workers to waive their right to compensation in the event of a second injury. Such waivers are now restricted or prohibited in most jurisdictions.

The method of financing the second-injury fund differs in the various state laws. The funds are

usually supported by an assessment against an employer or his insurance carrier in "no dependency" death cases. In some states the funds are supported by an assessment based on total premiums collected or total compensation payments. In a few states an appropriation by the State legislature is made to the fund.

### HOW SHOULD THE LIABILITY BE APPORTIONED IN CASES WHERE THE DISABLED WORKER WAS EXPOSED IN A NUMBER OF PLANTS TO THE HAZARDS CREATING HIS DISABILITY?

Two principal methods of determining liability in occupational disease cases are followed by the states where a disease, such as silicosis, may have been contracted through employment by more than one employer. In some states, the liability may be apportioned among all the employers involved. In other states, the liability may be charged entirely against the last employer who permitted the worker to be exposed to the hazard.

The apportionment of liability among a number of employers creates difficult administrative problems. The problems of how much should be apportioned to each employer, what to do in case of uninsured employers who may have moved out of the state or gone out of business, and how to obtain the necessary evidence to show causal relationship where the exposure occurred many years ago are among those to be solved where this method of handling occupational disease cases is followed. On the other hand, the charging of compensation only against the last employer may be unfair where the injured worker was employed for a relatively short time in comparison to the period employed by other employers. Consideration should be given to use of second-injury funds in such cases to reduce the cost assessed against the last employer.

Radiation diseases are among the slowly developing occupational diseases. The exposure in such cases may occur, therefore, in more than one employment and the problem of apportioning liability should be included in any study to be made of the workmen's compensation aspects of such diseases.

### WHAT METHODS SHOULD BE USED TO GO FORWARD WITH INTERNATIONAL DISCUSSION OF THESE PROBLEMS?

The questions raised in this paper point up some of the problems which we anticipate will come under the workmen's compensation laws in the field of atomic energy. In order to provide the answers to these questions, an exchange and pooling of ideas and experience between the various countries on the administration and application of workmen's compensation laws would be desirable. For this purpose, the ILO is the logical agency to make the necessary arrangements for such an exchange. It is suggested, therefore, that a resolution requesting this service on part of ILO be considered by this Conference, as a means of promoting future international discussion of these highly important problems.

# Industrial Training Aspects of the Peaceful Uses of Atomic Energy

By Fred W. Erhard,* USA

The progress being made in the production and application of a variety of radioactive materials has provided new research techniques for the sciences and new application that will possibly affect many areas of activity. With a more adequate and more versatile supply of radioactive material, uses will be expanded in its known applications, where the past limitation has been the expense or scarcity of material or equipment. In addition, the many new applications being developed that will affect, directly or indirectly, every phase of the economy indicates that there will be a growing demand for training.

New developments depend upon the existing knowledge and skill available in the application and use of the available equipment and the experimental designs of new equipment which often involves new applications of materials and, in some instances, the development of new materials. The development of new applications and methods in many instances is limited by the availability of personnel with the knowledge and skill required for the safe handling of the material and its application.

One of the limitations in expanding the use of atomic energy will be the availability of skilled personnel in research and in application of new developments. This makes it necessary to consider the training needed with adequate provisions for identifying and executing the training to expand existing applications and to develop and operate new facilities.

The research and development of radioactive materials and their applications have created a team of scientists, engineers, and practitioners in the fields of agriculture, medicine, and industry. These teams, in their specific fields, are providing the talent needed to bridge the gap between scientific discoveries and practical application. The scientists are, in many instances, selected because of their specialized knowledge in agriculture, medicine, or an industrial specialty. The engineer, operator, and craftsmen are required to have a broad training in their field with a high degree of competency. In addition, training is provided in those areas of the work which are peculiar to the production and use of radioactive materials.

The training that is offered at this time is, with few exceptions, organized to provide the additional knowledge and experience for personnel who have completed training in one of the sciences or as an engineer. In addition, other training has been conducted that provides the information needed and develops the skills that will be used in varied assignments in the production of fissionable materials and the installation and maintenance of equipment. Skilled personnel in the operation of chemical plants, skilled construction and maintenance craftsmen are selected because of their versatility and competency in a related industry or in a specific craft.

A cadre of skilled operators from chemical plants was selected to start one operation. While this group was familiar with most of the equipment that was to be used, they had to learn a new process and new application of the available equipment, as well as new equipment which had to be developed. In providing this training, it was necessary to establish exacting standards of performance and safety and health practices that are essential for their own protection, as well as their fellow workers. It is estimated that ninety per cent of the jobs are counterparts of those found in private industry. Ten per cent of the jobs represent those areas of activity, in a plant producing fissionable material, that are peculiar to the operation. These jobs are in the special hazardous department, which is responsible for protective devices, thickness of shields and placement of shields to control radiation, also laboratory technicians who check all incoming materials and those shipped, and production operators preparing isotopes for shipment. Another activity known as Health Physics works with medical and biological scientists in the determination of the amount of radiation which is safe to subject personnel. They also examine all localities to determine the radiation level and check some jobs to determine the dosage the individual has been subjected to. A Health and Medical program is staffed by industrial and medical doctors and nurses and is responsible for conducting physical examinations at regular intervals and medical services for the work force.

## TRAINING ACTIVITIES

The original training conducted when atomic plants were being constructed consisted of the training needed by selected personnel experienced in related industries or with basic scientific backgrounds

* US Department of Labor, Bureau of Apprenticeship.

that would prepare them for the varied assignments and duties involved in an activity still in developmental stages. Experience with the operation will more clearly define the duties and identify responsibilities which will develop as a result of increased knowledge and improved methods. There is, as would be expected, every indication that the training plans will be more comprehensive and will, in the future, provide all of the training needed to acquire competency in each area of assignment. The training was organized to provide skilled personnel to serve as a cadre for starting new activities in research and development, as well as in new industrial applications. This training is organized so it also supplements the basic training that is considered preparatory for the development of scientists and engineers and the operating and maintenance jobs.

All of the training has followed a similar pattern. It provides the theoretical and related knowledge, which is then followed by practical experience or laboratory practice conducted or observed under actual operating conditions. In some areas of employment, it has been found to be an advantage to provide all of the preparatory training in an operating unit in connection with employment. In other areas of activity, as the research and development projects, the development of scientists and engineers is accomplished by providing training beyond the academic degrees provided by the universities. This portion of the training is so organized in connection with the colleges and universities that it can be used in meeting the requirements of advanced degrees.

## ADVANCED STUDIES

The Atomic Energy Commission has contracts with companies who operate its plants. To provide the advanced training needed, these contractors have been paired with one or more universities or colleges which provides a cooperative arrangement for keeping training up-to-date with advancing technology. In addition, the Atomic Energy Commission has provided fellowships for advanced studies.

The Atomic Energy Commission Industrial Hygiene Fellowship is to provide, through the Institute of Nuclear Studies, training in industrial hygiene—a specialized field closely related to the atomic energy program. Fellows must have a bachelor's degree with a major in physics, chemistry or engineering and must be acceptable for graduate work at the university to which they are assigned.

Atomic Energy Commission's Radiological Physics Fellowship (Radiological Physics, often termed health physics) deals with the principles of radiation protection, the techniques and the underlying physics involved in radiation detection, measurement, and shielding, particularly as they apply to the protection of personnel from dangerous exposure to radiation.

Fellows must have a bachelor's degree in physics, chemistry, or engineering (usually electrical or chemical) with minors in mathematics, biophysics or similar fields, and be acceptable for graduate work in the university to which they are assigned. Applicants with other qualifications may be considered in special circumstances.

Oak Ridge School of Reactor Technology is operated for the Atomic Energy Commission by Carbide and Carbon Chemicals Company. The Oak Ridge School of Reactor Technology provides advanced education in reactor theory and technology to engineers and scientists who will engage directly in reactor research, design and development.

Students are selected by the committee on admissions to Oak Ridge School of Reactor Technology. The committee is composed of three representatives from the Atomic Energy Commission and two from the Oak Ridge National Laboratory. All applicants must have at least a bachelor's degree in chemistry, engineering, metallurgy, physics, or engineering physics and outstanding academic records.

Oak Ridge School of Reactor Training is a terminal school. Students considering work toward an advanced degree are advised to complete their university work prior to applying for entrance. Application for admission to ORSORT implies the intent of the applicant to enter directly into reactor-development work upon his graduation.

Selections are made from two categories of students, briefly described as follows:

Category A—Recent graduates with no employment commitments fall in Category A. These students will join the Oak Ridge National Laboratory as full-time student-employees and will be paid a monthly stipend for the duration of the session. At the end of the course, Category A students will be available for regular employment by the Atomic Energy Commission, its contractors, or other organizations actively interested in reactor development.

Category B—Experienced development or design engineers or scientists who are employees of the Atomic Energy Commission or its contractors, other appropriate government agencies or their contractors, or industrial organizations having an active interest in the development of reactors and their components fall in Category B. Category B students will remain on the payroll of their home organization during the session and will return to their sponsors upon completion of the course.†

## OTHER AREAS OF ADVANCED TRAINING

Some of the universities have recognized the necessity of bringing the school to the plant in order to deal more effectively with advanced studies in the sciences and engineering related to nuclear reactions. One example is the Massachusetts Institute of Technology Engineering Practice School operated in cooperation with the Carbide and Carbon Chemicals Company for the Atomic Energy Commission. This training provides the equivalent of one semester

† The preceding five paragraphs taken from 1955-56 Bulletin of "The Oak Ridge School of Reactor Technology".

of graduate work in connection with an established operation. The objective is to provide an education by practical application of basic fundamentals to technical problems encountered in industry. The advanced study is in the fields of chemical engineering.

## INSTITUTE OF NUCLEAR STUDIES

The Oak Ridge Institute of Nuclear Studies is sponsored by twenty-nine colleges and universities in the South and Southwest section of the United States. The Institute has a Board of Directors of nine members selected from the cooperating universities and a resident staff of seven.

The Institute is a non-profit educational corporation chartered under the laws of the State of Tennessee to conduct a program of research and training in the Nuclear Sciences. The Institute operates under a contract with the Atomic Energy Commission through which all of its programs are carried out and financed. It likewise maintains a number of formal agreements with the Commission's industrial and other contractors in Oak Ridge, which permits the use of research and training facilities in programs of interest to the universities.

## PROGRAMS OF THE INSTITUTE‡

### Research Participation Program

"Through this program, university scientists participate in the Oak Ridge research program for periods of three months or longer. In addition to assisting with the atomic energy research program, they return to their university with new ideas and techniques for the research program there and a fresh viewpoint for teaching."

### Radioisotope Training Program

"Radioisotopes, or so-called 'tracer atoms,' have been called the most important research tool to be developed since the invention of the microscope. In Oak Ridge—world center of radioisotope training and production—the Institute has trained more than 800 scientists from universities, medical schools, and industrial research organizations from throughout this country and from many foreign countries in the safe and efficient use of this new research tool."

### Oak Ridge Graduate Program

"Selected graduate students from any university in the country may carry out their research for the doctoral degree in the sciences in the splendid research laboratories of Oak Ridge. Limited opportunities also exist for master's degree candidates."

### Cancer Research Program

"The Institute is conducting a long-range study of the effects of atomic energy materials on cancer, utilizing a 30-bed hospital and laboratory unit. This is a joint program with the Southern medical schools. Patients are admitted for treatment only through a medical school."

### AEC Fellowship Program

"Several hundred AEC-sponsored predoctoral and postdoctoral fellowships in the physical, biological, and medical sciences are being administered by the Institute for the current academic year. These fellowships are intended to encourage the training of scientists along lines of interest to the nation's atomic energy program. Research under these fellowships is carried out in the university of the student's choice."

### Radiological Physics Fellowship Program

"This is another AEC fellowship program for the specialized training of personnel to guard against the hazards of radiation on the atomic energy project, in medical schools, hospitals and other installations using radiations or radioactive materials."

### Traveling Lecture Program

"The outstanding scientific staff of Oak Ridge is one of the South's major resources. Arrangements are made whereby these scientists lecture, conduct seminars, and otherwise take part in activities on university campuses."

### Summer Symposia

"The Institute and the Oak Ridge National Laboratory sponsor each summer a two-weeks symposium for university teachers in some field related to the atomic energy program. One of the recent symposia was in nuclear engineering."

### Resident Graduate Program

"This is a program conducted by the University of Tennessee through a sub-contract with the Institute as a service to Oak Ridge employees of the Commission and its contractors. Through this program, Oak Ridge research workers may continue their formal education by attending graduate school at night and on Saturdays."

### American Museum of Atomic Energy

"An impressive array of artifacts, exhibits, and other materials concerning the atomic energy program is accumulating. The American Museum of Atomic Energy is the custodian of many of these materials. It is operated by the Institute as a public service. In addition to the Museum at Oak Ridge, the Institute provides traveling exhibits for state fairs, scientific meetings, and similar activities."

## OPERATOR AND CRAFT TRAINING

The training of process operators and skilled craftsmen will have to be considered from the viewpoint of the production of radioactive material and industrial applications of these materials. In dealing with both these areas, it will be necessary to deal with experiences to date while considering developments and future projections that will create jobs or affect existing jobs, thus creating a need for training

‡ This section taken from Bulletin dated November, 1951, of "Oak Ridge Institute of Nuclear Studies, Inc."

## TRAINING FOR THE PRODUCTION OF RADIOACTIVE MATERIALS

It is estimated that ninety per cent of the jobs in the production of radioactive material have a counterpart in other industries. For this reason, it is often the practice, especially for a large company, to select a cadre from other plants of skilled operators and craftsmen who have many of the skills and experiences needed in the operation of a plant producing radioactive materials. Following this careful selection, it has been necessary to provide the training needed in the operation of a new process and the development of new skills and knowledge that are peculiar to the production of radioactive material. The exacting demands of the work to be done by the crafts requires that training be provided to perfect the application of the skills of the trade and in the use of new materials and new applications. As the activity grows, a cadre of skilled people will be selected from operating plants who will be thoroughly trained in the operation and skills required to operate a new activity. With the potential growth that is indicated, the present operating plants are faced with the responsibility of training operators and craftsmen that are prepared to progress to more responsible jobs in new undertakings. Operating plants recruit from the local labor market and provide the training needed to develop future operators and craftsmen. Those selected for training will be high school graduates and, in some instances, will have had some college training.

## PROCESS OPERATOR TRAINING

Process operator training is organized in two sections. The first section is conducted away from the job. This portion of the training deals with the process, equipment and controls used, safety devices to protect personnel against radiation, safe work habits, and other related information required on the job.

The second phase of the process operator training program is organized on a carefully prepared work schedule where those in training work under the direction of experienced operators. The work schedule usually starts with the identification of equipment, piping and controls, and is so designed to provide progression in the training to the more exacting assignments, as the skill and knowledge of the individual is demonstrated.

## TRAINING FOR THE SKILLED CRAFTS

The construction of the plants, installation of equipment, conversions and maintenance is done by skilled craftsmen. Some of the crafts employed as carpenters and bricklayers do not require any specialized training pertaining to job skills and job knowledge other than that required for safety and health in connection with assignments in areas subject to exposure to radioactive material.

The mechanical crafts, as the machinists, pipefitters, electricians, and boilermakers, require a broad background of experience and versatility in the work of the trade. More exacting standards of performance than required by present industrial practice are necessary because of new materials, new methods and new applications. These factors create a need for training for even the most experienced craftsman.

New plants were manned by experienced craftsmen that were recognized for their skill and knowledge of the craft. These craftsmen, with the selected group of skilled process operators, provided the experience and knowledge needed when working with the scientists and engineers in the conversion of scientific discoveries into an operating production plant. The approach is no different from that used in modern industry except for the intensity of the research and development with the accompanying changes, improved methods and broadened applications of skills. To meet these requirements of the developments, it is necessary to maintain a well-trained, versatile work force.

In addition to the training conducted for craftsmen, provisions are made for the selection and training, from the local labor market, of candidates who meet specific age and educational qualifications to be employed as apprentices.

The apprentice training program is organized for the gradual development of job skills through employment which is supplemented with related technical training. The apprentice works on the job under the guidance of a skilled worker. His job training is organized so he will have training and experience in all phases of his trade. The work schedule and the work assignments made on the job are arranged so as to progress the apprentice to new and more exacting work as he demonstrates his ability to do the work, thus keeping the apprentice in an active learning situation throughout his apprentice period.

The job training schedule lists all of the major processes of the trade and provides an estimated time for the training in each process. Work schedules are prepared for each apprentice to provide a diversity of experience and adequate time to develop the skills and trade judgment required of each phase of the work. This method for scheduling job skill training is usually used, as it provides more flexibility in scheduling and rotating apprentices in different work activities. The work of the apprentice will be arranged so he will be a productive worker in an established work activity.

Training periods for apprentices vary from three to five years. Apprenticeship provides the preparatory training in skills and trade knowledge required for entrance into a skilled trade.

Apprentices in atomic plants are given a period of off-job training which prepares them for assignments in the plant. It is considered necessary to provide this instruction because every employee is trained in safety and health practices and given background information on the over-all operation. Following this pre-assignment training, apprentices

are first assigned to the maintenance shops and are not permitted to work in the plant. As the apprentice progresses in his training in the shops and is considered sufficiently advanced, he is assigned to selected jobs in the plant to work under the direction of a craftsman.

The related trade and technical knowledge deals with those phases of the training that can best be given through more formal arrangements away from the job. Usually this phase of the training is scheduled for four to six hours a week in two class periods during regular working hours. Related training is conducted throughout the period of apprenticeship. The related instruction portion of the training is organized so it progresses with the job skill training schedule in the plant. The objective is to take advantage of the interest factor in providing technical training needed by the apprentice in his daily work. Since it is generally required that an apprentice must be a high school graduate, all of the off-job instruction is directed toward meeting the needs of the craft and its application in the industry.

Apprentices are trained in all of the construction and metal trades. The work of some of these trades is the same as that found in other industrial activities. In those trades that produce and maintain the equipment, there is a need for additional training that is peculiar to the operation. In one atomic energy plant, apprentices are being trained in the following trades: Auto Mechanic, Carpenter, Electrician, Instrument Mechanic, Leadburner, Machinist, Painter, Millwright, Pipefitter, and Sheet Metal Worker.

## FUTURE TRAINING

New developments and new applications of radioactive materials will be accompanied by the training required in industry. There will be a period of transition between research and development and industrial use. Industry will use this period to simplify and master the technical problems that have to be dealt with effectively before extensive industrial applications become a reality. The training needed will be of short duration, providing the individual is competent in the general field of endeavor and only has to master a few new techniques. The use of isotopes as tracer elements in agriculture, medicine, and industry is one example where skilled technicians have been provided new avenues of research and applications that have been limited in the past.

Industrial uses of isotopes in radiography, thickness gauges, ionization, acceleration of chemical reactions, cold sterilization, flow rate measurement, detection of leaks in pressure vessels and others, all represent specific applications that will be handled by skilled personnel who will require training in these new methods. This type of training will be a requirement for each new development and will be continued until such time as it is made a part of the preparatory training for the occupation.

The mechanical trades recognize the impact that growth of atomic activities will have and are studying how it will affect their trades. The Boilermakers, Pipefitters, and Electricians' Unions have a staff assignment with the responsibility of keeping abreast of this activity, so they will be prepared to advise their membership on the training needed to do this work.

The Boilermakers' Union has, in one industrial center, conducted some training for their members that deals with the skills and related technical material they need to make and install heat exchangers that use atomic energy as a source of heat. This craft union has a full time representative studying the training needs for this type of equipment. Their first job is to identify the skills involved, the standards of performance required for such exacting work, new tools and equipment to be used, and the processes developed to fabricate new materials. As the requirements for this craft training are defined and the training material is organized, plans are being made to conduct this training away from the job as preparatory to actual job experience when demands increase. This development will also become a part of the regular training for apprentices as soon as it can be made available.

As the demands increase on the crafts for skilled personnel to work on atomic installations in industry, management and labor will cooperate in identifying the training needed and in arranging for the development of new job skills in the present work force. This training will be conducted on the job, supplemented by such preparatory training and related technical instruction as is needed to prepare the individual to work in this new phase of the craft's activity.

## SUMMARY

The use of atomic energy in industry will progress in proportion to the available skilled personnel who are equipped to make the new applications and methods. The training will be a gradual development in step with the progress being made in industry. New activities will be started with experienced personnel in key jobs who will be responsible for the training of the work force.

All of the training will be organized to provide the theoretical training which will be combined with practical experience where the individual will demonstrate his competency to handle radioactive material. The standards of training and the competency of the individual, because of the hazards involved, will be carefully evaluated in connection with the licensing provisions of the US Atomic Energy Commission.

The research and development work, which is well established and will be expanded as industrial activities increase, will depend upon the training provided by the universities in their regular science and engineering courses and the cooperative ar-

rangements made with the Atomic Energy Commission and their contractors for advanced studies and research activities.

The scientists, engineers, and skilled craftsmen have been and will continue to be secured from the universities and related industries to provide the key personnel for this development. The universities are expanding and keeping up to date the training needed for the future development of technical personnel. Carefully planned apprenticeship will continue to be the method used to provide the skills and knowledge needed for future craftsmen who will produce and maintain the equipment used in new applications of atomic energy in industry.

# Boiler Safety Codes for Power Reactors

**By E. C. Miller,* USA**

Many new problems in materials application and design result from the requirements of nuclear power reactors. Existing boiler safety codes do not in all cases cover these novel requirements. Establishment of specific codes with adequate engineering design standards to insure safety in reactor performance, requires recognition of all the problems involved plus further research aimed at the solution of these problems. The purpose of this paper is to present some of these problems, not to suggest the solutions, which will ultimately come from code-writing bodies.

In the United States and in the Dominion of Canada, the ASME Boiler and Pressure Vessel Code is the safety standard generally accepted by industry, insurance companies, and state, provincial, and municipal regulatory bodies. This code, which was developed by the Boiler Code Committee of the American Society of Mechanical Engineers, will, in all probability, serve as the framework for an industrial power reactor safety code. Other agencies whose codes and specifications may contribute to the safety standards for a complete reactor system include the American Society for Testing Materials, the American Standards Association Code for Pressure Piping, the American Welding Society, various industry standardizing groups, the Atomic Energy Commission and other federal agencies.

Major factors which contribute to the unusual problems of reactor design and safety include the frequent use of unconventional structural materials selected because of specific nuclear properties, the need for large diameter pressure vessels, the possibility of damage to structural materials through radiation effects, the difficulty of periodic inspection of a radioactive engineering system, and the need for leak-tight minimum maintenance systems. These unusual factors, which are more or less peculiar to nuclear power systems, have resulted in some of the problems listed below. These problems have been recognized in the course of providing metallurgical engineering service—including the approximation of code equivalent safety standards—to an organization actively engaged in the design and construction of a nuclear power reactor.

Reactor systems may involve the use of relatively unconventional materials, both metallic and nonmetallic, whose choices are dictated by specific nuclear requirements. Examples of these are: zirconium, uranium, thorium, beryllium and hafnium, plus alloys and compounds developed from them. Considerable nuclear and mechanical property data have been accumulated regarding these materials, and specifications may have been prepared for their use in reactor system applications which do not require them to hold pressure or contribute appreciably to the structural integrity of the system. There is, however, a probability that some reactor types may make substantial structural as well as nuclear demands upon some of these materials. In such event, Code approval will require that mechanical property data be adequate to establish permissible design values and that approved standard materials' specifications ensure that the required properties are met consistently and reproducibly. Test data are required over the entire range of potential operating temperatures, covering short-time tensile strength, elongation and reduction of area; creep and bend testing; and elastic moduli, Poisson's ratios, thermal conductivities and thermal expansion coefficients. The anisotropy of the noncubic crystal structures of some of these materials, and preferred orientation resulting from mechanical working, may produce marked and variable directionality in the mechanical and physical properties which will have to be taken into account in establishing design criteria. Also, the properties of several of these metals are profoundly influenced by minute quantities of contaminants—particularly the interstitial elements: carbon, hydrogen, oxygen, and nitrogen. Properties of several of these materials are subject to further modifications through metallographic changes resulting from allotropic transformations occurring during thermal cycling in fabrication.

The nature of the circulating coolant or fuel may make it necessary to keep the volume of the circulating fluid at a minimum for reasons of investment in high cost coolants or the hazards associated with handling large quantities of coolants or fuels. These hazards include criticality, radioactivity, toxicity and inflammability. High heat transfer rates may have to be effected through thick sections of materials with low thermal conductivity whose selection has been dictated by nuclear requirements, corrosion resistance, or high temperature mechanical strength. It may be necessary to remove substantial quantities of heat generated inside vessel walls or structural components by radiation. These factors contribute to high thermal gradients and high thermal stresses,

* Oak Ridge National Laboratory.

complicate the stress pattern of the system, and may tend to produce warpage, thermal fatigue, or stress corrosion cracking.

To provide corrosion resistance in pressure containing components it may be necessary to clad or to use loose or intermittently bonded linings of a material not necessarily metallurgically compatible with the structural backing. Present codes make little provision for completely unattached linings, nor do they offer detailed guidance for the welding of lined or clad components, except in a few instances, or for the insertion of nozzles in such systems where the continuity of the corrosion-protective lining must be maintained without impairing the structural integrity of the pressure-containing backing material. Code-accepted practices are available and used industrially for the joining of cladding or linings of nickel alloys or stainless steel on carbon steels. Since these materials are relatively compatible, a metallurgically sound transition weld can be effected without impairment of the essential properties of either the lining or the backing material. Except for such materials and components whose geometry and joint design are very simple, satisfactory standards for making Code-acceptable welded joints in bi-metallic components are not yet available for use in the more complex engineering systems, particularly if linings of zirconium, titanium, aluminum, or certain of the precious metals, for example, are to be used in contact with carbon steels, low alloy steels, or high alloy stainless steels.

The combinations of temperature and pressure contemplated for industrial power reactors are, in general, not excessive by pressure vessel code standards. However, when comparable pressures and temperature are anticipated for conventional chemical plant service, the pressure vessels are usually of small diameter and the desired volume is achieved by an increase in the length of the vessel in proportion to its diameter. In the case of nuclear reactors, however, it is generally desirable for the major dimensions to be approximately equal—whether the shape actually approaches cubic, spherical, or cylindrical. This means that, operating at even moderate pressures, the diameters and wall thickness of a reactor vessel may be increased to the point of seriously complicating the problems of material selection and vessel fabrication. Whether these problems are solved by accepting heavy-walled sections, by using laminated vessel construction, or by making the vessels of very high strength materials, expansion of the present code coverage will be required to provide adequate design and safety standards for such construction.

The effects of long-time, high-intensity, irradiation of reactor structural materials are by no means completely understood. Other papers presented at this Conference will describe in greater detail what is known of these radiation effects. It is well to mention, however, that some metallic materials may be hardened, strengthened, and made less ductile by neutron bombardment at relatively low temperatures. At higher temperatures these effects may tend to anneal out; but, if stressed during this irradiation, the material may undergo accelerated creep. Elements with high neutron absorption cross sections may, within an entirely feasible time for some reactor types, undergo substantial transmutation, as in the case of the conversion of gold to mercury. More actual experience and radiation-effects test data are required before design limitations can be set with certainty that safety factors are even approximately equivalent to those used in conventional boiler code practice. In the meantime, some overdesign will have to be tolerated; and, in some cases, added physical safeguards and protective devices should be installed to provide for the possibility of unanticipated failures. The problem is further complicated by the difficulties involved in effecting periodic inspection after a system may have become radioactive.

Not all of the problems listed above apply to all reactor types. Nevertheless, most of them do occur in one or the other of the five power reactor types included in the current five-year power reactor development program of the Atomic Energy Commission. These are the pressurized water reactor, the fast plutonium breeder, the boiling reactor, the homogeneous reactor, and the sodium-graphite reactor. Information which will be useful in establishing an eventual power reactor code has been obtained from the successful operation of the small scale Homogeneous Reactor Experiment, the Experimental Breeder Reactor, and the Boiling Reactor Experiment. Similarly, valuable experience and data should result from the eventual operation of two or more full-scale power reactors now being built —the Pressurized Water Reactor and the Submarine Intermediate Reactor—and the intermediate scale Homogeneous Reactor Test.

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# Research Reactor Planning

**By B. Karlik, Austria**

Austria is considering the construction of a research reactor as a joint project of science and industry and is at present engaged in the preparations therefore. It is expected that within a period of one year it will be possible to solve the major problems raised such as the juridical form of the co-operation of the partners in the project, the financial problem, the coordination of the research programmes as well as the choice of the reactor type, the setting up of a time schedule, etc. The construction of a power reactor, however, is, at the moment, not taken into consideration.

# Record of Proceedings of Session 4.3

**TUESDAY AFTERNOON, 9 AUGUST 1955**

*Chairman:* **Mr. F. Perrin (France)**

*Scientific Secretaries:* **Messrs. J. Goedkoop and J. Gaunt**

**PROGRAMME**

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|---|---|---|
| P/1063 | Administrative and legal problems on the use of radio-isotopes in Japan | K. Suzue |
| P/400 | Administrative and legal problems of widespread use of high-level radiation sources—industrial health and safety. Radiological health-safety codes | W. Binks |
| P/778 | The general problems of protection against radiations from the public health point of view | WHO |
| P/319 | The achievement of radiation protection by legislative and other means | L. S. Taylor |

DISCUSSION

Mr. Y. FUJIOKA (Japan) presented paper P/1063.

Mr. W. BINKS (UK) presented paper P/400.

Mr. P. DOROLLE (WHO) presented paper P/778 as follows: The world is now facing a change in the problem of protection against radiation. In the past, only radiologists and, more recently, atomic energy workers were exposed to the hazards of radiation: this is no longer the case. With the widespread use of nuclear power, whole populations are threatened with exposure to those hazards. Consequently, while remaining an important aspect of industrial hygiene, radiological protection has now become part of the very broad domain of public health. That is why the World Health Organization has thought it desirable to submit to this Conference paper P/778 which I have the honour to present to you.

The World Health Organization, whose constitution has been recognized by eighty-one member States as well as by four associate members, is a specialized agency of the United Nations. Its function is to help the peoples of the world to reach the highest possible level of health by assisting Governments, at their request, in developing their public health programmes and by directing and co-ordinating world efforts in this connexion.

Accordingly, the document now before you attempts to deal with the problem of radiological protection in the broadest possible way, adopting what must be the viewpoint of public health administrators throughout the world. Although it was prepared with the help of specialists of recognized experience, the document does not aim at originality. Many of the ideas expressed in it will certainly be found, and undoubtedly in a more detailed and precise form, in documents presented by national delegations.

The very fact that this document is presented to you today by a public health administrator who is not a specialist clearly shows that what we have tried to do is to draw the attention both of national public health authorities and of nuclear energy specialists to the extent and magnitude of the public health problems created by the development of the peaceful uses of the new source of energy, the various aspects of which are being studied here.

What, to begin with, is the scope of the problem?

Man has of course always been exposed to radiation from cosmic and other natural sources, but present developments in nuclear energy are such that, with the passage of time, the ambient radiation will be raised significantly by radiation sources of man's own making, if these are not controlled. Everything that is already known about the biological effects of radiation indicates that an intensification of background radiation will certainly have somatic and genetic effects on man.

It is therefore the responsibility of the public health authorities to do as much as possible to prevent the background radiation from rising unnecessarily fast and to too high a value. This problem exists now, on a small scale, as a result of the operation of nuclear energy plants, and the protective aspect is now being adequately dealt with at the industrial level; but very soon this will no longer be possible.

The present codes of practice for dealing with radioactive effluent from atomic energy plants are compiled on the supposition that an insignificant proportion of the world's population is involved. When the exposed proportion becomes more significant, these codes may have to be made more exacting. The limiting factor in compiling standards is, and is likely to remain for some time, knowledge of human genetics, a subject on which there are few relevant quantitative data.

But time is short, and the public health services must fulfill their obligations before—as might soon happen—general exposure to background radiation reaches levels from which there is no return.

This lack of sufficient numerical data on background radiation and its effects on the world's population may open the way to the two extreme courses of excessive caution and undue laxity. Both are equally dangerous.

The public health services must now attempt to profit from our present knowledge, which is based on the experience acquired by the nuclear energy industry during the last ten years. They must not be accused of hindering the development of nuclear power and thus seeking to deprive the world of its benefits; on the contrary, there must be positive and constructive co-operation now, between the public health administrators and the specialists responsible for the technical development of nuclear power. Such co-operation can only be advantageous in every way. The industrial experience of the past has shown that the public health problems which have been foreseen and attended to ahead of time have proved less costly than those discovered too late.

In the paper before you, an attempt has been made to indicate briefly the principal aspects of the public health problem, the scope of which I have endeavoured to define. One of these is a human aspect, and is concerned with professional training. It is essential that responsible public health officers should acquire a scientific knowledge of the problems of nuclear energy so that they may collaborate usefully with engineers and physicists in studying protection against radiation and make the engineers and physicists realize how important these health questions are.

It is also important that public health officers should be able to keep pace with the constant progress in the study of nuclear energy; to enable them to do so, an effort should be made to sort out from the vast and daily growing body of literature those findings which are of importance for public health.

As I have already intimated, the task of the public health services would be lightened considerably by the provision of adequate protection in new installations at the start, and for this reason collaboration must, I repeat, begin at the early design stage. This involves an important problem of co-ordination.

Another and equally important matter of interest to the public health services will be the siting of nuclear energy plants. There again, a problem of co-ordination is involved.

At the moment the problem of the protection of the neighbouring communities is dealt with by the technical staffs of the plants themselves. When these plants become more numerous, this protection will clearly have to be the responsibility of the public health authorities in most cases. Codes of practice will have to be worked out with the plant technicians and must be of an eminently practical nature if they are to be effective. This applies to the problem of effluents, whether liquid or gaseous, or in the form of radioactive dusts; and also to the serious problem of the elimination of waste, especially long-lived highly radioactive waste.

In this connexion, no doubt, a problem will arise which goes beyond national boundaries. It would seem that, in order to avoid the dispersal of such waste in a manner which would cause an unacceptable spread of radioactivity over the surface of the globe, some kind of international agreement will be necessary to determine waste-disposal sites.

I shall not dwell on these technical aspects of the problem, for they are dealt with in the body of the document before you, and my present purpose is not to repeat the substance of that document, but to give you a broad outline of it.

I come now to the public health programme which the World Health Organization advocates. It is in the form of a very general programme of work which, we think, could logically be carried out *pari passu* with the development of nuclear power. This programme is not intended to be all-inclusive and would certainly be subject to modification in the light of review by technically competent bodies such as the International Commission on Radiological Protection.

The intention is to create a starting point for discussion in the hope of stimulating action in the right direction. The following items among others might be included in such a programme:

First of all, the training of public health personnel. That, if you will forgive the repetition, is one of the immediate important requirements, since, if their views are to command respect, public health personnel must receive adequate training. The World Health Organization attaches particular importance to this problem, and has studied ways and means of adapting its methods and general professional training programme to the requirements of the new field of study. By the grant of fellowships, assistance to educational institutions, the organization of demonstration projects, the assignment of visiting professors and lastly, the exchange of scientific information, the World Health Organization will be able to help Governments which desire such help both in supplementing the scientific training of doctors and their assistants in the use of radioisotopes, whether for diagnosis or treatment, and in providing the necessary specialized training for the public health officers, public health engineers, industrial health doctors and biophysicists who will have an increasingly important part to play in protection against radiation. WHO has dealt with this part of its programme in a paper on education and training (P/779, Session 24, Vol. 16, these Proceedings), which will not be presented orally, but interested delegations may refer to it.

In the second place, the programme should include the dissemination of pertinent public health infor-

mation. While there is at present a vast body of knowledge concerning the radiological health problems associated with nuclear energy, this knowledge is for various reasons not available in a co-ordinated and condensed form. If this state of affairs is to be corrected effectively, the present information must be co-ordinated and made available to those requiring it. This would entail a considerable effort, and it is a task which deserves the attention of an international organization such as the World Health Organization. With the help of the world authorities with which it is attempting to establish a network of communications, WHO proposes to select important public health subjects and to have them discussed amongst groups of experts working in the respective fields. Such an effort requires close collaboration with the existing national or international scientific institutions. In this matter as in so many others, therefore, WHO's function will have to be that of stimulation and co-ordination. By means of its publications it will also be able to help in disseminating the information thus collected.

A third part of the programme would be the promotion of a world-wide scientific study of the somatic effects of low-level radiation on humans in relation to the increase of the background radiation.

Lastly, as the fourth item in the programme, we suggest the inception of a similar study of the genetic effects of radiation. While this would certainly be a long term project, it should be started without delay. Whether the subject of study is the somatic or the genetic effects of radiation, we consider it essential that uniform methods and common standards should be adopted, so that all results may be compared.

The document before you deals fully with this aspect and with the details of the proposed programme of work.

In this statement, which I should have preferred to make shorter, I have outlined the programme of work which we consider essential for the effective protection of the world's population against radiation. As we explain in the conclusion of the document before you, we have to admit that though work on radiations has proceeded for some fifty years, knowledge of their effects on the human species is still deficient; and yet it is the duty of our generation, which will bequeath nuclear science to posterity, to hand down to future generations, not only the technique of applying the new source of energy, but also the knowledge of how to deal with the hazards inherent in it.

Mr. L. S. TAYLOR (USA) presented paper P/319, preceding it with the following words: I am afraid Mr. Binks and I think so much alike, that I am going to present a paper which will not sound too different from the one he presented earlier.

This report will consist mainly of a brief outline of radiation control measures, with particular reference to practices as they appear to be developing in the United States. A more detailed report is to be found in the Proceedings of this Conference.

## DISCUSSION OF P/1063, P/400, P/778 AND P/319

Mr. S. P. COBB, JR. (USA): I should like to address my question to Dr. Fujioka. Do I correctly understand that item 4 of article 35 of the Labor Standard Act gives detailed descriptions of medical symptoms that automatically warrant compensation?

Mr. FUJIOKA (Japan): Yes, that is right.

Mr. COBB (USA): Then how do you prevent abuse of the parts of this article?

Mr. FUJIOKA (Japan): It is a rather difficult problem, but we are now thinking about the establishment of the new Act, as I said, and we are giving that matter consideration.

Mr. COBB (USA): In delayed cases of radiation damage, how do you distinguish between occupationally induced damage and damage which might occur from natural causes?

Mr. FUJIOKA (Japan): It is a very difficult problem, but it depends upon the circumstances—how long the person has been engaged in his occupation. Let me say that I am not a physician, but a physicist, and I am very much afraid that I have not been able to give you exactly the answer you want. Mr. Tsuzuki, who is a member of my delegation, is a physician, and perhaps he can explain it to you in greater detail.

Mr. COBB (USA): I had one additional question, which I believe you have answered in connection with one previously asked. That question was: Are you considering plans to revise your Labor Standard Law, in view of the increasing use of radiation emitters in Japan, possibly to have provisions similar to those which you are going to have in the Radioactive Substances Control Act? I believe you have answered that by indicating that you were taking that under consideration.

Mr. FUJIOKA (Japan): That is correct.

Mr. TAYLOR (USA): I ask this question somewhat in the frame of reference that Mr. Binks and I are both interested in the same problems. I would be interested to have him bring out, if he would, a little bit about the possible plans that they may have in the United Kingdom with regard to compulsory inspection of radiation sources. Perhaps he could speculate—and I understand that it would have to be complete speculation—as to the number of such inspections that he might guess would have to be undertaken if this program were entered upon within, say, the next year or so.

Mr. BINKS (UK): In my contribution, I tried to show the trends of thought in the United Kingdom in planning control. We have already accomplished a certain amount; namely, we have various codes of practice under preparation. One, for hospitals, has just been completed and will be released shortly. As regards licensing, we all realize that this is a

very tricky matter indeed, and it is still under discussion in the United Kingdom. It is not possible to say at this stage just what degree of actual inspection will be required before permission is granted to use ionising radiations. Personally, I would hope that in the majority of cases we might be satisfied if we were assured that the organization which is planning to use ionising radiations has the necessary knowledge about the radiation hazards and will carry out the necessary measures advocated in the codes. I therefore think that it is fair to sum up at this stage by saying that we have to feel our way in regard to licensing.

Mrs. P. CAMBEL (Turkey): I should like to ask Mr. Fujioka which department takes care of the blood counts of the working people. Is it a Department of Health Physics? And how often do they check the blood counts of the workers? Are there any differential counts made, and are any shifts to the left in the blood count taken into account? Further, I should like to know whether there is any compensation for scientific and technical personnel: are they included among the working men, and are there any special salaries for scientific and technical personnel exposed to radiation hazards?

Mr. FUJIOKA (Japan): I am in charge of the establishment of the law as a member of the related committee and therefore I read the paper. But here is Mr. Tsuzuki, who is a specialist in medicine, and he can answer this medical question better than I.

Mr. TSUZUKI (Japan): The first question is: "How often are blood counts made to supplement the health physics check, and by what department are differential counts made?" My answer is that the blood count will be made twice by the health physicist belonging to that organization, and at that time the differential count should also be made.

As to the second question, the answer is that they can have the same annual leave.

The third question was whether there is compensation for scientific and technical personnel. The answer is that we do have such a regulation for the scientific and technical personnel also.

Mrs. CAMBEL (Turkey): I would like to know from Mr. Dorolle whether radioactive courses in muds for rheumatism and arthritis patients will also be controlled. Many patients go to these muds without having special indications for their treatment, and it may be that women with fibroids in the uterus may have haemorrhages due to this treatment. These muds used to be considered inoffensive, but I think we should now consider whether they should be under radiation protection.

Mr. DOROLLE (WHO): I must confess that Mrs. Cambel's question finds me somewhat at a loss. We have so far dealt here only with the question of artificial radiation, excluding completely the problem of natural radiations, especially when used for therapeutic purposes. I do not know of any examples of special rules for the use of radioactive waters and muds. It seems to me, at any rate from what we know of national regulations, that none of them has dealt with this problem, which seems in all cases to be one of clinical practice and therapeutic indication and has never been considered from the point of view of protection against radiation.

That is all I can say in reply to the question raised. We should, however, be glad to obtain information outside the Conference, if this would be of service to the delegation of Turkey.

Mr. V. ORTIZ (El Salvador): I think that one of the most difficult problems is what to do with radioactive reactor waste. Since this waste will have to be buried or dispersed in the depths of the sea, contamination problems affecting fish will undoubtedly arise. How will this situation be dealt with? I address this question to the representative of WHO.

Mr. DOROLLE (WHO): I am greatly flattered to be asked such a question, for it is essentially a technical question which certainly deserves the most careful study. It is being studied in a number of countries and will undoubtedly be the subject of international studies.

The question of the disposal of waste by burying it at sea is an important problem with bearings on ecology in general, on the question of fisheries, and, both directly and indirectly, on the question of the protection of human life. When we said just now in presenting our paper that there would have to be some kind of international agreement on sites for the disposal of long-lived highly radioactive waste, we certainly had in mind the hazard which the indiscriminate dumping of highly radioactive material into the sea might present.

That is certainly a problem of international concern to which particular attention will have to be given.

# Session 6.2

## REACTOR SAFETY AND LOCATION OF POWER REACTORS

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# The Safety of Nuclear Reactors

**By C. Rogers McCullough,* Mark M. Mills,† and Edward Teller,‡ USA**

## REACTOR TECHNOLOGY

In any new field of technology, it is important to investigate, quantitatively if possible, as many features of the field as seem pertinent for human welfare. Nuclear reactor technology is such a field, and no one looks to it with hope for many material benefits for mankind. Among these benefits are the possibility of electric power generation, propulsion by nuclear energy, and the utilization of reactors as research tools in many branches of science and medicine.

Along with a long list of possible attractive features of reactors, there are, unfortunately, certain dangerous characteristics. The Advisory Committee on Reactor Safeguards (see Appendix) has the responsibility of looking at the hazards connected with nuclear reactors. The members of this committee are exceedingly anxious to see rapid and fruitful development of reactor technology, but because of the nature of the hazards involved, and because they have been specifically requested to look at hazard problems, they feel it important that no undue risks be taken in the development of nuclear reactors.

## REACTOR SAFETY

Immediately, when one attempts to evaluate reactor hazards, there is encountered the necessity for attempting to define the notion of reactor safety, and what this notion shall include. Of course, absolute safety is not possible and what is really meant in connection with reactor hazards is the minimization of hazards until one has an acceptable calculated risk.

The operation of nuclear reactors appears safe and it is, in fact, deceptively safe. A nuclear reactor will not run away unless a number of serious mistakes of planning and operation should be committed. It is, however, impossible to conduct extensive operations over a long time without occasional occurrences of such mistakes. We have been exceedingly lucky so far that nobody has as yet been killed by a runaway reactor. It is not possible to count on indefinite continuation of such good luck.

One of the current difficulties in evaluating reactor hazards is this lack of experience with reactor accidents. So far, there have been essentially no reactor accidents leading to serious consequences. For this reason, statistical information about reactor accidents, although all favorable, does not suffice to give useful statistical information of the type needed by insurance companies, for example, in evaluating the nature of hazards. In other words, to determine what is an acceptable risk, a certain amount of judgment, detailed technical evaluation of a given reactor, and caution must be employed.

With all the inherent safeguards that can be put into a reactor, there is still no fool-proof system. Any system can be defeated by a great enough fool. The real danger occurs when a false sense of security causes a relaxation of caution.[1]

Problems of reliability, adequate control, adequate supervision, must all be included. It is convenient to look upon the concepts of reactor safety in the following ways:

One important concept is the division of safety problems into on-site and off-site problems. The on-site problems have to do with the protection of reactor operating personnel and other people who may be at the reactor site in order to make use of it, and the protection of the economic investment in the reactor facility. Off-site problems have to do with the protection of the general public, or persons who are not more or less directly connected with the operation of the reactor. One way to minimize off-site hazards is simply to locate the reactor at a remote and unpopulated place. In terms of reactor utilization and economics, this solution is often unsatisfactory. The economic utilization of electric power generated by reactors, for example, nearly always requires that the reactor be located reasonably close to potential users of this power. This means that for economic reasons the reactor should be located near populous, industrial areas.

Substantial moral and ethical problems are involved in connection with reactor hazards. On-site personnel, like persons working in other industries, knowingly and willingly submit themselves to whatever hazards are associated with working near a reactor because of salary requirements, special working conditions, or personal interest.

For off-site people, on the other hand, who have no knowledge or interest in the operation of the reactor, it seems that prevention of danger to their persons or damage to their property is a mandatory

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moral obligation in the operation of a reactor. This problem is more severe than in the case of dangerous chemical or explosives plants, because the radioactivity contained in a reactor can constitute a hazard to a wide area if it escapes from a machine and becomes dispersed.[2] This public hazard has been one of the main concerns of the Advisory Committee on Reactor Safeguards.

From another point of view, the safety of a nuclear reactor can be said to depend upon two things: The intrinsic built-in stability and reliability of the machine, and administrative control of the machine and its operation. For example, the reactivity may decrease rapidly with increasing temperature. In this case, it may be practically impossible to exceed some safe limit in temperature. This intrinsic stability is very desirable. In fact, one may say that a machine with large intrinsic stability can be so stable, because of fundamental physical characteristics, that only a Maxwell demon can make it misbehave. An ordinary machine, which depends on the operation of the control system to set its power level, can be upset by a mere gremlin! One would like to minimize the dependence upon administrative control for safe operation of a reactor. However, as a matter of practical fact, most reactors will nearly always require a certain dependence upon administrative control for safe and reliable operation. This means that problems arise connected with the loading and unloading of fuel, the startup and shutdown of the reactor, proper manipulation of controls, and adequate accounting for all materials made radioactive by the reactor, including both intentionally irradiated material and any radioactive effluent associated with the operation. Thus the normal, as well as the abnormal operation and behavior of the reactor must be carefully considered. It is clear that a reactor which in normal operation is well run and under complete and precise control is much less likely to behave in an abnormal fashion leading to a serious accident.

## THE CONTAINED RADIOACTIVITY

The most serious continuing hazard associated with nuclear reactors is due to the large amount of radioactivity which they contain. Large reactors may contain hundreds of pounds of radioactive fission products which correspond to many tons of radium in conventional radioactive measure. Not all of these fission products are as hazardous as radium, but nearly all of them contribute substantially to the hazard.[3] There are two ways in which the hazard of contained fission products may be minimized: One is to remove fission products during the operation of the reactor in such a way as to maintain a minimum concentration of such material in the machine. This continuous removal of fission products requires some type of fluid fuel, either liquid or gaseous, in order to continue cleanup operations on the fuel during the operation of the machine. The other way to minimize the hazard is to minimize the possibility of the escape of these fission products from the machine. The potential ability of a reactor to run away makes it possible for this radioactive material to escape to the surrounding areas. The hazard is crudely analogous to conducting both explosive and virulent poison production under the same roof.[4]

Until really safe nuclear machines of the future become available, we have to construct our reactors with extreme circumspection and we must continue to operate them with the same caution after ten years of safe running as on the very first day when they were started up.[4]

In order to emphasize the characteristic of the special hazard due to radioactive materials in the reactor, a list of tolerances is presented in Table I.[5] Although there has been a substantial effort in the assessment of the effects of radiation on biological systems, particularly systems resembling people, there is still a great deal to be learned.[3] However, even allowing for considerable error in the quantitative assessment of this problem, it is still evident from Table I that radioactive poisons are more hazardous than chemical poisons by a factor of something like $10^6$ to $10^9$. This is such an enormous factor that radioactive poisons essentially must be considered a qualitative new kind of problem. Furthermore, this implies

**Table I.[5] Comparison of Toxic Substances in Air* (concentration in mg/m³)**

| *Substance* | *Tolerance* † | *"Fatal dose"* ‡ | *Ratio fatal to tolerance* |
|---|---|---|---|
| **Chemical Poisons** | | | |
| Chlorine | 2.9 ¶ | 290 § | 100 |
| Arsine | 0.16 ¶ | 800 § | 5000 |
| Beryllium | $1.5 \times 10^{-5}$ | ? | ? |
| **Radioactive Poisons**** | | | |
| $U^{233}$ (insol) | $1690 \times 10^{-9}$ | $1690 \times 10^{-5}$ | 10,000 |
| $Pu^{239}$ | $32 \times 10^{-9}$ | $32 \times 10^{-5}$ | 10,000 |
| $Sr^{90}$ | $1.3 \times 10^{-9}$ | $1.3 \times 10^{-5}$ | 10,000 |

* It should be remembered that industrial poisons are usually in many ton quantities, whereas radioactive poisons are in 100-kilogram quantities.

† "Tolerance" for chemical poisons is defined as the maximum tolerable level for 8 hours per day exposure. In the case of radioactive poisons tolerance is the maximum level which can be tolerated every day for 8 hours equivalent to 0.043 rem per day.

‡ "Fatal Dose" in the case of chemical poisons is defined as the "rapidly fatal" dose when the given concentration in air is inhaled for 30 minutes to one hour. In the case of radioactive material this means about 50% survival if the dose is acquired quite rapidly, for example, over a minute or perhaps during an 8-hour day. This is equivalent to about 400 rem.††

¶ Adopted at meeting of the American Conference of Governmental and Industrial Hygienists in Atlantic City, N. J., in April 1951.

§ Industrial Hygiene and Toxicology, Frank H. Patty, Editor, Interscience Publishers, Inc., 1949.

** Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water, Handbook 52, National Bureau of Standards, March 20, 1953.

†† The Effects of Atomic Weapons, US Government Printing Office, Revised September 1950.

that the problem of keeping radioactive materials within the reactor and preventing the spread of radioactive materials over populous areas is very serious.

In Table II there is a summary of delayed heat production and the corresponding radioactivity from fission products. For a machine of 250,000-kw heat power (60,000-kw electric power), something like 300 million curies of activity remains at the end of one day after shutdown. This corresponds to 300 tons of radium in terms of radioactivity. The sheer quantity of radioactivity is enormous.

Operation of this reactor for one year produces about 100 kilograms of fission products. On the basis of $10^{-7}$ mg/cm$^3$ this can contaminate $10^6$ cubic kilometers of air to tolerance. Said another way, a layer of air one km deep covering an area 1000 km on a side could be brought to tolerance level.

Another feature of radioactive poisons is that a lethal level is not detectable by human senses. Furthermore, very serious injury may not be detected for some years after exposure.[2]

Table II. Delayed Heat Power and Radioactivity* after Normal Shutdown

| Time after shutdown | | Activity level from a previous steady heat power of: 300 kw activity as: | | 250,000 kw activity as: | |
|---|---|---|---|---|---|
| sec | | kw | curies | kw | curies |
| 10 | 10 sec | 12.9 | $2.1 \times 10^6$ | 11,000 | $1.8 \times 10^9$ |
| $10^2$ | 1.7 min | 8.0 | $1.3 \times 10^6$ | 6800 | $1.1 \times 10^9$ |
| $10^3$ | 16.7 min | 5.2 | $8.4 \times 10^5$ | 4300 | $7.0 \times 10^8$ |
| $10^4$ | 2.8 hr | 3.3 | $5.3 \times 10^5$ | 2700 | $4.4 \times 10^8$ |
| $10^5$ | 28 hr | 2.0 | $3.3 \times 10^5$ | 1700 | $2.8 \times 10^8$ |

* The radioactivity figures are for fission products only (do not include radioactive fuels or components). It is assumed that the mean decay event corresponds to 1.0 Mev in converting from kw to curies.

## ESCAPE OF RADIOACTIVITY

The way in which reactors can malfunction and lead to the escape of fission products may be classified as follows: (1) a super-critical nuclear excursion or nuclear runaway; (2) melt-down of reactor components, even with the chain reaction shut down, because of the delayed heat produced by the radioactive fission products; and (3) possible exothermic chemical reactions among the components of the reactor itself. The latter, although it is clearly not present if the machine is operating normally, may be initiated by a runaway nuclear chain reaction or by delayed heat melting.

These problems will be discussed in more detail below. The first two are unique to nuclear reactors as compared to other power sources, and have no true analogue in other areas of technology. They are discussed in some detail, for research reactors,[6] and nuclear power plants[7] elsewhere.

## THE PROBLEM OF NUCLEAR RUNAWAY

An outstanding characteristic of nuclear reactors is their potential ability to achieve extremely high power levels in a short time if adequate control of the machine is lost. A typical nuclear runaway accident may start and be over in times appreciably less than a second. In this respect they are different from any other large-scale machines, and it is this extremely short time that makes it quite important that automatic control and safety systems be available, be reliable, and be relatively rapid in their operation.

Another feature of a possible nuclear runaway is that it does not seem to be very violent. A comparison between a nuclear reactor and an atomic bomb is very misleading and certainly not to the point. From a number of studies of possible reactor accidents of this type, it must be concluded that even though reactor accidents could happen quite rapidly in terms of human reaction times and conventional external emergency human actions, nevertheless, a nuclear reactor is a very sluggish device and does not produce a nuclear explosion even remotely approximating that of an atomic bomb. Indeed, for the large thermal reactors, nothing like an explosion really occurs. For very fast reactors with a non-thermal neutron spectrum and heavily loaded with enriched uranium, it does appear possible to have an accident which is fast enough so that portions of the machine may be propelled with velocities of a few meters per second. This again does not resemble an atomic bomb explosion, or even the explosion of ordinary chemical explosives; rather it is similar to the events that might occur in an automobile accident. Therefore a nuclear runaway, in itself, does not represent a serious hazard to off-site people.

However, as pointed out above, a nuclear runaway can serve to do two things: It may disrupt the structure of the reactor sufficiently so that radioactive poisons may escape, or it may lead to exothermic chemical reactions between different components of the reactor core, and a chemical explosion of considerable violence. Indeed, for certain types of reactor structures, it would appear that the chemical reaction that might follow a nuclear runaway would produce substantially greater energy and violence than the runaway which preceded it.

In order to make some of these notions more quantitative, it is convenient to talk about the rising period of a nuclear reactor. A nuclear reactor which is supercritical increases in power level by a factor of $e$ at each interval of time corresponding to the so-called $e$-folding time. In turn, the $e$-folding time is related to the intrinsic neutron generation time of the reactor, and the degree of supercriticality, by the so-called in-hour equation. In Fig. 1, a number of curves are shown connecting the rising period of the reactor with its excess reactivity, i.e., the fraction of excess neutrons produced in one generation. The $e$-folding times shown in the figure are relatively long. This is due to the delayed neutrons. As you all know, the fission event produces certain fission products which, in turn, after periods ranging up to 80 seconds,

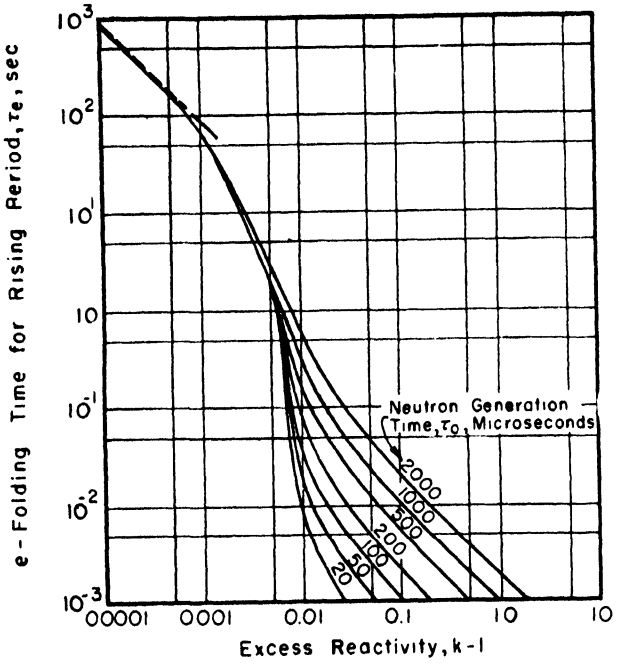



**Figure 1. Variation of rising period with excess reactivity from the inhour equation:**

$$k - 1 = \frac{\tau_e}{\tau_e} + k \sum_i \frac{\beta_i \tau_i}{\tau_e + \tau_i}$$

emit delayed neutrons. The fraction of these delayed neutrons produced in $U^{235}$ fission amounts to something like ¾% of all neutrons produced. If one makes the excess reactivity of the reactor so great that the chain can proceed without the delayed neutrons, then the reactor is said to be in a prompt critical condition, that is it is critical or even supercritical on prompt neutrons alone. In this condition the *e*-folding time becomes short and one may estimate it by means of the equation:

$$\frac{1}{\tau_e} = \frac{k_{ex}}{\tau_0}$$

In this equation, $\tau_e$ is the *e*-folding time, $\tau_0$ is the intrinsic neutron generation time which depends on the type of reactor, and $k_{ex}$ is the excess reactivity above prompt criticality.

Typically, neutron generation times are about one millisecond for large thermal reactors, something like ⅒ millisecond for water boilers and small thermal reactors, and may be as short as a microsecond or less for fast-spectrum epithermal machines. The value that one may assign to $k_{ex}$ depends on the type of machine and its requirements for excess reactivity in order to conduct experiments, overcome temperature effects, allow for burnup of the fissionable material, or override fission-product poisons. However, it seems reasonable to assume an excess *k*-value of about 0.01 fraction for terms of discussion. If this is done, then an *e*-folding time for the large thermal machines of ⅒ second is obtained. Only seven *e*-folding times, that is 7/10 second, are required in order to increase the power level of the machine by a factor of 1000. For machines with shorter neutron generation times, the *e*-folding times are correspondingly reduced. Here one may say that nuclear reactors represent a genuine departure from conventional power sources in that enormous power level increases are possible in the event of mal-operation in remarkably short times.

The Safeguard Committee has urged the development of automatic fuses which would prevent a nuclear runaway in case the reactor gets out of control. These fuses are expected to have two characteristics. First of all, they should be self-contained and wholly automatic so that they are not subject to error of adjustment or maintenance and are not subject to intentional tampering. Second, these fuses are to be activated by changes in the power level, essentially changes in the flux level of the nuclear reactor, and have rapid enough response so that they will introduce a substantial negative reactivity in the reactor in a time of the order of one second or less. One of the continuing difficulties in the development of these fuses is this latter requirement for short-time operation. It appears that successful development of such a fuse will soon be achieved, but because of the short-time requirement this development is neither easy nor simple.

As a matter of practical fact, one must consider how a large excess reactivity might be achieved in a nuclear reactor.[6] First of all, it is clear that it can not really be achieved instantaneously although something analogous to instantaneous excess reactivity can be obtained on startup of a reactor if only a weak source of neutrons is used during the startup procedure. It is then conceivable that through some error, rapid removal of control rods would allow the reactor to be highly supercritical before the power level had risen to something approaching the normal power range. For this reason, startup accidents are particularly to be avoided.

In any event, one must consider not only the possible degree of excess reactivity but also the rate at which reactivity may be added to the machine. For this reason, one would like safety rods and shim rods that move out rather slowly but which could be re-inserted rapidly at any point during withdrawal.§

One would also like the degree of control residing in the control and safety system to have a graded weight so that as the reactor becomes nearly critical, only smaller amounts of reactivity are introduced by the withdrawal of control rods. Safety rods which must be completely withdrawn and cocked before they may be re-inserted are particularly undesirable.

There is another point which is quite pertinent in the serious consideration of how rapidly excess reactivity might really be added to a given nuclear reactor. Extremely fast reactors, for example, look

§ In order to avoid circumlocution, all remarks concerning control and safety systems will be made as though these were conventional absorber systems. Of course it is entirely possible to increase the reactivity of a reactor by inserting fissionable material instead of withdrawing an absorber or by changing the characteristics of a reflector. Our discussion will assume that all controls are of an absorbing type.

particularly dangerous because of the very short *e*-folding time that one can achieve with modest excess reactivity. However, part of this danger is spurious because the reactor will become supercritical enough to run through a complete runaway accident before very much excess reactivity can be added by ordinary methods of operation of controls. Only very sudden motion of the control rods, motion so rapid that it would have to be induced by special pneumatic systems could lead to a rapid, explosive type of accident with these fast reactors. For this reason, a careful study of the possible rate of reactivity increase, rather than the total potential excess reactivity available, should be carried out when the nuclear runaway problem is considered.

It seems that the prevention of nuclear runaway accidents is very closely associated with the problem of excess reactivity and the rate at which excess reactivity might be added to a given machine. This in turn depends on the technical details of any given machine, both in its neutronic behavior and in the operation of control devices and possible other ways of changing the reactivity, perhaps because of the presence of experimental irradiation facilities. This is not a problem that can be generally solved for all machines, but each machine must be studied on its own merits.

We will now turn to the characteristics of a nuclear runaway, assuming that it is actually underway. As pointed out above, a nuclear runaway is not particularly violent but it does take place in a remarkably short time. The runaway will proceed according to the following steps. First of all, excess reactivity is inserted, the reactor then rises exponentially in power level with a nearly constant *e*-folding time until enough energy is accumulated in the structure to affect the behavior of neutrons. These early effects are characterized by the term, temperature coefficients of the reactivity. These may be either positive, that is making the reactor more reactive, or negative, making the reactor less reactive and tending to shut it down. If an increase in power level tends to make the reactor more reactive and increase the power level not only further but make the further increase more rapid, one sometimes says that this is an autocatalytic reactor. Such a reactor appears to be particularly dangerous, and can possibly achieve really short *e*-folding times. A few strongly autocatalytic reactors are known. For most reactors, negative reactivity coefficients will take effect and lead to a lengthening in the *e*-folding time.

Finally, a third phase of the runaway will occur when enough of the reactor structure is actually melted, vaporized or otherwise affected (in most cases reactivity coefficients will not be adequate to shut the reactor down without destructive effects, although for some reactors this will indeed be the case), and these destructive effects will shut down the nuclear chain reaction and stop the runaway.

Since the negative reactivity coefficients can lead to shutting off the nuclear accident without destructive effects, a few words about these coefficients may be desirable. First of all, large negative reactivity coefficients are clearly wanted. However, these coefficients must be quick acting, able to take effect and shut down the reactor during the transient conditions of a runaway. Primary changes in temperature are caused by the generation of fission heat in the fuel elements. Heating of the fuel elements may change the reactivity of the machine negatively if the fuel elements contain large quantities of $U^{238}$. This negative change is due to the increased absorption of resonance energy neutrons by Doppler broadening of the $U^{238}$ absorption resonances. For large lumped thermal reactors, this effect amounts to about $10^{-5}$ fraction of reactivity per degree C temperature rise.

A secondary reason for the temperature coefficient is the heating of the moderator. In many reactors, this will beneficially reduce reactivity. However, the time for heat to flow from the hot fuel elements to the moderated portion may be sufficiently long, several seconds to a minute in some cases, so that although the moderated temperature coefficient is favorable, it does not have time to come into play during a nuclear runaway. For example, the thermal diffusion time across a four-inch thickness of graphite-moderator in a large lumped thermal machine is nearly a minute. This time is so long that the coefficient associated with moderator heating plays no part during the runaway.

One of the important technical areas associated with understanding nuclear runaway behavior of a reactor is that of heat transfer under transient conditions. Relatively little knowledge in this area has been available because most heat transfer studies are conducted under steady-state conditions.

It appears likely that a nuclear runaway will cause enough disruption of the reactor structure so that fission products will start to leak out of the reactor into the surrounding area. How fast this escape of fission products will be depends upon the type of reactor and type of reactor accident. It may be possible to show that there will be very little mechanical violence outside the reactor shield, so that if the building which houses the reactor can be made gas-tight, then escape of fission products to areas outside the reactor building can be greatly reduced. We wish to emphasize that in most cases the building need only be gas-tight and not explosion-proof.

## DELAYED ENERGY PRODUCTION

Nuclear reactors have another somewhat unfavorable characteristic. Because of the accumulated fission products, and the accompanying exothermic radioactive transformation of these fission products, a nuclear reactor will continue to produce heat even when the nuclear chain reaction is shut down. The energy produced by the fission products has been studied, and the result for power production in the

reactor which has been operating for a long time may be summarized in the following equation:

$$P_{delayed} = 0.07\, P_{normal}\, [t(\text{sec})]^{-0.2},\ t > 1 \text{ sec}$$

Here $P_{normal}$ is the normal operating power level of the reactor, $P_{delayed}$ is the delayed heat power level of the reactor in the same units as the normal power level, $t$ is the time in seconds, and the 0.07 is an experimentally determined coefficient. Although the fission products individually decay exponentially, the result of their statistical production is to make this delayed heat decay with the relatively weak power law indicated. For about one second after the reactor is shut down the delayed power level is approximately 7% of the normal power level.

The delayed heat problem is clearly a serious one. If some failure in the cooling system should occur, breakdown of pumps, loss of pumping power, mechanical failure of cooling piping, then even if the nuclear chain reaction is immediately shut down by inserting control or safety rods, there will still be left a substantial heat load which must somehow be disposed of.

For example, if the fuel elements from the Materials Testing Reactor were suddenly removed from the reactor and left standing in the open air, they would melt down by themselves by delayed heat production. If they were suddenly immersed in water, probably this melting would not take place.

One may consider the delayed heat problem from the point of view of suddenly stopping forced cooling in the reactor and also suddenly stopping the chain reaction. The fuel elements, and other materials in the reactor in close thermal contact with the fuel elements will then start to increase in temperature. The rate of temperature rise will be proportional to the preceding steady power level of the machine, and the rate of temperature rise will be reduced if there is a large heat capacity in intimate thermal contact with the fuel elements. In fact, since the rule of du Long and Petit indicates that the heat capacity of solid materials is proportional to the number of atoms they contain, a crude rule of thumb would state that the rate of temperature rise is proportional to the power of the reactor per atom of material in good thermal contact with the fuel elements. The simple expression we have given for the delayed power indicates that the time rate of temperature rise should be proportional to the 0.8 power of the time. In Fig. 2 are curves showing the rate of temperature rise following uncooled shutdown from normal operation for a few reactors.[9] One concludes that this temperature rise, although not so rapid as to constitute a sudden event in terms of human reaction times, is nevertheless rapid enough to be quite troublesome. In Table II are summarized some delayed heat power levels.

Three types of preventive designs are suggested. One is to have a standby emergency cooling system which works either by gravity flow of coolant or by natural convection. Another is to have standby emergency arrangements of some sort which might be rather analogous to fire-fighting equipment. The "fire-fighters" would then approach the reactor and make suitable connections and force through emergency cooling. A third possibility is to have standby forced convection cooling similar to the main cooling system but connected to a special power supply and with special separate piping.

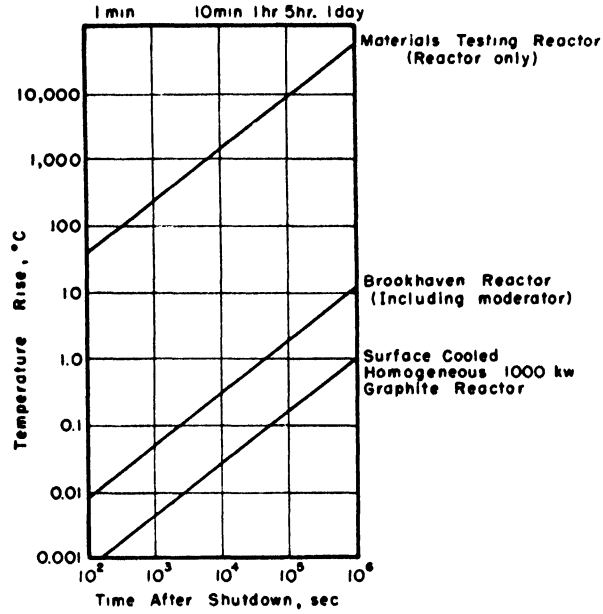



**Figure 2. Calculated temperature rise after shutdown of the chain reaction. (Heat capacity assumptions are indicated.) (Courtesy of W. H. Zinn, Argonne National Laboratory)**

It is clear that a delayed accident, if it could not be brought under control, might very well lead to sufficient disruption of the reactor core to allow fission products to escape. It is also clear that this again will not be of itself a very violent event, and again as in the case of the nuclear runaway it is probable that an accident of this kind can be minimized a good deal by providing a gas-tight building around the reactor.

## CHEMICAL REACTIONS

Either a nuclear runaway or a delayed heat accident may cause considerable melting and mixing of reactor components and lead to exothermic chemical reactions between these components. A simple example of this type is that of an air-cooled graphite reactor. A sudden temperature rise in the uranium fuel may be sufficient to cause it to melt and heat the adjacent graphite so that both the uranium and graphite can burn in the cooling air. If the air supply is not turned off, it is likely that a substantial portion of the reactor could be consumed in this way. This would then disperse radioactive fission products into the surrounding area through the exhaust portion of the cooling system.

Another example is that of a heavy-water-moderated-and-cooled natural uranium reactor. In a machine of this sort a runaway accident could melt the

uranium and allow it to mix intimately with the water. In this case the thermodynamic potential indicates that an exothermic chemical reaction can take place. Whether or not such a reaction would be rapid and violent is not clearly known. In this case one has to deal with the chemical kinetics of a heterogeneously reacting chemical system (among other things the probable degree of dispersion of the uranium into the water is not known). Presumably, the rate will depend upon the intrinsic molecular kinetic process in the conventional chemical sense, but it will also depend upon the degree of dispersion of the uranium into the water, the rate at which reacting molecules can diffuse through the uranium oxide layer that would be formed between the uranium and the water, and the degree of turbulent mixing and scrubbing of the two reactants against each other. This latter effect might be generated by the reaction itself. This is clearly a complex problem and a great deal more needs to be learned. However, one can say this: If an exothermic reaction of this type goes to completion, the resulting energy release will nearly always be substantially greater than the energy generated in a preceding nuclear runaway. Thus it is important to determine the possible chemical reactions. A substantial increase in reactor safety can be achieved by the elimination of possible reacting components in the reactor structure.

## SAFE DESIGNS

It seems worth while to summarize the preceding discussion with a few remarks concerning the approach to safe reactor designs. First of all, it is desirable to provide a large negative reactivity coefficient. This can usually be achieved by thermal coupling of the fuel elements to those portions of the reactor which give a substantial reduction to the neutron multiplication when heated. For example, in the case of enriched, water-moderated reactors, close thermal contact between the fuel elements and the moderated water can lead to enough heating and vaporization of the water to reduce the water density in the event of a nuclear runaway, and shut down the reactor before serious damage is done. Successful tests of this sort have been made.[10] A design of this sort must be thought through carefully in order to make sure that enough heat transfer surface is available and rapid enough heat flow will take place to shut down a machine during an accident.

The control system must be carefully designed so that in the event of too high a power level, too high a rate of rise of power level, a serious reduction in coolant flow, or any major failure of fuel elements, the reactor will shut down in a time interval small enough to minimize damage. All potentially dangerous failures should be monitored by instruments and control channels leading to shut-down or "scram." These monitors and channels should be at least in duplicate, independent of each other, and preferably of different types. In addition, particular care should be taken that a failure cannot put the controls out of operation.

In order to prevent a delayed heat accident, it is important that enough natural convection heat transfer can take place in the overheated core to dispose of the delayed heat, perhaps just into the ground. Even if the structure is damaged, one must try to keep the temperature lower than that temperature which would start a substantial pressure rise in the reactor structure. In that case, the fission products may be kept inside the reactor shield. This means that some coolant contained in the core should have a large surface to which it can transfer heat by natural convection or by boiling convection, and that this degree of cooling should be sufficient to keep the bulk of all volatile materials below their boiling points. It may be remarked that boiling heat transfer is known to be especially efficient, so that in any event there will tend to be a ceiling put on the temperature rise at about the boiling temperature of the original coolant employed. This in turn implies that by appropriate construction one may limit the pressure inside the shield to a few atmospheres. Thus it may be rather easy to make sure that the fission products are kept inside the shield.

Finally, the problem of chemical reaction among reactor components can often be minimized. For example, already-reacted components might be used in some cases. Uranium oxide rather than uranium metal in an air- or water-cooled reactor may serve as an example.

The other general conclusion that the Safeguard Committee has come to is that explosive hazard in reactor accidents is minor, at least for people not at the reactor site. Indeed, for many reactors, it appears unlikely that there will be much mechanical violence external to the reactor shield. For this reason, a gas-tight building, or a moderately gas-tight building which may confine the fission products during a cooling period and from which the fission products are exhausted into scrubbers and out a high stack, may serve to prevent the spread of fission products following a reactor accident. For some reactors the confining building will have to be a gas-tight pressure vessel. Safe-design procedures represent an important field of nuclear reactor development.

## ADMINISTRATIVE CONTROL

Although good administrative control of the reactor does not lead to the same degree of confidence in the good behavior of the machine that intrinsic gremlin-free built-in stability does, nevertheless, good administrative control does enhance the safety and reliability of reactor operation. Indeed, good administrative control is mandatory for those people who have an economic stake in the reactor. From the point of view of public hazard, careful reactor operation and maintenance makes it very much less likely that there will be a reactor accident.

However, this administrative control should not start just when the reactor is put into operation. Throughout the design and construction of the reactor, thorough supervision, careful design for reliability, and thorough testing of all reactor components including coolant system, off-gas systems, shims, safety and control mechanisms, and all control and operating instrumentation should be carried through. All these components should be given systematic and thorough shakedown testing before the reactor is put into operation and before it becomes radioactive so that modification, correction, and maintenance can be done with less difficulty. Indeed, it is extremely difficult to emphasize how important it is to have complete, thorough, systematic shakedown of all portions of reactor control and instrumentation.

In the design of the reactor, careful attention should be given to the problem of maintenance after it is placed in operation. It should be possible to enter all instrument areas, most of the control areas, and obviously the central control room, after the machine has started up and been operating for some time. Fuel-element failure, a continuing problem, may allow radioactivity to enter portions of the reactor structure which normally would be expected to be radiation-free. This should be taken into account in the original design.

Once the reactor is placed into operation, continuing close supervision is essential. Maintenance procedures should be carefully followed and maintenance checks should be scheduled in an appropriate way. The period of reactor startup is a particularly critical one, and should be followed very closely. Reactor loading and unloading are delicate operations, particularly the unloading of now-radioactive fuel elements. Startup of a reloaded reactor must be carefully considered since the reactivity may have been affected by a new fuel loading. The normal, or routine, day-to-day operation requires close supervision so that troubles may be detected at an early date and corrective measures taken. Clearly, careless operation of the controls may lead to a supercriticality accident and the manipulation of the controls should be carried out only by people who are thoroughly familiar with the characteristics of the reactor and its associated equipment.

If a reactor is employed as an irradiation facility, it is possible for experiments to give rise to sudden changes of reactivity. Experiments should be planned, the plan reviewed by the administrative staff, and suitable emergency procedures decided upon, before inserting experiments into the reactor.

Finally, there is one phase of the administratively controlled reactor which is usually taken for granted but may require a word or so: The careful accounting for all materials which have been irradiated in the machine. There is usually available a number of test holes in which experimental irradiations may be carried out. The samples so irradiated can be highly radioactive and should not be allowed to accumulate unduly, or to be lost, or to be handled in an irresponsible manner.

## CONSEQUENCES OF AN ACCIDENT

We believe the following discussion outlines the main features which can make a nuclear incident dangerous. In the event of a reactor accident, there will probably result a release of radioactive material from the reactor. Operating personnel may be seriously injured or perhaps even killed. The reactor itself may be damaged beyond repair or recovery. The reactor building and its associated equipment are very likely to be heavily contaminated and indeed, it may not be possible to clean up the building sufficiently to put it into operation again. Design of the building so that possible cleanup operations are as easy as possible is desirable.[11] Smooth, clean surfaces, perhaps clad in stainless steel, would make cleanup operations easier. Finally, radioactive materials can escape from the reactor site altogether. Fission products may be carried in the wind and spread over adjacent populated areas and constitute an acute hazard. Radioactive material may escape into the ground and be carried by the percolating ground water to adjacent rivers or other water supplies. Although a great deal needs to be known about the character of radioactive material that might escape from a reactor, whether it is in large or small particles, whether it is indeed gaseous, whether it would rise high into the air or seep slowly along and into the ground, nevertheless, some notion of the possible spread of the hazard could be obtained by study of the meteorology, and hydrology at the reactor site.[12,13,14] It is desirable, for example, to have the prevailing wind to blow from the reactor to uninhabited areas. It is also desirable to have the reactor site not be located on a main watershed. From the point of view of the hazard alone, it is of course desirable to have the reactor site far from populous or vital industrial areas. It will not always be possible to obtain this remote location and still obtain economic utility from the reactor. For this reason, the Safeguard Committee is continuing to emphasize the importance of safe reactor designs, the development of contained fuses to minimize the possibility of a runaway accident, and the use of gas-tight containing vessels and buildings.

Perhaps it is important again to emphasize the degree of public hazard that might follow a reactor accident. Assuming that good luck prevails and no one is killed, it may nevertheless be necessary to evacuate a large city, to abandon a major watershed, and very probably it would be necessary to make the reactor site itself a forbidden area for some years to come.

Despite all these possible dire consequences, it is the belief of the Advisory Committee on Reactor Safeguards that nuclear reactors will soon start to produce substantially increasing material benefits for

humanity. We believe that useful electric power in large quantities can be generated by nuclear reactors. It is our concern that rapid progress shall be made but that enough caution be observed so that no catastrophic event will delay the fruition of reactor development.

## APPENDIX. THE ADVISORY COMMITTEE ON REACTOR SAFEGUARDS TO THE UNITED STATES ATOMIC ENERGY COMMISSION

The Advisory Committee on Reactor Safeguards was formed by combining the Reactor Safeguard Committee and the Industrial Committee on Reactor Location Problems. At this time members are: M. Benedict, Massachusetts Institute of Technology; H. Brooks, Harvard University; W. P. Conner, Jr., Hercules Powder Company; R. L. Doan, Phillips Petroleum Company; H. Friedell, Western Reserve University; I. B. Johns, Monsanto Chemical Company; C. R. McCullough, Chairman, ACRS; M. M. Mills, University of California Radiation Laboratory; K. R. Osborn, Allied Chemical and Dye Corporation; D. A. Rogers, Allied Chemical and Dye Corporation; C. R. Russell, Secretary, ACRS; R. C. Stratton, Travelers Insurance Company; E. Teller, Department of Physics, University of California; H. Wexler, United States Weather Bureau; and A. Wolman, The Johns Hopkins University.

The Reactor Safeguard Committee was formed in 1947, and the Industrial Committee on Reactor Location Problems was formed in 1949. The following were also associated with these committees for prolonged periods: Cmdr. J. Dunford, US Atomic Energy Commission; Col. B. Holzman, US Air Force; J. Kennedy, Washington University, St. Louis; F. Seitz, University of Illinois; G. Weil, formerly Division of Reactor Development, US AEC; and J. A. Wheeler, Princeton University.

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# Experimental Determinations of the Self-Regulation and Safety of Operating Water-Moderated Reactors

By J. R. Dietrich,* USA

## INTRODUCTION

One of the important characteristics of a nuclear reactor is the degree of hazard which it creates in the surrounding area. If the usefulness of reactors, either for research or for power production, is to be exploited effectively, the hazard must be minimized, since isolation of the reactor compromises its utility and increases its cost. It is, therefore, important to find means for evaluating the hazards of specific reactors and methods of improving the safety of reactors in general.

The ultimate question in an evaluation of reactor safety is the question of what will happen if the reactor is inadvertently made supercritical and allowed to "run away" without any artificial limitation of its power. For, although safety devices which impose artificial limitations will certainly be provided for in the reactor design, the possibility of their failure as well as the consequences of their finite speed of operation must be recognized.

In general it can be said that the reactivity of a reactor will be related to its power level once the power has become sufficiently high to cause significant changes in the temperatures of the reactor parts. At moderate power levels this dependence can be such as to cause the reactivity either to decrease or to increase with power level, depending on the design of the specific reactor, but eventually at some power level any runaway reactor will become subcritical, through some degree of disassembly of itself if not by other means (Fig. 1). The safety question has to do with how violent the energy release becomes before the eventual shutdown is achieved.

The more important ways by which increasing power can cause a reactor to lose reactivity are by expansion of the fuel, by heating and expansion of the moderator, and, if strong resonance absorbers are present, by Doppler broadening of the resonances. In many cases the unknowns in the magnitudes of the applicable effects and in their speeds of operation make safety evaluations quite uncertain.

The most straightforward way of evaluating the unknown aspects of the reactor shutdown process is by observing experimental runaways of actual reactors. Some experiments of this kind on two reactor types which have particularly favorable power-limitation characteristics, the solid-fuel, water-moderated reactor and the water-moderated homogeneous reactor, are reported here. In addition to the instrumental measurements reported here, motion picture records, which add materially to the information on the safety characteristics of the reactors, are available.

Homogeneous reactors can, in general, be made to have negative temperature coefficients of reactivity. The negative coefficient results primarily from thermal expansion of the fuel solution, which decreases not only the density of the moderator, but that of the fuel as well. The coefficient is quite large for small reactors with high neutron leakage. Furthermore, since the heat of fission is liberated directly in the fuel solution, the action of the negative coefficient is very rapid, and insofar as such reactors can limit their power by temperature coefficient alone, they can be expected to be quite effectively protected against destructive runaways.

The solid-fuel water-moderated reactor may get a certain degree of power limitation from the Doppler coefficient if it contains a large fraction of $U^{238}$. Beyond this, power limitation comes from the moderator temperature coefficient and expulsion of moderator from the reactor core by the formation of steam at the hot fuel element surfaces. The bulk temperature of the moderator does not change rapidly enough

* Argonne National Laboratory. Including work by C. C. Bigelow—Pratt and Whitney Aircraft Corp., R. A. Cameron—Argonne National Laboratory, B. C. Cerutti—Argonne National Laboratory, R. E. Coté—Pratt and Whitney Aircraft Corp., W. R. Davis—University of Washington, J. J. Dickson—Argonne National Laboratory, J. R. Dietrich—Argonne National Laboratory, J. M. Harrer—Argonne National Laboratory, R. O. Haroldsen—Argonne National Laboratory, P. R. Kasten—Oak Ridge National Laboratory, L. D. P. King—Los Alamos Scientific Laboratory, N. L. Krisberg—US Atomic Energy Commission, D. C. Layman—Argonne National Laboratory, H. V. Lichtenberger—Argonne National Laboratory, W. C. Lipinski—Argonne National Laboratory, R. N. Lyon—Oak Ridge National Laboratory, M. Novick—Argonne National Laboratory, V. K. Paré—Oak Ridge National Laboratory, R. L. Ramp—Argonne National Laboratory, W. M. Sandstrom—University of Washington, R. J. Schiltz—Argonne National Laboratory, O. A. Schulze—Argonne National Laboratory, A. R. Snider—California Research and Development Co., G. H. Stonehocker—Argonne National Laboratory, J. A. Thie—Argonne National Laboratory, A. J. Ulrich—Argonne National Laboratory, S. Untermyer—Argonne National Laboratory, S. Visner—Oak Ridge National Laboratory, G. K. Whitham—Argonne National Laboratory, C. Zabel—Los Alamos Scientific Laboratory, W. H. Zinn—Argonne National Laboratory, and C. B. Zitek—Commonwealth-Edison Company.

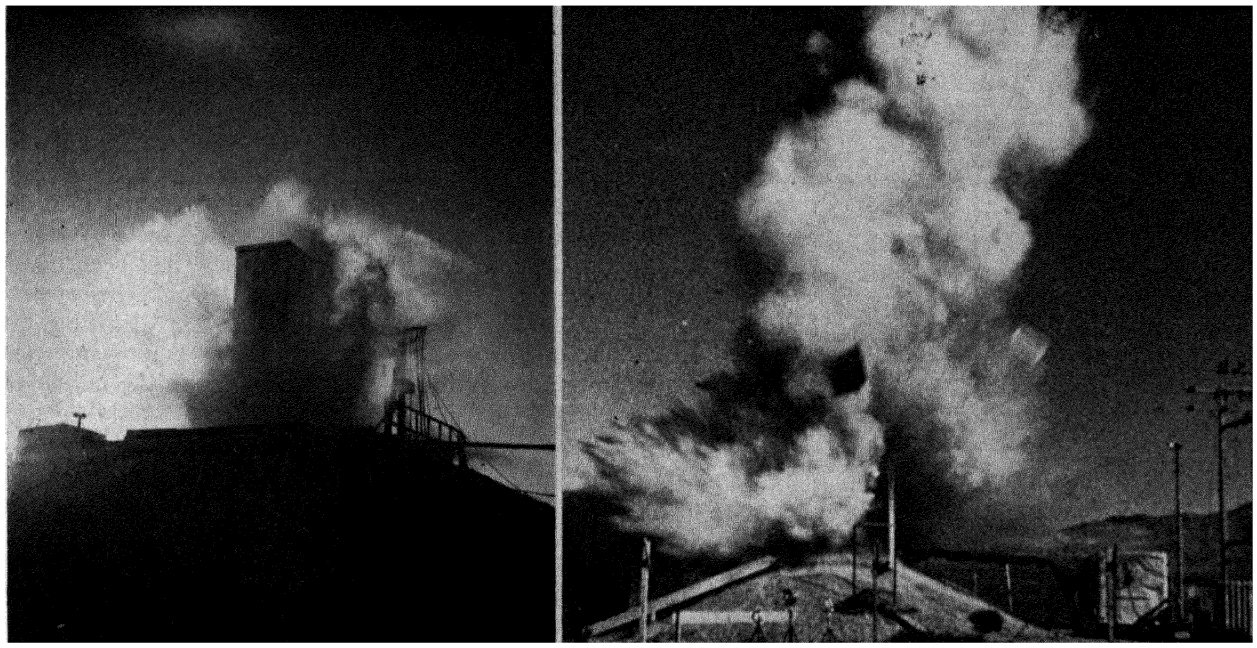


Figure 1. Effects of large reactivity additions to a water-moderated reactor; left, reactor shutting itself down safely by water expulsion after being made supercritical by 2.1% $k_{eff}$; right, reactor destroyed by melting of fuel plates after being made supercritical by 3.3% $k_{eff}$

to make the temperature coefficient effective against rapid power increases, but early laboratory experiments by Untermyer and later ones by West, Weills, Hooker, and Schiltz showed that the expulsion of moderator by steam can be very rapid.[1,2] The reactor experiments, initiated at the suggestion of Untermyer, confirmed the effectiveness of the process. They were begun in the early summer of 1953 by Argonne National Laboratory as part of an experimental program on boiling water-moderated reactors. The effectiveness of boiling as a safety process was proved, and the following year the severity of the experiments was increased to the point of planned destruction of the reactor. A new reactor was built in 1954 with which similar experiments were made at elevated pressures.

In July and August of 1953 safety experiments were run on the Los Alamos "Supo" reactor by Lyon, Kasten, and others of Oak Ridge National Laboratory and King, Zabel, and others of Los Alamos Scientific Laboratory. Shortly thereafter a program of more drastic safety experiments was run on the Homogeneous Reactor Experiment at Oak Ridge National Laboratory by Paré, Visner and others. Although these experiments were not carried to conditions as severe as those used for the experiments on solid-fuel reactors, they demonstrated a high degree of inherent self-limitation of power in the homogeneous systems.

## EXPERIMENTS WITH HOMOGENEOUS REACTORS

The runaway behavior of the non-boiling homogeneous reactor, whose power is limited by the temperature coefficient of reactivity, is the most straightforward of those investigated. Reactors of this type may have very high negative temperature coefficients of reactivity which result primarily from the expansion of fuel solution out of the reactor proper as the temperature increases.

Figure 2 is a diagram of the Homogeneous Reactor Experiment, which was used for the tests described here. The core is normally full of the fuel solution of enriched uranium in water. If the solution heats and expands, the displaced fraction of the solution is accommodated by the pressurizer tank, where it contributes nothing to reactivity. The temperature coefficient of the reactor was about $-0.1\%$ $k_{eff}$ per degree C. The volume of the core was 50 liters, and the effective prompt neutron lifetime was $7.5 \times 10^{-5}$

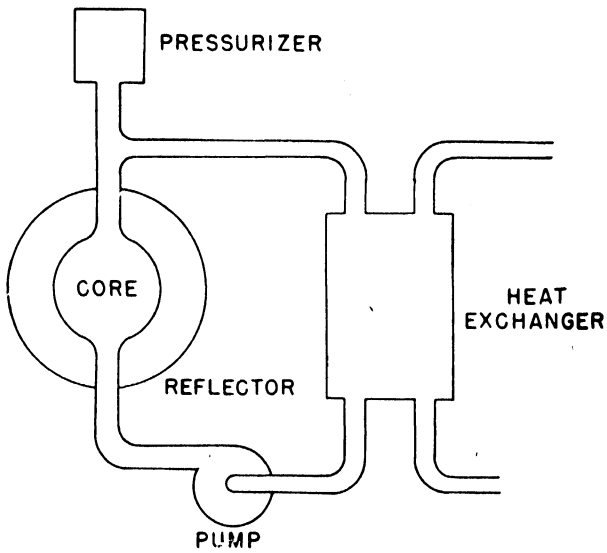



Figure 2. Diagram of homogeneous reactor system

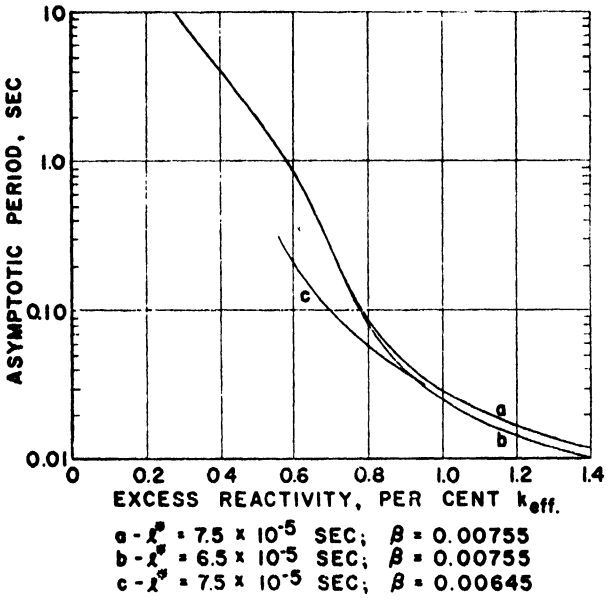



**Figure 3. Relation between excess reactivity and reactor period**

sec. The relation between excess reactivity and asymptotic reactor period is given by curve *c*, Fig. 3. The deficiency in delayed neutron fraction results from the circulation of the fuel solution. The reactor was pressurized to a pressure of 1000 psi.

If reactivity is added to such a reactor according to some law of time variation, $k_a(t)$, resulting in a period which is short compared to the residence time of the fuel solution in the reactor, then the law which expresses the variation of reactivity with time is simply

$$k(t) = k_0 + k_a(t) + C \int_0^t P(\tau)d\tau \qquad (1)$$

where $C$ is a characteristic constant of the reactor which includes the heat capacity and the (negative) temperature coefficient of reactivity, and $P(\tau)$ is the instantaneous reactor power. This equation must, of course, be coupled with the usual differential equations characteristic of the kinetics of the neutron chain reaction to specify the variation of reactor power with time. If the power increase is fast enough that the compressibility of the fuel solution is important, still further relations must be included to describe the dynamics of the system. Kasten has treated these considerations at some length.[3] Regardless of the complications which may occur in specific reactors, the safety experiments which have been made indicate that the fundamental situation is reasonably well understood.

The reactor used for the experiments was not provided with special means for increasing reactivity rapidly, and hence the experimental situations were those relatively complex ones which would be characteristic of practical reactor accidents. Reactivity was increased experimentally by several methods: withdrawal of a weak control rod; increase of fuel concentration; raising of the reflector level; rapid cooling of the circulating fuel; and pumping of precooled fuel solution into the reactor proper. The latter method gave the largest and fastest reactivity changes, and the results obtained by it are the ones reproduced here.

The experiments were made by stopping the circulating pump (Fig. 2), cooling the fuel solution in the heat exchanger to about 100°C, and then restarting the pump to inject the cooled solution rapidly into the reactor core, which had been maintained at a temperature of about 180°C. The severity of the experiment was adjusted by adjusting the initial power level of the reactor before the cold solution was injected. Figure 4 shows the measured power variations for two different initial power levels. Naturally, the lower initial level allows the greater increase in reactivity before the reactor begins to shut itself down and results in the higher maximum power. The temporary power decrease immediately after the pump starts is a result of the initial loss of delayed neutron emitters as the old fuel solution is displaced by the new.

This type of power transient can be characterized by specifying both the rate of addition of reactivity by the inflow of cold solution and the minimum reactor period reached during the transient. Figure 5 contains a set of theoretical curves giving the maximum power reached as a function of the minimum period reached during the transient, with rate of reactivity addition as the parameter. The experimental results are plotted in the same figure, with the rate of reactivity addition indicated by each. In view of the relatively complex experimental conditions, the agreement with theory is satisfactory.

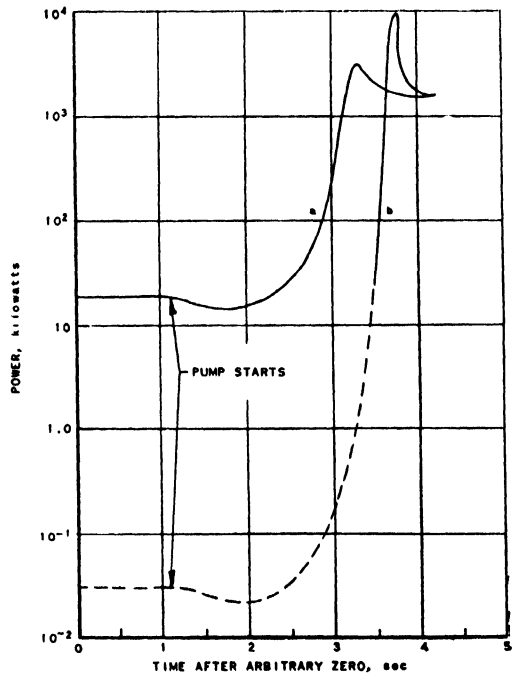
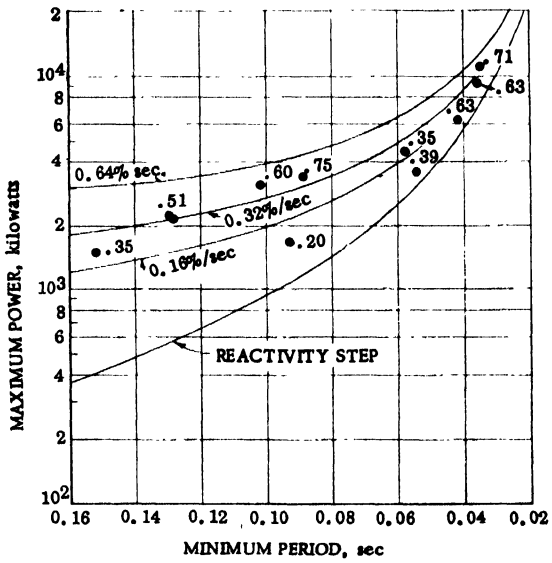



**Figure 4. Power variations during injection of cold fuel solution in homogeneous reactor experiment**




THE CURVES ARE COMPUTED FOR VARIOUS RATES OF REACTIVITY ADDITION. THE RATES OF ADDITION USED ARE MARKED BY THE EXPERIMENTAL POINTS.

**Figure 5. Maximum power as a function of minimum period for homogeneous reactor power transients**

The experiments indicated, further, that for the conditions tested the reactor would settle down to steady operation after the transient. The power level would then be determined simply by the rate of heat removal from the heat exchanger: the reactor power would be self-regulating via the temperature coefficient of reactivity.

The Supo reactor differs from the Homogeneous Reactor Experiment in that its fuel solution is not circulated; heat is removed by a cooling coil in the reactor vessel. Furthermore, the reactor vessel is not completely full of solution. The temperature coefficient is nevertheless strongly negative: about $-0.024\%\ k_{eff}$ per degree C. The reactor has been described by King.[4]

The safety experiments were made in connection with an investigation of boiling operation of the reactor. The reactor was operated at powers of several kilowatts as a boiler, and the power under this condition fluctuated, but was self-regulating. Sudden reactivity additions up to about $0.4\%\ k_{eff}$ were made, under conditions of both boiling and non-boiling operation. In both cases the reactor power was self-limiting, but the excursion was terminated more rapidly under boiling conditions. The experiments indicated that there is no very long time delay in the formation of steam bubbles in a homogeneous reactor once the solution has reached saturation temperature. Once this is known, it is to be expected that steam would be by far the more effective shutdown agent for long-period power excursions at atmospheric pressure. For whereas about 5000 calories of heat are required to produce 1 $cm^3$ of effective void in liquid water by thermal expansion, only about 0.3 calorie is required to evaporate sufficient water to produce 1 $cm^3$ of steam at atmospheric pressure. It is by no means evident, however, that the same situation would hold for very short period transients or at very high pressure.

## EXPERIMENTS ON SOLID FUEL REACTORS

In the experiments made with solid-fuel, water-moderated reactors the expulsion of water by steam formation was the important process in transient limitation of the power. Since a quantitative theory of the process has not been developed, it is necessary to present the results and the experimental conditions in some detail.

The experiments were made in two different reactors which were also used for investigation of the steady-state characteristics of boiling reactors. The pertinent differences between the two reactors lay in their core characteristics. These differences will be described, but differences in the mechanical details of the two reactors will be ignored.

### Description of the Reactors

Figure 6 is a cutaway drawing of the first reactor, which was constructed outside and which was operated remotely from a control station half a mile away. The reactor tank was contained in a larger shield tank of ten-foot diameter which was sunk part-way into the ground and had earth piled around it for additional shielding. Adjacent to the shield tank was a pit with concrete walls in which was installed equipment for filling and emptying the reactor and shield tanks, and for preheating the water in the reactor tank. The reactor tank, four feet in diameter and about thirteen feet high, contained the reactor core, which consisted of an adjustable number of plate-type fuel elements held at the bottom by a supporting grid and at the top by a removable cover grid.

In operation the reactor tank was filled with water to a height of three to four and one-half feet above the top of the core; this water constituted the reflector, moderator, and coolant. The shield tank was filled with water only when the reactor was shut down.

The reactor contained five cadmium control rods which were operated by drive mechanisms located in the rectangular housing above the shield tank. The connection from the mechanism to the rods was through spring-loaded magnetic couplings. These couplings could be released in unison or individually, allowing the rods to drop freely downward under the acceleration of the springs plus gravity. When released, the center control rod dropped out of the reactor core to apply the excess reactivity used for the experiments. The other four rods when released dropped into the reactor core to terminate the experiments. Each rod traversed the length of the core in about 0.2 sec.

The fuel elements were made of aluminum-clad, aluminum-uranium alloy plates, of 60 mils total thickness, fastened into aluminum side plates to make boxes roughly 3 inches square. Figure 7 is a

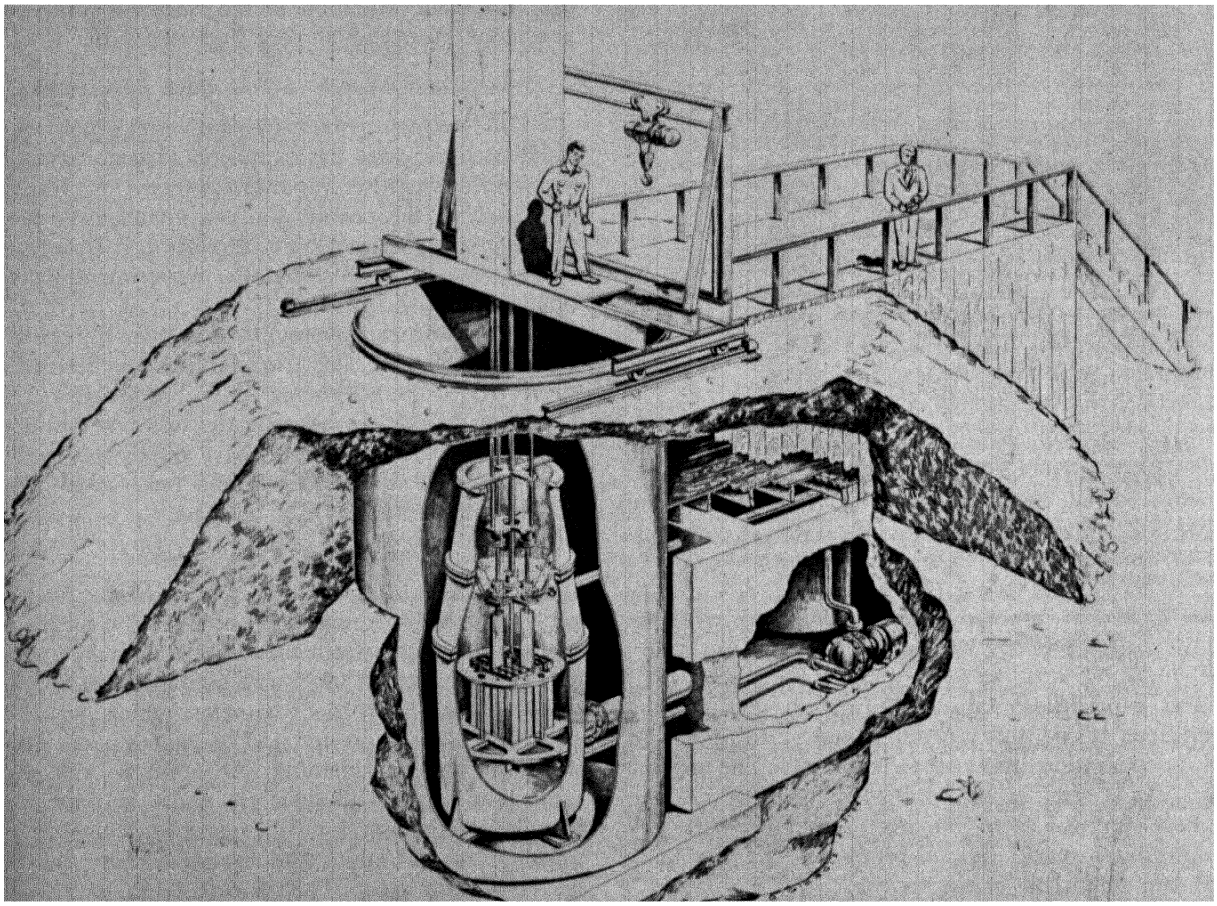


Figure 6. Cutaway drawing of reactor I

drawing of a fuel element for one of the reactors. The other reactor used elements of identical outside dimensions, but each element contained only ten of the fuel plates.

The two reactors will be designated I and II. Each reactor was loaded, for any given experiment, with the number of fuel elements which would give a convenient amount of reactivity. Typical loadings for the two reactors are diagrammed in Figure 8. Reactor II contained several elements of higher uranium content around its periphery to flatten the power distribution.

The relationship between asymptotic reactor period and excess reactivity for the reactors is given in Fig. 3 (curves *a* and *b*). For larger excess reactivities the period ($\tau$) is given by

$$\tau = \frac{l^*}{k_{ex}(1-\beta)-\beta} \qquad (2)$$

where $\beta$ is the total delayed neutron fraction and $l^*$ the effective neutron lifetime. Other characteristics of the two reactors are summarized in Table I.

### Typical Transient Behavior of the Reactors

The experiments were made by the following procedure. The reactor water temperature was adjusted to the desired value, and the reactor was made critical at a low power (about 1 watt) by appropriate positioning of the control rods. The center control rod was then dropped out of the reactor core. The initial power was sufficiently low and the speed of rod ejection was sufficiently high so that in almost all cases the rod was completely out of the core and the reactor period reached its stable value before

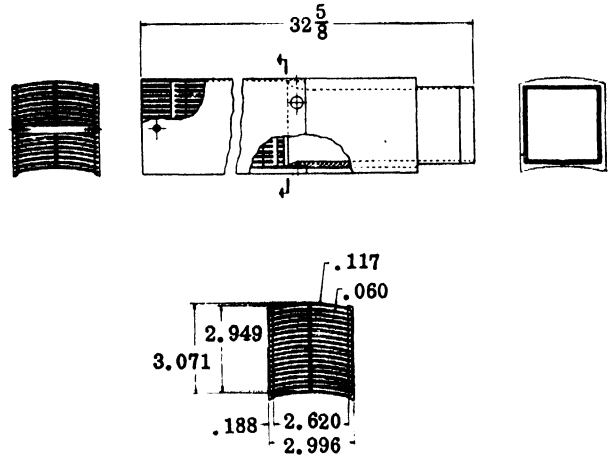
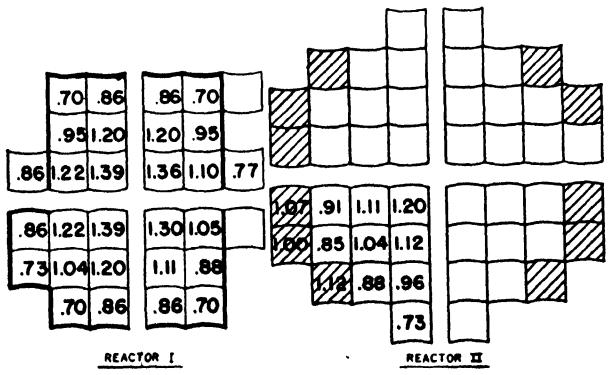



Figure 7. Standard fuel assembly

the reactor power had risen high enough to produce significant thermal effects. The power was allowed to continue to rise until the formation of steam in the reactor core reduced the reactivity below criticality and caused the power to fall to a low value. After it was evident that the power had been safely limited by the formation of steam, the remaining four control rods were dropped into the reactor to terminate the experiment, referred to as a power excursion. By proper adjustment of the number of fuel elements in the reactor core and of the positions of the four outer control rods, the reactor could be made critical with the center control rod inserted to any desired degree in the core. The magnitude of excess reactivity applied by ejection of the center rod could thus be adjusted at will.

Figure 9 is a reproduction of a typical chart from the multichannel magnetic oscillograph which recorded the data on the experiments. In this case the applied excess reactivity was 1.4% $k_{eff}$ and reactor I was used. The neutron flux (proportional to reactor power) was recorded over about three decades by three different neutron-sensitive ion chambers working through logarithmic amplifiers. The stable reactor period ($\tau$) is indicated by the three ion chamber records as 0.0096, 0.0107, and 0.0109 second, respectively. The temperature of one of the fuel plates, which was situated at roughly the highest flux position in the core, is recorded by two fast-response thermocouples. Both of the couples were located near the position of maximum neutron flux; one was installed on the surface of the plate, the other at its central plane. There is little difference between the two temperatures, because of the high thermal conductivity of the thin plate.

The ion chambers, which were calibrated in terms of absolute power by thermal methods, indicate that the reactor power reached a maximum value of 220 megawatts before the formation of steam checked the rise. Further generation of steam reduced the reactivity below the critical value, and caused the power to decrease very rapidly to a value of about 0.2 megawatt.




EACH BOX REPRESENTS A FUEL ELEMENT.

THE SHADED BOXES IN REACTOR II CONTAINED FUEL PLATES OF HIGHER FUEL LOADING (SEE TABLE I)

THE NUMBERS IN EACH BOX INDICATE THE RATIO OF THE TOTAL POWER PRODUCED IN THAT ELEMENT TO THE AVERAGE POWER PER ELEMENT. THE MEASUREMENTS ON REACTOR I WERE MADE WITH A SLIGHTLY DIFFERENT LOADING THAN THAT DIAGRAMMED.

THE HEAVY OUTLINE IN REACTOR I ENCLOSES THE CLEAN COLD CRITICAL LOADING. REACTOR II WAS CRITICAL AT ROOM TEMPERATURE WITH A 6 BY 8 ELEMENT ARRAY OF THE MORE LIGHTLY LOADED ELEMENTS (TABLE I)

THE AXIAL MAXIMUM/AVERAGE POWER RATIO IS 1.3 FOR BOTH CORES

**Figure 8. Core loading diagrams for reactors I and II**

**Table I. Comparison of Reactors I and II**

| | *Reactor I* | *Reactor II* |
|---|---|---|
| Ratio, $\frac{\text{volume aluminum in core}}{\text{volume water in core}}$ | 0.626 | 0.422 |
| $U^{235}$ content per fuel element | 138.6 gm | 93.4 gm or 157.3 gm |
| Number of fuel plates per element | 18 | 10 |
| Plate spacing (between center lines) | 0.177 inch | 0.324 inch |
| Measured reactivity loss with temperature increase: | | |
| 80°F to 200°F | 0.82% $k_{eff}$ | 0.45% $k_{eff}$* |
| 80°F to 280°F | 1.93% $k_{eff}$ | 0.76% $k_{eff}$* |
| 80°F to 420°F | -- | 1.57% $k_{eff}$* |
| Calculated loss of reactivity caused by replacement of 10% of core water by steam, at 200°F | 2.4% $k_{eff}$ | 1.0% $k_{eff}$* |
| Effective neutron lifetime ($l^*$) | $6.5 \times 10^{-5}$ sec | $7.5 \times 10^{-5}$ sec* |

* These values apply to the case in which the reactor is loaded only with fuel elements of low uranium content.

Once the initial power excursion has been checked by boiling in the reactor the specific power variation depends both quantitatively and qualitatively upon the amount of excess reactivity to which the reactor was initially subjected, and upon the bulk temperature of the reactor water. Figure 10 contains tracings from excursion records for reactor I similar to those of Fig. 9, but of longer duration. They summarize the typical behavior of the reactor for various amounts of applied excess reactivity when the reactor water is initially at saturation temperature. When the excess reactivity applied is low, corresponding to a reactor period of about 0.03 sec or longer, the reactor power after the initial surge settles down to a relatively steady value in the neighborhood of half a megawatt (top curve, Fig. 10). For this type of excursion the self-regulating characteristic of the reactor operates rapidly enough to stabilize the power at a steady value characteristic of the amount of applied excess reactivity. After the power has reached this steady value, further reactivity could, of course, be applied, and the reactor would continue to operate stably in steady boiling at a higher power.

If the excess reactivity which is applied by ejection of the control rod exceeds that corresponding to a period of 0.02 or 0.03 sec, the initial power excursion is followed by a series of qualitatively similar excursions of smaller amplitude, which occur at intervals of about 1 sec (second curve, Fig. 10). The amplitudes of the successive excursions, although they vary in an irregular manner, have no sustained tendency to increase or decrease. This

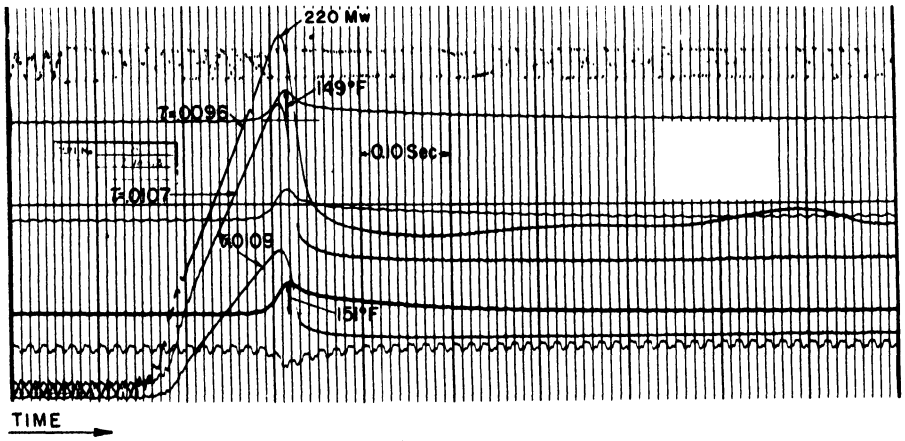



Figure 9. Typical record of power excursion

type of operation will hereafter be referred to as *"chugging."*

When the applied excess reactivity was greater than that corresponding to about a 0.01-sec period, the chugging was no longer observed, and the power after the first surge remained at a low value. This permanent shutdown was no doubt the result of expulsion of sufficient water from the reactor tank to partially uncover the reactor core. The occurrence of this behavior in other reactors would, of course, depend upon the specific design of the reactor in question.

When the applied excess reactivity was increased to about 2% $k_{eff}$ to give periods in the 0.005-sec range, the qualitative behavior of the reactor power remained the same, but the fuel plate temperature did not drop immediately after the power surge (bottom curve, Fig. 10). The fuel plate temperature remained high for almost a second after the power surge and then decreased by small jumps, as though the plate had been blanketed by steam for some time after the power excursion.

Experiments of this type were not carried to periods shorter than about 0.013 sec when the reactor water was cooler than the saturation temperature. With this condition, which will hereafter be called the subcooled condition, chugging was never experienced. In all cases after the initial power excursion the reactor power stabilized at some more or less steady value. Figure 11 is a record of such an experiment on reactor I, in which the initial period was 0.014 sec. Although the reactor power oscillated after the excursion, the oscillation amplitude was very much less than that for typical chugging operation.

Some of the details of the nuclear and thermal behavior of the reactor during a power excursion are illustrated by Figs. 12a and 12b, which apply to the subcooled reactor I for excursions of two different periods. The curves show the time variation of reactor power, on a linear scale, and the fuel plate surface temperature. In these experiments the fuel plate, with thermocouple attached, was coated with a thermally insulating plastic over a section of its length. The temperature of this section of the plate is also included in the figures. This temperature, except for the effect of a small heat loss through the thermal insulation, is proportional to the total nuclear energy liberated in the plate. The point at which there is a sharp deviation between the temperature of the bare section and that of the insulated section evidently marks the beginning of rapid steam formation at the bare plate surface. Up until this time the temperature of the bare plate, like that of the insulated plate, is roughly proportional to the time integral of the reactor power.

Since no single fuel plate can produce sufficient steam to shut the reactor down it is evident that the time relationship between peak reactor power and peak temperature of the bare fuel plate, as well as the ratio of maximum bare plate temperature to maximum insulated plate temperature, will depend upon the local value of power density at the plate location relative to the power density elsewhere in the reactor. The plate used for these measurements was located at or near the maximum power density, and the thermocouples were installed near the point of maximum power density in the plate. Nevertheless, Fig. 12a indicates that for long-period excur-

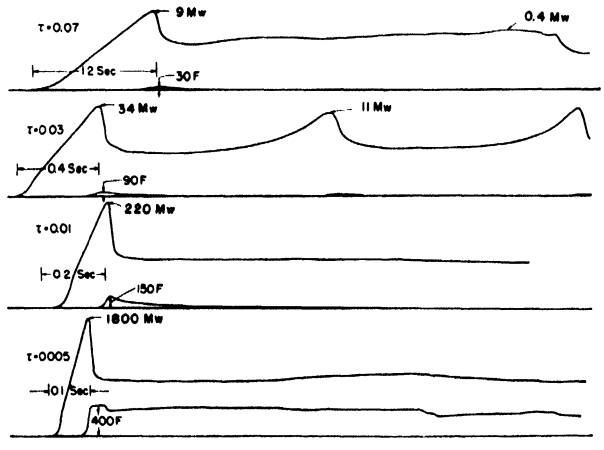



Figure 10. Representative records of excursions at saturation temperature with various excess reactivities

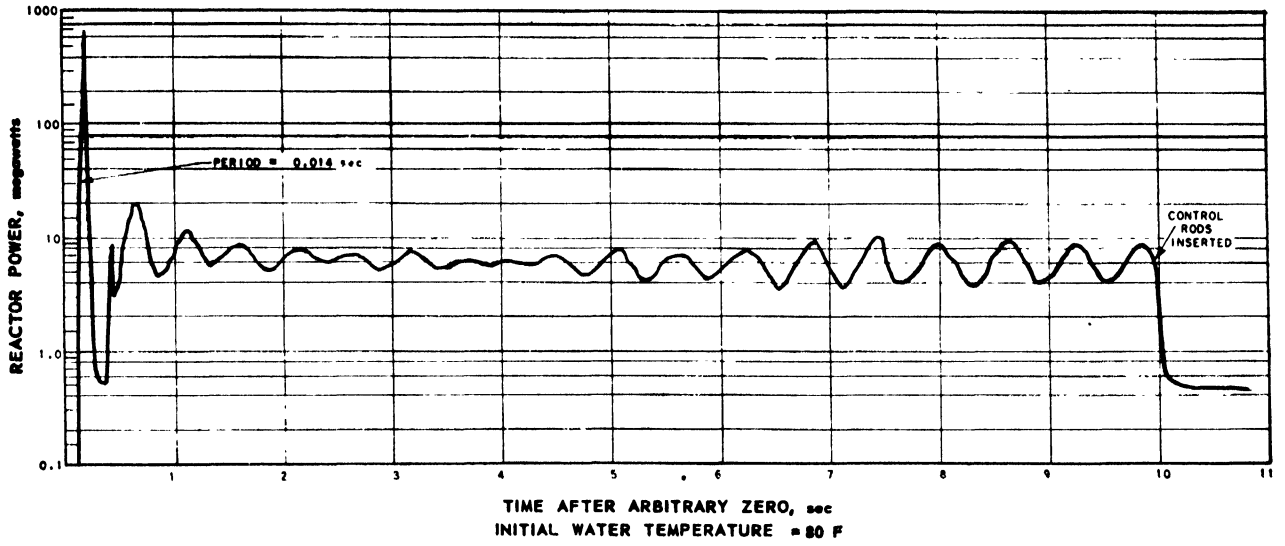



Figure 11. Reactor power variation during 10-second run following initial excursion of 14-millisecond period

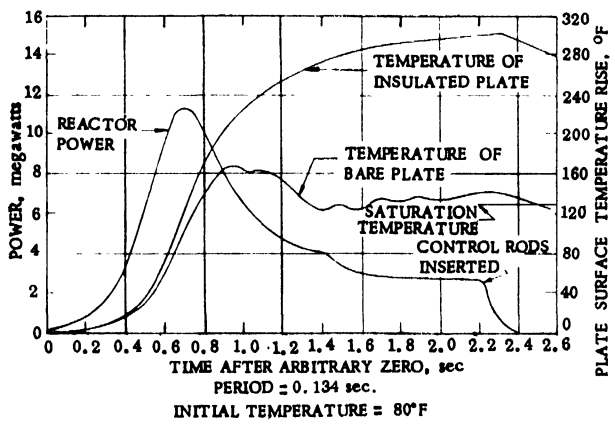
sions other plates were responsible for the first formation of steam, since power began to decrease before plate temperature reached saturation.

### Effects of Several Variables on Power, Energy and Fuel Plate Temperature

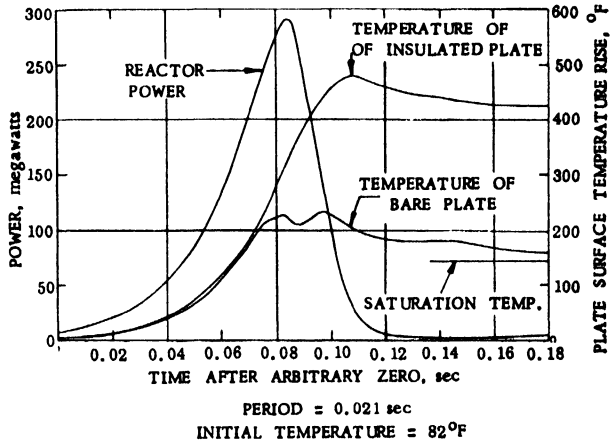
Both the total nuclear energy liberation and the maximum fuel plate temperature reached during a power excursion depend upon the amount of excess reactivity involved in the excursion. In the following presentation, the reciprocal of the stable reactor period is used to characterize the excess reactivity. The relation between the two is given in Fig. 3. In Fig. 13 the total energy† liberated by the power excursion and the maximum fuel plate surface temperature are plotted as functions of the reciprocal period for the case in which the reactor water was at saturation temperature before the excursion began. The shapes of the two curves are quite similar; in fact, the maximum fuel plate temperature rise is roughly proportional to the energy of the excursion for all periods shorter than about 0.03 sec, and the peak temperature corresponds to the temporary storage in the fuel plate of 60 to 70 per cent of the total energy of the excursion.

The energy liberation and maximum fuel plate temperature for the condition in which the reactor was initially at room temperature are given, for reactor I, in Fig. 14. The plotted temperature is the maximum above saturation temperature at atmospheric pressure rather than the total temperature rise. The fuel plate temperature is somewhat higher (and the energy release is much higher) for a given reactor period than in the case of saturated reactor water.

† Throughout the section on solid fuel reactors the term energy will be used to refer to the prompt fission energy which is converted to heat in the fuel plates. The total energy release, including delayed emission and energy converted to heat directly in the reactor water, is about 15% higher. A consistent definition is used for reactor power. By the total energy of a power excursion is meant the energy liberation up to the time of the first minimum in reactor power.




a




b

Figure 12. Power and fuel plate surface temperature rise during power excursions

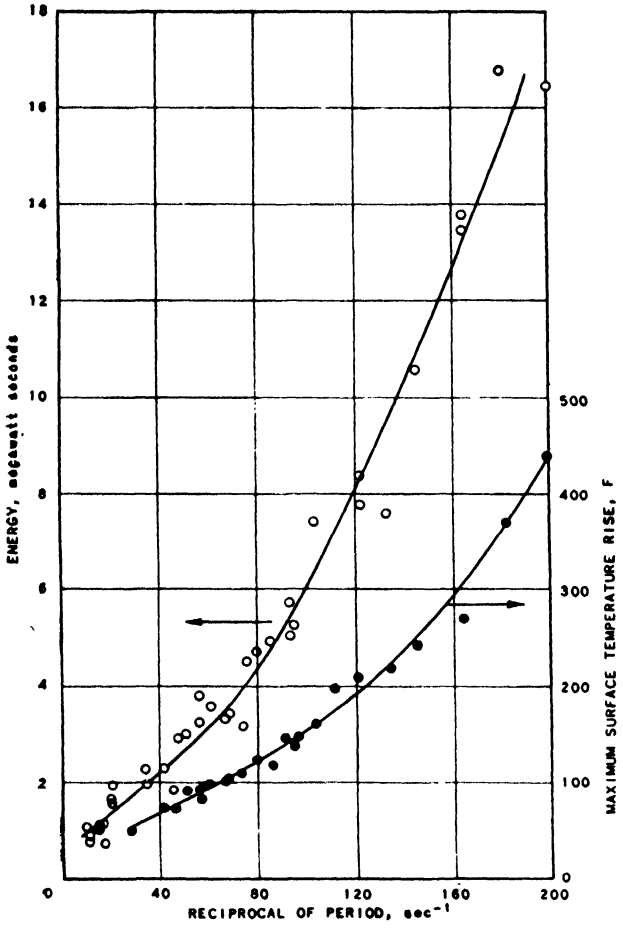



Figure 13. Maximum temperature rise of fuel plate surface and total energy release during power excursions of various exponential periods. Reactor I, at saturation temperature and atmospheric pressure

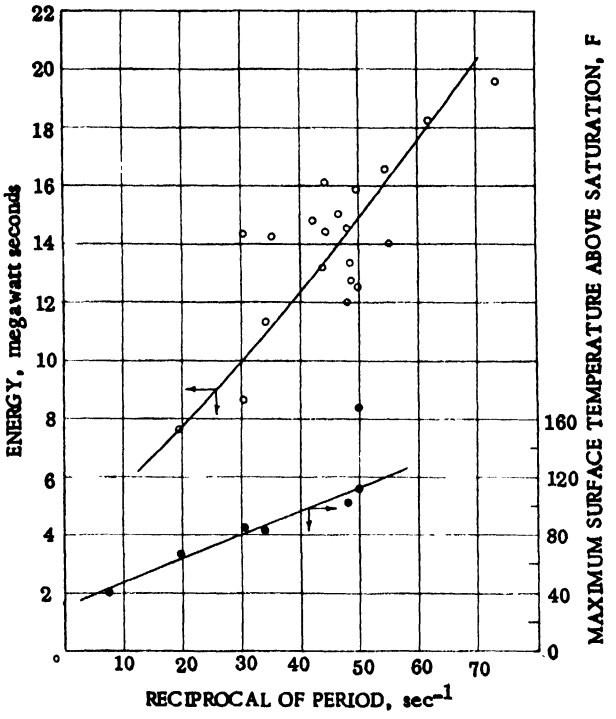



Figure 14. Maximum temperature of fuel plate surface and total energy release during power excursions of various exponential periods. Reactor I, at room temperature (approx. 80°F) and atmospheric pressure

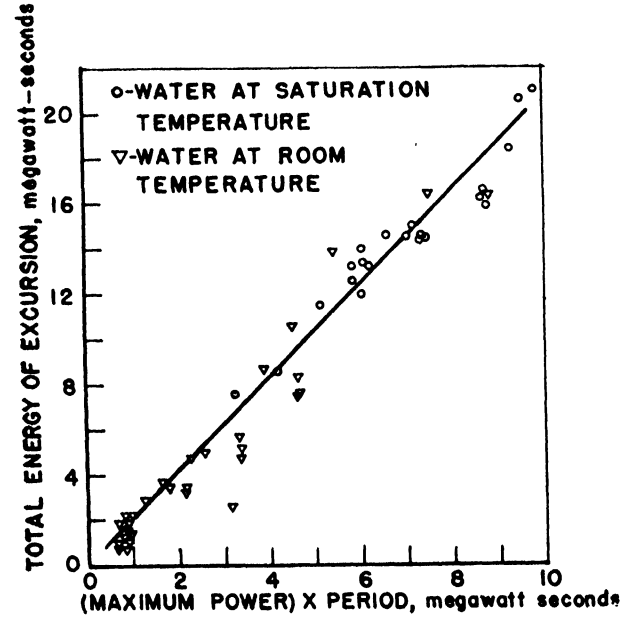
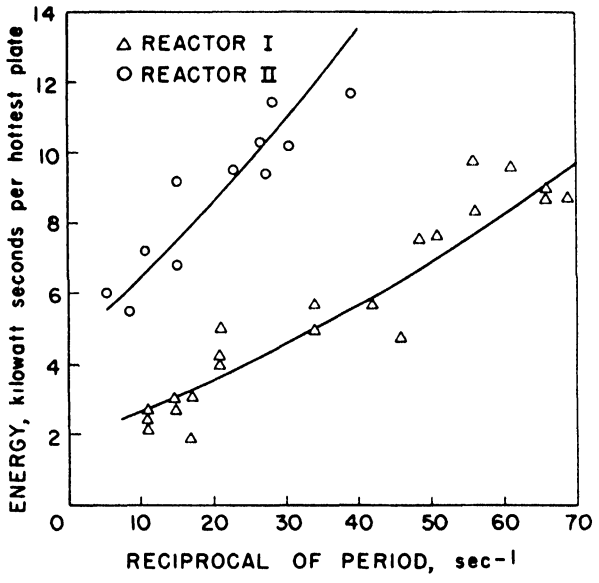



Figure 15. Relation of total energy, maximum power, and period. Reactor I

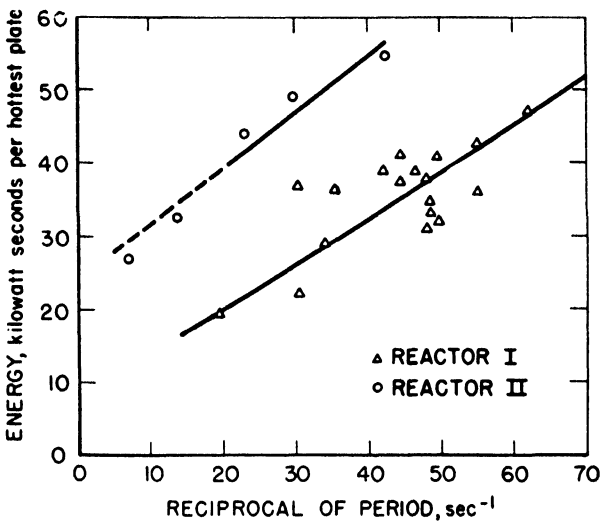
For the cases of both saturation temperature and room temperature water the total energy liberated during the power excursion was nearly proportional to the product of the maximum power and the period. These quantities are plotted in Fig. 15 for the experiments made with reactor I. The slope of the line is about two.

In comparing the behavior of the two reactors, I and II, which differed in core size, it is more informative to compare energy density or energy per fuel plate than to compare total energy. The latter comparison has been made. In Fig. 16 the energy release per fuel plate is compared, as a function of reciprocal period, for the two reactors: Fig. 16a is for the case of saturated reactor water, and Fig. 16b for the case of room temperature water. The dashed portion of Fig. 16b is an upper limit only. The plotted energy release is that of the fuel plate in the position of highest neutron flux. In comparing the behavior of the two reactors, reference should be made to Table I. Note that the volume of water associated with each fuel plate is about twice as great for reactor II as for reactor I. If the energy release per unit volume of water is compared, the values for reactor II are only slightly higher than those for reactor I, despite the fact that the steam coefficient of reactivity is more than twice as great for reactor I as for reactor II.

In reactor II the investigation of power transients was extended to reactor pressures as high as 300 psi. The pressurization of the reactor was by the vapor from the reactor water. Consequently, only the satu-



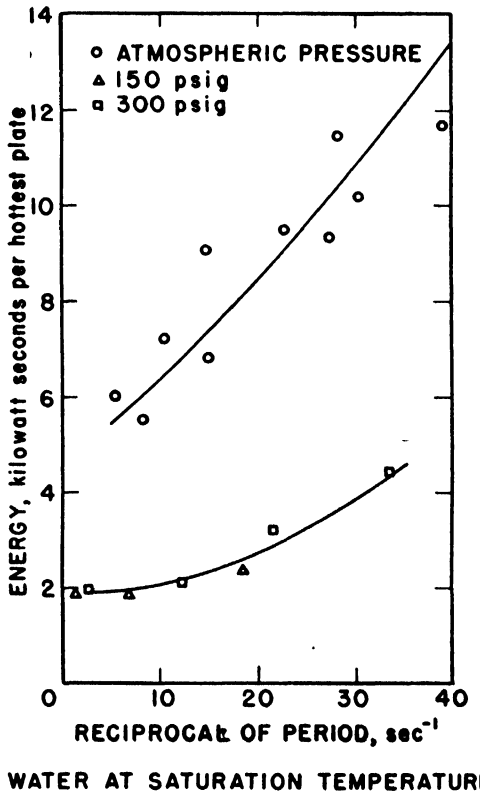

REACTOR WATER AT SATURATION TEMPERATURE AND ATMOSPHERIC PRESSURE




REACTOR WATER AT ROOM TEMPERATURE AND ATMOSPHERIC PRESSURE

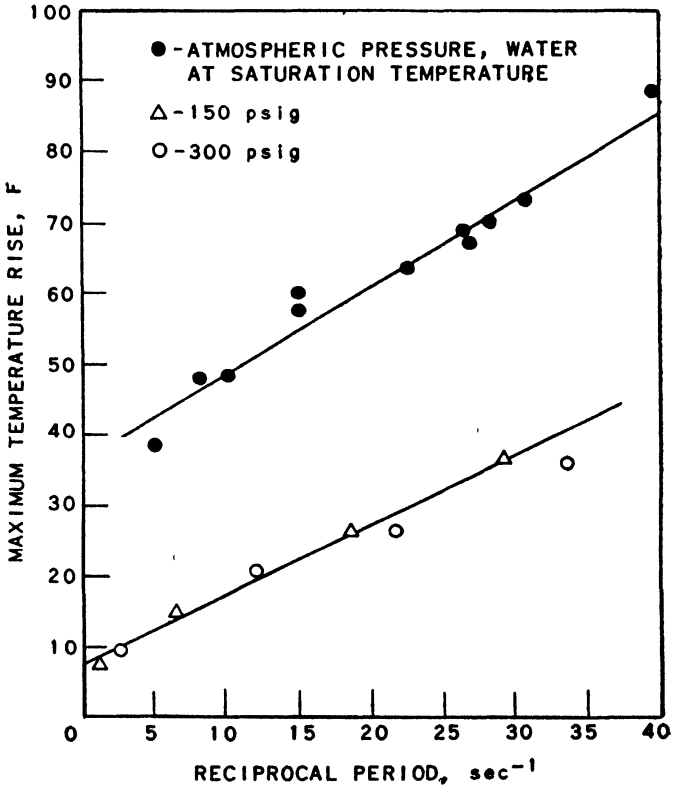
Figure 16. Energy release of hottest fuel plate during power excursions in reactors I and II; (a) left, reactor water at saturation temperature and atmospheric pressure; (b) right, reactor water at room temperature and atmospheric pressure

rated condition could be investigated. The power excursions were run with the reactor tank completely closed. In no case did the pressure in the steam space above the reactor water rise by more than about 5 psi as a result of an excursion. The effect of pressurization is to decrease both the energy released in an excursion of given period and the maximum temperature rise of the fuel plates (Fig. 17).




WATER AT SATURATION TEMPERATURE

REACTOR II




REACTOR III

Figure 17. a, left, energy release of hottest fuel plate during power excursions at different pressures; b, right, maximum fuel plate temperature rise during power excursions: Comparison of unpressurized and pressurized cases

### The Destructive Experiment

In the short period experiments with reactor I at atmospheric pressure, the steam pressure which built up in forcing the water rapidly from the reactor resulted in permanent deformation of the fuel plates. Because of this effect it was not possible to extend the experiments to periods shorter than about 0.005 sec without damaging the reactor to the point where it became unusable. Despite this mechanical damage the maximum temperatures reached by the fuel plates did not approach the melting temperature. It was decided that the reactor, which by this time had fulfilled its other purposes, should be sacrificed in an experiment which was violent enough to melt the fuel plates. For this purpose a control rod worth 4% $k_{eff}$ was completely ejected from the reactor core. To increase the severity of the experiment it was run with the reactor water at room temperature. Although the ejection of the rod required only about 0.2 sec, the rod was only about 80 per cent out of the core when the reactor power reached its peak value. The minimum period resulting from the ejection was 0.0026 sec.

The power excursion melted most of the fuel plates. The pressure resulting from the molten metal in contact with the reactor water burst the reactor tank and ejected most of the contents of the shield tank into the air. The sound of the explosion at the control station, half a mile away, was comparable to that resulting from the explosion of 1 to 2 pounds of 40 per cent dynamite on the bare ground at the same distance. Figure 1b, taken from motion picture records of the experiment, shows one stage of the explosion, as compared to the nondestructive ejection of water from the reactor when the period of the excursion was 0.005 sec.

The total energy release during the excursion as determined by calibrated cobalt foils in the reactor core, was 135 megawatt seconds. Other data taken during the excursion are less reliable because of the violent mechanical effects of the explosion. Figure 18 is a reproduction of the transient record. The absolute values on the power curve ($A$) may be in error by 30 or 40 per cent, and the shape of the decreasing portion may not be correct. The fuel-plate thermocouple (curve $B$) failed long before peak power was reached. Curve $C$ is a temperature record from an insulated fuel plate installed some distance outside the reactor core. It was used for an auxiliary determination of total energy and does not give good transient information, as it was connected to a relatively slow recorder. A pressure transducer, which was installed in the reactor tank adjacent to the reactor core, failed before it recorded a pressure of significant magnitude (curve $D$). Analysis of the mechanical damage of the transducer, however, indicated that the peak pressure was at least as high as 6000 psi, and was probably higher than 10,000 psi.

It was evident from examination of the reactor debris that many of the fuel plates had been prac-

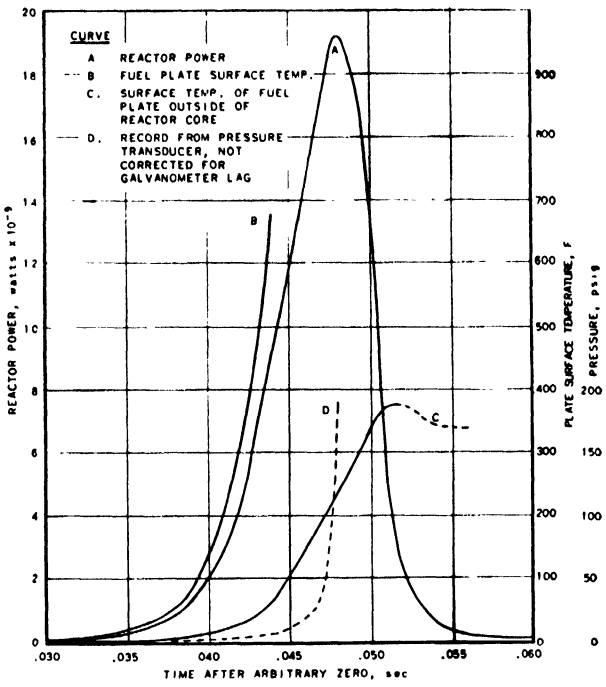


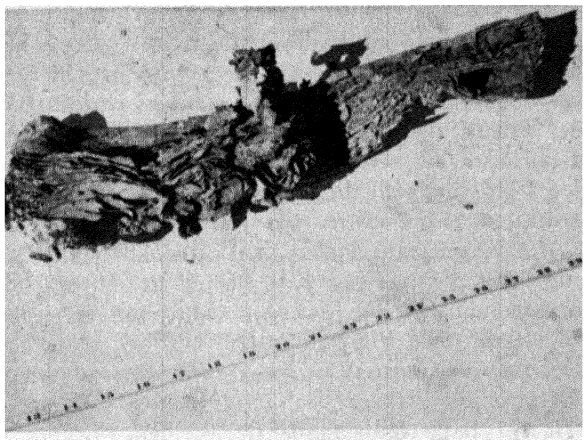

Figure 18. Replot of records from destructive experiment

tically completely melted. Others, evidently those at the edge of the core, had been only partly melted, and portions of them remained fastened to the side plates of the fuel elements (Fig. 19). Some of the fragments which had evidently been molten appeared as spongy metallic globules (Fig. 20). Other fragments appeared to have been molten inside, while the outside clad remained solid (Fig. 21).

Most of the heavy debris fell to the ground near the shield pit. The control rod drive mechanism, which weighed about a ton, fell on the side of the earth shield, after having been thrown about 30 feet into the air. Recognizable fuel plate fragments were thrown as far as 200 feet from the reactor site. Surveys of the total fission-product radioactivity of all the debris indicated that practically all of the fuel originally in the reactor could be accounted for within a radius of 350 feet around the original reactor location. Although these measurements necessarily lacked precision, they showed that no large fraction of the reactor core material left the site in the form of airborne material. At the time of the experiment, the wind velocity was 8 miles per hour at ground level, and 20 miles per hour at 250 feet above ground. Fifteen minutes after the experiment the total beta plus gamma activity level, 3 feet above ground, at a point 0.8 mile directly downwind of the reactor, was 5 mr/hr. At all points farther from the reactor the effects of fall-out were less than this value. Momentarily during the explosion, a gamma dose rate in excess of 400 mr/hr was indicated on a survey meter half a mile from the reactor. This indication decayed rapidly; the total dose received at the half-mile point (cross wind) was less than 10 mr.

Both the observed radiation intensities and the mechanical damage were roughly consistent with the measured nuclear energy release of 135 megawatt seconds. Although the explosion was spectacular, its effects were comparable to those which could be caused by a moderate amount of chemical explosive. The destruction of the reactor tank was not surprising, since it was constructed of relatively thin (½-inch) steel. Most of the equipment outside the shield tank was either undamaged or repairable, and much of it, including the control rod drive mechanism, was decontaminated, reconditioned, and re-used on reactor II.

There was no evidence that the power-limitation process in the destructive experiment differed qual-



gure 19. Fuel element side plate with attached cluster of fuel plat fragments

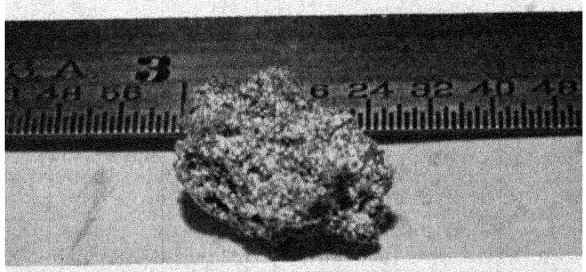
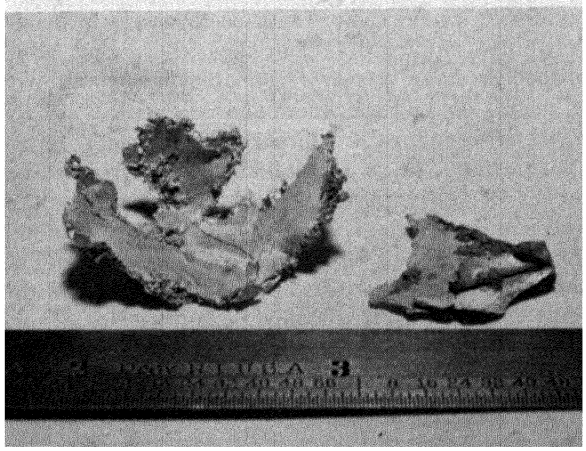


Figure 20. Pellet of spongy aluminum-uranium mixture



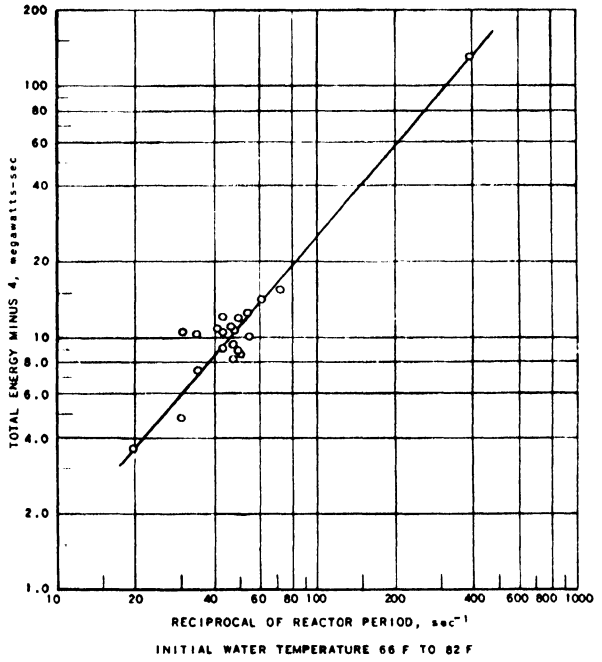



**Figure 22. Total energy of excursion minus energy required (4 Mw-Sec) to raise temperature of center of average plate to the boiling point**

itatively from that which was effective in the earlier, nondestructive experiments. It was quite evident that the nuclear power release was terminated at an early stage of the explosion; indeed, high-speed motion pictures recorded the light flash emitted by the reactor as it reached high power and showed that it was extinguished before any ejected material appeared above the top of the shield tank. The flash lasted about 0.003 second. The energy stored in the fuel plates as sensible heat and latent heat of fusion during the relatively short nuclear power burst was, of course, released during the much longer explosion process.

Figure 22 is a plot of energy released as a function of reciprocal period for all the power excursions made with cold water in reactor I. In plotting the curve, a constant energy of 4 megawatt-seconds has been subtracted from each value. This is the quantity of energy which has been released up to the time when the center temperature of the average fuel plate has reached the atmospheric boiling point of water. The highest point on the curve, which refers to the destructive experiment, does not appear inconsistent with the other values.

## Discussion

The experiments that have been described prove that the reactors investigated possess a high degree of inherent safety, and indicate that it is possible to design practical reactors of these types which will be safe against any reactivity accident which can occur in practice. In this connection, the consistent and regular behavior of the reactors during the experiments is reassuring. Although such behavior would be expected in the case of the homogeneous

non-boiling reactor, anomalies would not have been surprising in the cases where shutdown depends on the rapid formation of steam. Actually, in the entire series of some two hundred experimental runaways, no inconsistencies of behavior were observed which could not be attributed to instrumental errors resulting from rather difficult experimental conditions.

A few general remarks may be made in connection with the application of the results to the estimation of the runaway behavior of other reactors. The behavior of the homogeneous, non-boiling reactor may be considered to be typical of the case in which the loss of reactivity is roughly proportional to the time integral of the transient reactor power. Thus the behavior of power as limited by a negative "prompt"‡ temperature coefficient in a solid-fuel reactor would be expected to be qualitatively quite similar. Quantitatively, of course, such "prompt" coefficients are generally small and their effectiveness is limited. In inferring the behavior of other steam-limited reactors from the results presented here, it should be remembered that the reactors used had fuel plates of such high thermal conductance that in all the non-destructive experiments the metal of the plate represented no important impedance to the transfer of heat to the water. Certainly the transient behavior will be strongly modified and the effectiveness of steam limitation of power will be decreased if a departure is made from this condition, as it may well be in certain power reactor designs. In such a case the difference between the behavior of the reactor with saturated water and with subcooled water may be a very important one, and reactors designed for operation as boiling reactors may have important safety advantages.

No complete theoretical treatment of the transient limitation of power by steam has yet been developed. Attention should be called to some of the general experimental results which will have important bearing on the formulation of such a theory. Perhaps the most obvious is that there is no apparent simple relation between the energy liberated during a transient and the energy content of a steam volume of the size necessary to remove the applied excess reactivity. For example, the heat of vaporization of sufficient steam to fill the entire core volume of reactor I at atmospheric pressure was only 0.087 megawatt-second, very much less than the energy generation in any of the experiments. Furthermore, in extending the experiments from reactor I to reactor II, the total energy release did not increase in proportion to the volume of steam required to produce a given decrease of reactivity. Finally, when the experiments were extended from atmospheric pressure to 300 psig (Fig. 17) the energy release for a transient of given period *decreased* by almost a factor of 3, whereas the energy content of unit volume of steam *increased* by a factor of 16.

Certainly one of the important considerations is that although the energy required to vaporize a significant volume of steam is small, the temperature differences required to transfer this heat to the water in the short time available during the transient may be large. The heat capacity of all the fuel plates of reactor I was 0.05 megawatt-second per degree F, and the heat capacity of all the water in the core was 0.15 megawatt-second per degree F. Thus the establishment of steep temperature gradients required the expenditure of significant quantities of energy. A further consideration is that relatively high steam pressures must be built up to expel the water from the reactor core rapidly enough to terminate the short-period transients. A few pressure measurements were made in the reactor core which indicated that the peak transient pressure increase was about 15 psi during a transient of period 0.034 sec, and of the order 100 psi during the transient of period 0.005 sec. Consequently, even when transients are run with the reactor water at the ambient saturation temperature, the water is effectively in the subcooled state during the power excursion. Not only must the fuel plates be heated to temperatures corresponding to the transient saturation condition, but steam, once it forms at the hot fuel plate surface, may recondense in the cooler water.

Approximate theoretical treatments of the steam transient have been made by various workers to extend the results of the experiments to other reactor designs. Golian *et al.*§ have assumed that steam is formed in a laminar layer immediately adjacent to the fuel plate, and that the thickness of the layer grows by conductive transfer of energy across the layer to the water boundary. Edlund and Noderer¶ employ a model in which the transient pressure rise plays an important part. It is assumed that the heat transferred to the water is contained in a layer adjacent to the fuel plate, the average temperature of which equals the transient saturation temperature. The layer, which contains water and steam, grows in thickness as though by a thermal conductive process, but with a thermal diffusivity determined empirically from the experiments. Both approaches have shown reasonable agreement with the results of the experiments on reactor I.

The semi-empirical approach to extension of the experimental data is facilitated by the circumstance that all of the experimental power transients have quite similar shapes if time is measured in the non-dimensional unit of asymptotic reactor period. This characteristic is illustrated by Fig. 15. Despite this regularity, however, detailed examination of the power curves reveals significant differences in behavior as the conditions of the experiment are changed. In Fig. 23 the power curves for three

‡ The "prompt" coefficient is that component of the temperature coefficient of reactivity which depends on fuel-element temperature alone. It results from thermal expansion of the fuel element and Doppler broadening of resonances.

§ US Naval Research Laboratory.

¶ Oak Ridge National Laboratory.

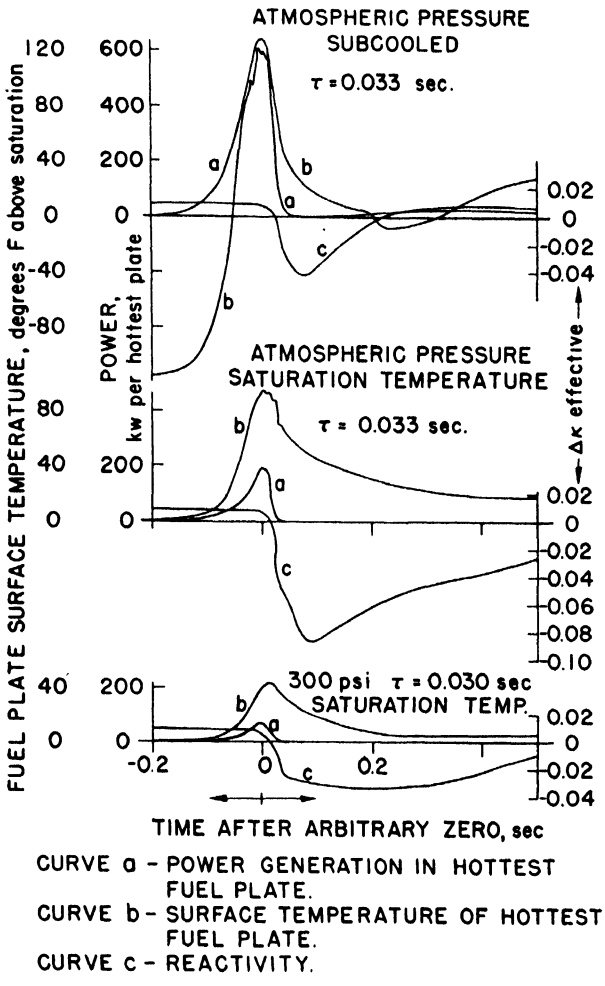



**Figure 23. Time variation of power, fuel plate temperature, and reactivity during power excursions on reactor II**

different reactor II transients of about the same period are reproduced along with the fuel-plate temperature records. The power curves have been analyzed to yield the variation of reactivity with time. The differences in magnitude of the power and temperature variations with subcooling and with pressure are striking, but it is interesting to note also the reactivity variations. At atmospheric pressure and saturation temperature the reactor shuts itself down by more than 8% $k_{eff}$. In the subcooled condition the degree of subcriticality achieved is considerably less, and the recovery to criticality is rapid, no doubt because of condensation of the steam. At 300 psi the degree of shutdown is again small, evidently because of the relatively low heat storage in the fuel plates. The recovery to criticality is, however, quite slow. A striking characteristic in all cases is the small fraction of the total reactivity change which suffices to stop the initial power rise. As is to be expected theoretically, this is equal to the initial prompt excess reactivity.

The limitation of power in fast transients is one aspect of the self-regulating behavior of reactors with strong negative power coefficients of reactivity. Experience with the solid-fuel boiling reactors (e.g., Fig. 10) has shown that the self-regulation is normally stable but that a type of instability (chugging) can result if the reactor is subjected to sufficiently large reactivity variations. Fortunately, even under chugging conditions the power is limited and does not reach a dangerous level. It is no doubt true for all self-regulated reactors that for some amplitude of reactivity excitation the characteristic rates of change of power are too rapid for the self-regulating process to maintain control, and instability will result. In the boiling reactors it is believed that the rate of escape of steam from the core, rather than the rate of formation of steam, represents the limiting time constant in the regulating process. Consequently, in subcooled boiling operation, where steam can leave by condensation, the range of stability is increased (Fig. 11). In any case, however, the range of stable self-regulation of the reactors investigated is more than adequate for practical use and probably exceeds that which can be attained practically with artificial control systems.

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# Radiological Hazards from an Escape of Fission Products and the Implications in Power Reactor Location

By W. G. Marley and T. M. Fry,* UK

The possible consequences of a large scale release of radioactivity from a nuclear reactor in the event of accident has long been recognised as an important factor in the development of nuclear power. The problem has become more pressing with the rise in total power and with the very high heat ratings in some of the designs now envisaged. The selection of suitable sites for nuclear power plants calls for an assessment of the radiological hazards from an escape of radioactivity and for the satisfactory limitation of the quantities of activity which might be released in the event of accident.

Direct experience has fortunately not yet shown the extent of the radiobiological effects of a massive release of radioactivity from a nuclear reactor. In the reactor incidents which have been reported so far the releases to the atmosphere and the consequential radiological effects have been trivial. Reliance must therefore be placed on theoretical estimates, though realistic calculation is difficult in view of the large number of imponderable factors. The validity of the results may be judged by applying similar reasoning to other hazardous materials for which long experience is available. An attempt has therefore been made to estimate the hazard ranges of a release of activity and to compare these with industrial experience in the large scale handling of the toxic gases, chlorine and phosgene ($COCl_2$).

## BIOLOGICAL IMPORTANCE OF VARIOUS RADIATION LEVELS UNDER ACCIDENT CONDITIONS

Levels have now been agreed internationally[1] for the permissible contamination of air and drinking water for consumption over a lifetime by persons exposed occupationally to radioactivity and safety factors have been introduced in evaluating these figures to ensure that no detectable injury will result. For the present purpose, however, it is required to know the levels, appropriate to a single emergency, at which definite injury is likely or at which drastic action, such as evacuation to avoid such injury, must be taken. The exposures envisaged would only occur in event of a major accident, and such events must necessarily for economic reasons be extremely rare.

A cloud of radioactivity released to the atmosphere may be effective biologically in two ways: first, by direct biological effects of beta and gamma radiation and inhalation of the activity during passage of the cloud; and secondly, by indirect biological effects due to deposition of the activity on to crops, herbage, drinking water sources and ground.

### Direct Biological Effects from the Cloud

It is unlikely that evasive action could be organised to avoid the direct effects of the radioactivity on persons in the open during passage of the cloud. The degree of injury sustained would depend on the duration and intensity of exposure and an exposure integral of 10 curie-sec/m$^3$ is taken to represent the limit of hazard and to be acceptable in an emergency. This exposure would result in: 5 r of total body radiation; 50 rep of beta radiation to the lung in the first day; 3 millicuries inhaled (quoted at 24 hours); 600 microcuries inhaled and retained for several days; 12 microcuries of $Sr^{89}$ retained; and 0.15 microcuries of $Sr^{90}$ retained.

The combined effects of these would probably be broadly comparable to that of a single dose of 25 r of gamma radiation alone, which would produce no detectable injury other than temporary slight blood changes. In making these assessments it has been assumed that exposure occurred within minutes of the accident, but the quantities of activity quoted are those which would be measured 24 hours after the release. On the same basis it is estimated that an exposure of 400 curie-sec/m$^3$ would produce injury which in most cases would prove fatal.

### Indirect Biological Effects of Ground Deposition

Since the deposition on crops, herbage, water and ground would take some time to become effective, there would be a strong case for the evacuation of persons in the potential danger area and the important issue is the emergency permissible level of ground contamination for continued normal habitation. Considering first the hazard of the gamma radiation from the deposited material, a dose of 25 r accumulated within three weeks is considered the maximum permissible in an emergency: this implies a total dose of 60 r in three months if the activity were to subside only by radioactive decay. Allowing for some shielding due to the roughness of the ground and due to house walls for persons generally in buildings, the

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gamma radiation dose alone would thus not necessitate evacuation for fission product contamination below 0.05 curie/$m^2$.

Normal habitation, however, also involves the risk from ingesting the deposited activity and particularly the bone-seeking isotopes. For this risk the limit of hazard is taken to be roughly equivalent to that of the ingestion of 120 microcuries of $Sr^{89}$ alone. The basis of the calculation is briefly: (1) the permissible dose to the bone over the decay period to be 80 r, believed equivalent to an instantaneous dose of 25 r; (2) 60 per cent absorption and deposition in bone; and (3) a safety factor of 5 to allow for uneven distribution in the bone (a factor shown to be necessary by biological experiments). This estimate agrees broadly with that by Morgan and Ford[2] when allowance is made for different uptake fractions assumed. The $Sr^{89}$ will always be accompanied by $Sr^{90}$-$Y^{90}$, $Ba^{140}$-$La^{140}$, and other bone-seeking isotopes. When allowance is made for the dose from these, the proportions of which would depend upon the duration of the irradiation, the emergency permissible ingestion level becomes about 30 $\mu$c $Sr^{89}$ together with the other associated bone-seeking activities (including about 0.3 $\mu$c of $Sr^{90}$ which has been considered acceptable). If the iodine isotopes are ingested in the same proportion there will also be considerable irradiation of the thyroid but this is considered acceptable in emergency since this organ is very insensitive.

The following criteria are therefore suggested for considerations of siting and the possible need for evacuation of members of the general public in the event of a nuclear reactor accident:

1. The exposure to gamma radiation should be limited to not more than 25 r in three weeks and 60 r in three months.

2. The inhalation or ingestion, due to continued occupation of a contaminated area, should not exceed 30 $\mu$c of $Sr^{89}$. This level allows for the other associated bone-seeking fission products.

3. The accumulation in the body of long-lived activity, such as $Sr^{90}$, should not be allowed to build up to a value exceeding the recommended peace-time permissible body burden for occupational exposure (1 $\mu$c for $Sr^{90}$ and 0.04 $\mu$c for $Pu^{239}$), since, if greatly exceeded, the activity would probably lead to serious incapacity in later life.

It is to be noted that these levels are the same as those accepted in the UK for use in connection with the control of emergency contamination of water supplies in Civil Defence.

### Limiting Levels of Fission-Product Contamination for Continued Habitation

The limiting degree of hazard from possible ingestion may be reached, in normal living in a rural community, at a contamination level of $10^{-3}$ curies/$m^2$ measured at one day. The consumption of contaminated crops would contribute much of the hazard, on the assumption that a man eats in one day brassica, lettuce, etc. from about 0.05 $m^2$ of ground, that half the activity is removed by washing and that the consumption continues for 40 days, after which the leaves are clean as a result of new growth or continued washing by rain. At this level also the limiting level for a child, playing on the ground, would be reached on the assumption that a child actually ingests the contamination from 0.1 $m^2$ of ground before the activity is removed or fixed by natural processes.

The long-term ingestion hazard from the build-up of activity in crops is estimated from the work of the University of California (Los Angeles) School of Medicine,[3] from which it appears that the edible parts of plants take up $Sr^{90}$ from contaminated soil so that the content per gram *dry* weight is about the same as that in the ground. The build-up of activity would thus be about limiting from contamination at the level of $10^{-2}$ curies/$m^2$ of the mixed fission products, measured at one day. At this level also it appears that open sources of drinking water would be contaminated to about limiting emergency ingestion levels, but in practice, there would be processes operating which would greatly clean up the drinking water. Temporary contamination of milk by $Sr^{89}$ may extend to zones where the fission product level is as low as $10^{-4}$ curies/$m^2$, as a result of the wide area of herbage grazed each day by a cow: supervision of the population would be needed down to this level in order to confirm that the basic tolerances were not being exceeded.

As a result of this brief analysis it appears that the action shown in Table I would have to be taken at the various levels of ground contamination stated, in order to prevent excessive exposure. It should be emphasized that the criteria adopted are fairly realistic and as far as can be seen do not contain significant hidden safety factors.

## THE SPREAD OF ACTIVITY FOLLOWING A REACTOR ACCIDENT

There is no satisfactory direct evidence of the spread of activity following a reactor accident. For the estimation of direct hazard from the cloud calculations have been based on Sutton's theory of diffusion for a ground level source with no thermal lift and yield values for the hazard ranges shown in the first two items of Table I for releases of the order of 1 megawatt of reactor power. Calculations of the possible maximum deposition of activity under conditions of fair weather or rain have been made by Chamberlain,[4] based on a study of the deposition of particles by turbulent diffusion in the atmosphere and also of the wash-out of particles by rain drops, using a theory by Langmuir. Assumptions are made concerning the particle size which are *a priori* likely but which also give the maximum deposition at a few kilometres and in this sense the contamination levels calculated are the maximum likely.

The maximum range downwind, $R$, to which various levels of ground contamination may be expected to extend is roughly proportional to the square root

**Table I. Estimated Maximum Hazard Ranges for Release of Reactor Fission Products under Lapse Conditions of Turbulence***

| *Description of hazard and action necessary* | *Ground contamination level at 1 day curies/m²* | *Dry weather value of B: km/(Mw)¼* |
|---|---|---|
| **Direct Effects** | | |
| Exposure to cloud lethal in the open: exposure integral 400 curie-sec/m³ (activity quoted at one day) | – | 0.2 |
| No serious hazard from cloud: exposure integral 10 curie-sec/m³ | – | 1.7 |
| **Indirect Effects** | | |
| Urgent evacuation within 12 hours. Persons in the open receive 25 r gammas in first 12 hr from ground contamination | 0.2 | 1.1 |
| Probable limit of external beta-gamma hazard from deposited material. Persons usually in buildings receive 25 r in three weeks and 60 r in three months | 0.05 | 2.3 |
| Evacuation necessary or severe restrictions on normal habits of living. Crops, milk, water and agricultural land contaminated. Rapid build up of isotopes in the body. Contamination level is 250 times that in hot laboratory at A.E.R.E. | $10^{-3}$ | 5.2 |
| Probable limit of temporary evacuation or restrictions. Build up of isotopes in the body from crops and water supplies probably not in excess of emergency limits. Milk temporarily heavily contaminated | $10^{-3}$ | 18 |
| Probable limit of temporary milk contamination. Build up of $Sr^{90}$ due to contaminated milk probably not in excess of emergency limit. | $10^{-4}$ | (60) |

* (1) Downwind range ($R$) of hazard under lapse dry weather conditions given by $R = BM^{½}$ where $M$ is the activity released expressed in megawatts. (2) In the worst conditions of rain, distances may be increased by a factor, with a maximum value of 2.8 at distances over 5 km. (3) In inversion conditions, formula does not apply and ranges may be larger.

of the amount of activity released and is thus given approximately by the expression

$$R = BM^{½}$$

where $M$ is the activity released expressed in megawatts, i.e., the total power of the reactor multiplied by the fractional release, and $B$ is a constant having values for the various levels as shown in Table I: the values given are those appropriate to ranges of the order of some kilometres for lapse conditions of turbulence in dry weather and wind speed 5 m/sec. The contamination zone would, of course, comprise only a narrow strip in the downwind direction, having an area about $R^2/14$ km². The maximum deposition possible in rain at any given distance is about eight times that in dry weather and the maximum distance at which a given deposition may occur in the most unfavourable rain is thus about 2.8 times the distance in dry weather. The intensity of rain necessary to produce this contamination depends on the distance downwind and the particle size and is unlikely to be reached in practice except for locations more than 5 km downwind. In inversion conditions the above simple formula no longer applies and greater ranges are possible. Following similar reasoning, Fitzgerald *et al.*[5] have derived formulae for the estimation of various hazards and Mesler and Widdoes[6] have made estimates of certain radiological effects. There is general agreement in principle with these authors when allowance is made for some differences in basic parameters.

It is clear from Table I that the release of a significant fraction of the fission products accumulated in a high-power reactor cannot be contemplated within many kilometres of a normally inhabited area. On the other hand, a release, under conditions of turbulence in dry weather, of fission products restricted to 0.1 megawatt of reactor power should not necessitate lengthy evacuation of civil population outside a narrow sector extending 1.7 km downwind: temporary evacuation or restriction on habits of living may be necessary out to 5.7 km, while seriously contaminated milk might be expected temporarily from pastures up to 20 km.

## COMPARISON WITH INDUSTRIAL EXPERIENCE OF TOXIC GASES

A direct comparison of the effects of inhalation of the cloud can be made between releases of fission products and of toxic gases such as chlorine and phosgene. In the event of 100 megawatts of fission products escaping it is seen from Table I that inhalation of the cloud would probably be lethal at 2 km and non-injurious at 17 km. The LD 50 (the dose for which, on the average, the chance of survival is 50 per cent) for chlorine is about ⅓ gm inhaled and for phosgene about 1/30 gm inhaled. Using these figures in a calculation similar to that on which Table I is based, it is found that a man, standing in the open, two kilometres downwind of the point of release of 10 tons of phosgene, would have a 50 per cent chance of survival. A 10-ton tank of phosgene thus represents a potential hazard which, as far as effects of inhalation are concerned, is comparable with a 100 Mw reactor.

A disaster occurred on the outskirts of Hamburg in 1928, when 8 tonnes of phosgene escaped from a storage tank. Fortunately, the gas was carried by the wind over open country before it reached a village. Five people, who were within 300 metres of the tank, were killed. Another five deaths and 30 other serious casualties occurred, mostly among those in the village, two to three kilometres away. Ill effects were felt by some hundreds of people up to 13 kilo-

metres downwind. These consequences are consistent with theoretical predictions. The number of casualties was so small only because the gas passed over an almost uninhabited area. It should be noted that the senses give little warning of phosgene, for an atmosphere with a musty smell, but in no way unpleasant to breathe, can prove fatal.

Chlorine was at one time considered so difficult to handle that it was transported and used in the form of bleaching powder. For 25 years phosgene was widely feared as an instrument of warfare. Despite the Hamburg disaster and a few others on the same scale, phosgene and chlorine are not regarded as so dangerous that they must be kept far from cities. In many densely populated areas, there are tanks each containing enough liquefied gas to kill hundreds and even thousands if it escaped. Reliance is placed on sound engineering for the safe custody of these materials. That this is justified is shown by the Report of the Chief Inspector of Factories for Great Britain for 1950, which records only three deaths due to chlorine gassing and one due to phosgene in the twelve previous years.

The preceding discussion gives no indication of the probability of an actual release of radioactivity from an atomic energy installation, but this is just as important as an assessment of the effects. The possible sequence of events leading to the release of activity from a reactor has, for instance, been considered by Hurwitz.[7] The engineering problems of nuclear reactors are more complex than those of the safe storage of liquefied gas. Moreover, the parallel between accidents involving chemical and radioactive poisons breaks down in regard to persistent effects. Most chemical poisons can be neutralised and disappear quite rapidly even if no counter-measures are taken but radioactivity cannot be neutralised and it is uncertain how much could be achieved in decontamination. A reactor disaster, though it would not cause more casualties than an accident in a chemical plant, might interfere, for a much longer time, with the livelihood and homes of those living in the neighbourhood. There are other differences between the two cases and it may be argued that a higher standard of safety is required for nuclear reactors than for toxic chemicals. It is especially important to avoid the setback which a major disaster at an early stage would impose on the atomic energy industry.

Accidents must necessarily be infrequent and, so long as they do not involve the release of more than a few tenths of a megawatt equivalent of fission products, remote siting of reactors should be unnecessary. It is considered that this degree of containment can be maintained by adherence to sound principles in the design, construction and operation of nuclear plant. It is therefore concluded that the satisfactory siting of high-power reactors with suitable engineering safeguards will not present an insuperable problem in the development of nuclear power.

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# Environmental Effects of a Major Reactor Disaster

**By H. M. Parker and J. W. Healy,* USA**

To achieve the economic advantage of locating nuclear power reactors close to large communities, it is essential that the potential environmental radiation hazards be unequivocally eliminated.

At the present time, it seems certain that inherently safe reactors can be constructed. Even if there is still a minute possibility of serious reactor accident, release of radioactive material to the environs can be preventive by a protective envelope, of which a large steel sphere is one feasible form.

The purpose in presenting a picture of what could happen in the event that such protection were not provided is to establish reasonable criteria for the degree of safety required. It will be readily seen that high engineering standards, and the accompanying expense, are fully justified for all reactors operating at high power levels.

Since practical experience with reactor failure has been minimal, estimates must rely on theoretical considerations. The estimates are sensitive to the exact mechanism of the initiating incident, the rate of energy release, the meteorological conditions and the associated deposition rates of the active materials. The uncertainties in each phase combine to give an answer which is probably only valid as to order of magnitude. However, it has been found that the total damage, evaluated by the somewhat arbitrary method proposed later, is relatively insensitive to the detailed characteristics of the model chosen for the incident. It seems likely that total damage estimates are reliable within a factor of about three.

## RELEASE OF CONTENTS

In the operation of power plants, practical reactors operating in the range of tens of thousands of kilowatts to several millions of kilowatts will be of eventual interest. In such reactors, with the resulting high specific power, the chief hazard in a major accident can be shown to arise from the fission products accumulated during the operating period. The quantities of the radioactive fission products are directly proportional to the reactor power level for a given pattern of operation, and show an increase in the longer lived components with increased time of exposure of the fuel elements. Figure 1 illustrates typical decay curves for fission product mixtures resulting from different irradiation times and shows the increase in the quantity of lingering radiation which would result from the dispersal of fission products from fuel elements which have operated for long periods of time.[1]

The curves do not include the additional fission products generated instantaneously in a runaway critical incident. This component is so low in long-lived fission products that it does not add significantly to the existing load of hazardous materials in the operating reactor.

The emphasis on the hazard of long-lived components stems both from the persistence of radiation from deposits on the ground and from the increased hazard following deposition of these isotopes in the body by inhalation or ingestion. As an example, the relative ingestion hazard for different fission product mixtures is shown in Fig. 2. The reference base is the bone-seeker hazard of the mixture characterized by one day irradiation and one hour post-incident decay.

Two basic models for release of material have been used. In one, an uncontrolled power burst is visualized to cause rapid vaporization of the fission products and to generate a high temperature radioactive

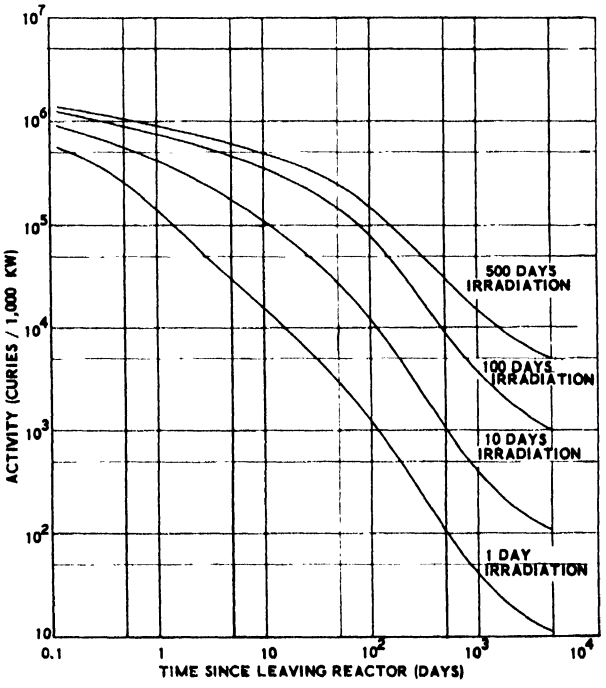



**Figure 1. Fission product activity**

* General Electric Company, Richland, Washington.

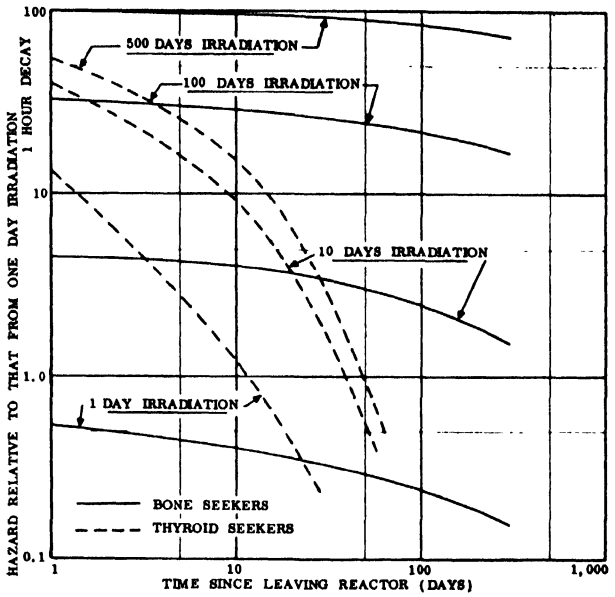



**Figure 2. Relative hazard for ingestion of fission product mixtures**

cloud that would rise to an elevation of several thousand feet and then travel downwind and disperse. In the other case, failure of the reactor coolant supply is visualized to lead to eventual melting of fuel elements and vaporization of fission products by the self-heating of the fission products. In this more leisurely process there is a much lower rate of heat production; a lower temperature cloud which would tend to travel close to ground level would result. An essentially similar picture would apply to a reactor consumed by fire.

In any of these models, it may be assumed that a portion of the primary escaping fission products will be retained in the reactor building structure. Only the fraction that escapes into the atmosphere generates an environmental hazard. What this release coefficient may be is best computed locally for each case. In the figures and tables:

$$\text{Power level equivalent to F.P. release} = (\text{Actual Power Level}) \times (\text{Release Coefficient})$$

In the limit, for a unit with a protective envelope of assured integrity, the release coefficient is zero, the equivalent power level is zero and environmental hazard does not occur. This is the real expected situation; however these data permit potential damage to be assessed for such pessimistic assumptions as a 1% or 10% leak from such a structure. Similarly, they may be applied to protective systems depending on large filters.

## CONTAMINATION LEVELS

The concentrations of fission products downwind will also be dependent upon the meteorological conditions and the surrounding terrain. Estimates of the spread for several cases were made by the use of Sutton's theory of turbulent diffusion.[2] Basic assumptions applied to the travel of the cloud over a level plain are given in Table I.

Calculations on other conditions have shown variations in detail and pattern but little difference in over-all damage estimates. A decrease in concentration additional to that by turbulent diffusion was computed using an empirical deposition constant equivalent to a settling rate of the material in the cloud of 2.8 cm per second.

The areas involved were computed on the basis of damage levels. These limits are given in Table II.

The distance at which lethal or damaging conditions would occur is strongly dependent upon both the time and elevation of release and the meteorological conditions. Present estimates indicate lethal conditions only within the immediate vicinity of the reactor for full escape from a 100,000 kw reactor and at distances of the order of 10–50 miles for reactors in the millions of kilowatts range.

Access to land downwind will be limited for a period of time because of radiation levels from the

**Table I. Assumptions Used in Calculation**

| | *Rapid release neutral atmosphere* | *Slow release neutral atmosphere* | *Slow release inversion atmosphere* |
|---|---|---|---|
| Height of release | 25% - 100 meters<br>25% - 500 meters<br>25% - 1000 meters<br>25% - 2000 meters | 70 meters | 70 meters |
| Time of release | Instantaneous | 10 hours | 10 hours |
| Angle covered by wind during release | | 45 deg | 30 deg |
| Wind speed | 100 meters-5.2 m/sec<br>500 meters-5.7 m/sec<br>1000 meters-6.4 m/sec<br>2000 meters-7.2 m/sec | 5 m/sec | 5 m/sec |

**Table II. Damage Limits**

| *Effect* | *Fallout limit* | *Resulting condition at boundary* |
|---|---|---|
| Lethal | | 190–350 r full body*<br>plus 800–1200 rads to lung-10 days<br>plus 150–250 rem to bone-10 days |
| Significant injury to humans | | 60–250 r full body*<br>plus 200–300 rads to lung-10 days<br>plus 30–70 rem to bone-10 days |
| Land unusable for 5 years | 5 mc/ft$^2$ | Dose rate falls to 300 mr/week in 5 years |
| Land unusable for 2 years | 2 mc/ft$^2$ | Dose rate falls to 300 mr/week in 2 years |
| Temporary evacuation | 0.5 mc/ft$^2$ | Gamma dose of 50 r in first year (30 r in 2 months; 40 r in 6 months) |
| Crops confiscated | 0.1 mc/ft$^2$ | $5 \times 10^{-5}$ $\mu$c Sr$^{90}$/gram of vegetation† |

* 20–30% from cloud passage; remainder from exposure to contaminated ground for 2–5 hr.

† Limit computed assuming ingestion of crop by humans for one year.

"fallout". The actual shape of these areas will again be strongly dependent upon the methods of release and dispersion with a rapid emission expected to result in a long, narrow band and a slow release resulting in a general spread over shorter distances but wider areas. Examination of typical cases and comparison with small-scale tracer tests have indicated that the areas involved are remarkably independent of the exact mechanism of release and dilution.

Average values of the contaminated areas resulting from the complete release of fission products after 100 days irradiation of the fuel are given in Fig. 3. Individual estimates for specific methods of release and rates of dilution in the atmosphere varied from these values by factors of two to three. As a rough rule of thumb, the area covered in square miles after one day decay is approximately given by the power level representative of the fraction of fission products escaping expressed in kilowatts multiplied by $2 \times 10^{-3}$ and divided by the contamination level contour of interest in millicuries per square foot.

These estimates of areas are strictly orders of magnitude for average depositions with specific locations within the area possibly more or less contaminated depending upon local conditions of terrain, wind patterns and nature of the surfaces. It is to be expected that personnel contamination will also occur

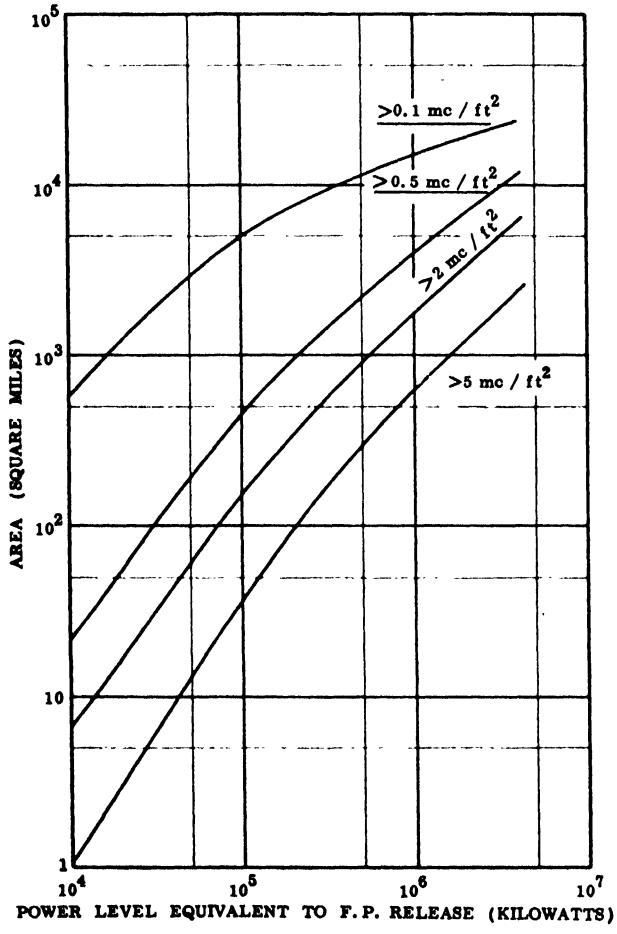



Figure 3. Average areas covered and contamination

within these areas with immediate decontamination necessary to prevent injury from the beta radiations.

The rate of decrease of this deposited material will be primarily influenced by radioactive decay although weathering and possibly incorporation into the top soil will occur. Gamma dosage rates at several feet above the ground will decrease to 300 mr/week in about five years for the five millicurie per square foot area, in about two years for the two millicurie per square foot area, and in about one year for the 0.5 millicurie per square foot area. The specific times required will be dependent upon the weathering and upon the time of irradiation of the fuel elements.

Precipitation during the evolution of fission products will change the estimates since the rate of deposition will be increased and the areas immediately surrounding the plant will be subjected to much higher contamination levels. This, of course, increases the probability of severe damage at close approaches but tends to decrease the damage at a distance and reduces the total areas involved.

Contamination of bodies of water will occur both by direct fallout and by secondary leaching of the materials into the streams. Direct fallout could render bodies of water reasonably close to the reactor unfit for use until the material is carried away. Present indications point toward leaching into the stream slow enough to cause little trouble although monitoring downstream is advisable.

The possibility of rainout into a stream near the reactor site exists with severe contamination resulting, depending upon the rate of washout from the cloud and the stream flow characteristics. Of more concern in at least some cases is the possibility of the escape of reactor coolant containing a significant quantity of fission products. Such a mishap would cause a band of grossly contaminated water, depending again upon the flow characteristics of the stream, which could cause severe contamination of water plants or other equipment downstream and in special cases could lead to serious radiation dosages to people using the water for sanitary purposes.

## DAMAGE ESTIMATES

As an illustration of the average expected cost of a major disaster, estimates were made by applying the areas of Fig. 3 to average census values[3] for several localities. The census values used are given in Table III.

Table III. Census Data

| Area | Population density people/mile² | Property values Rural $/mile² | Property values Urban $/mile² | Crop values $/mile² |
|---|---|---|---|---|
| Industrial Middle Atlantic States | 231 | 33,000 | 240,000 | 19,000 |
| Agricultural Middle West States | 154 | 95,000 | 150,000 | 36,000 |
| Agricultural Western States | 35 | 22,000 | 33,000 | 9000 |

It was assumed that property contaminated to greater than 2 mc/ft² would be purchased outright at market value while property contaminated to 0.5–2 mc/ft² would cost about 10% of the market value for rental or decontamination. Crops on land contaminated between 0.1 and 0.5 mc/ft² were assumed purchased for the first year with no allowance for later crops. A closer breakdown was not felt to be warranted in view of other uncertainties in the estimates. Figure 4 illustrates one series of average property damages expected for a reactor located in these three areas.

In order to illustrate the variation in contributions to the assumed damages, a breakdown by areas is given in Table IV for full release from several power reactors.

In addition it is estimated that for very large releases damage to humans could be extensive. As an example, for full release from a 1,000,000 kw reactor between 200 and 500 people could be killed in a region of population density of 200–500 people per square mile with perhaps 3000–5000 exposed to possibly damaging levels even with fairly prompt evaluation.

It may seem surprising that the estimated damage in Fig. 4 and Table IV is greater for a rich agricultural region than for an industrial area. This develops from the mode of averaging census values over a whole state. For a specific heavily industrialized zone, the property damage could be considerably higher.

To give proper perspective to these data, one must reiterate that the probability of a major reactor accident, although currently indeterminate, is obviously vanishingly small, and that the essential integrity of protective envelopes can be assured from conventional engineering data. The results should in no way be interpreted as a deterrent to the advancement of commercial power reactor technology. They do indicate the need for high standards of engineering and operation in all cases. More importantly, they show the high value of protective systems that are not quite perfect. Thus, for an actual power level of 100,000 kw and a protective system with a release coefficient of 0.01, the equivalent power level of 1000 kw is below the range in which it is relevant to compute estimated average environmental hazards. Over the small areas then involved, it is possible to apply decontamination methods that would be impracticable for the hypothetical large-scale disasters. The real damage in this case would therefore be even lower than is found by a plausible extrapolation of Fig. 4. This perhaps possible disaster is well within the range of disaster damage experienced from time to time in older industries.

**Table IV. Composition of Property Damage Estimates (millions of dollars)**

| *Category* | *10,000 kw* | *100,000 kw* | *1,000,000 kw* |
|---|---|---|---|
| Industrial area – Middle Atlantic States | | | |
| Purchase – Rural | 0.2 | 7 | 50 |
| Urban | 1.2 | 50 | 380 |
| Rent | 0.4 | 10 | 70 |
| Crops | 11 | 90 | 210 |
| Total | 13 | 160 | 710 |
| Agricultural area – Midwest States | | | |
| Purchase – Rural | 0.5 | 20 | 170 |
| Urban | 0.7 | 30 | 270 |
| Rent | 0.4 | 10 | 60 |
| Crops | 20 | 160 | 450 |
| Total | 22 | 220 | 950 |
| Agricultural area – Far West States | | | |
| Purchase – Rural | 0.1 | 4 | 35 |
| Urban | 0.2 | 7 | 50 |
| Rent | 0.1 | 2 | 12 |
| Crops | 5 | 43 | 100 |
| Total | 5 | 56 | 200 |

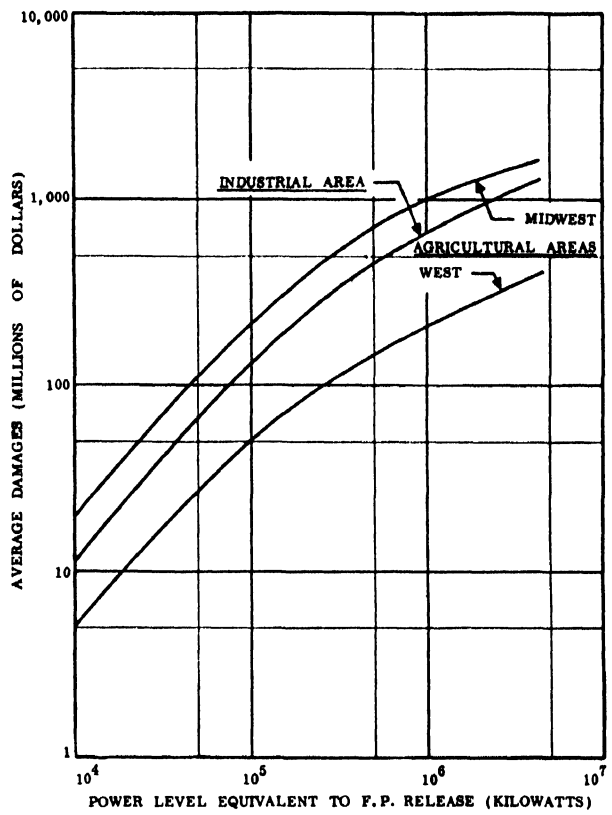



**Figure 4. Estimated average damages from release of fission products**

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# Radiation from Clouds of Reactor Debris

**By J. Z. Holland,* USA**

In the design and, especially, in the selection of sites for nuclear reactors, the problem arises of estimating the degree of hazard which might result to nearby populations from the accidental discharge of radioactive fission products to the atmosphere. In order to make such estimates it is necessary to postulate the type and amount of radioactive products released, the speed of movement and rate of spread of the resulting cloud, and the manner of exposure of people in its path, *i.e.*, whether by direct external irradiation, irradiation by material on the ground or on the skin, by inhalation, ingestion, etc.

The composition of the fission products in the source can be classified in three convenient categories: (1) the mixture resulting from a brief power excursion; (2) the mixture resulting from long-time operation of the nuclear reactor at a relatively steady power level; and (3) any particular radioisotope or arbitrary combination thereof.

In the first two cases the large range of decay rates of the individual fission products results in an approximate power-law decay rate of the mixture, roughly as $t^{-1.2}$ in the first case and as $t^{-0.2}$ in the second. In the third case the decay of each radioisotope is an exponential function of time with a constant coefficient. The fission products are beta and gamma emitters of a wide range of energies. Beta rays have a range of the order of millimeters in tissue and meters in air. Gamma rays have a mean free path in air of the order of hundreds of meters and would essentially irradiate the whole body uniformly under conditions of immersion in a radioactive cloud.

Meteorological factors which influence the mode and degree of exposure are the wind velocity, its variation with height through the layer containing the bulk of the cloud, the intensity and directional partitioning of turbulent eddy diffusion, and the occurrence, duration and intensity of precipitation. In the case of precipitation scavenging or gravitational settling of particulate material, the possible chains of events leading to external or internal irradiation of human beings are of such complexity and variety that even order-of-magnitude estimates as to the deposition rate would be acceptable in relation to the other uncertainties of the problem. In the case of external irradiation by the passing cloud, on the other hand, the problem is more readily expressed in mathematical terms, so that it is possible to estimate within more narrow limits the dosage to which a person at the ground might be subjected.

The theory of surface deposition of material from diffusing clouds has been treated by Chamberlain.[1] The relation between surface contamination and external irradiation has been presented by Cohen and Plesset[2] and by Fitzgerald, Hurwitz and Tonks.[3]

The inhalation dosage is adequately evaluated simply by integrating the concentration at the receptor with respect to time (e.g. reference 4). This result can be combined with breathing rate, critical organ concentration and retention parameters as outlined in reference 5 to obtain estimates of the internal radiation hazard.

Beta particles which would affect a receptor can be treated as originating within a small, approximately homogenous portion of the cloud. The time integral of local concentration (again as in reference 4) can be multiplied by a conversion constant to obtain a satisfactory estimate of the beta dosage.

The present paper deals primarily with the calculation of external gamma radiation dosage. For practical working formulas and conversion factors for all the modes of exposure described above, the reader is referred to reference 6.

## BACKGROUND OF THE CLOUD GAMMA DOSAGE PROBLEM

Of all the cloud dosage problems that of gamma radiation from a cloud of mixed fission products is perhaps the most difficult. This is because of two characteristics of the physical system:

1. The mean free path of the radiation is of the same order of magnitude as the cloud size, distance of travel or height of rise in some portion of the working range of each of these parameters.

2. The magnitude and non-linearity of the decay are also significant in time intervals of the same order as the travel time and exposure time. Thus the decay of the radioactivity, gradual spread of the cloud, spatial distribution of material in the cloud, and absorption and scattering of the radiation by the air, must all be retained in the equation. Each of these factors has a major influence on the result under some plausible combination of conditions.

Instructive approximate solutions have been obtained by previous investigators under various as-

* US Weather Bureau, Washington, D. C.

sumptions. Luckow *et al.*[7] assumed uniform concentration in a cloud travelling along the ground, whose dimensions were determined by Sutton's diffusion formula. Absorption and scattering of the gamma rays by the air were neglected, leading to largely pessimistic results. Taylor of the Westinghouse Electric Corporation[8] obtained the spatially integrated dose rate from the cloud at the moment when it is centered over the receptor. The distribution of material in space was given by Sutton's formula. Taylor then multiplied this dose rate by a time of passage determined from Sutton's formula and the wind speed. Multiple scattering of the gamma rays was ignored. Fitzgerald *et al.* at the Knolls Atomic Power Laboratory of the General Electric Company[3] carried out the space and time integrations for a cloud passing along the ground, without change in cloud size during irradiation, and without considering multiple scattering or decay during the time of exposure. Waterfield of the General Electric Company[9] carried out a complete numerical space-time integration for elevated as well as surface clouds, still, however, assuming simple exponential absorption and holding decay constant during passage of the cloud. He also presented a decay-integral correction factor for the case $f(t) = t^{-1.21}$, which tends to improve the accuracy considerably at intermediate, but not at small ( $< 300$ meters) distances. Gamertsfelder of the same laboratory has carried Waterfield's work further by including the dose buildup effect due to multiple scattering, but has not included the effects of non-linear decay during passage of the cloud. His results are quite satisfactory for distances greater than a few hundred meters from the point of origin.

In the present development the only restrictive assumptions are that the cloud particles all travel with approximately the mean wind speed during irradiation of the receptor, and that the turbulence responsible for the cloud spread is isotropic. As in all previous treatments, the dosage calculated for the average gamma photon energy is assumed to approximate the integrated effect of the actual gamma energy spectrum.

The resulting method does not depend upon any particular form of the diffusion law as long as it gives a Gaussian distribution of concentration with respect to distance from the cloud center. Results can be readily obtained for a wide variety of assumed meteorological conditions and for heights of rise up to 500 m. The initial source may be taken either as a point or as a puff of finite size. The computation method employs a nomogram whose maximum error is believed to be about $\pm$ 20%.

## MATHEMATICAL STATEMENT OF THE GENERAL CLOUD GAMMA DOSAGE PROBLEM

The conventional unit of gamma ray dosage is the roentgen.[10] One roentgen is equivalent to 83 ergs or $5.24 \times 10^7$ Mev (million electric volts) of energy absorbed per gram of matter. The dosage received in an infinitesimally small time interval $dt$ in a small volume of matter is thus

$$dD = \frac{\mu_m \Phi \, dt}{5.24 \times 10^7 \text{ Mev gm}^{-1} \text{ roentgen}^{-1}} \qquad (1)$$

where $D$ = dosage (roentgens), $\mu_m$ = mass absorption coefficient for gamma radiation ($m^2 gm^{-1}$), $\Phi$ = gamma energy flux per unit area into the outer surface of the receiving volume (Mev $sec^{-1}$ $m^{-2}$), and $t$ = time (sec).

Note that the dimensions of the receiving volume must be much less than $(\mu_m \rho)^{-1}$ where $\rho$ is the density of the absorbing medium (gm $m^{-3}$) for Equation 1 to be applicable. Now $\mu_m$ is nearly the same for both air and tissue for a given gamma photon energy.[11] The linear absorption coefficient $\mu_a$ ($m^{-1}$) is equal to $\mu_m \rho$. Thus $\mu_a/\rho$ for air can be substituted for $\mu_m$ to obtain the dosage in either air or tissue. At elevations near sea level, $\rho$ is approximately $1.29 \times 10^3$ gm $m^{-3}$, so that the total gamma dosage received in the small stationary receptor during passage of a cloud of fission products is

$$D = \frac{\mu_a \rho_0/\rho}{6.8 \times 10^{10} \text{ Mev m}^{-3} \text{ roentgen}^{-1}} \int_0^\infty \Phi \, dt \qquad (2)$$

where $\rho_0$ = sea level air density = $1.29 \times 10^3$ gm-$m^{-3}$.

In the remainder of this paper $\rho_0/\rho$ will be assumed to be approximately unity. For high altitudes (greater than about 2 km above sea level) the computed dosage must be multiplied by this factor in order to preserve the accuracy of the method.

One curie of radioactivity is equivalent to $3.7 \times 10^{10}$ disintegrations/sec. Therefore each element of the cloud emits gamma energy at the rate $3.7 \times 10^{10}$ disintegrations $sec^{-1}$ $curie^{-1}$ $\chi_c E \, dV$, where $\chi_c$ is the concentration of radioactivity (curie $m^{-3}$), $E$ is the gamma photon energy (Mev per disintegration) and $dV$ is the cloud volume element. If the space between the emitting volume element and the receptor is compltely empty, the part of the flux received at the surface of the receptor from this volume element is:

$$d\Phi = \frac{\chi_E dV}{4\pi r^2} \equiv \frac{3.7 \times 10^{10} \text{ dis sec}^{-1} \text{ curie}^{-1} \chi_c E \, dV}{4\pi r^2} \qquad (3)$$

where $\chi_E$ = concentration in energy emission units (Mev $sec^{-1}$ $m^{-3}$) $r$ = distance between the emitting element and the receptor (m)

In an absorbing but non-scattering medium an exponential absorption factor is introduced:

$$d\Phi = \frac{\chi_E e^{-\mu_a}}{4\pi r^2} \qquad (4)$$

If only the direct ray is considered (e.g., if the beam were collimated), the effect of scattering also is to attenuate the radiation exponentially so that

$$d\Phi = \frac{\chi_E e^{-(\mu_a + \mu_s) r} dV}{4\pi r^2} \qquad (5)$$

where $\mu_a$ is the linear scattering coefficient ($m^{-1}$). Finally, if the scattered radiation which reaches the receptor from all directions is included, the flux becomes

$$d\Phi = \frac{B\chi_E\, e^{-\mu r}\, dV}{4\pi r^2} \tag{6}$$

where $B$ = multiple scattering buildup factor, a function of $\mu r$ and $E$, and $\mu = \mu_a + \mu_s$, the total absorption-scattering coefficient ($m^{-1}$).

In Table I, values of $B$ are given for a range of values of $\mu r$ and $E$. These values of $B$ were calculated by Goldstein and Wilkins, Jr. of Nuclear Development Associated, Inc.[12] by numerical solution of the complicated integro-differential equation of energy transfer for gamma ray scattering.

For the range 0.5 to 2 Mev $B$ can be approximately represented by the three-term series:

$$B \cong 1 + \mu r + \frac{(\mu r)^2}{7E^{2.4}} \tag{7}$$

This formula is used in the subsequent calculations. An exponential formula for approximating $B$ has been developed by Taylor.[8]

The attenuation function $G(r)$ is here defined as:

$$G(r) \equiv \frac{B\, e^{-\mu r}}{r^2} \tag{8}$$

The "mean free path" $\lambda$, is defined as $1/\mu$. It is seen that for $E$ of the order of 1 Mev, when $r << \lambda$, $G(r) \cong 1/r^2$. That is, absorption and scattering can be neglected when the source and receptor are very close together.

The total flux of energy from each element of the cloud is:

$$d\Phi = \frac{G(r)}{4\pi}\chi_E dV \tag{9}$$

The concentration factor $\chi_E$ is the product of the total source strength $Q_E$ and a diffusion function $\chi$ representing the relative spatial distribution. The source strength, in turn, can be expressed as the product of an initial source strength $Q_0$ and a decay function $f(t)$, such that $Q_E = Q_0$ when $t = 0$, i.e., at the moment of release of the cloud. Then

$$d\Phi = \frac{1}{4\pi} Q_0 f(t) G(r) \chi(x,y,z,t) dx\, dy\, dz \tag{10}$$

where $x$ = distance horizontally downwind from the center of the cloud (m); $y$ = distance horizontally across wind from the x-axis (m), and $z$ = height above the ground (m).

The diffusion function $\chi$ ($x$, $y$, $z$, $t$) is, in general assuming no deposition on the ground):

$$\chi(x,y,z,t) = \frac{\left[e^{-\frac{(z+h)^2}{\sigma_z^2}} + e^{-\frac{(z-h)^2}{\sigma_z^2}}\right] e^{-\left(\frac{X^2}{\sigma_x^2} + \frac{y^2}{\sigma_y^2}\right)}}{\pi^{3/2}\,\sigma_x\sigma_y\sigma_z} \tag{11}$$

where $h$ = height of the center of the cloud above ground (m), $\sigma_x$, $\sigma_y$, $\sigma_z$ = cloud size parameters for the downwind, crosswind and vertical directions, respectively (m). The parameters $\sigma_x$, $\sigma_y$, and $\sigma_z$ depend upon the initial size and travel time of the puff and upon the spectrum and intensity of turbulent diffusion. Roberts[13] treated the case where the coefficients of turbulent diffusion in the $x$, $y$, and $z$ directions are considered to be constant. Taylor[14] showed the dependence of the diffusion upon the statistical properties of the atmospheric turbulence. Sutton[15] has solved the equations of diffusion by assuming for the Lagrangian autocorrelation function of the turbulent velocity components a mathematical form which behaves in a plausible manner. Sutton's solution, with parameters determined empirically, has been relatively successful in predicting concentrations of atmospheric contaminants from known sources. Frenkiel[16] has given more general methods for calculating the rate of spread of the cloud by means of observations of atmospheric turbulence. For the present purposes however the experience which has been gained in the choice of parameters in Sutton's and Roberts' formulas provides sufficient accuracy in relation to the uncertainties of the source strength and conditions of exposure. Roberts' and Sutton's methods of obtaining $\sigma_x$, $\sigma_y$ and $\sigma_z$ for the cloud resulting from an instantaneous point source are summarized in Table II.

**Table II. Expressions for $\sigma_x^2$, $\sigma_y^2$ and $\sigma_z^2$**

| | *Roberts** | *Sutton** |
|---|---|---|
| $\sigma_x^2$ | $2K_x t$ | $C_x^2(\bar{u}t)^{2-n}$ |
| $\sigma_y^2$ | $2K_y t$ | $C_y^2(\bar{u}t)^{2-n}$ |
| $\sigma_z^2$ | $2K_z t$ | $C_z^2(\bar{u}t)^{2-n}$ |

* $K_x$, $K_y$, $K_z$ = constant diffusion coefficients ($m^2$ $sec^{-1}$), $t$ = time elapsed since release (sec), $C_x$, $C_y$, $C_z$ = virtual diffusion coefficients ($m^{n/2}$), $u$ = mean wind speed or speed of movement of the center of the cloud (m $sec^{-1}$), $n$ = stability index (dimensionless).

In determining the intensity of gamma radiation from the entire cloud, the anisotropy of the turbulence can ordinarily be expected to have a second-order

**Table I. Multiple-Scattering Buildup Factor B**

| *E(Mev)* | $\mu r = 0$ | *1* | *2* | *4* | *7* | *10* | *15* | *20* |
|---|---|---|---|---|---|---|---|---|
| 0.255 | 1 | 3.09 | 7.14 | 23.0 | 72.9 | 166 | 456 | 982 |
| 0.5 | 1 | 2.52 | 5.14 | 14.3 | 38.8 | 77.6 | 178 | 334 |
| 1.0 | 1 | 2.13 | 3.71 | 7.68 | 16.2 | 27.1 | 50.4 | 82.2 |
| 2.0 | 1 | 1.83 | 2.77 | 4.88 | 8.46 | 12.4 | 19.5 | 27.7 |
| 3.0 | 1 | 1.69 | 2.42 | 3.91 | 6.23 | 8.63 | 12.8 | 17.0 |

effect; that is, it can be assumed that $K_x=K_y=K_z\equiv K$, $C_x=C_y=C_z\equiv C$, and hence $\sigma_x=\sigma_y=\sigma_z\equiv\sigma$. The integral to be evaluated is now:

$$D=\frac{\mu_a Q_0}{4\pi(6.8\times 10^{10}\ \text{Mev m}^{-3}\ \text{roentgen}^{-1})}$$
$$\times\int_0^\infty\int_0^\infty\int_{-\infty}^\infty\int_{-\infty}^\infty f(t)G(r)\chi(x,y,z,t)\,dx\,dy\,dz\,dt \tag{12}$$

where $f(t)$ = decay function, $G(r)$ = attenuation function, $\chi(x,y,z,t)$ = diffusion function, $r=\sqrt{(\bar{u}t+x-d)^2+y^2+z^2}$ (assuming the receptor is downwind from the point of release), $d$ = distance of receptor downwind from point of release (m).

A number of simplifications can be made:

1. The exponential of $-(z+h)^2/\sigma^2$ (the ground reflection term) in the diffusion function can be eliminated by changing the lower limit of $z$ from 0 to $-\infty$.

2. For the purpose of computing the integrated gamma dosage, the growth of the cloud by diffusion must be taken into account insofar as it affects the variation of dosage with distance of the receptor from the point of release of the puff. However, the cloud particles can be assumed to be moving approximately with the mean wind during irradiation of the receptor. This permits $t$ to be replaced by the constant $d/\bar{u}$ in the diffusion function. $\sigma$ then becomes a constant parameter in each integration.

3. $x$ can be neglected in the attenuation function since (1) $x$ is simply added to $\bar{u}t$, (2) the diffusion function is symmetrical in $x$ and (3) the values of $x$ which are small in relation to $\sigma$ and hence in relation to $\bar{u}t$ are most heavily weighted by the diffusion function. Then the $x$ integration now involves only the Gaussian function and can be carried out explicitly:

$$\int_{-\infty}^\infty f(t)G(r)\chi(x,y,z,d/\bar{u})\,dx=f(t)G(r)\frac{e^{-\frac{(z+h)^2+y^2}{\sigma^2}}}{\pi\sigma^2} \tag{13}$$

where $r=\sqrt{(\bar{u}t-d)^2+y^2+z^2}$

$$\sigma^2=C^2d^{2-n}\ \text{(Sutton)}$$

or

$$2Kd/\bar{u}\ \text{(Roberts)}$$

Now let $s=\sqrt{y^2+z^2}$ and $\theta=\tan^{-1}(z/y)$. Then $r=\sqrt{\bar{u}t-d)^2+s^2}$ and both $G(r)$ and $f(t)$ are independent of $\theta$. The integral becomes

$$D=\frac{A}{2\pi}\int_0^\infty\int_0^\infty\left[\int_0^{2\pi}e^{2\,sh\sin\theta/\sigma^2}d\theta\right]$$
$$\left[e^{-(S^2+h^2)/\sigma^2}f(t)G(r)\right]s\,ds\,dt \tag{14}$$

where

$$A=\frac{\mu_a Q_0}{2\pi\sigma^2\ (6.8\times 10^{10}\ \text{Mev m}^{-3}\ \text{roentgen}^{-1})}$$

But

$$\int_0^{2\pi}e^{2\,sh\sin\theta/\sigma^2}\,d\theta=2\pi\,J_0(2ish/\sigma^2) \tag{15}$$

where $J_0$ = zero-order Bessel function and $i=\sqrt{-1}$.

Finally the gamma dosage integral can be written:

$$D=A\int_0^\infty\left[J_0(\alpha)e^{-\beta}\right]\left[\int_0^\infty f(t)G(r)dt\right]s\,ds \tag{16}$$

where $\alpha=2ish/\sigma^2$ and $\beta=(s^2+h^2)/\sigma^2$

Thus the problem is reduced essentially to two steps:

1. Integrating the product of the attenuation and decay functions with respect to time for each perpendicular distance of passage, $s$, of the cloud particles. This, when multiplied by a conversion factor, gives the dosage from a unit point source of radiation travelling with the velocity $\bar{u}$ and passing at a perpendicular distance $s$ from a receptor located at a distance $d$ downwind from the point of origin.

2. Integrating the product of the point-source dosage determined in step 1 and the distribution function of particle concentration with respect to the distance $s$ of the particle trajectories from the receptor. This gives the integrated dosage received from all the cloud particles.

## CLOUD GAMMA DOSAGE IN THE SPECIAL CASE OF A NUCLEAR POWER EXCURSION

A short-lived nuclear reactor power excursion of 1 Mw-sec (1 megawatt-second = $10^6$ joules) would result in the production of radioactive fission products which would emit approximately $4.8\times 10^{16}\ (t/1\ \text{sec})^{-1.21}$ Mev/sec of gamma energy with an average energy of 0.7 Mev/disintegration.[8] This formula overestimates the source strength for $t<10$ sec. However, the time integral of the actual source strength from 0 to 10 sec is very nearly equal to the integral of the formula from 1 to 10 sec. Since integrated dosages are desired, and since the variation of the associated attenuation factor between $t=0$ and $t=1$ sec can generally be neglected in the range of distances and wind speeds of meteorological interest, the lower time limit can be taken as 1 sec, $Q_0$ as $4.8\times 10^{16}\ Q_E$ where $Q_E$ is the nuclear energy release in Mw-sec, and $f(t)$ as $(t/1\ \text{sec})^{-1.21}$.

The time integration of the product of the attenuation and decay factors of Equation 16 was carried out numerically for a selection of values of $s$, $d$ and $u$ sufficient to permit graphical interpolation to two decimal places. From these results a graph was prepared showing the gamma dosage in roentgens received at the ground at a distance $d$ downwind from the point of release, assuming that all the radioactivity produced in a power excursion of 1 Mw-sec is concentrated in a point source which passes the receptor at distance $s$ with $s$ interpreted as a height $h'$. This "unit point-source dosage" graph is shown in Fig. 1. The

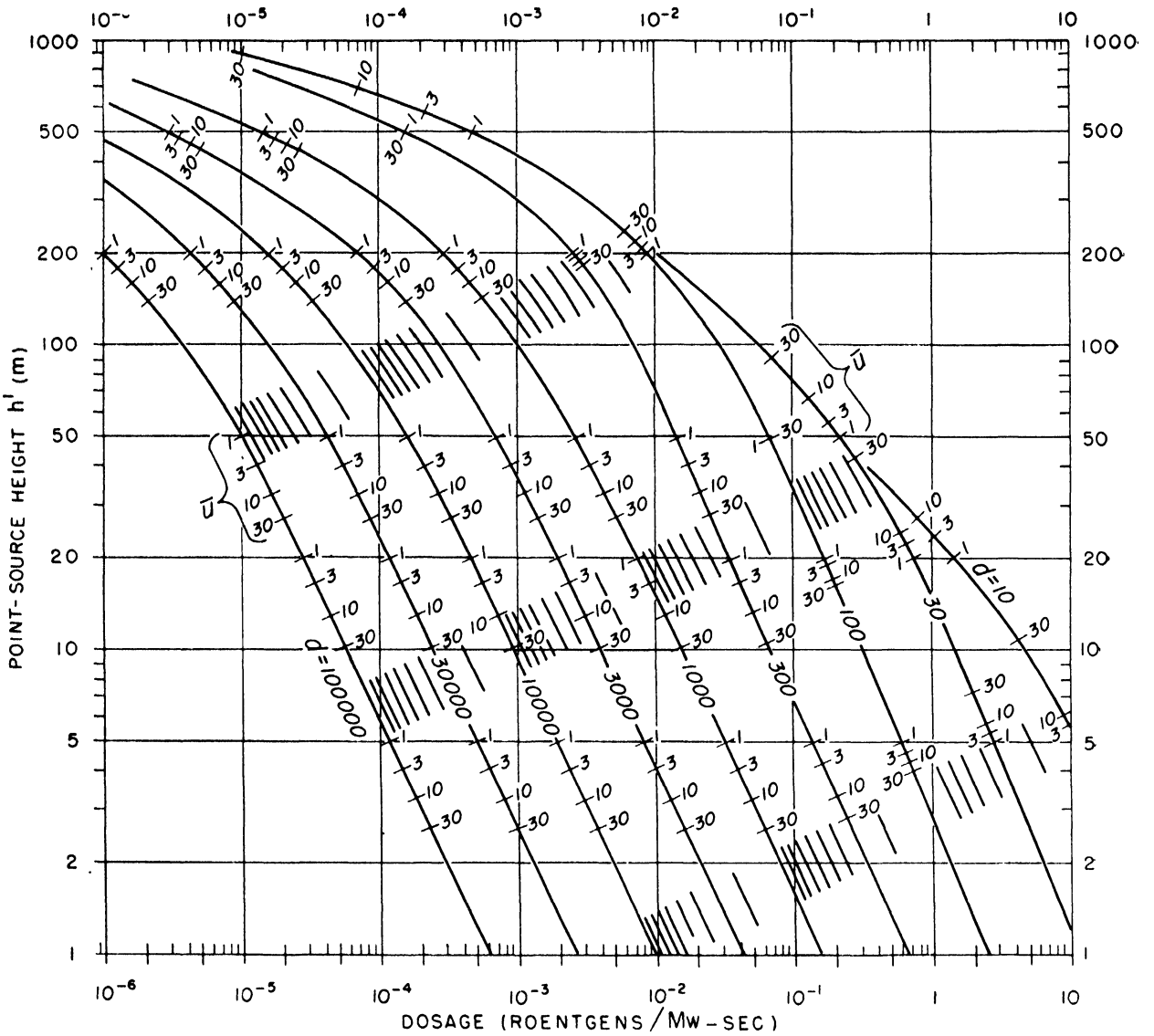



Figure 1. Unit point-source gamma dosage due to power excursion products, received at distance $d$ (m) downwind from initial source with wind speed $u$ (m/sec) shown as displacement from 1 m/sec mark along lines of constant distance

curves are for constant distance $d$. The wind speed is introduced as a displacement along the distance curves.

The spatial distribution function was computed for a selection of values of $s$, $h$ and $\sigma$, then multiplied by the "unit point-source dosage." The product was integrated numerically with respect to $s$ for values of $\sigma$ corresponding to "neutral" meteorological conditions (Sutton's stability parameter $n = 0.25$). These conditions and the resulting dosages are summarized in Table III.

The computed dosage from a cloud at the height $h$ whose size is described by the parameter $\sigma$ is equal to the dosage from a point source of the same total strength but passing at a different height $h'$. The "equivalent point-source height" $h'$ can be considered to be dependent only on $h$ and $\sigma$. From the graphs of point-source dosage (Fig. 1) and cloud dosage (Table III) which had been derived so far, a graph

Table III. Cloud Gamma Dosage, Power Excursion Products, "Neutral" Conditions

| $h$ (m) | $n$ | $C$ ($m^{n/2}$) | $u$ (m/sec) | $d$ (m) | $\sigma$ (m) | $D$ (r/Mw-sec) |
|---|---|---|---|---|---|---|
| 0 | 0.25 | 0.20 | 3 | 30 | 3.9 | 6.4 |
| | | | | 300 | 22 | $5.1 \times 10^{-2}$ |
| | | | | 3000 | 221 | $2.35 \times 10^{-4}$ |
| | | | | 30,000 | 1654 | $3.52 \times 10^{-7}$ |
| 50 | 0.25 | 0.15 | 5 | 300 | 22 | $1.96 \times 10^{-2}$ |
| | | | | 563 | 38 | $1.07 \times 10^{-2}$ |
| | | | | 3000 | 166 | $3.88 \times 10^{-4}$ |
| | | | | 30,000 | 1240 | $7.44 \times 10^{-7}$ |
| 200 | 0.25 | 0.10 | 7 | 300 | 15 | $2.74 \times 10^{-3}$ |
| | | | | 3000 | 110 | $1.43 \times 10^{-4}$ |
| | | | | 4380 | 154 | $1.31 \times 10^{-4}$ |
| | | | | 30,000 | 827 | $1.73 \times 10^{-6}$ |
| 500 | 0.25 | 0.08 | 9 | 300 | 12 | $1.57 \times 10^{-4}$ |
| | | | | 3000 | 88 | $5.56 \times 10^{-6}$ |
| | | | | 16,000 | 382 | $4.05 \times 10^{-6}$ |
| | | | | 30,000 | 661 | $1.73 \times 10^{-6}$ |

of the "equivalent point-source height" was constructed (Fig. 2). The coordinates are $\sigma$ and $h'$, with curves as constant values of actual cloud height $h$.

The "equivalent point-source height" graph and the "unit point-source dosage" graph were combined into a nomogram for computing the actual unit-source dosage, given the cloud-size parameter and height $h$. Alignment-type nomograms were added to permit the computation of $\sigma$ by either Sutton's or Roberts' formula, and to multiply the unit-source dosage by the actual source strength. The complete nomogram, with instructions for its use, is contained in reference 6.

When $\sigma$ exceeds 2000 m the dosage can be computed on the assumption of approximate radiative equilibrium. The formula for a receptor at the ground is then:

$$D = \frac{(4.8 \times 10^{16}\ \text{Mev/Mw-sec})\, Q_B\, (\bar{u}/d)^{1.21}\, e^{-h^2/\sigma^2}}{(6.8 \times 10^{10}\ \text{Mev/m}^3\ \text{roentgen})\, \pi\, u\, \sigma^2} \qquad (17)$$

However, in this case as well as that of $h > 500$ m a power excursion of at least 2000 Mw-sec would be required to produce a dosage as large as 1 roentgen anywhere on the ground.

A few interesting features of Fig. 2 can be pointed out. For example, elevated clouds behave essentially as point sources as long as $\sigma < h/5$. The maximum dosage, or minimum equivalent point-source height for a cloud of given strength and height occurs when $\sigma$ is about $3\ h/4$. When $\sigma$ increases beyond about $2h$, the cloud height has no further effect on the dosage.

In order to show the range of values to be expected under a plausible variety of meteorological conditions, dosages have been computed by means of the nomograms for the cases given in Table IV.

Computations were carried out in each case except "trapping" both for an initial point source and for an initial volume source for which the value of $\sigma$ at the origin was taken as $\sigma_0 = 10 + 0.3h$. This rate of cloud spread corresponds roughly to that given by Sutton's hot puff formula[17] with $C = 0.6$. The "trapping" case was designed to approximate a situation in which diffusion is confined to the downward direction (as by an inversion) and horizontal spread is also confined (as by a valley). To give a maximum dosage at each distance, $\sigma$ was held constant at $0.75\ h$ and the computed dosages were doubled to account for downward reflection by an inversion. Selected results are shown in Fig. 3.

When $h = 0$ the initial point source gives significantly larger dosages at small distances than does the initial volume source. The dosages increase with increasing atmospheric stability at all distances. Dosages exceeding 1 r/Mw-sec are confined to the closest 100 m except in stable conditions with very small initial volumes. The strong wind case (not shown) gives dosages slightly higher than the neutral case.

The elevated sources show less variability of the dosage with the meteorological variables. Trapping, of course, always gives the highest dosages. Of the other cases, the stable case results in the highest dosages at great distances while the strong-wind case is worse at intermediate heights and distances. At greater heights instability, with more rapid downward spread of the cloud, results in the highest dosages at small and intermediate distances, except for trapping.

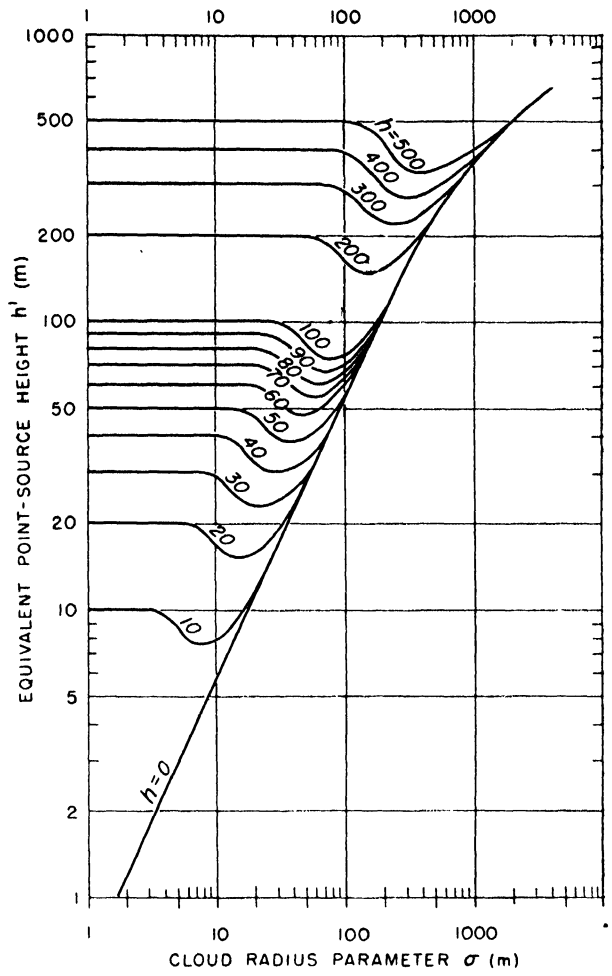
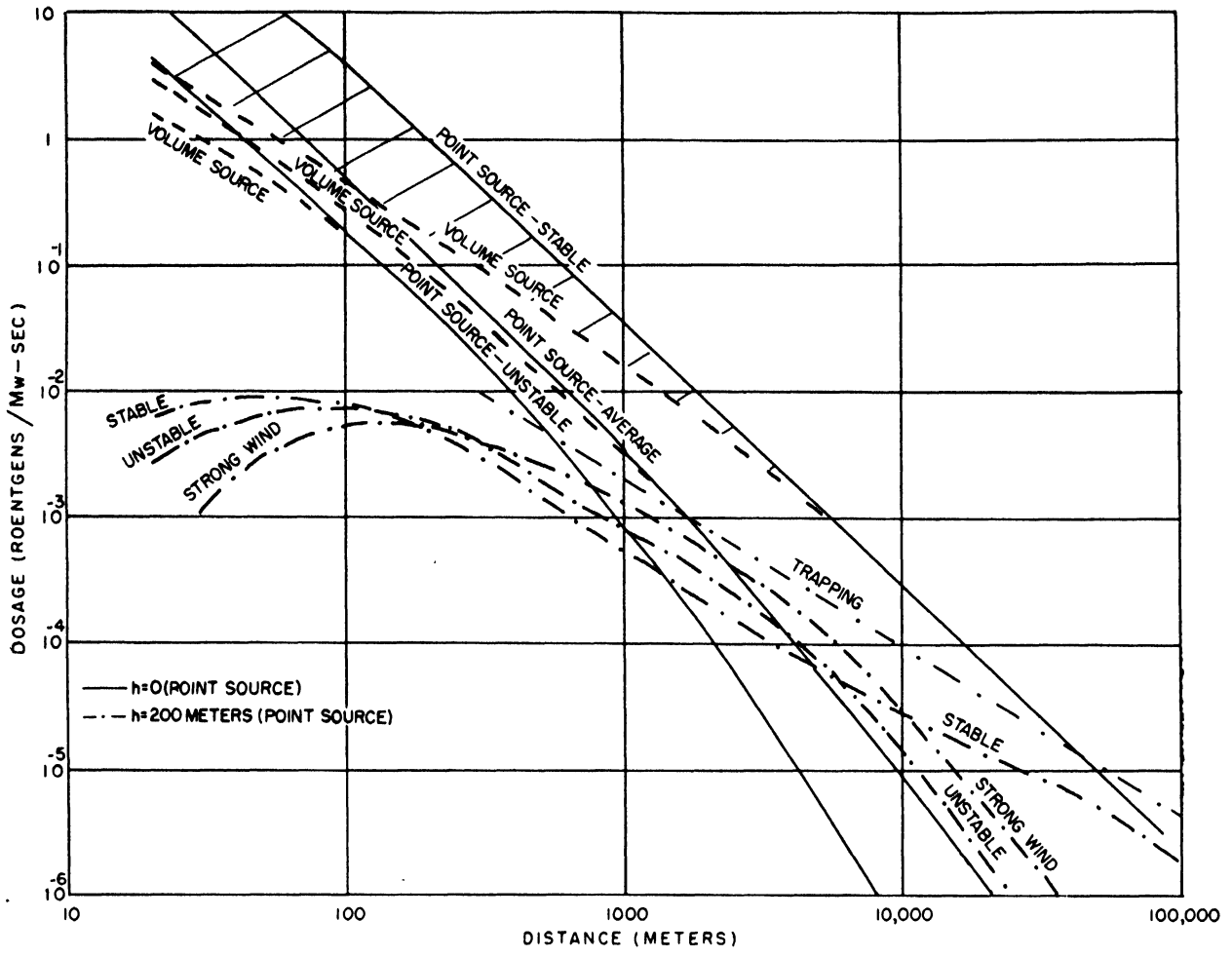



**Figure 2. Equivalent point-source height for cloud at height h(m)**

**Table IV. Meteorological Parameters for Sample Cases**

| Case | $n$ | $C$ ($m^{n/2}$) $h = 0$ | 50 | 200 | 500 | $a$ (m/sec) 0 | 50 | 200 | 500 |
|---|---|---|---|---|---|---|---|---|---|
| Neutral | 0.25 | 0.20 | 0.15 | 0.10 | 0.08 | 3 | 5 | 7 | 9 |
| Stable | 0.50 | 0.05 | 0.03 | 0.02 | 0.01 | 1 | 3 | 6 | 9 |
| Unstable | 0.20 | 0.50 | 0.20 | 0.15 | 0.10 | 7 | 10 | 12 | 13 |
| Strong wind | 0.25 | 0.20 | 0.15 | 0.10 | 0.08 | 15 | 22 | 30 | 35 |
| Trapping | —— | —— | $= 0.75\ h$ | —— | —— | -- | 5 | 7 | 9 |




GAMMA DOSAGE VARIABILITY — POWER EXCURSION

**Figure 3. Gamma dosage with various initial heights, volume and meteorological conditions (power-excursion case)**

A comparison of the results obtained by actual integration in the "neutral" case with those obtained by Waterfield,[9] Taylor[8] and Fitzgerald *et al.*[3] is given in Table V for an initial point source. It is seen that while the agreement is generally within a factor of two for surface clouds, simplifications in the assumed attenuation introduce greater differences as the elevation is increased.

## CLOUD GAMMA DOSAGE IN THE CASE OF STEADY-POWER FISSION PRODUCTS

If the contents of a nuclear reactor which has been operating at a steady-power level were suddenly ejected into the atmosphere, the gamma source strength in Mev/sec[18] would be:

$$Q = 2.3 \times 10^{14} P\,[t^{-0.21} - (t_0 + t)^{-0.21}] \qquad (18)$$

where $P$ = previous steady power of the reactor (kw), $t$ = time after shutdown (sec), and $t_0$ = duration of steady operation (sec).

If $t_0$ is greater than a few months this can be approximated by $Q = 2.3 \times 10^{14} P t^{-0.21}$ without serious error. In view of the slow decay rate, it is justifiable for the purpose of estimating hazards to hold $Q$ constant at the value corresponding to $t = d/u$ in carrying out the space-time integration of radiation flux from the cloud.

**Table V. Comparison with Previous Authors, Power Excursion Case, "Neutral" Conditions (roentgens per Mw-sec)**

| $h$ | $d$, m | *Holland* | *Waterfield* | *Taylor* | *Fitzgerald et al.* |
|---|---|---|---|---|---|
| 0 | 30 | 6.4 | 5.9 | 5.6 | 8.7 |
| | 300 | $5.1 \times 10^{-2}$ | $3.1 \times 10^{-2}$ | $4.2 \times 10^{-2}$ | $6.2 \times 10^{-2}$ |
| | 3000 | $2.35 \times 10^{-4}$ | $1.1 \times 10^{-4}$ | $1.4 \times 10^{-4}$ | $2.7 \times 10^{-4}$ |
| | 30,000 | $3.52 \times 10^{-7}$ | $2.9 \times 10^{-7}$ | $3.8 \times 10^{-7}$ | $4.4 \times 10^{-7}$ |
| 50 | 30 | 0.17 | 0.1 | | |
| | 300 | $1.96 \times 10^{-2}$ | $8.1 \times 10^{-3}$ | | |
| | 3000 | $3.88 \times 10^{-4}$ | $1.84 \times 10^{-4}$ | | |
| | 30,000 | $7.45 \times 10^{-7}$ | $5.1 \times 10^{-7}$ | | |
| 200 | 30 | $6.1 \times 10^{-2}$ | $4.9 \times 10^{-2}$ | | |
| | 300 | $2.74 \times 10^{-2}$ | $3.5 \times 10^{-4}$ | | |
| | 3000 | $1.43 \times 10^{-4}$ | $4.2 \times 10^{-5}$ | | |
| | 30,000 | $1.73 \times 10^{-6}$ | $8.8 \times 10^{-7}$ | | |
| 500 | 30 | $1.2 \times 10^{-4}$ | | | |
| | 300 | $1.57 \times 10^{-4}$ | | | |
| | 3000 | $5.56 \times 10^{-6}$ | | | |
| | 30,000 | $1.73 \times 10^{-6}$ | | | |

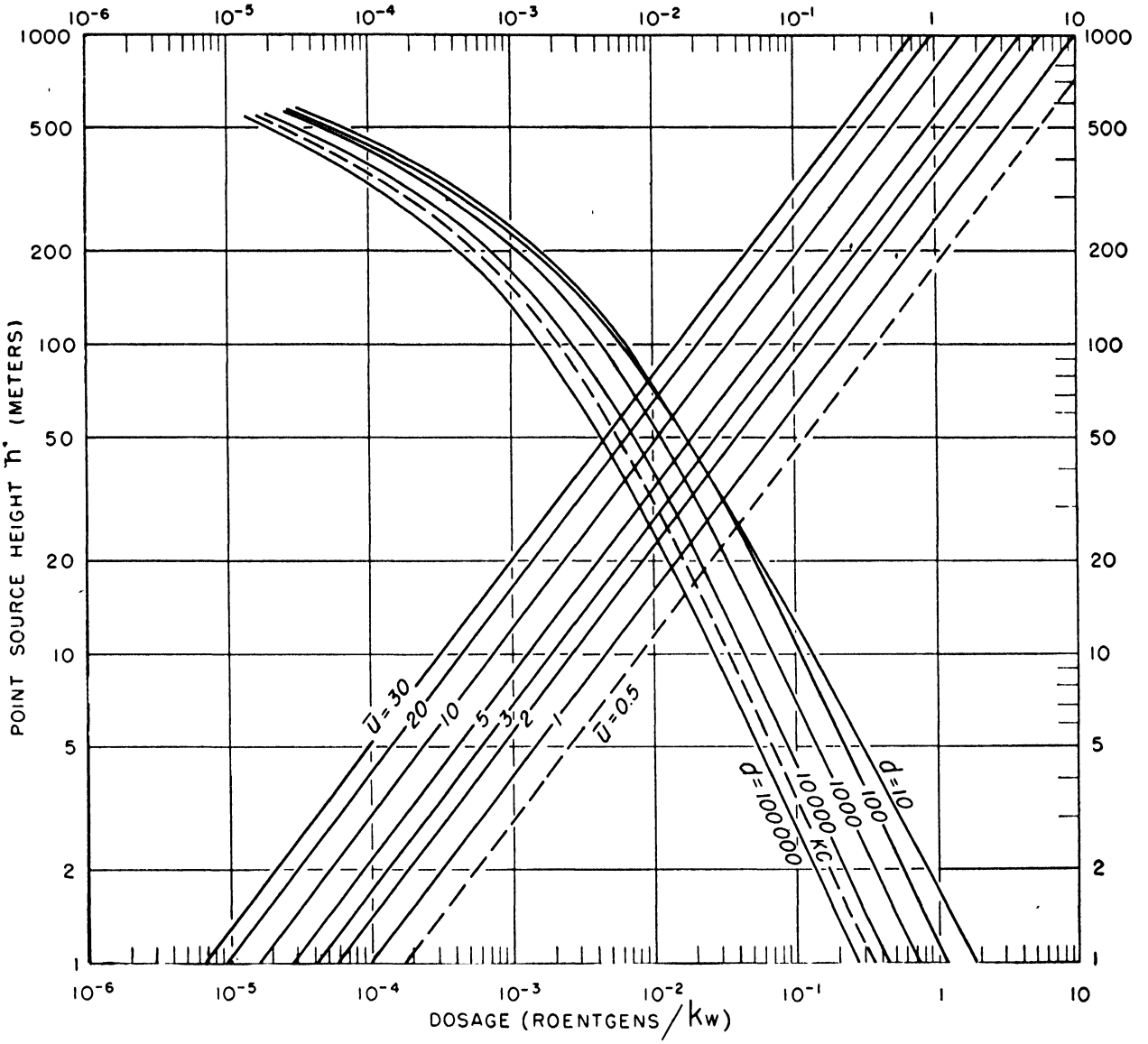



**Figure 4. Unit point source gamma dosage due to steady power products, received at distance d (m) downwind from initial source with wind speed u (m/sec) shown as horizontal displacement from 1 m/sec line**

The unit point-source dosages for this case were computed and graphed (Fig. 4).The curves for constant distance $d$ are drawn for a wind speed of 1 m/sec. The dosage now decreases with increasing wind speed at all distances. The wind speed is introduced as a horizontal displacement indicated by the spacing between the sloping wind speed lines. A nomogram for computing cloud gamma dosages for the steady-power case has been constructed by substituting the graph of Fig. 4 for Fig. 1 in the power-excursion nomogram. The steady-power nomogram with instructions is also contained in reference 6.

## GAMMA DOSAGE DUE TO CLOUDS OF ARBITRARY RADIOISOTOPES

The dashed curved marked "*KC*" in Fig. 4 is included to permit computation of the gamma dosage due to the passage of a cloud of any specific radioisotope whose gamma energy is of the order of 0.5 to 1 Mev and whose half-life is large compared to the cloud passage time (say 10 $\sigma/\bar{u}$). This curve gives the point source dosage due to 1 kilocurie of an isotope emitting a gamma photon of 0.7 Mev energy and passing at the height $h$ with the speed 1 m/sec. This dosage should be multiplied by

$$\frac{Q_i E e^{-0.693d/\bar{u}T}}{0.7\bar{u}} \qquad (19)$$

where $Q_i$ = the initial source strength (kilocuries), $E$ = gamma photon energy (Mev), and $T$ = half-life (sec).

The "*KC*" curve can also be used in this same manner for continuous source problems. Substitution of the emission rate $dQ_i/dt$ in kc/hr for $Q_i$ yields the gamma dose rate $dD/dt$ in r/hr.

## CONCLUSION

With the solution of the cloud gamma dosage problem, the nuclear reactor engineer now has a set of tools for analyzing the environmental consequences of hypothetical accidents. While imperfect, these tools are believed to be comparable in precision to those which are available for analyzing the possible failures of the reactor assemblies themselves.

## ACKNOWLEDGEMENT

The author gratefully acknowledges the assistance of David A. Lawson in carrying out the numerical integrations required in obtaining the results presented here.

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# Radiological Monitoring of a Nuclear Release

**By Percy Griffiths, Claude W. Sill and Mack Wilhelmsen,* USA**

At the National Reactor Testing Station in the State of Idaho, a network of radiation detection and sample collection devices is operated on a continuous basis in order to provide complete radiological control of local operations. Particular emphasis is placed upon downwind locations from all operating installations. Types of radiation monitoring devices at present in use are: continuous air monitors, gamma scintillation scanners, particle collectors and fall-out collectors. In addition to physical observation of meters and recorders utilized with certain of these devices, radiochemical and radioautographic analyses are performed.

Personnel of the various health physics groups support the established radiation monitoring network. In the event of a mishap at any of the experimental sites, or when the nature of experimentation is such that hazardous conditions could develop, these persons are alerted. In some extreme cases, personnel and equipment are based in close proximity to the work in progress.

During the summer of 1953 and spring of 1954, experiments were conducted by Argonne National Laboratory utilizing a small remotely-operated, water-cooled and moderated reactor. During an intential short period excursion on July 22, 1954, a violent release of energy occurred which destroyed the reactor core and some associated equipment. As this possibility had been anticipated, all precautions had been taken to minimize any resultant hazard. Five mobile radiation-monitoring teams were based around the reactor area at distances greater than 800 meters. Highway barricades were readied for closure of traveled roads should the necessity arise. All operational and observation personnel were based at the control area, a distance of 800 meters from the reactor. In addition all travel had been restricted within this distance (Fig. 1).

Immediately following the short excursion, a column of dark grey smoke and dust was observed blowing out through the top of the reactor to a height of approximately 25 meters. A short, sharp explosion was heard, followed by a slight shock wave, felt at the control area. At this time the control mechanism, tower, and various other objects were blown into the air, then fell off to the sides of the earth dike shielding the reactor. Following the initial emission the cloud drifted slowly toward the southwest at a speed of about 8 kilometers per hour. At a distance of approximately 1.2 kilometers, the cloud began to rise in a thermal, and at an elevation of about 300 meters above the ground gradually diffused until it was no longer visible.

Simultaneously with the release several portable radiation detection instruments located 800 meters from the reactor indicated short radiation exposure rates beyond the limit of the range of the instruments being used at this time, or an exposure rate greater than 500 mr per hour. Subsequent development and interpretation of X-ray film indicated a total dosage of approximately 50 mr at 440 meters, and 30 mr at 580 meters.

Within a few minutes of the release all mobile teams began monitoring downwind from the reactor taking readings 1 meter above ground level. Fifteen minutes after the release the maximum measurement was 6 mr per hour at 1.6 kilometers from the reactor; 25 minutes after the release 2.0 mr per hour at 3.2 kilometers. Meteorological data indicated that the cloud travelled to the southwest for approximately 1 hour at a speed of about 8 kilometers per hour. The wind then reversed as the temperature distribution in the lower layers of the air changed from inversion to lapse, and the cloud travelled toward the northwest at speeds up to 10 kilometers per hour (Fig. 2). This trajectory was confirmed when an increase of radiation amounting to 6 times the prevailing background was detected by a continuous air monitoring device located 13 kilometers from the point where the wind shift had occurred 3 hours earlier. Stations ranging from 8 to 110 kilometers further downwind did not indicate any radiation as a consequence of the experiment.

The only indication of radioactive fallout was obtained from direct readings of the ground following the cloud passage to the southwest up to a distance of 3.2 kilometers from the reactor. The maximum was determined at a distance of 1.6 kilometers where the readings were 6 mr per hour at 1 hour after the release, 0.5 mr per hour at 6 hours, and 0.05 mr per hour at 26 hours.

Within the first hour it was established that there was no radiation hazard to anyone outside the 800 meter perimeter as a result of the experiment. Radi-

* US Atomic Energy Commission. Prepared by Percy Griffiths, Claude W. Sill; including work by Health & Safety Division, Idaho Operations Office, US Atomic Energy Commission and US Weather Bureau.

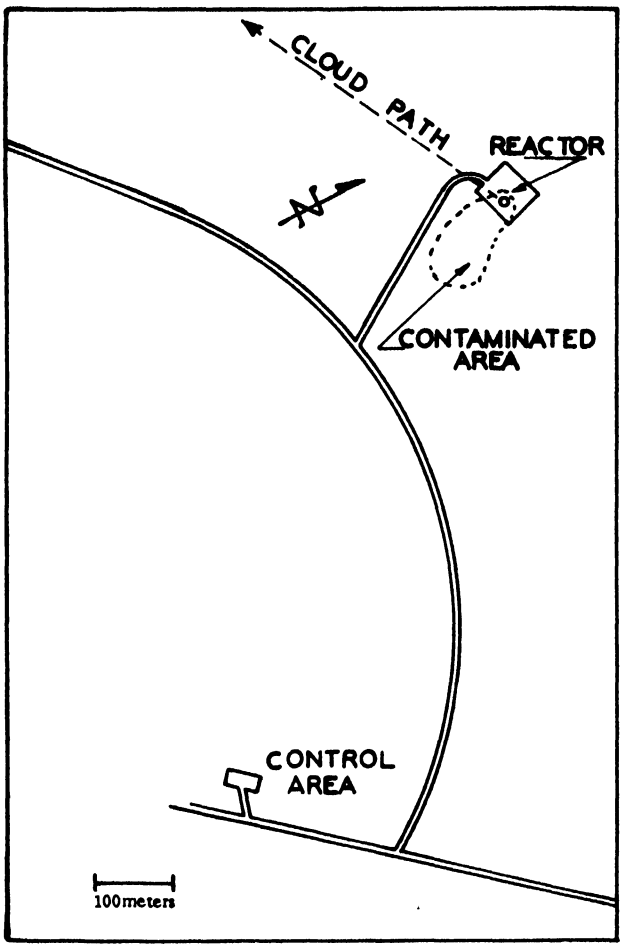



Figure 1. Reactor location and adjacent area

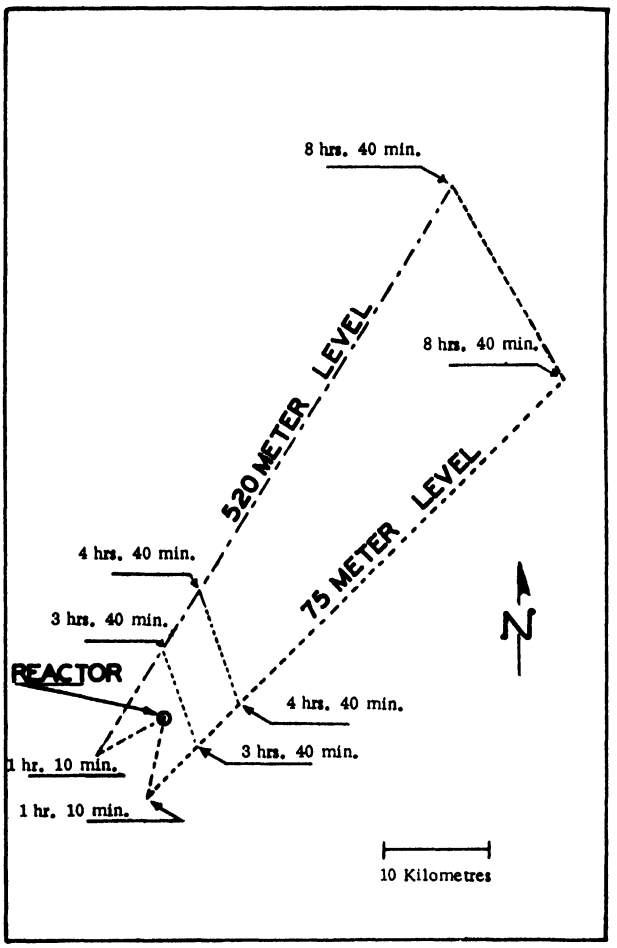



Figure 2. Meteorological trajectories

ation protection activity was then directed toward the reactor area proper in order to establish safe time limits for the purpose of making inspections, and for removing photographic film before it became overexposed from the high level of radiation present. The first attempt to enter the reactor area by way of the access road from the south failed when radiation exposure rates up to 40 roentgens per hour were encountered at 60 meters from the reactor. Further investigation proved that the south side of the reactor area to a distance of approximately 120 meters contained a large number of fuel plate and reactor fragments. However, entry to the reactor area proper was accomplished from the east side in a radiation field of 1 roentgen per hour. Removal of film and initial inspections were possible in radiation fields up to 30 roentgens per hour. At this time, 5½ hours after the release, the radiation level over the top of the reactor pit was 8 roentgens per hour, 10 roentgens per hour at the west end of the pump pit, and 30 roentgens per hour 15 meters south of the reactor from fuel plate fragments scattered on the ground.

Five days after the experiment, radiation levels had decreased sufficiently to permit commencement of salvage and decontamination. A change area was set up at the west side of the reactor at a distance of 75 meters, where radiation background levels were less than 1 mr per hour. Instruments and salvageable materials were removed to this location and decontamination performed with a minimum of difficulty. Exposures of persons working on this operation were kept below 3.9 roentgens over the entire period.

The affected area south of the reactor was of particular interest. Five days after the release, expeditions were started into this area to remove and store all visible articles of radioactive debris. Eleven days after the release all of these items had been removed but the radiation level had not been lowered significantly. The area was set up on a 3-meter grid pattern and surveyed at intersections. At each location 2 types of readings were taken with an open window ionization type instrument. One set of readings was obtained at ground level, the other at 1 meter above. The resultant readings were then plotted according to levels of 1 to 10 mr per hour, 10 to 50 mr per hour, and greater than 50 mr per hour (Fig. 3a and 3b). An area of approximately 7800 square meters was proven to be contaminated in a strip approximately 60 meters in width and 130 meters in length. It appears that as a result of the explosion, the base plate of the reactor control rod

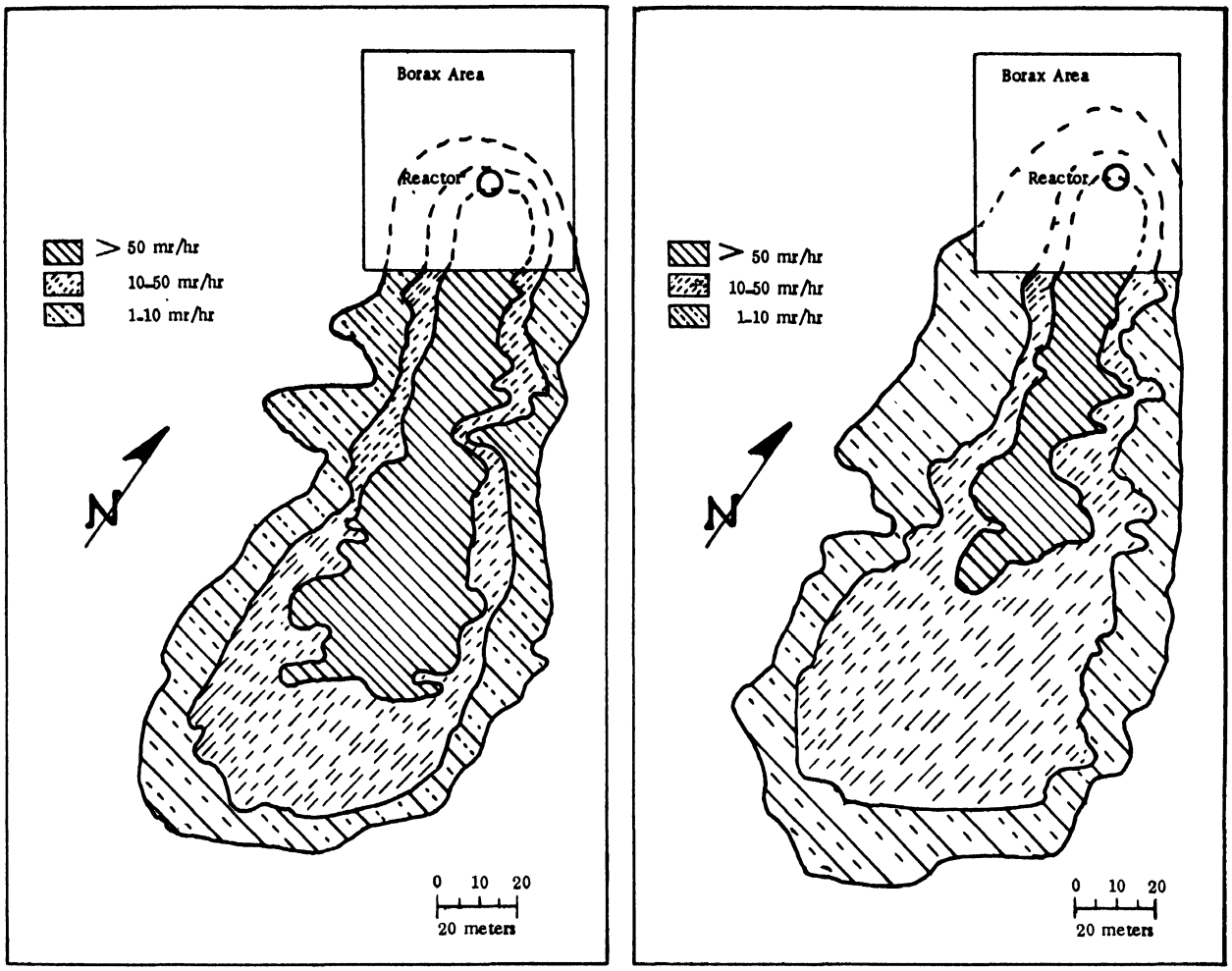



Figure 3. (Left) Survey of contaminated area taken at ground level. (Right) Survey of contaminated area taken at 1 meter above ground

mechanism had tilted as it broke loose from the platform, forming an approximate 45 degree angle. Fuel elements and associated debris were deflected toward the south by the obstruction. The entire affected area was later covered with a layer of gravel to a minimum depth of 15 centimeters which reduced the radiation dosage rate to less than 5 mr per hour.

Because the reactor was remotely operated with an established safety distance of 800 meters, no hazard to operating personnel or to others at the testing station was present. The entire experiment was accomplished with no radiation exposure to personnel above permissible levels. A similar release involving a directly operated reactor of similar size would undoubtedly subject personnel within the immediate vicinity to radiation exposures and airborne contaminants of serious magnitude unless adequate shielding and associated safeguards comprised part of the installation.

It would appear that, in design criteria for similar experimental reactors, that provision should be made to control the direction of debris blowing out as a consequence of a release such as occurred in this instance. Entry and cleanup would be facilitated and the possibility of contaminating expensive equipment and property minimized.

The health physics function in respect to this experiment was primarily personnel protection. Unfortunately, expenditure for scientific data had not been considered. As a result, much information of value to nuclear science was not obtained.

# Prospective Uses of Atomic Energy from the Viewpoint of Radiology

**By F. Herčik, Czechoslovakia**

The use of atomic energy for peaceful purposes confronts us with very important problems. In a relatively short period of time the level of radiation, which is of certain importance in man's life, will increase. Until now man, as well as all other living organisms, has been subjected to the action of cosmic rays, earth's radioactivity, action of radioactive matter contained in air and water, and action of radioactive potassium within his body. There is no doubt that living organisms have adapted themselves to this radiation. I do not propose to discuss the part that this radiation may have played in the phylogenetic development of organisms; however, there is no doubt that the basic doses of radiation from the above sources have conditioned and are continuously conditioning the existence of all living things. The total radiation dose received by a person in the course of one year from the above sources is about 0.1 r. According to our present day knowledge of these hazards, the amount is so small that it should not be of any significance.

The problem will assume a very different aspect when the sources of atomic energy become more numerous. There is no doubt that this will cause an increase in the dose, both as a result of external radiation from various sources, which, in one way or another, will work themselves into subterranean waters, will increase the quantity of radioactive matter present in drinking water. It appears that the problem of the increase of natural radioactivity in drinking water will become one of the most serious ones which require a solution. Furthermore, some attention will have to be given to the problem of the external radiation arising as a result of the use of radioactive sources in their most varied technological forms. Here, the most important thing will be some form of protection against radiation which will be not only conveniently realizable technically but also economically feasible.

In order properly to evaluate the radiation hazard, the question of the permissible doses will have to be subject to revision. In my opinion, two sorts of errors are committed at this point. On the one hand, the permissible dose is unduly lowered on the basis of experimental facts obtained with test subjects phylogenetically far removed from man. In this category, for instance, belongs the generalization of data obtained by tests on plant cells or on several forms of animal life such as *Drosophila*. It seems to me that here, the error is made by all those who forget that the human body is regulated, to a great extent, by neuro-humoral mechanisms, as a result of which, most probably, the recovery reactions are much more extensive than in simple biological subjects. Moreover, Darlington's and Koller's experiments have shown to us that, even in subjects taken in the plant world, there is a repair of chromosome damage after irradiation. The cells that have been damaged do not multiply, but are replaced by healthy cells which continue to multiply. The more active the tissue, the more it is in accordance with Lacassagne's rule, sensitive to irradiation; but also the more vigorously do the recuperating processes take place. In the highest types of mammals, as in man, each cell evidently is controlled, in some way, by a neuro-humoral regulation. As an example of this, we might mention the central control of cell permeability, and adjustment of the fluid balance in the body. Thus, we are well entitled to assume that, following irradiation, the recovery processes in the human body are centrally controlled, and that they are probably equipped to repair more serious damage than would the simpler organisms. On the other hand, a similar error is often committed by those who, falling into the other extreme, propose to increase the permissible doses unduly, without factual information.

I think that the effect of irradiation on the whole of the organism must be evaluated on the basis of the nature of functional integration. Plants, in most instances, have humoral integration, while animals have various degrees of neuro-humoral regulation. The neural component continues to increase, as the highest phylogenetic forms are reached.

It must further be borne in mind that it is impossible to assume that, in biological objects of any degree of complexity, the effect of irradiation on any biological function is continuous, in other words, that the changes brought about by small doses will only increase quantitatively as the doses are increased. In fact, depending on the size of the dose, the changes are *qualitatively* different. For a given biological object, small doses cause a set of reactions whose nature is entirely different from those caused

Original language: Russian.

by large doses. It is also known that partial irradiation of an organism calls forth changes which are different from those due to general irradiation. In view of all this it is necessary to verify the doses which have heretofore been considered permissible; this in turn involves the necessity of explaining the basic radiobiological processes arising in the irradiated organism. In particular, it is necessary to explain, on the one hand, the primary effect of irradiation on living matter, and, on the other hand, the processes whose development takes place as a secondary effect.

In this connection, it must be admitted that, although experimental work has been going on for more than 50 years, our knowledge of the effects of irradiation on the living organisms is still insufficient. This applies even to our knowledge of the primary process. I do not propose to deal here with the advantages and shortcomings of the target theory and radical hypothesis. I should only like to stress that, even during the interaction of hard radiation photons with matter, not only does ionization take place, as is usually assumed, but also atom excitation. It is known that one half the energy of the hard X-rays is dissipated in the form of atom excitation, from which it follows that, in these cases, a portion of the radiant energy changes into an effect similar or identical to the effect of ultra-violet radiation. In other words, X-rays, and in all likelihood also, other forms of penetrating radiation, exhibit not only an ionization effect, but also an effect of the excitational type which, in turn, creates changes in the chemical forces which bind the atoms of a molecule. Both these processes can be noticed during an experiment, and it is thus possible to investigate the value of each of the two components. Possibly, this new standpoint will have an effect on the explanation of primary processes taking place in living matter following penetrating irradiation.

At the present time, there is no doubt that the primary processes in irradiated living matter primarily affect the nucleo-proteins. Here, the effects of irradiation are discontinuous, as I have already indicated above; i.e., the effects of the small irradiation doses are different from those of the medium and large doses. Irradiation acts primarily on nucleo-protein metabolism. I submit that small irradiation doses only bring about a deviation of nucleo-protein metabolism, of a form which results in the creation of a protein foreign to the body, to which the organism can react by forming some protective material, leading to the creation of a protective barrier around this foreign protein. In the case of an incomplete blockade, this can be of a carcinogenic nature, or cause some other undesirable effect. A large dose leads to the slowing down of the nucleo-protein metabolism, which is evidenced by the lowering of the phosphorus exchange rate in the nucleic acids. The latter action is connected with full blocking of the nucleic acid exchange. Still larger doses lead to deamination and dephosphorylation of the proteins, and thus to their degradation. Thus, different doses of irradiation most probably cause qualitatively different processes in the organism, which call forth changes of differing macroscopic appearance.

On the basis of the above, it becomes clear that the action of the protective material must also be analyzed discontinuously. It is quite legitimate to assume that the material which is blocking the action of small irradiation doses must have properties completely different from those of the substances which must block the reaction to large irradiation doses. For this reason, in the search for protective materials, one must turn mainly to the search and identification of the protective material of the organism itself, since irradiation of a low intensity undoubtedly is a biological factor which existed even in ancient times, probably in much larger quantities than now. It is possible that some organisms have a regulating apparatus which permits them to block the effect of irradiation doses stronger than the normal level of adaptation of the organism. In this regulating apparatus, an important part is played by anti-radiation material generated by the organism, whose identification can play a large part in protecting against irradiation. All of this leads us to the important question which we touched upon at the beginning of this report; namely, to the question of regenerative action.

On the basis of the radiological experiments which we have been carrying out, it was established that living tissue is capable of overcoming the effects of irradiation, especially if the whole system participates in this process. Unfortunately, no data from the literature bearing on this question can be quoted for lack of time. I think it is sufficient to mention that the time factor, which is so significant for the therapeutic uses of irradiation, is based on the regenerative actions which are known to us. Some geneticists, basing themselves on experiments carried out, most often with *Drosophila,* less frequently with mice, express the opinion that, under the influence of irradiation, there is irreparable disruption of the chromosomes, mainly in the form of a mutation of genes which may show up only in future generations. One must remember that there are no experiments on people to which one could possibly refer; further, that these conclusions, drawn in most cases on the strength of experiments with the lowest forms of life, have been bodily applied to people; and finally, and perhaps most importantly, that all of the conclusions arise from the mechanistic gene concept, which assumes the existence of a special germ matter, isolated from the rest of the body, which has changeless ability to pass on from generation to generation, and which is not subject to any influence, either on the part of the system, or of the environment. Even the basis of this concept is incorrect, since it denies the plasticity and adaptability of live beings. I think that this concept is harmful in its

very social results; which, for instance, becomes clear from the conclusions of several geneticists who state that mutation and chromosome breakage due to irradiation may be attended by a lowering of the ability to conceive. Since, in the opinion of these geneticists, undesirable mutations may arise from irradiation, and social institutions permit those mutants to remain alive, they consider it more proper to lower the fertility of the people. Permit me in this connection to quote the words of geneticist Darlington: "In general, it may be said that general irradiation of humanity (should such be possible) sufficiently strong to decrease the rate at which people are multiplying, may in these times be considered desirable." It seems that a more crass example cannot be found, which would show the mistakenness of the mechanistic hypothesis in relation to society. Also, in the field of inheritable changes caused by irradiation, we must carry out research of such nature as to be applicable to people with a high degree of probability.

From all of the above, it becomes clear that the permissible doses must again be subject to a review. I assume that all of us are agreed that the most harmful radiation, for the human body, is that consisting of sudden doses of penetrating radiation resulting from accidents, or generated by tests with nuclear weapons. Biologists, united in the International Union of Biologists, at a conference which took place in Rome in April of this year, unanimously passed a resolution calling for the discontinuance of all types of atomic and hydrogen bombs, since, due to these tests, the level of radioactive radiation is increased, which constitutes a danger that may cause serious damage to mankind. We feel that our meeting should add the warning voice of the biologists, by means of a resolution to be passed here. We are gathered to seek new avenues for the peaceful use of atomic energy, and therefore we cannot be insensitive to the harmful use of atomic energy for military purposes; on the contrary, we must strongly come out against such harmful use, and pursue our protest to the very limits.

# Record of Proceedings of Session 6.2

## WEDNESDAY AFTERNOON, 10 AUGUST 1955

*Chairman:* Mr. W. B. Lewis (Canada)

*Scientific Secretaries:* Messrs. E. O. Hughes, I. D. Rojanski and F. de Hoffmann

### PROGRAMME

P/853 The safety of nuclear reactors ..... C. R. McCullough *et al.*

P/481 Experimental determinations of the self-regulation and safety of operating water-moderated reactors ..... J. R. Dietrich

P/394 Radiological hazards from an escape of fission products and the implications in power reactor location.. W. G. Marley and T. M. Fry

DISCUSSION

The CHAIRMAN: I strongly suspect that the reason why the Secretariat and the President suggested that I might be Chairman of this session is that we had one occasion at Chalk River on which we decided that we should evacuate the site temporarily. This was a matter of reactor safety. We did not suffer any injury from that incident. We believe that the question of reactor safety and the siting of power reactors is one which, although deserving serious attention, will be taken in our stride in the further development towards nuclear power generation.

Mr. C. R. MCCULLOUGH (USA) presented paper P/853.

Mr. J. R. DIETRICH (USA) presented paper P/481.

Mr. W. G. MARLEY (UK) presented paper P/394.

### DISCUSSION OF P/853, P/481, and P/394

The CHAIRMAN: The first question, from Sir John Cockroft (UK), is addressed to Mr. McCullough and reads: "What kind of fuses are being developed for reactor safeguard?"

Mr. MCCULLOUGH (USA): A number of types are being studied. These mainly depend upon a neutron poison which is compressed and which can be released when the power level of the reactor gets above a certain point. The release mechanism is dependent upon heating by the neutron flux. When this release occurs, the gas expands. This poisons the reactor and reduces reactivity.

The CHAIRMAN: The next question is addressed to Mr. Marley by Mr. K. Z. Morgan (USA) and it reads: "Have you considered the advantages of short fuel cycling such as might be practical with a homogeneous reactor?"

Mr. MARLEY (UK): We have given qualitative consideration to short fuel cycles. The advantage of short fuel cycles in respect to the hazards of deposition is only realized if you can shorten the fuel cycle to an extremely short period. Moderate reduction of the irradiation period reduces the hazards only very slightly in respect of most of the constituents apart from strontium-90 which, of course, is reduced in proportion to the irradiation time. If, however, one reduces drastically the irradiation time the deposition problem becomes negligible and the irradiation from the cloud is perhaps the limiting factor.

The CHAIRMAN: The next question is from Mr. M. J. Lavigne of Canada, addressed to Mr. Dietrich: "In the reactor excursion in which fuel plates were melted, was there any evidence of chemical reaction of aluminium and water?"

Mr. DIETRICH (USA): There was no conclusive evidence. I can give the following observations which have some bearing on this question. The destructive effects seemed to be entirely consistent with the amount of nuclear energy liberated, and it does not seem necessary to postulate any chemical release of energy in order to explain the effects. Secondly, I might say that certainly most of the fuel was left in the immediate vicinity of the reactor. This was established by radioactive surveys after the accident. No large fraction of the aluminum reacted and blew away. At least, if it did it left its fuel content behind. There did seem to be aluminum oxide on the particles that were found on the ground. Whether this was formed by reaction with the water or by reaction with the air I cannot say. You perhaps noted in the movie some bright spots in the dark cloud of smoke. We can only attribute these to fragments of aluminum burning in the air; and this, of course, would account for much oxide.

The CHAIRMAN: The next question is from Mr. P. W. Mummery, United Kingdom, and is addressed to Mr. Dietrich: "Most reactor designs suitable for large-scale power plants will differ from those used in the experiments described by Mr. Dietrich.

For example, different geometry of fuel and coolant; dependence on resonance absorption in uranium or thorium instead of leakage as the mechanism for reducing reactivity, or use of heavy water. Would Mr. Dietrich comment on the application of his results to these designs?"

Mr. DIETRICH (USA): First I will say, with respect to the geometry of the core and the disposal of the fuel elements, that we did make one other set of experiments, whose results I did not show, with more widely spaced fuel elements, and in this case the energy release per fuel plate just about doubled and we had just about doubled the spacing of the fuel plates: this might lead one to conclude that you must supply a given amount of energy to each unit volume of water. On the other hand the second reactor also had a much smaller negative steam coefficient of reactivity and I would say the energy increase was less than one would expect as the combination of these two effects on the simple picture that the energy generation is always proportional to the energy content of an amount of steam necessary to counteract the reactivity.

With regard to $D_2O$ as opposed to $H_2O$: my impression is, from the results we have, that the destructiveness of the excursion depends much more upon the rate of increase of power than upon the intrinsic value of the excess reactivity; so that we expect that in $D_2O$ reactors with their longer neutron lifetimes the self-protection will be more effective provided the reactor can be designed with the same coefficient of reactivity. With regard to the negative coefficient being determined by the resonance absorption rather than leakage: we can see no neutron effect which makes this situation any different from the leakage one except that you must be sure that it is there (and this is reactor physics), but, of course, this does mean the fuel elements will be made heavier and they will be made differently, and we do not believe that the results can be simply applied to any kind of a fuel element without taking into account its heat capacity and its thermal conductivity, and so on. I wish we had enough results to give a complete story on this question now, but we do not yet have them.

The CHAIRMAN: The next question is from Mr. J. J. Went of The Netherlands. It reads as follows: "With respect to the great importance for the countries now considering building reactors, I should like to put the following question to Mr. McCullough: What has been the policy of the United States Atomic Energy Commission regarding the degree of isolation required for nuclear reactors?"

Mr. MCCULLOUGH (USA): In reply to this question I would say that in our country where it was possible and feasible to locate reactors far from inhabited areas, we did so. However, if you are considering research reactors, power reactors, I think it is obvious that you cannot put them out in deserts. Therefore, we are working on a policy now which attempts to balance perhaps four factors: first, careful design of the machine in all of its parts; second, adequate administrative control; third, containment; and fourth, location. I think you will agree on reflection that location and containment can be balanced one with the other. Mr. Marley's remarks are extremely pertinent to this point; namely, if you can reduce the amount of reactivity which can escape to a low value, then comparatively small distances are quite adequate.

The CHAIRMAN: I have another question from Mr. Went to Mr. McCullough. It reads as follows: "You mentioned that a gas-tight building is desirable, but that it does not need to be explosion-proof. Is that also true for the destruction of a pressurized water reactor?"

Mr. MCCULLOUGH (USA): I think we are getting into a definition of explosion here. We feel that it is possible to contain the release of the pressure under reasonable conditions of a pressurized water reactor. This takes careful figuring and designing of the final vapor container.

# Session 17C

## SAFETY STANDARDS AND HEALTH ASPECTS OF LARGE-SCALE USE OF ATOMIC ENERGY

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# Radiation Injury and Protection—Maximum Permissible Exposure Standards

By W. Binks,* UK

## CONCEPTION OF "MAXIMUM PERMISSIBLE WEEKLY DOSE"

It is normal for human beings to live in an environment of ionizing radiations at very low levels of intensity, for they have always been subjected to "natural" radiation from cosmic and terrestrial sources. Since all types of ionizing radiations produce changes in living cells and some changes, at least, are regarded as being deleterious, it is believed that any significant radiation exposure in excess of the natural level may produce deleterious effects even though, for small additional doses, such effects may not be detectable. Having examined the evidence on radiation effects on human beings and on animals, the International Commission on Radiological Protection[1] (ICRP) suggests that a worker may be occupationally exposed throughout his working lifetime to a practical average level, for each type of radiation, without incurring appreciably bodily injury.† Indeed the risk involved may be less than that from many common and, at times, voluntarily accepted, conditions of life. This level is called the "maximum permissible weekly dose."

## RELATIVE BIOLOGICAL EFFECTIVENESS OF RADIATIONS

Whilst all ionizing radiations produce the same qualitative effects in their interaction with human tissue, quantitative differences may occur since another factor, besides the total ionization produced per gram of irradiated substance, is operative. A heavy particle, such as an $\alpha$-particle, has a very much shorter range in a medium than an electron or $\beta$-particle of like energy. Accordingly the "specific ionization" or number of ions produced per cm length of track in a medium, is very much greater in the case of the $\alpha$-particle than of the lighter particles. It has been observed that, dose for dose, radiation of high specific ionization produces a greater biological effect than radiation of low specific ionization. As most of the evidence accumulated on the biological effects of radiation concerns X-rays generated at voltages of the order of 250 kv, this type of radiation is used as the base-line and is said to have a biological effectiveness of 1.0. The ratio of the biological effectiveness of radiation which produces an appreciably different specific ionization to that of the radiation used as the point of reference is called the "relative biological effectiveness" (RBE). It is usual to relate the RBE factors to specific biological effects. Reference should be made to the report of the ICRP for the recommended RBE factors.

## VALUES OF MAXIMUM PERMISSIBLE WEEKLY DOSES OF EXTERNAL AND INTERNAL RADIATION

According to the Recommendations of the International Commission on Radiological Units (1954), "absorbed dose"—or amount of energy imparted to matter by ionizing particles per unit mass of irradiated material at the place of interest—shall be expressed in terms of the "rad," which has the value of 100 ergs per gram. For X-rays and $\gamma$-rays of quantum energy up to 3 Mev, the more familiar unit, the röntgen (r), can be used. Accepting these proposals, the ICRP recommends that, for whole-body exposure to external radiation measurable in röntgens, the maximum permissible weekly dose shall be 0.3 r for the blood-forming organs, eyes and gonads and 0.6 for the skin. For external radiation measurable in rad in soft tissue, the röntgen is replaced by the rad, in the above. To assist conversion to radiations having RBE factors different from those of X-rays and $\gamma$-rays, the ICRP suggests a convenient unit called the "rem", the dose in rem being equal to the dose in rad multiplied by the appropriate RBE factor. With the exception of radiation of low penetrating power, having a half-value layer of less than 1 mm of water, a generalized statement can thus be made that the basic permissible weekly dose for whole-body exposure to external ionizing radiation of any type is 0.3 rem in the blood-forming organs, eyes and gonads, and 0.6 rem in the skin. For partial irradiation of the body, for example, of the hands and forearms, feet and ankles, head and neck, the value is 1.5 rem in the skin and less in deeper tissues with a limitation that, in the case of irradiation of the head, the weekly dose to the lens of the eye shall not exceed 0.3 rem.

The ICRP has also recommended values for the maximum permissible body burden and for the maximum permissible concentration in air and water for a large number of radioisotopes which are now in use and which may, therefore, be inhaled or ingested. The

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† "Appreciable bodily injury" is defined as any bodily injury or effect that a person would regard as being objectionable and/or competent medical authorities would regard as being deleterious to the health and well-being of the individual.

starting point for the assessment of these values is radium, for which there is valuable pathological evidence on the permissible body burden, from a case now famous in medical annals. The figure at present accepted for radium is 0.1 $\mu$c. On this basis, the corresponding body burden for any other bone-seeking isotope can be derived if evidence is available from animal experiments on the biological effect of the isotope relative to that of radium, dose for dose. For bone-seeking isotopes whose biological effectiveness relative to radium is unknown, and for the remaining isotopes which have no affinity for bone and about which relatively little is known of their effects on human beings, the permissible body burden is regarded as that amount of the isotope in question which, when distributed in the body, produces a weekly dose of 0.3 rem in the so-called "critical organ" in which maximum concentration of the isotope occurs as a result of selective action. The critical organ for each isotope is judged on the basis of limited animal experiments with radioisotopes and of metabolic studies on human beings of the distribution within the body of a stable element which simulates the isotope in question. In the case of the bone-seeking isotopes, a safety factor of 5 is applied to the calculated body burden to allow for possible uneven distribution of the isotope within the bone.

The fractions of inhaled or ingested isotopes which are retained in the body and the fractions which are deposited in certain body organs depend not only on the isotope itself but on such other factors as the type of compound (for example, valency and degree of solubility) and the nature of the compound (for example, size of dust particle). From information on these fractions and on the rate of loss of activity of the isotope in the body, either because of excretion or of radioactive decay, a calculation can be made of the daily rates of intakes into the body of the isotope in air and in water which would result, sooner or later, in the maximum permissible body burden being reached. Then from a knowledge of the total daily intake of air or water into the body, the maximum permissible concentration of the isotope in air or in water can be fixed.

In this way, the ICRP has evaluated the maximum permissible body burdens and maximum permissible concentrations in air and in water of some 86 isotopes, a few of which are treated both for soluble and insoluble compounds. Already requests have reached the Commission to evaluate the levels for a further 50 or so isotopes.

The mode of derivation and presentation by the Commission of the levels for radioactive isotopes is apt to be misleading in that there is an apparent exactitude in the data and formulae which does not really exist. This weakness has to be admitted. The assumptions made so far are somewhat crude in nature and should serve to indicate the limitation of the present ICRP values and to point the need for more extensive researches to improve our knowledge. Meantime, as the various isotopes listed are already in use and the number of persons exposed is increasing the need for authoritative guidance is obvious. The Commission believes its report meets this need.

## MEASUREMENT OF BODY BURDENS

In assessing the maximum permissible concentrations of isotopes in air and in water, the ICRP has assumed that the biological elimination from the critical organ follows a simple exponential law, with a biological half-life of, say, $T_b$. If this is combined with the radioactive half-life, $T_r$, the effective half-life, $T$, for the elimination of the isotope from the critical organ is given by $T = T_b T_r/(T_b + T_r)$. The daily elimination from the critical organ is then $0.693\ qf_2/T$, where $q$ is the body burden, $f_2$ is the fraction in the critical organ of that in the total body, and $T$ is expressed in days. The measurement of the body burden $q$ by means of the activity of the excreta is thus dependent on $T$, that is, on the assumption of an exponential law. The elimination process seems, however, to be much more complex than this, and the exponential relationship holds only over limited periods of time. In the case of a long-term study on the elimination of radium from the bodies of human beings, Norris and co-workers[2] at the Argonne Laboratory state that the concept of a well-defined biological half-life of radium in bone is inconsistent with the data presented. They find that the coefficient of elimination (fraction of the total body burden excreted per day) can be represented by a power function of the form $-0.52/t$, where $t$ is the time in days after injection of the radium. Had the elimination followed a simple exponential function, the fraction of the body burden excreted per day would have been a constant.

When the isotope involved emits $\gamma$-rays or, if a $\beta$-emitter, is present in sufficient quantities that it gives rise to appreciable bremsstrahlung, direct determinations of the body burden can be made by measuring these radiations external to the body. As, however, elaborate and expensive equipment is required for this purpose, it is the more usual practice to estimate body burden indirectly from measurements of the radioactivity of the excreta. In view of the complexity of the elimination function it will be clear that more extensive investigations must be made of the relationship between body burden and activity of excreta before the former can be reliably assessed indirectly via the latter. The information gained from studies of this relationship for the various isotopes may well react upon the calculated values for the maximum permissible concentrations in air and in water, for it should be realised that effective half-life, $T$, enters into the formulae concerned.

The difficulties of interpreting "excretion" data are well illustrated in the case of exhaled radon in breath. There has been long experience in radon measurements over the past 20 years or so, and the difficulty in estimating the radium content from the exhaled radon would not appear to lie in the actual

estimation of the radon present in the sample, but in the wide variation in breathing rate from one person to another (a range of 4.0 to 13.2 litres per minute was found by the Argonne Laboratory workers) and in the variation, with time, of the exhaled fraction of the radon produced in the body. For long-standing cases of radium deposited in the body, the earlier value of Evans[3] of 45 per cent radon exhaled was based on an assumed average breathing rate of 5.0 litres per minute. If this is corrected to the average of 7.5 litres per minute found by Norris and co-workers it becomes 67.5 per cent, which is in close agreement with Norris's latest value of 70 per cent. For shorter-term depositions of radium, it has been noted[4] in tests on five persons involved in an accident with radium sulphate dust that the percentage of radon exhaled fell in the early stages from an average of about 20 per cent at 2 days after the accident to an average of about 10 per cent at 10 days, thereafter rising gradually to an average of about 30 per cent at 100 days and of about 45 per cent at 1 year. If continual inhalation of radium dust occurs, as in radium-luminizing departments, it is difficult to know what figure to take for the exhaled radon fraction.

## MAXIMUM PERMISSIBLE LEVELS FOR NON-OCCUPATIONAL GROUPS

Although initially the concept of maximum permissible dose applies to those occupationally exposed to ionizing radiations, consideration must also be given to patients undergoing treatment (and, perhaps, diagnostic examinations) and to the population as a whole for whom the integrated total of radiation from all causes may have genetic consequences.

Radiation treatment of patients should be planned to minimise the risk of producing late effects such as carcinogenesis and leukaemogenesis. In this connection, for internally administered isotopes, knowledge is required of the doses to different organs from given depositions acting over given periods, bearing in mind that the "critical organ" may differ according to the integrated time of effective action of the radiation.

Suggestions have been made from time to time that a figure should be agreed upon for the total dose which a radiological worker should receive during his lifetime. Generally one thinks in terms of a few hundred röntgens in this respect. If it should come about that a maximum permissible lifetime dose is fixed for workers, then it would seem logical to apply this figure to the total doses from radiation treatment of non-malignant cases and from radiodiagnostic procedures. But even if no lifetime dose can be assessed, it is reasonable to reduce the radiation exposure of patients as much as is compatible with successful treatment or diagnostic investigation.

Lifetime doses of the order of a few hundred röntgens seem to be unacceptable, on genetic grounds, if more than a small fraction of the population is so irradiated. The amount which is required to double the spontaneous mutation rate in Man is not known. If a value of the order of 50 r which has been found for a few other species is applicable to human beings, there is a strong case for keeping the dose *per capita* per generation well below this figure. But, in view of the extreme importance of a figure for the doubling rate for human beings, more intensified researches on genetic effects are called for. In the meantime, it may be of interest to note that in the United Kingdom, estimates have been made of the total dose received by all radiological workers and of the total dose received by the gonads of all males and females during diagnostic examinations which take place before the age of about 30 years. Averaging these totals over the entire population, it is found that the dose *per capita* per generation of 30 years is less than 0.1 r, or less than $\frac{1}{30}$th of the natural radiation level from cosmic and terrestrial sources. The additional genetic load is thus insignificant at the moment.

Suggestions have been made in one or two quarters that a certificate might be introduced in which details of all radiation exposure (medical and occupational), which an individual receives during his lifetime, are recorded. A scheme of this nature would ultimately permit, on the one hand, a correlation between the doses received by individuals and any early or late radiation effects such as induced carcinoma, leukaemia, or shortening of the life span and, on the other hand, an appraisal of the total dose received by the community and the possible genetic consequences.

With reference to prolonged exposure of "large populations", the ICRP recommends that the maximum permissible levels should be reduced by a factor of 10 below those accepted for occupational workers. A distinction should be drawn between "large populations" and "whole populations". If the whole population were involved and a figure of 0.03 r per week (that is, $\frac{1}{10}$th of the level for occupational workers) were allowed, the total dose per capita per generation of 30 years would be about 47 röntgens. This is of the same order as the present figure assumed for doubling the spontaneous rate. Obviously one ought not to apply the ICRP rule to whole populations. "Large populations" should not, indeed, on the basis of the above calculations, exceed 10 per cent of the whole population; alternatively, a greater reduction factor than 10 should be applied if a higher fraction of the population is at risk.

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2. Norris, W. P. *et al., Studies of the Metabolism of Radium in Man*, Amer. J. Roent., Rad. Therapy and Nuclear Med., 73 (No. 5), 785-802 (1955).
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4. Marinelli, L. D. *et al., Transport of Radium Sulfate from the Lung and its Elimination from the Human Body following Single Accidental Exposures.* Radiology, 61 (No. 6), 903-914 (1953).

# Maximum Permissible Exposure Standards

**By Robert S. Stone,* USA**

## HISTORY OF IDEA OF TOLERANCE

Professor Roentgen discovered X-rays and investigated their characteristics in the latter part of 1895. In December of that year when he announced his discovery he told of his use of heavy metals to confine the newly discovered rays. By the end of January, 1896, Grubbé,[1] who was building and experimenting with the new "X-ray" tubes, had developed a new and peculiar dermatitis of his hands which was determined to be due to the action of the roentgen rays. In a relatively short time it was found that X-rays produced many changes in people who worked with them and in animals used for experimental purposes. The injuries recorded include the following: erythematous dermatitis; smarting of the eyes; epilation of hair; constitutional symptoms such as tiredness, weakness, nausea, and vomiting; cancer in chronic X-ray ulcer; inhibition of bone growth; sterilization of male rabbits and guinea pigs; reduction of white blood cells; alteration of bone marrow, etc. By 1906 it had been found that most of the organs of the body could be altered by the X-rays.

While the need for protection against the ionizing radiation was obvious from the very beginning because of the injuries, the idea of trying to define an amount of exposure that could be tolerated by the human body was slow in developing. When the British X-Ray and Radium Protection Committee was formed in 1921, it discussed "establishing a maximum tolerance dose in terms of a specifiable and reproducible biological standard, and if possible expressing this biological standard in physical units."[2] In 1925, the problem was stated more explicitly by Mutscheller:[3] "In order to be able to calculate the thickness of the protective shield there must be known the dose which an operator can for a prolonged period of time, tolerate without ultimately suffering injury." In an attempt to find such a "dose," he and others selected some people who had been occupationally exposed for some years without showing evidence of any deleterious effect. The amount of radiation to which these people had been exposed was then determined in retrospect. When the units used are translated into roentgens, the doses the different workers advocated as tolerable nearly all fall in the range of 0.1 to 0.2 r per day.

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**Table I. Historical Landmarks: Radiation Injuries**

| | | |
|---|---|---|
| 1895 | Discovery of X-rays | Roentgen |
| 1896 | Dermatitis of hands (January) | Grubbé |
| | Smarting of eyes (March) | Edison |
| | Epilation (April) | Daniel |
| 1897 | Constitutional symptoms | Walsh |
| 1898 | Paraplegia and spasmodic muscular contraction | Rodet and Bertin |
| 1899 | Degeneration of vascular endothelium | Gassman |
| 1902 | Cancer in X-ray ulcer | Frieben |
| 1903 | Bone growth inhibited | Perthes |
| | Sterilization produced | Albers-Schönberg |
| 1904 | Blood changes produced | Milchner and Mosse |
| | Leukopenia produced | Heineke |
| 1906 | Bone marrow changes demonstrated | Warthin |
| 1912 | Anemia found in two X-ray workers | Béclère |

**Table II. Historical Landmarks: Tolerance**

| | |
|---|---|
| 1902 | Rollins photographic indication of "safe" intensity |
| 1921 | British X-ray and Radium Protection Committee considered "establishing a maximum tolerance dose" |
| 1925 | First attempt at defining a "tolerance dose" by Mutscheller |
| 1931 | US Advisory Committee on X-ray and Radium Protection recommends tolerance dose of 0.2 r per day |
| 1934 | International X-ray and Radium Committee endorses tolerance dose of 0.2 r per day |
| 1936 | US Advisory Committee reduces tolerance dose to 0.1 r per day |
| 1942 | Metallurgical (Plutonium) Project adopts tolerance dose of 0.1 r per day |
| 1950 | International Commission on Radiological Protection recommends 0.3 r (air) per week = 0.05 r per day |

In 1931 the Advisory Committee on X-Ray and Radium Protection in the United States, and in 1934 the International Committee on X-Ray and Radium Protection both endorsed a tolerance dose of 0.2 r per day. In 1936 the United States Committee reduced the tolerance dose to 0.1 r per day, apparently for two reasons. First the practice in Europe was to measure the dose on the skin with backscatter, whereas the practice in the United States was to measure the dose in air without backscatter. For the quality of radiations then available and the size of the fields involved in whole-body exposure, the agreement between 0.1 r in air and 0.2 r on the skin was very good. Second, by 1936 there were many more 200-kv X-ray machines in operation producing more penetrating radiations than before, and therefore a greater percentage of the surface dose was reaching the deeper organs. The main basis on

Table III. Estimation of Tolerance Doses

| Date | Author | Days for dose* | Calculated r per day† |
|---|---|---|---|
| 1902 | Rollins | (Photographic) | 10 |
| 1925 | Mutscheller | 3 | 0.2 |
| 1925 | Sievert | 3 | 0.2 |
| 1926 | Solomon | 0.3 | 2.0 |
| 1927 | Dutch Board | 15.0 | 0.04 |
| 1928 | Barclay and Cox | 3.5 | 0.17 |
| 1928 | Kaye | 5 | 0.12 |
| 1931 | Advisory Committee, USA | .. | 0.2 |
| 1932 | Failla (γ-rays) | 30.0 | 0.1 |
| 1932 | Stenstrom | 3.7 | 0.16 |
| 1936 | Advisory Committee, USA | .. | 0.1 |
| 1950 | International Commission | .. | 0.05‡ |

* Number of days over which 1/1000 of an erythema dose may be received.

† Calculated on basis of erythema dose = 600 r, except Failla γ-ray erythema = 1800 γr.

‡ ICRP recommended 0.3 r (air) per week.

which these tolerance doses were established was still the observation of those occupationally exposed.

These "tolerance doses" were largely of interest to medical radiologists until 1942 when the first nuclear chain-reacting pile started to operate. Since then, the matter of radiation protection and safe amounts of exposure have become of interest to the citizen in the street as well as to the many thousands of workers occupationally exposed. The need for more accurate information on which to base the tolerance dose became known to many scientists and money for experiments was made available. New tests of various kinds were tried in attempts to find the earliest possible changes as a result of exposure to ionizing radiations with the hope of avoiding more serious effects. Several changes were found, such as bilobed lymphocytes, changes in intracellular structure, more immature leucocytes, etc. The new problem that is not yet solved is: Are these minor changes of real significance to the individual?

## PERMISSIBLE DOSE

The problem of how to define a permissible dose became more difficult than that of specifying what dose had been tolerated by a few people. After much deliberation the International Commission on Radiation Protection[4] defined permissible dose as "a dose of ionizing radiation that in the light of present knowledge is not expected to cause appreciable bodily injury to a person at any time during his lifetime." "Appreciable bodily injury" is defined as "any bodily injury or effect that a person would regard as being objectionable and/or competent medical authorities would regard as being deleterious to the health and well-being of the individual." In attempting to give a numerical value to the permissible dose the fact that man is and always has been bombarded by the ionizing cosmic rays and the radiations from the natural radioactive elements must be taken into consideration. These radiations provide a lower limit that is inescapable (about 10 mr per week).

The upper limit of permissible exposure must be based on the experience of radiation workers (mostly medical) over the past 60 years, and on such animal experiments as seem to be pertinent. There are several factors that make the interpretation of the human experience difficult. First, we have no real measure of the level of exposures prior to about 1942. Second, the long latent period between cause and effect—sometimes 25 years—makes it difficult to connect the two. Third, the biological variability of man is so great that some people known to have been overexposed have had no complications while others not known to have been overexposed have died of such conditions as leukemia that probably were due to such exposures as they suffered.

## CRITICAL ORGANS

Reviewing the experiences of man and the results of animal experimentation revealed clearly that some organs of the body are critical to the problem of permissible exposure. The skin is the most exposed organ of the body. It was the first known to be injured and is the one most frequently involved. In the damaged skin, cancer sometimes develops and causes the death of the person. By 1911 Hesse[5] was able to collect 94 cases of X-ray induced human tumors. The great length of time between cause and effect makes it difficult to set a permissible dose for the skin. Some doctors have developed detectable skin changes as long as 25 years after stopping the practice of exposing their hands while doing fluoroscopic work. By estimating the probable doses received by the skin of the hands of radiologists who showed no skin changes, the International Commission set 1500 mr per week as the maximum permissible dose when only the hands and forearms are exposed, as in handling radioisotopes. When the whole body is exposed, the limit for the skin is 300 mr.

It was recognized early in the study of radiation effects that any peripheral blood change was one of the first objective signs of the effect of X-rays. The hemopoietic tissue was thus established as a critical organ, the changes in which can be followed by studies of the peripheral blood. Before there were physical instruments capable of making reproducible measurements, blood counts were used as a biological monitor of exposure. In the period before personal and area monitoring was easily possible, many if not most radiologists, radium workers, and technicians were overexposed, as was demonstrated by the first few weeks of accurate monitoring in the radiology departments of many hospitals. Hence the reports of Carman and Miller in 1924[6] and Goodfellow in 1935[7] dealt with marked changes in the blood counts due to real overexposures. With the establishing of tolerance doses and the development of means to measure exposures, hematologists have had to search for new criteria of damage. Now, Ingram[8] and others have found bilobed lymphocytes in the blood of cyclotron workers presumably ex-

posed to very little radiation. Helde and Wahlberg[9] have associated "changes" with very low exposures in very short times. Mayneord[10] has found "trends" in serial counts which he considers significant. The counts were done on workers exposed to below maximum permissible levels. On the other hand, Jacobson and co-workers[11] have found no correlation of counts with exposure at low levels, and Chamberlain, Turner, and Williams,[12] while finding some evidence of detectable radiation effects on the average count of large groups of workers even at low levels of exposure, conclude the white cell counting does not give much of a measure of protection to the individual. What really is of concern is whether or not these changes are significant and can be considered as "appreciable bodily injury." The real danger that threatens is the development of leukemia, a universally fatal disease. March[13] found that 14 of 299 radiologists, 4.68 per cent, dying between 1928 and 1948 died of leukemia while only 334 of 65,922 non-radiologist physicians, 0.51 per cent, died of the disease. No one has detected the premonitory signs of leukemia. It is well established that many people who have had severe blood changes from radiation exposure have not developed leukemia. Experimental studies using animals have not helped us to date except to prove that in susceptible animals leukemia occurs earlier and more frequently following whole-body irradiation.

This author believes that while variations from the normal must be studied and all avenues of studying blood changes investigated, slight deviation from an admittedly unstable "normal" should not be classed as "appreciable bodily injury."

The National Committee on Radiation Protection of the USA made a careful study of the value of blood counts and reached the following conclusions: (1) Provided that radiation monitoring of personnel, and where applicable of sites, is carried out by instruments (film badges, pocket meters, etc.) in all circumstances involving potential exposure to penetrating ionizing radiations, blood counts should no longer be required as a *method of monitoring.* (2) Blood counts as a part of pre-employment, interval, and terminal examinations are good medical practice —to be done at the discretion of the medical officer in charge—but not as a part of a monitoring service. (3) Blood counts are a necessary part of the medical examination of anyone overexposed to penetrating ionizing radiations.

The International Commission on Radiological Protection has recommended that the permissible weekly dose in the blood forming organs be 300 mr. The lowering of this dose from essentially 600 mr per week was an attempt to be on the safe side and was not based on proof that 600 mr was too high a dose. The 300 mr is to be measured in the cubic centimeter that receives the highest dose. Many experiments suggest that the whole body must be exposed before leukemia becomes a problem.

Another tissue that has entered the list of critical tissues is the lens of the eye. It was known that treatments with large doses of X-rays could cause cataracts. Cataracts had not been seen as a result of long-continued small doses in radiologists. The fact that neutrons seem to have a highly selective action on the lens as evidenced by the changes in the eyes of some physicists working with them and of some of the Japanese exposed at Hiroshima and Nagasaki has placed the lens tissue in the critical list. However, it should be noted that exposure of adult animals to levels of X-ray dosage much above permissible levels did not produce cataracts. The International Commission decided to set 300 mr per week as the permissible dose, mainly to have a basis for dose from neutrons.

The effects of ionizing radiation on the gonads have been under investigation since at least 1903. However, except for human experience, little was known about the effects of long-term low-level exposure until Lorenz and Heston started genetic experiments in 1941.[14] Exposure of the gonads to ionizing radiations can produce sterility, abnormal children in the first generation, and long-term genetic effects.

Recent surveys[15, 16] of radiologists and pathologists (as controls) show that the exposed group reported more offspring than the unexposed group. While many factors are involved, at least a loss of fertility of those occupationally exposed was not demonstrated. However, one of the surveys[15] revealed that while the normal offspring of the unexposed group constituted 83.23 per cent, the normal offspring of the exposed group constituted only 80.42 per cent. This difference is not large and should not be viewed with alarm. The other survey[16] showed that the amount of radiation received by radiologists under usual conditions does not have any very large effect on fetal and infant death rates in their children. The animal experiments of Lorenz showed that mice could live, breed, and die while exposed constantly to 1.1 r per day.[14] Experiments reported from the University of Rochester[17] indicate that the sperm count of dogs was reduced by daily exposures of 0.5 r. Insofar as long-term genetic effects are concerned, those who are competent to discuss this subject are doing so at this Conference. In determining the permissible *occupational* exposure of a limited number of people, the genetic factor is not considered to be the determining one.

All things considered, insofar as the gonads are concerned, the recommendation of the International Committee "that exposure to radiation be kept at the lowest practicable level in all cases" should be followed. Based solely on considerations involving avoidance of damage to the exposed individual himself, the basic permissible weekly dose in the gonads was set at 300 mr.

## LIFESPAN

While the idea of critical organs is clearly a direct method of attack on the problem of permissible

dose, it must be remembered that a shortening of the lifespan is possible without the development of any of the specific conditions mentioned above. This effect became apparent when large numbers of animals of selected strains were studied under conditions of prolonged daily low-level exposure to radiations. The average life of the group of animals was shorter when numbers of them developed anemia, leukemia, or cancer, but there appeared to be another factor of an indefinite character which many have chosen to call premature aging. In the case of man, the biological variation is so great that it is difficult if not impossible to establish any small difference in length of life that might exist between those who have been occupationally exposed to radiation and the rest of the population. In the case of animals it has been shown with certainty insofar as mice are concerned that exposure to daily doses of X-rays of slightly greater than 1 r cause a reduction in lifespan.[14] Boche[17] has shown that the lifespan of the rat is definitely shortened by daily exposures of 0.5 r and probably 0.1 r, the exposures starting at the time of maturity. On the basis of such figures it was felt that for whole-body exposure the permissible dose should not exceed 0.05 r per day. Even this provides a factor of safety of only 2.

## RECOVERY FROM INJURY AND PROTRACTION OF EXPOSURE

One of the biological facts that makes it difficult to establish permissible doses for long-term exposures is that recovery from radiation injury takes place in certain instances even while the radiation is continuing. Other changes produced by radiation are irreversible and no recovery takes place. There probably are injuries that show all gradations of response between the extremes of complete recovery and of no recovery. Enough facts with regard to these different rates of recovery are not yet known to allow the projection of the various reactions over the lifetime of man. It is known that some of the acute reactions to radiation take just as long in a mouse whose total life is from two to three years as in a man whose total life is about 70 years. Thus, it is not possible to simply multiply the time for certain reactions to occur in laboratory animals by factors relating their total life to man's total life.

Damaging events in the genes producing genetic change have been shown to be totally additive; that is, there is no recovery. The changes causing epilation in man have been shown to have little relation to the protraction of a given total dose over the period of a week, but there is some recovery in a period as long as a month. The reaction known as the erythema reaction of the skin requires about 50 per cent more radiation when given on two successive days than when given all at one time. The amount of radiation to produce a given weight loss in the mouse testis is the same whether given in one day or in five days.[18] The amount of radiation required to kill 50 per cent of a group of mice increases with the intensity of the radiation and with the fractionation of the dose over days, weeks, or months. Henshaw,[19] using mice, found that 51.6 r given at one session once each week was more effective as judged by the $LD_{50}$ at 18 weeks than when given each week in six daily sessions of 8.6 r.

These studies have been cited to emphasize the problem of deciding whether the permissible dose should be stated as a certain dose per second, per minute, per hour, per week, per month, per year, or per lifetime. The maximum permissible dose of radiation to the whole body is stated as being 0.3 r per week. If an individual were exposed to the maximum amount for 50 weeks of each year for the period from 25 to 65 years of age, he would receive a total of 600 r in the forty years. If subjected to 600 r in one day, he would probably die very quickly. If exposed to 15 r in one day each year for 40 years, what would happen is not known because of lack of experience with animals or people exposed in that way. There is experience with daily or almost daily exposures of radiologists, physicists, and technicians. A decision had to be made on somewhat arbitrary grounds as to the length of time over which a permissible dose could be averaged no matter how it was received. The International Commission decided on the week as the unit of time. From the point of view of occupational exposure it is more convenient to monitor exposures each week than each day, and experience to date indicates little difference of biological effect at such low levels between daily and weekly distribution of dose. The Commission defined the permissible weekly dose as "a dose of ionizing radiation accumulated in one week of such magnitude that, in the light of present knowledge, exposure at this weekly rate for an indefinite period of time is not expected to cause appreciable bodily injury to a person at any time in his lifetime. One week as used here is any seven consecutive days." Inferred in this definition is the belief that such a dose could be received in a matter of a few minutes during the week or spread over the whole seven days. No one has sufficient information as yet to know whether such a dose would still be "permissible" if received in a small fraction of a second.

## RELATIVE BIOLOGICAL EFFECTIVENESS

The Relative Biological Effectiveness (RBE) of radiations of various types and energies is a necessary factor in determining the permissible dose of radiation for other than X-rays up to 250 kv. The biological effectiveness of a radiation is the relationship between a given biological reaction and the dose of radiation required to produce it. Since for both man and animals the effects of X-rays up to 250 kv are known with greater precision than for other types, the biological effectiveness of such rays is arbitrarily established as the base line. The relative biological effectiveness of a given radiation is the factor that relates its biological effectiveness to that of 250 kv X-rays.

Table IV. Effects of Small Weekly Doses of Ionizing Radiation

| *Dose (mr/week)* | *Subject* | *Effect produced* | *Reported by:* |
|---|---|---|---|
| 100 | Man | Questionable slight decrease of mean total white cell count of group | Mayneord |
| 100 | Man | Total number of blood abnormalities (group av.) increased | Sievert |
| 500 | Dog | Sperm count slightly reduced | Boche |
| 500 | Rat | Leukemia induced | Barnett |
| 600 | Man | Blood, no significant changes | Pearlman and Sacher |
| 770 | Mice, $LAF_1$ | Mean survival time, no significant decrease | Lorenz |
| 770 | Guinea pig | Lymphocyte and platelet counts reduced | Lorenz |
| 770 | Mouse | Ovarian, mammary, and lung tumors induced | Lorenz |
| 2500 | Dog | Lymphocyte count reduced | Ingram |
| 2500 | Rat | Mean survival time decreased | Boche |
| 4000 | Man | Skin of hand, minimal changes | Parker |
| 5000 | Dog | 50% of group aspermic after 9 months | Boche |
| 7700 | Mouse | Mammary cancer induced | Lorenz |
| 7700 | Mouse, bda | Breeding, no effect | Lorenz and Heston |
| 7700 | Mouse | Ovarian follicles, decreased after 18 months | Lorenz |
| 7700 | Mouse | Decrease of spermatogonia in mitosis | Eschenbrenner |
| 7700 | Mouse | Testicular weight decreased | |
| | Guinea pig | | Lorenz |
| 7700 | Guinea pig | Lung tumors increased in number | Lorenz |
| 15400 | Guinea pig | Platelet, heterophil, and red cell counts reduced | Lorenz |
| 15400 | Mouse | Lymphocyte count reduced | Lorenz |
| 15400 | Mouse | Mean survival time reduced | Lorenz |
| 30800 | Mouse | Leukemia induced | Lorenz |
| 30800 | Mouse | Sterility induced | Lorenz and Heston |

Since the biological effectiveness is known to be related to the density of ionization along the paths of the ionizing particles, this characteristic can be used to determine the RBE. The linear ion density per unit of path is called the specific ionization. Various combinations of photon or particle radiations with differing energies may have similar specific ionizations. Thus, a very high energy proton may have the same specific ionization as a low energy electron. It is thus better to relate the RBE to the specific ionization rather than to any particular type of radiation. The average specific ionization of 250 kv X-rays is 100 ion pairs per micron of water and by definition the RBE is 1.

The problem of relative biological effectiveness is complicated by the fact that, when determined by different biological reactions, even in the same organism the RBE is not always the same. When mammals only are used the differences are not great enough to seriously complicate protection problems.

In practice, if one wishes to determine the permissible dose of a given radiation, he first finds its specific ionization. From the data collected by the International Commission on Radiological Protection[20] and reproduced below he obtains the RBE for that radiation. The basic permissible dose in tissue roentgens for 250 kv X-rays multiplied by this RBE factor gives the permissible dose in rems.

The *rem* is a unit defined as "the absorbed dose of any ionizing radiation which has the same biological effectiveness as 1 rad of X-radiation with an average specific ionization of 100 ion pairs per micron of water, in terms of its air equivalent, in the same region." In practice, a dose in rems is equal to the dose in rads multiplied by the appropriate RBE. Its greatest usefulness is in integrating exposures involving various kinds of radiations.

Table V. RBE Values

| *Average specific ionization (ion pairs/$\mu$ of water)* | *RBE* |
|---|---|
| 100 or less | 1 |
| 100 to 200 | 1 to 2 |
| 200 to 650 | 2 to 5 |
| 650 to 1500 | 5 to 10 |
| 1500 to 5000 | 10 to 20 |

## PERMISSIBLE DOSE FROM INTERNALLY DEPOSITED RADIOACTIVE MATERIAL

A discussion of this subject could occupy an entire volume. Suffice it to say here that it is assumed that the absorption of a given quantity of radiation uniformly throughout the volume of a critical tissue should produce the same biological reaction whether that radiation comes from radioactive materials deposited in the body or from a beam of radiation coming from outside the body. The actual spacial distribution of radioactive materials within any critical tissue is usually such that some parts of the tissue become very much more heavily irradiated than others. This is especially true of materials emitting beta rays with low penetrating power, but it is by no means limited to such materials. Others at this Conference will discuss the permissible amounts of radioactive materials in the body. The extreme difficulty of applying the permissible exposure stand-

ards developed for external radiation to the permissible limits for radiations originating from radioactive materials within the tissues is readily apparent to all who understand the distribution of chemicals in biological tissue.

## OTHER FACTORS AFFECTING THE PERMISSIBLE DOSES

The age of a person occupationally exposed to radiation should have some influence on the amount of radiation that he can tolerate because (1) genetic and sterility considerations apply only during the normal reproductive period, and (2) the latent period for the appearance of some injuries may be longer than the remaining lifetime of the individual. For this and other reasons the National Committee on Radiation Protection (USA)[21] states that those over 45 years of age could with safety be given a permissible weekly dose double that for those under 45.

The location, kind, and amount of tissue exposed also have a bearing on the permissible dose. Most of the specified doses have been based upon an exposure of the entire body. When only limited areas are exposed, the dose to these might conceivably be increased. This can apply to exposure of the skin of the hands and forearms, of the feet and ankles, or of the head and neck, or to other areas when the exposure is to low energy radiations with poor penetrating power. The effect of irradiating small volumes of internal organs is not sufficiently known to justify increasing the maximum permissible weekly dose to such organs.

There are other conditions that might be considered as modifying the rules, such as the frequency of exposure, the urgency of the work to be done, and the number of individuals to be exposed. All of these subjects are considered at length in Bureau of Standards Handbook 59 of the US Department of Commerce, "Permissible Dose from External Sources of Ionizing Radiation."* [21]

## PROTECTION RULES

The International Commission on Radiological Protection has published those rules[21] that have been agreed upon internationally, and the various national committees and commissions have undoubtedly made known rules applying in their own borders.

It is no longer possible to state simply that the maximum permissible dose is 0.1 roentgen. The roentgen is applicable only to the measurement of X-rays with quantum energies below 3 Mev. Modifying factors are necessary for the penetrating power of the radiation in question, the average specific ionization (ion pairs per micron of water), the portion of the body exposed, the weekly dose fluctuation, and, above all, for non-occupational exposure of large segments of the population.

* Obtainable from the Superintendent of Documents, Washington 25, D. C., USA, price 30 cents.

The basic permissible weekly doses for whole-body exposure, from which all other permissible doses are calculated, are established for X-rays with photon energies below 3 Mev and are expressed in roentgens as tissue dose. These doses are 600 mr in the skin, and 300 mr in a significant volume of the blood forming organs, the gonads, or the lenses of the eyes.

A discussion of the biological bases for maximum permissible exposures by Cantril[22] appeared in 1951, and the subject was reviewed by Stone[23] in 1952, taking into account the clinical and biological data acquired by the Manhattan Project.

## SUMMARY

It has been shown that radiation injuries were produced as early as the first month after the announcement of the discovery of X-rays but that the idea of a permissible (tolerance) dose was slow in developing.

The basis for establishing the first permissible dose was the experience of workers in medical radiology.

The fact that the effects of radiation may not appear for many years after exposure has emphasized the need for establishing conservative standards.

The need for specifying the permissible doses in critical organs has developed because of the various effects of different radiations on these organs.

The irreversible and inescapable effects of radiation on the genes requires that future generations be taken into consideration.

Radiations differing in biological effectiveness from "ordinary" X-rays have come into widespread use. This has made it necessary to establish permissible doses for such radiations based on comparisons with the better known X-rays. The transfer of permissible doses in tissue roentgens to rems expressed in absorbed energy is accomplished through the RBE and the specific ionization.

Maximum permissible exposures have been established that take into account the factors of types of exposure and the age of the person.

## CONCLUSION

No better conclusion to the above review can be given than that of the International Commission on Radiological Protection, 1950: "While the values proposed for maximum permissible exposures are such as to involve a risk that is small compared to the other hazards of life, nevertheless in view of the unsatisfactory nature of much of the evidence on which our judgments must be based, coupled with the knowledge that certain radiation effects are irreversible and cumulative, *it is strongly recommended that every effort be made to reduce exposures to all types of ionizing radiations to the lowest possible level.*"

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# Maximum Permissible Concentration of Radioisotopes in Air and Water for Short Period Exposure

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The National[1] and International[2] Committees on radiation protection have agreed upon maximum permissible concentrations (MPC) of radioisotopes in air and water for continuous exposure. For most radioisotopes, a state of equilibrium is reached between the concentration of the radioactive material in the environment and in the body organs as a result of continuous exposure. Although most of the radioisotopes reach this state of equilibrium in a few weeks or months, natural thorium, $Pu^{239}$, $Sm^{151}$, and $Ra^{226}$ are exceptions, reaching 34%, 34%, 36%, and 67% of equilibrium, respectively, during the maximum period of exposure, assumed to be 70 years. Many radioactive materials are relatively insoluble, and the fraction entering the blood stream following ingestion or inhalation is small so that the gastrointestinal (GI) tract becomes the critical body tissue rather than the kidney, bone, liver, or other body organ.[2] Of the 355 MPC values listed in the ICRP Handbook,[2] 71% of those for ingestion, and 41% of those for inhalation refer to the GI tract as the critical organ—meaning that the GI tract receives the maximum permissible dose rate of 0.3 rem/wk at a lower MPC value than that for any other body organ. It was assumed in these calculations that the radioactive material remains in the GI tract only 31 hours, so a condition of equilibrium is reached between the environment and the GI tract in a few days.

The purpose of this report is to extend the method of calculation used in the ICRP Handbook[2] to estimate the MPC values or the maximum permissible intake (MPI) of these same radioactive materials for a single exposure. Since the concentration of material in the GI tract reaches equilibrium with that in air and water in a few days, one might suspect that the GI tract frequently would become the critical body organ for short periods of exposure. Another body organ likely to receive a high exposure and as a consequence become the critical body organ for single exposure is the lung. A preliminary investigation[3] indicated that for short periods of exposure the lung is the critical body organ for about 50% of the radioisotopes when the permissible dose is set as 0.3 rem during the week following the exposure and for 83% of the radioisotopes when the permissible dose is set as $0.3 \times 52$ or 15.7 rem during the ensuing year. In most of the other cases the GI tract is the critical body organ. Therefore, in this study considerable attention is given to the lung and the GI tract as the most probable critical body organs in establishing MPI values for a single exposure.

Since no official MPI values for single exposures have been agreed upon, a number of criteria will be examined and several tables of values based on various assumptions will be given in the hope that the reader will examine these critically and offer suggestions which will be helpful in making the final selection of the MPI values for single exposure.

The MPC values for continuous exposure have been useful in setting the average maximum permissible concentration of the radionuclides in air and water for occupational exposure, and following a recommendation of the National[1] and International[2] Committees on radiation protection, it has been common practice to set the MPC for environmental exposure at $\frac{1}{10}$th of the occupational exposure values. Much of the biological data used in establishing the continuous exposure MPC values were incomplete, and in many cases animal data were of questionable application to human exposure. Also many of the assumptions made in these calculations were at best poor approximations; nevertheless, these MPC values are serving a useful purpose in that they indicate what are considered to be safe values based on the most reliable information presently available. Since thousands of people are working regularly with radioactive materials, these values are invaluable as a day-to-day guide in maintaining the concentrations of radioactive materials in air and water at what are considered to be safe occupational levels, and the radioactive wastes discharged from these operations into the environment are kept at an average concentration less than $\frac{1}{10}$th of these values. Also upper limits are given in the handbooks[1,2] for the maximum permissible body burden of these radioactive materials corresponding to the maximum permissible concentrations (MPC) for continuous occupational and environmental exposure.

Although continuous exposure (MPC) values are necessary for all work with radioactive material, they do not answer many questions relative to the hazards associated with the occasional accident or spill of a radioactive material, and the resultant exposure of short duration—usually referred to as a *single exposure*. Also, in the design of laboratories

* Oak Ridge National Laboratory.

for work with radioactive materials[4]; in the passage of legislation for the control of radiation hazards[5]; in the establishment of regulations for the shipment of radioactive materials[6]; and in many other similar areas of interest, there has been a considerable need for a reliable criterion to serve as a basis for grouping the various radioactive isotopes into categories of *Very Dangerous, Moderately Dangerous,* and *Slightly Hazardous* materials.

There are many factors that determine the relative hazards associated with the accidental spill of a radioactive material such as the physical state (solid, liquid, or gas) the solubility, the volatility, the specific activity, the biological damage that may result from the inhalation or ingestion of the material, etc. This account is limited to a discussion of the relative biological hazards associated with the various radioisotopes following an accidental spill, assuming each to be present in a common physical and chemical form. The calculations are based only on inhalation of the radioactive material or passage into the body as the result of a contaminated wound. Ingestion is not considered since preliminary studies indicated a much greater internal exposure hazard from inhalation and injection than from ingestion. In event of an inhalation accident, it is assumed that part of the material is retained in the respiratory tract where it presents a lung hazard and a portion is swallowed directly or brought up from the bronchus by the cilia and swallowed where it presents a hazard either to the GI tract itself or some other body organ such as kidney, thyroid, bone, etc. One of these body organs may become the critical body tissue as a result of the absorption of the radioactive material through the intestinal wall and deposition in the organ in a large concentration so that it receives the major absorbed dose of radiation. Another type of serious accident with radioactive material may occur as a result of getting radioactive material into the body by way of a puncture wound, an open sore, or in some cases through the intact pores of the skin. One of the more common single exposure accidents is the result of puncture wounds with contaminated glassware. It is to be hoped that the values given in these tables may aid in the selection and establishment of suitable maximum permissible single exposure values that are applicable to most accidents with radioactive materials, and that this grouping of the radionuclides in the order of increasing radiation hazard may serve as the basis for properly placing them in categories of *Very Dangerous, Moderately Dangerous,* and *Slightly Hazardous.*

## GENERAL ASSUMPTIONS AND NOMENCLATURE

In estimating the maximum permissible single exposure values for the various radionuclides, the characteristics of the standard man assumed are those which are outlined in the National[1] and International[2] Handbooks on internal dose and summarized as follows:

### 1. Mass *m* of the Body Organs in Grams and Effective Radius, X, in cm

| | gm | cm |
|---|---|---|
| Total body | $7 \times 10^4$ | 30 |
| Muscle | $3 \times 10^4$ | 30 |
| Fat | $1 \times 10^4$ | 20 |
| Bone | $7 \times 10^3$ | 5 |
| Blood | $5.4 \times 10^3$ | — |
| Skin | $2 \times 10^3$ | 0.1 |
| Liver | $1.7 \times 10^3$ | 10 |
| GI tract | $1.6 \times 10^3$ | — |
| Small intestine | 1100 | 30 |
| Stomach | 250 | 10 |
| Lower large intestine | 150 | 5 |
| Upper large intestine | 135 | 5 |
| Lungs | $1 \times 10^3$ | 30 |
| Kidneys | 300 | 7 |
| Spleen | 150 | 7 |
| Thyroid | 20 | 3 |

### 2. Time

| | |
|---|---|
| Period of inhalation | $t_1 = 8$ hours |
| Period of injection | $t_1 = 0$ |
| Period of permissible exposure | $t_2$ |

The following three cases are considered: $t_2 = 7$ days; $t_2 = 1$ year; and $t_2 = 70$ years.

When the GI tract is the critical organ, only the one exposure period is considered, e.g., $t_2 = 7$ days.

### 3. Permissible Dose, *D*

Three cases are considered: $D = 0.3$ rem for $t_2 = 7$ days; $D = 15.7$ rem for $t_2 = 1$ year; and $D = 150$ rem* for $t_2 = 70$ years.

### 4. Absorbed Energy per Disintegration, $\Sigma(bE)$(RBE) *N*

In calculating the values of $\Sigma(bE)(\mathrm{RBE})N$, the relative biological effectiveness (RBE) was set equal to 1 for $\beta^-$, $\beta^+$ $e^-$, X- and $\gamma$-radiation, 10 for $\alpha$, and 20 for the recoil ions. The nonuniform distribution factor, $N$, was set equal to 1 in all cases except for the ionizing particles emitted in the bone, in which case it was set equal to 5. $P^{32}$ and $Ra^{226}$ were exceptions to this rule in that $N$ was taken as 1. The value of $\Sigma(bE)(\mathrm{RBE})N$ was reduced by a factor of 2 for all calculations based on the GI tract in order to account for the fact that the walls of the GI tract are irradiated mostly from only one-half the solid angle. The calculation of $\Sigma(bE)(\mathrm{RBE})N$ for the GI tract makes correction for the absorption of some of the radioactive material through the wall of the small intestine and for the decay of the parent and growth of a radioactive daughter—if there is one—as the contaminated material passes through the GI tract during a 31-hour period. (See Appendix for details).

The values of $\Sigma(bE)(\mathrm{RBE})N$ may be found as follows:

1. For Case 1 dealing with inhalation of soluble radioactive material, use Table 5 of ICRP Handbook,[2] and also column 5 (the values outside parenthesis) to find $\Sigma(bE)(\mathrm{RBE})N$ for organ listed in column 4.

* It is to be observed that this value is considerably less than $15.7 \times 70$. This reduction is deliberate and based on the assumption of the authors that the lifetime dose should not exceed 150 rem.

2. For Case 2 dealing with injection of soluble radioactive material, use same references as for Case 1 above.

3. For Case 3 dealing with injection of insoluble radioactive material, it is assumed the effective diameter, $X$, of the contaminated tissue in the wound is 0.1 cm. The value of $X$ determines the fraction of the $\gamma$-energy absorbed in the wound and values of $\Sigma(bE)(\mathrm{RBE})N$ have not been given in the ICRP Handbook[2] in this case. Therefore, they are calculated, using Equation 5 of the ICRP Handbook.[2]

4. For Case 4 dealing with inhalation of insoluble radioactive material and the lung as the critical body organ, use Table 5 of the ICRP Handbook,[2] column 5, second value in parenthesis.

5. For Case 5 dealing with inhalation of insoluble radioactive material, in which case a portion of the GI tract is the critical body organ, use Table 5 of the ICRP Handbook,[2] column 5, first value in parenthesis, for the upper and lower large intestines, each of which is assumed to have an effective diameter $X = 5$ cm.

Use Table 5 of the ICRP Handbook,[2] column 5, second value in parenthesis for small intestine, which is assumed to have a value of $X = 30$ cm.

The values of $\Sigma(bE)(\mathrm{RBE})N$ have not been calculated for the stomach with an assumed value of $X = 10$ cm. Therefore, they are calculated for this report, using Equation 5 of the ICRP Handbook.[2]

#### 5. Coefficients of Elimination and Decay and Half-Lives

$\lambda_r$ = coefficient of radioactive decay
$\lambda_b$ = coefficient of biological elimination
$\lambda = \lambda_r + \lambda_b$ = effective coefficient
$T_r$ = radioactive half-life
$T_b$ = biological half-life
$T$ = effective half-life
$T = T_b T_r / T_b + T_r$.

The half-lives used are taken from the ICRP Handbook[2] except where more recent measurements are available; the values of $T_r$ are given in column 3 of Table 5; $T_b$ for the soluble radioactive material is given in column 13 of Table 4; and $T$ is given in column 6 of Table 5. For the insoluble radioactive material, $T_b$ is taken as 120 days, both for the lungs and for the tissue in a wound. For the case where the GI tract is the critical organ, $T_b$ is calculated from the values of $f_1$ (see Appendix) in which $f_1$, the fraction going from the GI tract to the blood, is given in column 5, Table 4, of the ICRP Handbook.[2]

#### 6. Fraction, F, retained in the Critical Body Organ

The value of $f$ for inhalation of soluble radioactive material is the same as given in the ICRP Handbook,[2] Table 4, column 12 for the body organs listed in column 9. For inhalation of insoluble material $f = 0.125$ when the lung is the critical organ, and $f = 0.625$ when the GI tract is the critical organ. The value of $f$ for soluble radioactive material taken into the body as a result of a puncture wound is the same as that given for $f_2$ in column 15 of Table 4 of the ICRP Handbook;[2] since $f_2$ is the fraction going from the blood to the critical organ, this is equivalent to assuming all the soluble radioactive material injected into the body in a wound is taken up in the blood. In the case of insoluble radioactive material taken into the body as a result of a puncture wound, $f$ is set equal to 1.

## CALCULATIONS AND RESULTS

The relative hazard of the various radionuclides is based on five different criteria as follows:

#### Case 1. The Inhalation of Soluble Radioactive Material

In this case the critical body organ is one of the following: total body; muscle; fat; bone; blood; skin; liver; kidneys; spleen; or thyroid.

#### Case 2. The Injection of Soluble Material into the Body by Way of a Puncture Wound

This case is similar to Case 1 except for the mode of entry into the body, and it is assumed the fraction, $f_2$, of the radioactive material arrives at the critical body organ immediately after the injection.

#### Case 3. The Injection of Insoluble Radioactive Material by Way of a Puncture Wound

It is assumed that $f = 1$, and that all of the radioactive material remains in a localized mass of tissue of $10^{-3}$ grams at the site of the wound.

#### Case 4. The Inhalation of Insoluble Radioactive Material with the Lung as the Critical Organ

#### Case 5. The Inhalation of Insoluble Radioactive Material where some Portion of the GI Tract is the Critical Body Tissue

The portions of the GI tract considered are the stomach, small intestine, upper large intestine, and lower large intestine. It is assumed some of the material is eliminated uniformly in the passage of the radioactive material through the small intestine and that radioactive daughter products—where present—contribute to the absorbed dose after they are produced along the GI tract.

The equations used in each of the above five cases are given in the next section of this report, and the derivations are given in the Appendix. Tables I, II, III, IV, and V of this report summarize the data for Cases 1, 2, 3, 4, and 5, respectively, and give the maximum permissible intake (MPI) in $\mu$c that will deliver the specified dose to the indicated critical body organ. In Table IV the lung is the critical organ; in Table III the wound site is the critical tissue; and in Table V the portion of the GI tract resulting in the smallest (MPI) value is the critical organ. In the case of the GI tract (Table V) the calculations are made only on the basis of a permissible dose of 0.3 rem delivered in one week, since the total time in the GI tract is 31 hours. The lowest value of MPI in each case is given also in these Tables in mg in order to indicate the

**Table 1. Single Exposure Values for Inhalation of Soluble Radioactive Material when Body Organs Other Than the GI Tract or Lungs Are the Critical Body Organs**

| (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) |
|---|---|---|---|---|---|---|---|---|
| | | | | MPI† or μc inhaled in 8 hours that will present a dose of: | | | | |
| *Z* | *Isotope* | *Critical organ* | *Fraction deposited in critical organ f* | *0.3 rem in 1 wk* | *15.7 rem in 1 yr* | *150 rem in 70 yrs* | *Smallest values in columns 5, 6 and 7 in mg* | *Relative hazard‡¶ H* |
| 1 | $H^3$ (HTO or $H^3_2O$) | T. body | 0.75 | *1.5 × 10⁴* | $1.7 \times 10^5$ | $1.7 \times 10^6$ | $1.6 \times 10^{-3}$ | $6.6 \times 10^{-5}$ |
| 4 | $Be^7$ | Bone | $9.0 \times 10^{-2}$ | *7.7 × 10³* | $4.0 \times 10^4$ | $3.7 \times 10^5$ | $2.2 \times 10^{-5}$ | $1.3 \times 10^{-4}$ |
| 6 | $C^{14}$ ($CO_2$) | Fat | 0.36 | *4.7 × 10²* | $3.2 \times 10^3$ | $3.0 \times 10^4$ | 0.10 | $9.6 \times 10^{-4}$ |
| 9 | $F^{18}$ | Bone | $7.5 \times 10^{-2}$ | *1.3 × 10⁴* | $6.7 \times 10^5$ | $6.3 \times 10^6$ | $1.4 \times 10^{-7}$ | $7.8 \times 10^{-5}$ |
| 11 | $Na^{24}$ | T. body | 0.73 | *2.8 × 10²* | $1.5 \times 10^4$ | $1.4 \times 10^5$ | $3.3 \times 10^{-8}$ | $3.6 \times 10^{-3}$ |
| 15 | $P^{32}$ | Bone | 0.2 | *53* | $8.0 \times 10^2$ | $7.7 \times 10^3$ | $1.9 \times 10^{-7}$ | $1.9 \times 10^{-2}$ |
| 16 | $S^{35}$ | Skin | $7.4 \times 10^{-2}$ | *4.7 × 10²* | $5.7 \times 10^3$ | $5.7 \times 10^4$ | $1.1 \times 10^{-5}$ | $2.1 \times 10^{-3}$ |
| 17 | $Cl^{36}$ | T. body | 0.73 | *3.7 × 10²* | $4.0 \times 10^3$ | $4.0 \times 10^4$ | 15 | $6.6 \times 10^{-6}$ |
| 19 | $K^{42}$ | Muscle | 0.53 | *3.3 × 10²* | $1.8 \times 10^4$ | $1.7 \times 10^4$ | $5.7 \times 10^{-8}$ | $3 \times 10^{-3}$ |
| 20 | $Ca^{45}$ | Bone | 0.41 | *33* | 67 | $5.0 \times 10^2$ | $2.0 \times 10^{-6}$ | $3 \times 10^{-2}$ |
| 21 | $Sc^{46}$ | Spleen | $7.5 \times 10^{-3}$ | 40 | $6.7 \times 10^2$ | $6.3 \times 10^3$ | $8.7 \times 10^{-7}$ | $3.3 \times 10^{-2}$ |
| | | Liver | 0.12 | *29* | $4.7 \times 10^2$ | $4.3 \times 10^3$ | | |
| 21 | $Sc^{47}$ | Spleen | $7.5 \times 10^{-3}$ | $1.8 \times 10^2$ | $7.3 \times 10^3$ | $7.0 \times 10^4$ | $1.5 \times 10^{-7}$ | $8.1 \times 10^{-3}$ |
| | | Liver | 0.12 | *1.2 × 10²* | $5.3 \times 10^3$ | $5.0 \times 10^4$ | | |
| 21 | $Sc^{48}$ | Spleen | $7.5 \times 10^{-3}$ | 87 | $4.3 \times 10^3$ | $4.3 \times 10^4$ | $3.3 \times 10^{-8}$ | $2.0 \times 10^{-2}$ |
| | | Liver | 0.12 | *50* | $2.5 \times 10^3$ | $2.4 \times 10^4$ | | |
| 23 | $V^{48}$ | Bone | 0.025 | *2.7 × 10²* | $4.7 \times 10^3$ | $4.3 \times 10^4$ | $1.6 \times 10^{-6}$ | $3.9 \times 10^{-3}$ |
| 24 | $Cr^{51}$ | Kidneys | $4 \times 10^{-3}$ | *10⁴* | $10^5$ | $10^6$ | $1.1 \times 10^{-4}$ | $9.9 \times 10^{-5}$ |
| 25 | $Mn^{56}$ | Kidneys | $2.2 \times 10^{-2}$ | *1.2 × 10³* | $6.0 \times 10^4$ | $6.0 \times 10^5$ | $5.3 \times 10^{-8}$ | $8.7 \times 10^{-4}$ |
| | | Liver | $9 \times 10^{-2}$ | $1.6 \times 10^3$ | $8.3 \times 10^4$ | $8.0 \times 10^5$ | | |
| 26 | $Fe^{55}$ | Blood | 0.65 | *1.2 × 10³* | $5.0 \times 10^3$ | $4.3 \times 10^4$ | $5.7 \times 10^{-4}$ | $8.4 \times 10^{-4}$ |
| 26 | $Fe^{59}$ | Blood | 0.65 | *14* | $1.2 \times 10^2$ | $1.2 \times 10^3$ | $2.8 \times 10^{-7}$ | $7.2 \times 10^{-2}$ |
| 27 | $Co^{60}$ | Liver | $7 \times 10^{-3}$ | *3.7 × 10²* | $8.7 \times 10^2$ | $8.3 \times 10^4$ | $3.2 \times 10^{-4}$ | $2.7 \times 10^{-3}$ |
| 28 | $Ni^{59}$ | Liver | $7 \times 10^{-3}$ | *5.7 × 10³* | $1.3 \times 10^5$ | $1.2 \times 10^6$ | 73 | $1.4 \times 10^{-6}$ |
| 29 | $Cu^{64}$ | Liver | 0.13 | *8.3 × 10²* | $4.3 \times 10^4$ | $4.0 \times 10^5$ | $2.2 \times 10^{-7}$ | $1.2 \times 10^{-3}$ |
| 30 | $Zn^{65}$ | Bone | $4.5 \times 10^{-2}$ | *1.5 × 10³* | $1.6 \times 10^4$ | $1.6 \times 10^5$ | $1.8 \times 10^{-4}$ | $6.6 \times 10^{-4}$ |
| 31 | $Ga^{72}$ | Bone | 0.1 | *2.2 × 10²* | $1.2 \times 10^4$ | $1.1 \times 10^5$ | $7.3 \times 10^{-8}$ | $4.5 \times 10^{-3}$ |
| 32 | $Ge^{71}$ | Kidneys | $5 \times 10^{-3}$ | *9.0 × 10³* | $3.3 \times 10^5$ | $3.2 \times 10^6$ | $5.7 \times 10^{-5}$ | $1.1 \times 10^{-4}$ |
| 33 | $As^{76}$ | Kidneys | $2.7 \times 10^{-3}$ | *4.3 × 10²* | $2.2 \times 10^4$ | $2.1 \times 10^5$ | $2.8 \times 10^{-7}$ | $2.3 \times 10^{-3}$ |
| 37 | $Rb^{86}$ | Muscle | 0.33 | *1.4 × 10²* | $3.3 \times 10^3$ | $3.2 \times 10^4$ | $1.8 \times 10^{-6}$ | $6.9 \times 10^{-3}$ |
| 38 | $Sr^{89}$ | Bone | 0.22 | *10* | 47 | $4.3 \times 10^2$ | $3.7 \times 10^{-7}$ | $9.9 \times 10^{-2}$ |
| 38 | $Sr^{90} + Y^{90}$ | Bone | 0.22 | 5.3 | 5.7 | *5* | $2.5 \times 10^{-5}$ | 0.20 |
| 39 | $Y^{91}$ | Bone | 0.14 | *16* | 73 | $7.0 \times 10^2$ | $6.3 \times 10^{-7}$ | $6.3 \times 10^{-2}$ |
| 40 | $Zr^{95} + Nb^{95}$ | Bone | $5.8 \times 10^{-2}$ | *1.1 × 10²* | $5.3 \times 10^2$ | $5.3 \times 10^3$ | $5.0 \times 10^{-4}$ | $9.0 \times 10^{-3}$ |
| 41 | $Nb^{95}$ | Bone | 0.12 | *1.7 × 10³* | $1.8 \times 10^3$ | $1.7 \times 10^4$ | $4.3 \times 10^{-6}$ | $6.0 \times 10^{-3}$ |
| 42 | $Mo^{99}$ | Bone | $2 \times 10^{-4}$ | *9.3 × 10⁴* | $4.0 \times 10^6$ | $4.0 \times 10^7$ | $2.0 \times 10^{-4}$ | $1.1 \times 10^{-5}$ |
| 43 | $Tc^{96}$ | Kidneys | $3 \times 10^{-3}$ | *4.7 × 10²* | $2.1 \times 10^4$ | $2.0 \times 10^5$ | $1.4 \times 10^{-6}$ | $2.1 \times 10^{-3}$ |
| 44 | $Ru^{106} + Rh^{106}$ | Kidneys | $10^{-2}$ | *20* | $2.4 \times 10^2$ | $2.3 \times 10^3$ | $6.0 \times 10^{-6}$ | $4.8 \times 10^{-2}$ |
| 45 | $Rh^{105}$ | Kidneys | $2 \times 10^{-2}$ | *1.4 × 10²* | $7.0 \times 10^3$ | $6.7 \times 10^4$ | $1.7 \times 10^{-7}$ | $7.2 \times 10^{-2}$ |
| 46 | $Pd^{103} + Rh^{103*}$ | Kidneys | $4 \times 10^{-2}$ | *1.4 × 10²* | $5.0 \times 10^3$ | $4.7 \times 10^4$ | $1.9 \times 10^{-6}$ | $6.9 \times 10^{-3}$ |
| 47 | $Ag^{105}$ | Liver | $2 \times 10^{-3}$ | *2.0 × 10³* | $8.7 \times 10^4$ | $8.3 \times 10^5$ | $7.3 \times 10^{-5}$ | $4.8 \times 10^{-4}$ |
| 47 | $Ag^{111}$ | Liver | $2 \times 10^{-3}$ | *5.0 × 10³* | $2.4 \times 10^5$ | $2.3 \times 10^6$ | $3.2 \times 10^{-5}$ | $2.0 \times 10^{-4}$ |
| 48 | $Cd^{109} + Ag^{109*}$ | Liver | 0.19 | *2.0 × 10²* | $6.0 \times 10^2$ | $5.7 \times 10^3$ | $7.7 \times 10^{-5}$ | $5.1 \times 10^{-3}$ |
| 50 | $Sn^{113}$ | Bone | $7.6 \times 10^{-2}$ | *9.3 × 10²* | $5.0 \times 10^3$ | $5.0 \times 10^4$ | $9.0 \times 10^{-5}$ | $1.1 \times 10^{-3}$ |
| 52 | $Te^{127}$ | Kidneys | $2 \times 10^{-2}$ | *53* | $8.7 \times 10^2$ | $8.3 \times 10^3$ | $6.0 \times 10^{-6}$ | $1.9 \times 10^{-2}$ |
| 52 | $Te^{129}$ | Kidneys | $2 \times 10^{-2}$ | *18* | $3.7 \times 10^2$ | $3.3 \times 10^3$ | $6.0 \times 10^{-7}$ | $5.4 \times 10^{-2}$ |

* The daughter products in these cases are isomers in an excited state.

† The MPI is the μc present initially in the volume of air to be breathed during the succeeding 8 hours and which then results in the indicated dose.

**Table 1. Single Exposure Values for Inhalation of Soluble Radioactive Material when Body Organs Other Than the GI Tract or Lungs Are the Critical Body Organs — *Continued***

| (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) |
|---|---|---|---|---|---|---|---|---|
| *Z* | *Isotope* | *Critical organ* | *Fraction deposited in critical organ f* | *MPI† or μc inhaled in 8 hours that will present a dose of:* 0.3 rem in 1 wk | 15.7 rem in 1 yr | 150 rem in 70 yrs | *Smallest values in columns 5, 6 and 7 in mg* | *Relative hazard ‡,¶ H* |
| 53 | $I^{131}$ | Thyroid | 0.15 | *0.70* | 17 | $1.7 \times 10^{2}$ | $5.7 \times 10^{-9}$ | 1.4 |
| 55 | $Cs^{137} + Ba^{137*}$ | Muscle | 0.36 | *$1.4 \times 10^{2}$* | $1.8 \times 10^{3}$ | $1.7 \times 10^{4}$ | $1.8 \times 10^{-3}$ | $6.9 \times 10^{-3}$ |
| 56 | $Ba^{140} + La^{140}$ | Bone | 0.2 | *8.7* | $1.5 \times 10^{2}$ | $1.4 \times 10^{3}$ | $1.2 \times 10^{-7}$ | 0.11 |
| 57 | $La^{140}$ | Bone | 0.1 | *70* | $3.3 \times 10^{3}$ | $3.3 \times 10^{4}$ | $1.2 \times 10^{-7}$ | $1.4 \times 10^{-2}$ |
| 58 | $Ce^{144} + Pr^{144}$ | Bone | 0.1 | *9.3* | 17 | $1.2 \times 10^{2}$ | $3.0 \times 10^{-6}$ | 0.11 |
| 59 | $Pr^{143}$ | Bone | $6.3 \times 10^{-2}$ | *73* | $1.4 \times 10^{3}$ | $1.3 \times 10^{4}$ | $1.1 \times 10^{-6}$ | $1.4 \times 10^{-2}$ |
| 61 | $Pm^{147}$ | Bone | $9 \times 10^{-2}$ | *$2.0 \times 10^{2}$* | $5.7 \times 10^{2}$ | $5.3 \times 10^{3}$ | $2.1 \times 10^{-4}$ | $5.1 \times 10^{-3}$ |
| 62 | $Sm^{151}$ | Bone | $5 \times 10^{-2}$ | $1.1 \times 10^{3}$ | $1.2 \times 10^{3}$ | *$2.4 \times 10^{2}$* | $10^{-2}$ | $4.2 \times 10^{-3}$ |
| 63 | $Eu^{154}$ | Bone | $9 \times 10^{-2}$ | *57* | 60 | $1.2 \times 10^{2}$ | $4.0 \times 10^{-4}$ | $1.8 \times 10^{-2}$ |
| 67 | $Ho^{166}$ | Bone | $7 \times 10^{-2}$ | *$1.3 \times 10^{2}$* | $6.7 \times 10^{3}$ | $6.3 \times 10^{4}$ | $1.9 \times 10^{-7}$ | $7.8 \times 10^{-3}$ |
| 69 | $Tm^{170}$ | Bone | 0.18 | *21* | 90 | $8.3 \times 10^{2}$ | $3.7 \times 10^{-6}$ | $4.8 \times 10^{-2}$ |
| 71 | $Lu^{177}$ | Bone | $7.5 \times 10^{-2}$ | *$2.4 \times 10^{2}$* | $9.7 \times 10^{3}$ | $9.3 \times 10^{4}$ | $2.2 \times 10^{-6}$ | $4.2 \times 10^{-3}$ |
| 73 | $Ta^{182}$ | Liver | $7.5 \times 10^{-2}$ | *47* | $1.8 \times 10^{2}$ | $1.7 \times 10^{3}$ | $7.0 \times 10^{-6}$ | $2.1 \times 10^{-2}$ |
| 74 | $W^{181}$ | Bone | $2.4 \times 10^{-2}$ | *$9.3 \times 10^{3}$* | $3.3 \times 10^{4}$ | $3.3 \times 10^{5}$ | $1.8 \times 10^{-3}$ | $1.1 \times 10^{-4}$ |
| 75 | $Re^{183}$ | Thyroid | $1.3 \times 10^{-3}$ | *$1.4 \times 10^{3}$* | $7.3 \times 10^{4}$ | $7.0 \times 10^{5}$ | $2.3 \times 10^{-4}$ | $7.2 \times 10^{-4}$ |
| | | Skin | 0.12 | $7.3 \times 10^{3}$ | $2.4 \times 10^{5}$ | $2.3 \times 10^{6}$ | | |
| 77 | $Ir^{190}$ | Kidneys | $1.7 \times 10^{-2}$ | *$2.9 \times 10^{2}$* | $7.3 \times 10^{3}$ | $7.0 \times 10^{4}$ | $4.7 \times 10^{-6}$ | $3.3 \times 10^{-3}$ |
| | | Spleen | $6 \times 10^{-3}$ | $4.0 \times 10^{2}$ | $7.7 \times 10^{3}$ | $7.3 \times 10^{4}$ | | |
| 77 | $Ir^{192}$ | Kidneys | $2 \times 10^{-2}$ | *31* | $4.0 \times 10^{2}$ | $3.7 \times 10^{3}$ | $3.3 \times 10^{-6}$ | $3.3 \times 10^{-2}$ |
| | | Spleen | $6 \times 10^{-3}$ | 47 | $2.5 \times 10^{2}$ | $2.3 \times 10^{3}$ | | |
| 78 | $Pt^{191}$ | Kidneys | $3.3 \times 10^{-2}$ | *43* | $1.8 \times 10^{3}$ | $1.7 \times 10^{4}$ | $1.9 \times 10^{-7}$ | $2.3 \times 10^{-2}$ |
| 78 | $Pt^{193}$ | Kidneys | $3.3 \times 10^{-2}$ | *40* | $1.4 \times 10^{3}$ | $1.4 \times 10^{4}$ | $2.5 \times 10^{-7}$ | $2.5 \times 10^{-2}$ |
| 79 | $Au^{196}$ | Liver | 0.25 | 53 | $1.7 \times 10^{3}$ | $1.7 \times 10^{4}$ | $3.7 \times 10^{-7}$ | $2.3 \times 10^{-2}$ |
| | | Kidneys | $7.2 \times 10^{-2}$ | *43* | $1.4 \times 10^{3}$ | $1.3 \times 10^{4}$ | | |
| 79 | $Au^{198}$ | Liver | 0.25 | 31 | $1.4 \times 10^{3}$ | $1.3 \times 10^{4}$ | $8.3 \times 10^{-8}$ | $4.8 \times 10^{-2}$ |
| | | Kidneys | $7.2 \times 10^{-2}$ | *20* | $9.0 \times 10^{2}$ | $8.7 \times 10^{3}$ | | |
| 79 | $Au^{199}$ | Liver | 0.25 | 73 | $3.0 \times 10^{3}$ | $2.9 \times 10^{4}$ | $2.7 \times 10^{-7}$ | $1.9 \times 10^{-2}$ |
| | | Kidneys | $7.2 \times 10^{-2}$ | *53* | $2.1 \times 10^{3}$ | $2.0 \times 10^{4}$ | | |
| 81 | $Tl^{200}$ | Muscle | 0.26 | *$4.0 \times 10^{2}$* | $2.1 \times 10^{4}$ | $2.0 \times 10^{5}$ | $7.0 \times 10^{-7}$ | $2.5 \times 10^{-3}$ |
| 81 | $Tl^{201}$ | Muscle | 0.26 | *$1.4 \times 10^{3}$* | $6.0 \times 10^{4}$ | $6.0 \times 10^{5}$ | $6.3 \times 10^{-6}$ | $7.2 \times 10^{-4}$ |
| 81 | $Tl^{202}$ | Muscle | 0.26 | *$6.7 \times 10^{2}$* | $1.7 \times 10^{4}$ | $1.6 \times 10^{5}$ | $1.2 \times 10^{-5}$ | $1.5 \times 10^{-3}$ |
| 81 | $Tl^{204}$ | Muscle | 0.26 | *$4.7 \times 10^{2}$* | $6.0 \times 10^{3}$ | $5.7 \times 10^{4}$ | $1.1 \times 10^{-3}$ | $2.1 \times 10^{-3}$ |
| 82 | $Pb^{203}$ | Bone | $9.8 \times 10^{-2}$ | *$1.3 \times 10^{3}$* | $6.3 \times 10^{4}$ | $6.0 \times 10^{5}$ | $4.3 \times 10^{-6}$ | $7.5 \times 10^{-4}$ |
| 82 | $Pb^{210}$ + dr | Bone | $9.8 \times 10^{-2}$ | *0.21* | 0.26 | 0.80 | $2.7 \times 10^{-6}$ | 4.8 |
| 84 | $Po^{210}$ | Spleen | $4 \times 10^{-3}$ | *0.60* | 3.7 | 33 | $1.3 \times 10^{-7}$ | 1.7 |
| 85 | $At^{211}$ | Thyroid | 0.05 | *0.10* | 5.3 | 53 | $5.0 \times 10^{-11}$ | 9.6 |
| 88 | $Ra^{226}$ + 55% dr | Bone | $2.6 \times 10^{-2}$ | 1.4 | 1.4 | *0.32* | $3.2 \times 10^{-4}$ | 0.30 |
| 89 | $Ac^{227}$ + dr | Bone | $7.5 \times 10^{-2}$ | *$4.7 \times 10^{-3}$* | $5.3 \times 10^{-2}$ | 0.11 | $6.3 \times 10^{-6}$ | $2.1 \times 10^{2}$ |
| 90 | $Th^{nat}$ | Bone | 0.2 | $1.6 \times 10^{-2}$ | $1.5 \times 10^{-2}$ | *$2.6 \times 10^{-3}$* | 23 | $4.2 \times 10^{-6}$ |
| 90 | $Th^{234} + Pa^{234}$ | Bone | 0.2 | *7.7* | 73 | $7.0 \times 10^{2}$ | $3.3 \times 10^{-7}$ | 0.13 |
| 92 | $U^{nat}$ | Kidneys | $8 \times 10^{-2}$ | *$3.7 \times 10^{-2}$* | 0.28 | 2.7 | $1.1 \times 10^{2}$ | $9.3 \times 10^{-7}$ |
| 92 | $U^{233}$ | Bone | $5 \times 10^{-2}$ | *0.43* | 0.70 | 3.7 | $4.7 \times 10^{-2}$ | $2.1 \times 10^{-3}$ |
| 94 | $Pu^{239}$ | Bone | 0.18 | 0.13 | 0.12 | *$2.0 \times 10^{-2}$* | $3.2 \times 10^{-4}$ | 0.30 |
| 95 | $Am^{241}$ | Bone | $6.3 \times 10^{-2}$ | *0.33* | 0.37 | 0.90 | $1.1 \times 10^{-4}$ | 0.90 |
| 96 | $Cm^{242}$ | Bone | $6.3 \times 10^{-2}$ | *0.29* | 0.67 | 5.7 | $8.7 \times 10^{-8}$ | 3.3 |

‡ The larger the number in this column the greater the hazard.

¶ Values of *H* in this column correspond to the smallest value in columns 5, 6 or 7.

**Table II. Single Exposure Values for Injection (or Puncture Wounds) when Soluble Radioactive Materials Are Taken into the Body**

| (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) |
|---|---|---|---|---|---|---|---|---|
| | | | | *MPI or μc injected that will result in a dose of:* | | | | |
| *Z* | *Isotope* | *Critical organ* | *Fraction going from blood to critical organ $f_2'$* | *0.3 rem in 1 wk* | *15.7 rem in 1 yr* | *150 rem in 70 yrs* | *Smallest values in columns 5, 6 or 7 in mg* | *Relative hazard † ‡ H* |
| 1 | $H^3$ (HTO or $H^3_2O$) | T. Body | 1.0 | $1.1 \times 10^4$ | $1.3 \times 10^5$ | $1.2 \times 10^6$ | $1.1 \times 10^{-3}$ | $9.1 \times 10^{-5}$ |
| 4 | $Be^7$ | Bone | 0.35 | $2.0 \times 10^3$ | $10^4$ | $9.6 \times 10^4$ | $5.7 \times 10^{-6}$ | $5.0 \times 10^{-4}$ |
| 6 | $C^{14}$ ($CO_2$) | Fat | 0.5 | $3.4 \times 10^2$ | $2.3 \times 10^3$ | $2.2 \times 10^4$ | $7.5 \times 10^{-2}$ | $1.3 \times 10^{-4}$ |
| 9 | $F^{18}$ | Bone | 0.1 | $3.0 \times 10^3$ | $1.6 \times 10^5$ | $1.5 \times 10^6$ | $3.2 \times 10^{-3}$ | $3.3 \times 10^{-4}$ |
| 11 | $Na^{24}$ | T. body | 1.0 | $1.7 \times 10^2$ | $9.0 \times 10^3$ | $8.6 \times 10^4$ | $2.0 \times 10^{-3}$ | $5.9 \times 10^{-3}$ |
| 15 | $P^{32}$ | Bone | 0.3 | 34 | $5.2 \times 10^2$ | $5.0 \times 10^3$ | $1.2 \times 10^{-7}$ | $2.9 \times 10^{-2}$ |
| 16 | $S^{35}$ | Skin | 0.14 | $2.5 \times 10^2$ | $3.0 \times 10^3$ | $2.9 \times 10^4$ | $5.8 \times 10^{-6}$ | $4.0 \times 10^{-3}$ |
| 17 | $Cl^{36}$ | T. body | 1.0 | $2.6 \times 10^2$ | $3.0 \times 10^3$ | $2.9 \times 10^4$ | 11 | $9.4 \times 10^{-7}$ |
| 19 | $K^{42}$ | Muscle | 0.75 | $1.9 \times 10^2$ | $10^4$ | $9.9 \times 10^4$ | $3.2 \times 10^{-3}$ | $5.3 \times 10^{-3}$ |
| 20 | $Ca^{45}$ | Bone | 0.58 | 24 | 46 | $3.5 \times 10^2$ | $1.3 \times 10^{-6}$ | $4.2 \times 10^{-3}$ |
| 21 | $Sc^{46}$ | Spleen | $3 \times 10^{-2}$ | 10 | $1.6 \times 10^2$ | $1.6 \times 10^3$ | $3.0 \times 10^{-7}$ | 0.10 |
| | | Liver | 0.6 | 57 | 92 | $8.8 \times 10^2$ | | |
| 21 | $Sc^{47}$ | Spleen | $3 \times 10^{-2}$ | 31 | $1.4 \times 10^3$ | $1.3 \times 10^4$ | $2.9 \times 10^{-3}$ | $4.2 \times 10^{-2}$ |
| | | Liver | 0.6 | 24 | $10^3$ | $9.8 \times 10^3$ | | |
| 21 | $Sc^{48}$ | Spleen | $3 \times 10^{-2}$ | 20 | $10^3$ | $9.7 \times 10^3$ | $6.3 \times 10^{-9}$ | 0.11 |
| | | Liver | 0.6 | 9.4 | $4.7 \times 10^2$ | $4.5 \times 10^3$ | | |
| 23 | $V^{48}$ | Bone | 0.1 | 64 | $1.1 \times 10^3$ | $1.1 \times 10^4$ | $3.7 \times 10^{-7}$ | $1.6 \times 10^{-2}$ |
| 24 | $Cr^{51}$ | Kidneys | $1.4 \times 10^{-2}$ | $2.8 \times 10^3$ | $2.9 \times 10^4$ | $2.8 \times 10^5$ | $3.0 \times 10^{-5}$ | $3.6 \times 10^{-4}$ |
| 25 | $Mn^{56}$ | Kidneys | $8 \times 10^{-2}$ | $1.3 \times 10^2$ | $6.9 \times 10^3$ | $6.6 \times 10^4$ | $6.0 \times 10^{-9}$ | $7.7 \times 10^{-3}$ |
| | | Liver | 0.16 | $3.7 \times 10^2$ | $1.9 \times 10^4$ | $1.8 \times 10^5$ | | |
| 26 | $Fe^{55}$ | Blood | 1.0 | $7.8 \times 10^2$ | $3.2 \times 10^3$ | $3.0 \times 10^4$ | $3.6 \times 10^{-4}$ | $1.3 \times 10^{-3}$ |
| 26 | $Fe^{59}$ | Blood | 1.0 | 9.1 | 79 | $7.5 \times 10^2$ | $1.8 \times 10^{-7}$ | 0.11 |
| 27 | $Co^{60}$ | Liver | $2 \times 10^{-2}$ | $1.3 \times 10^2$ | $3.0 \times 10^3$ | $2.8 \times 10^4$ | $1.1 \times 10^{-4}$ | $7.7 \times 10^{-3}$ |
| 28 | $Ni^{59}$ | Liver | $2 \times 10^{-2}$ | $1.9 \times 10^3$ | $4.5 \times 10^4$ | $4.3 \times 10^5$ | 25 | $4.0 \times 10^{-7}$ |
| 29 | $Cu^{64}$ | Liver | 0.33 | $2.6 \times 10^2$ | $1.4 \times 10^4$ | $1.3 \times 10^5$ | $6.8 \times 10^{-3}$ | $3.8 \times 10^{-3}$ |
| 30 | $Zn^{65}$ | Bone | 0.15 | $4.5 \times 10^2$ | $4.8 \times 10^3$ | $4.6 \times 10^4$ | $5.4 \times 10^{-5}$ | $2.2 \times 10^{-3}$ |
| 31 | $Ga^{72}$ | Bone | 0.4 | 46 | $2.4 \times 10^3$ | $2.3 \times 10^4$ | $1.5 \times 10^{-3}$ | $2.2 \times 10^{-2}$ |
| 32 | $Ge^{71}$ | Kidneys | $2 \times 10^{-2}$ | $2.2 \times 10^3$ | $8.2 \times 10^4$ | $7.8 \times 10^5$ | $1.4 \times 10^{-5}$ | $4.5 \times 10^{-4}$ |
| 33 | $As^{76}$ | Kidneys | $1 \times 10^{-2}$ | $10^2$ | $5.3 \times 10^3$ | $5.1 \times 10^4$ | $6.4 \times 10^{-3}$ | $10^{-2}$ |
| 37 | $Rb^{86}$ | Muscle | 0.44 | $10^2$ | $2.5 \times 10^3$ | $2.4 \times 10^4$ | $1.3 \times 10^{-6}$ | $10^{-2}$ |
| 38 | $Sr^{89}$ | Bone | 0.4 | 5.5 | 26 | $2.4 \times 10^2$ | $2.0 \times 10^{-7}$ | 0.18 |
| 38 | $Sr^{90} + Y^{90}$ | Bone | 0.4 | 2.9 | 3.0 | 2.7 | $1.3 \times 10^{-5}$ | 0.34 |
| 39 | $Y^{91}$ | Bone | 0.55 | 4.0 | 19 | $1.8 \times 10^2$ | $1.6 \times 10^{-7}$ | 0.25 |
| 40 | $Zr^{95} + Nb^{95}$ | Bone | 0.23 | 27 | $1.4 \times 10^2$ | $1.3 \times 10^3$ | $1.3 \times 10^{-6}$ | $3.7 \times 10^{-2}$ |
| 41 | $Nb^{95}$ | Bone | 0.25 | 79 | $8.6 \times 10^2$ | $8.2 \times 10^3$ | $2.0 \times 10^{-6}$ | $1.3 \times 10^{-2}$ |
| 42 | $Mo^{99}$ | Bone | $3 \times 10^{-4}$ | $5.9 \times 10^4$ | $2.6 \times 10^6$ | $2.4 \times 10^7$ | $1.3 \times 10^{-4}$ | $1.7 \times 10^{-5}$ |
| 43 | $Tc^{96}$ | Kidneys | $5 \times 10^{-3}$ | $2.6 \times 10^2$ | $1.2 \times 10^4$ | $1.2 \times 10^5$ | $8.0 \times 10^{-7}$ | $3.8 \times 10^{-3}$ |
| 44 | $Ru^{106} + Rh^{106}$ | Kidneys | $4 \times 10^{-2}$ | 5.1 | 60 | $5.7 \times 10^2$ | $1.5 \times 10^{-6}$ | 0.2 |
| 45 | $Rh^{105}$ | Kidneys | $5 \times 10^{-2}$ | 51 | $2.6 \times 10^3$ | $2.4 \times 10^4$ | $6.2 \times 10^{-3}$ | $2.0 \times 10^{-2}$ |
| 46 | $Pd^{108} + Rh^{108*}$ | Kidneys | 0.1 | 56 | $2.0 \times 10^3$ | $1.9 \times 10^4$ | $7.5 \times 10^{-7}$ | $1.8 \times 10^{-2}$ |
| 47 | $Ag^{105}$ | Liver | $6 \times 10^{-3}$ | $6.8 \times 10^2$ | $2.9 \times 10^4$ | $2.8 \times 10^5$ | $2.4 \times 10^{-5}$ | $1.5 \times 10^{-3}$ |
| 47 | $Ag^{111}$ | Liver | $6 \times 10^{-3}$ | $1.6 \times 10^3$ | $7.7 \times 10^4$ | $7.4 \times 10^5$ | $10^{-5}$ | $6.3 \times 10^{-4}$ |
| 48 | $Cd^{109} + Ag^{109*}$ | Liver | 0.75 | 49 | $1.5 \times 10^2$ | $1.4 \times 10^3$ | $1.9 \times 10^{-5}$ | $2.0 \times 10^{-2}$ |
| 50 | $Sn^{113}$ | Bone | 0.3 | $2.4 \times 10^2$ | $1.3 \times 10^3$ | $1.2 \times 10^4$ | $2.3 \times 10^{-5}$ | $4.2 \times 10^{-3}$ |
| 52 | $Te^{127}$ | Kidneys | $6 \times 10^{-2}$ | 18 | $2.9 \times 10^2$ | $2.8 \times 10^3$ | $2.0 \times 10^{-6}$ | $5.6 \times 10^{-2}$ |

* The daughter products in these cases are isomers in an excited state.

† The larger the number in this column the greater the hazard.

**Table II. Single Exposure Values for Injection (or Puncture Wounds) when Soluble Radioactive Materials Are Taken into the Body — *Continued***

| (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) |
|---|---|---|---|---|---|---|---|---|
| | | | | *MPI or μc injected that will result in a dose of:* | | | | |
| *Z* | *Isotope* | *Critical organ* | *Fraction going from blood to critical organ $f_2$* | *0.3 rem in 1 wk* | *15.7 rem in 1 yr* | *150 rem in 70 yrs* | *Smallest values in columns 5, 6 or 7 in mg* | *Relative hazard* † ‡ *H* |
| 52 | $Te^{129}$ | Kidneys | $6 \times 10^{-2}$ | *5.9* | $1.2 \times 10^{2}$ | $1.1 \times 10^{3}$ | $1.9 \times 10^{-7}$ | 0.17 |
| 53 | $I^{131}$ | Thyroid | 0.2 | *0.52* | 13 | $1.2 \times 10^{2}$ | $4.1 \times 10^{-9}$ | 1.9 |
| 55 | $Cs^{137} + Ba^{137*}$ | Muscle | 0.48 | *1.1 × 10²* | $1.4 \times 10^{3}$ | $1.3 \times 10^{4}$ | $1.4 \times 10^{-3}$ | $7.3 \times 10^{-3}$ |
| 56 | $Ba^{140} + La^{140}$ | Bone | 0.7 | *2.4* | 41 | $3.9 \times 10^{2}$ | $3.3 \times 10^{-8}$ | 0.42 |
| 57 | $La^{140}$ | Bone | 0.4 | *16* | $8.0 \times 10^{2}$ | $7.6 \times 10^{3}$ | $2.8 \times 10^{-8}$ | $6.3 \times 10^{-2}$ |
| 58 | $Ce^{144} + Pr^{144}$ | Bone | 0.4 | *2.3* | 4.3 | 31 | $7.3 \times 10^{-7}$ | 0.43 |
| 59 | $Pr^{143}$ | Bone | 0.25 | *18* | $3.4 \times 10^{2}$ | $3.2 \times 10^{3}$ | $2.7 \times 10^{-7}$ | $5.6 \times 10^{-2}$ |
| 61 | $Pm^{147}$ | Bone | 0.35 | *50* | $1.5 \times 10^{2}$ | $1.3 \times 10^{3}$ | $5.3 \times 10^{-5}$ | $2.0 \times 10^{-2}$ |
| 62 | $Sm^{151}$ | Bone | 0.2 | $3.0 \times 10^{2}$ | $2.9 \times 10^{2}$ | *60* | $2.5 \times 10^{-3}$ | $3.9 \times 10^{-3}$ |
| 63 | $Eu^{154}$ | Bone | 0.35 | *14* | 16 | 31 | $9.5 \times 10^{-5}$ | $7.1 \times 10^{-2}$ |
| 67 | $Ho^{166}$ | Bone | 0.22 | *37* | $1.9 \times 10^{3}$ | $1.8 \times 10^{4}$ | $5.3 \times 10^{-8}$ | $2.7 \times 10^{-2}$ |
| 69 | $Tm^{170}$ | Bone | 0.7 | *5.4* | 23 | $2.1 \times 10^{2}$ | $8.9 \times 10^{-7}$ | 0.19 |
| 71 | $Lu^{177}$ | Bone | 0.3 | *58* | $2.4 \times 10^{3}$ | $2.3 \times 10^{4}$ | $5.3 \times 10^{-7}$ | $1.7 \times 10^{-2}$ |
| 73 | $Ta^{182}$ | Liver | 0.3 | *11* | 46 | $4.3 \times 10^{2}$ | $1.7 \times 10^{-6}$ | $9.1 \times 10^{-3}$ |
| 74 | $W^{181}$ | Bone | $8 \times 10^{-2}$ | *2.1 × 10³* | $10^{4}$ | $10^{5}$ | $4.0 \times 10^{-4}$ | $4.8 \times 10^{-4}$ |
| 75 | $Re^{183}$ | Thyroid | $2.5 \times 10^{-3}$ | *7.2 × 10²* | $3.8 \times 10^{4}$ | $3.6 \times 10^{5}$ | $1.2 \times 10^{-4}$ | $1.4 \times 10^{-3}$ |
| | | Skin | 0.8 | $1.1 \times 10^{3}$ | $3.5 \times 10^{4}$ | $3.4 \times 10^{5}$ | | |
| 77 | $Ir^{190}$ | Kidneys | $5 \times 10^{-2}$ | 98 | $2.5 \times 10^{3}$ | $2.4 \times 10^{4}$ | $4.3 \times 10^{-7}$ | $3.6 \times 10^{-2}$ |
| | | Spleen | $8 \times 10^{-2}$ | *28* | $5.7 \times 10^{2}$ | $5.4 \times 10^{3}$ | | |
| 77 | $Ir^{192}$ | Kidneys | $5 \times 10^{-2}$ | 12 | $1.5 \times 10^{2}$ | $1.5 \times 10^{3}$ | $3.9 \times 10^{-7}$ | 0.28 |
| | | Spleen | $8 \times 10^{-2}$ | *3.6* | 18 | $1.8 \times 10^{2}$ | | |
| 78 | $Pt^{191}$ | Kidneys | 0.11 | *12* | $5.1 \times 10^{2}$ | $4.9 \times 10^{3}$ | $5.2 \times 10^{-8}$ | $8.3 \times 10^{-2}$ |
| 78 | $Pt^{193}$ | Kidneys | 0.11 | *11* | $4.1 \times 10^{2}$ | $3.9 \times 10^{3}$ | $6.9 \times 10^{-8}$ | $9.1 \times 10^{-2}$ |
| 79 | $Au^{196}$ | Liver | 0.98 | *13* | $4.3 \times 10^{2}$ | $4.1 \times 10^{3}$ | $1.1 \times 10^{-7}$ | $7.7 \times 10^{-2}$ |
| | | Kidneys | 0.24 | 13 | $4.1 \times 10^{2}$ | $3.9 \times 10^{3}$ | | |
| 79 | $Au^{198}$ | Liver | 0.98 | 7.4 | $3.3 \times 10^{2}$ | $3.1 \times 10^{3}$ | $2.3 \times 10^{-8}$ | 0.17 |
| | | Kidneys | 0.24 | *5.8* | $2.5 \times 10^{2}$ | $2.4 \times 10^{3}$ | | |
| 79 | $Au^{199}$ | Liver | 0.98 | 18 | $7.4 \times 10^{2}$ | $7.1 \times 10^{3}$ | $7.5 \times 10^{-8}$ | $6.7 \times 10^{-2}$ |
| | | Kidneys | 0.24 | *15* | $6.1 \times 10^{2}$ | $5.8 \times 10^{3}$ | | |
| 81 | $Tl^{200}$ | Muscle | $6 \times 10^{-2}$ | *1.6 × 10³* | $8.1 \times 10^{4}$ | $7.7 \times 10^{5}$ | $2.7 \times 10^{-6}$ | $6.3 \times 10^{-4}$ |
| 81 | $Tl^{201}$ | Muscle | $6 \times 10^{-2}$ | *5.8 × 10³* | $2.5 \times 10^{5}$ | $2.4 \times 10^{6}$ | $2.7 \times 10^{-5}$ | $1.7 \times 10^{-4}$ |
| 81 | $Tl^{202}$ | Muscle | $6 \times 10^{-2}$ | *2.8 × 10³* | $7.2 \times 10^{4}$ | $6.9 \times 10^{5}$ | $5.1 \times 10^{-5}$ | $3.6 \times 10^{-4}$ |
| 81 | $Tl^{204}$ | Muscle | $6 \times 10^{-2}$ | *1.9 × 10³* | $2.5 \times 10^{4}$ | $2.4 \times 10^{5}$ | $4.4 \times 10^{-3}$ | $5.3 \times 10^{-4}$ |
| 82 | $Pb^{203}$ | Bone | 0.3 | *4.1 × 10²* | $1.9 \times 10^{4}$ | $1.8 \times 10^{5}$ | $1.4 \times 10^{-6}$ | $2.4 \times 10^{-3}$ |
| 82 | $Pb^{210}$ | Bone | 0.3 | *7.1 × 10⁻²* | $8.5 \times 10^{-2}$ | 0.25 | $9.1 \times 10^{-7}$ | 11 |
| 84 | $Po^{210}$ | Spleen | $2 \times 10^{-2}$ | *0.12* | 0.72 | 6.9 | $2.7 \times 10^{-8}$ | 8.3 |
| 85 | $At^{211}$ | Thyroid | $7 \times 10^{-2}$ | *5.3 × 10⁻²* | 2.8 | 26 | $2.6 \times 10^{-11}$ | 19 |
| 88 | $Ra^{226} + 55\%$ dr | Bone | $7.5 \times 10^{-2}$ | 0.49 | 0.49 | *0.11* | $1.1 \times 10^{-4}$ | $9.0 \times 10^{-2}$ |
| 89 | $Ac^{227}$ + dr | Bone | 0.3 | *1.2 × 10⁻²* | $1.3 \times 10^{-2}$ | $2.8 \times 10^{-2}$ | $1.7 \times 10^{-7}$ | 60 |
| 90 | $Th^{nat}$ | Bone | 0.78 | $4.5 \times 10^{-3}$ | $4.0 \times 10^{-3}$ | *6.7 × 10⁻⁴* | 5.9 | $1.7 \times 10^{-6}$ |
| 90 | $Th^{234} + Pa^{234}$ | Bone | 0.78 | *1.9* | 18 | $1.8 \times 10^{2}$ | $8.1 \times 10^{-8}$ | 0.53 |
| 92 | $U^{nat}$ | Kidneys | 0.33 | *8.7 × 10⁻³* | $6.8 \times 10^{-2}$ | 0.65 | 26 | $3.9 \times 10^{-7}$ |
| 92 | $U^{233}$ | Bone | 0.33 | *7.1 × 10⁻²* | 0.11 | 0.57 | $7.4 \times 10^{-3}$ | $1.3 \times 10^{-3}$ |
| 94 | $Pu^{239}$ | Bone | 0.7 | $3.5 \times 10^{-2}$ | $3.1 \times 10^{-2}$ | *5.2 × 10⁻³* | $8.3 \times 10^{-5}$ | 0.12 |
| 95 | $Am^{241}$ | Bone | 0.25 | *8.4 × 10⁻²* | $9.6 \times 10^{-2}$ | 0.23 | $2.8 \times 10^{-5}$ | 0.36 |
| 96 | $Cm^{242}$ | Bone | 0.25 | *7.6 × 10⁻²* | 0.17 | 0.14 | $2.3 \times 10^{-8}$ | 13 |

‡ Values of *H* in this column correspond to the smallest value in columns 5, 6 or 7.

**Table III. Single Exposure Values for Injection (or Puncture Wounds) when Insoluble Radioactive Material Is Deposited in the Wound**

| | (1) | (2) | (3) | (4) | (5) | (6) |
|---|---|---|---|---|---|---|
| | | *MPI or μc injected that will result in a dose of:* | | | | |
| *Z* | *Isotope* | *0.3 rem in 1 wk* | *15.7 rem in 1 yr* | *150 rem in 70 yrs* | *mg corresponding to smallest value in columns 2, 3, or 4* | *Relative hazard H* |
| 1 | $H^3$ (HTO or $H^3_2O$) | $1.4 \times 10^{-4}$ | $3.3 \times 10^{-4}$ | $2.9 \times 10^{-3}$ | $1.4 \times 10^{-11}$ | $7.1 \times 10^3$ |
| 4 | $Be^7$ | $8.1 \times 10^{-4}$ | $5.0 \times 10^{-3}$ | $5.0 \times 10^{-2}$ | $2.3 \times 10^{-12}$ | $1.2 \times 10^3$ |
| 6 | $C^{14}$ ($CO_2$) | $1.6 \times 10^{-5}$ | $3.6 \times 10^{-5}$ | $3.2 \times 10^{-4}$ | $3.5 \times 10^{-9}$ | $2.9 \times 10^3$ |
| 9 | $F^{18}$ | $2.2 \times 10^{-4}$ | $1.1 \times 10^{-2}$ | 0.11 | $2.3 \times 10^{-15}$ | $4.5 \times 10^3$ |
| 11 | $Na^{24}$ | $9.9 \times 10^{-6}$ | $4.9 \times 10^{-4}$ | $4.9 \times 10^{-3}$ | $1.1 \times 10^{-15}$ | $10^5$ |
| 15 | $P^{32}$ | $1.5 \times 10^{-6}$ | $2.3 \times 10^{-5}$ | $2.3 \times 10^{-4}$ | $5.2 \times 10^{-15}$ | $6.7 \times 10^5$ |
| 16 | $S^{35}$ | $1.6 \times 10^{-5}$ | $7.4 \times 10^{-5}$ | $7.4 \times 10^{-4}$ | $3.7 \times 10^{-13}$ | $6.3 \times 10^4$ |
| 17 | $Cl^{36}$ | $3.3 \times 10^{-6}$ | $7.4 \times 10^{-6}$ | $6.5 \times 10^{-5}$ | $1.4 \times 10^{-7}$ | $7.1 \times 10^1$ |
| 19 | $K^{42}$ | $5.6 \times 10^{-6}$ | $2.8 \times 10^{-4}$ | $2.8 \times 10^{-3}$ | $9.3 \times 10^{-16}$ | $1.8 \times 10^5$ |
| 20 | $Ca^{45}$ | $10^{-5}$ | $3.5 \times 10^{-5}$ | $3.4 \times 10^{-4}$ | $5.6 \times 10^{-13}$ | $10^5$ |
| 21 | $Sc^{46}$ | $6.3 \times 10^{-6}$ | $2.9 \times 10^{-5}$ | $2.9 \times 10^{-4}$ | $1.9 \times 10^{-13}$ | $1.6 \times 10^5$ |
| 21 | $Sc^{47}$ | $7.5 \times 10^{-6}$ | $2.9 \times 10^{-4}$ | $2.9 \times 10^{-3}$ | $9.2 \times 10^{-15}$ | $1.3 \times 10^5$ |
| 21 | $Sc^{48}$ | $10^{-5}$ | $4.9 \times 10^{-4}$ | $4.9 \times 10^{-3}$ | $6.7 \times 10^{-15}$ | $10^5$ |
| 23 | $V^{48}$ | $6.6 \times 10^{-6}$ | $9.6 \times 10^{-5}$ | $9.6 \times 10^{-4}$ | $3.9 \times 10^{-14}$ | $1.5 \times 10^5$ |
| 24 | $Cr^{51}$ | $4.6 \times 10^{-4}$ | $4.4 \times 10^{-3}$ | $4.4 \times 10^{-2}$ | $5.0 \times 10^{-12}$ | $2.2 \times 10^3$ |
| 25 | $Mn^{56}$ | $4.4 \times 10^{-5}$ | $2.2 \times 10^{-3}$ | $2.2 \times 10^{-2}$ | $2.0 \times 10^{-15}$ | $2.3 \times 10^4$ |
| 26 | $Fe^{55}$ | $1.4 \times 10^{-4}$ | $3.5 \times 10^{-4}$ | $3.1 \times 10^{-3}$ | $6.4 \times 10^{-11}$ | $7.1 \times 10^3$ |
| 26 | $Fe^{59}$ | $7.5 \times 10^{-6}$ | $5.1 \times 10^{-5}$ | $5.1 \times 10^{-4}$ | $1.7 \times 10^{-13}$ | $1.3 \times 10^5$ |
| 27 | $Co^{60}$ | $7.8 \times 10^{-6}$ | $1.8 \times 10^{-5}$ | $1.6 \times 10^{-4}$ | $6.8 \times 10^{-12}$ | $1.3 \times 10^5$ |
| 28 | $Ni^{59}$ | $1.7 \times 10^{-5}$ | $3.8 \times 10^{-5}$ | $3.4 \times 10^{-4}$ | $2.2 \times 10^{-7}$ | $4.5 \times 10^1$ |
| 29 | $Cu^{64}$ | $7.5 \times 10^{-5}$ | $3.8 \times 10^{-3}$ | $3.8 \times 10^{-2}$ | $2.0 \times 10^{-14}$ | $1.3 \times 10^4$ |
| 30 | $Zn^{65}$ | $6.6 \times 10^{-5}$ | $2.0 \times 10^{-4}$ | $1.9 \times 10^{-3}$ | $8.0 \times 10^{-12}$ | $1.5 \times 10^4$ |
| 31 | $Ga^{72}$ | $1.5 \times 10^{-5}$ | $7.6 \times 10^{-4}$ | $7.6 \times 10^{-3}$ | $4.8 \times 10^{-15}$ | $6.7 \times 10^4$ |
| 32 | $Ge^{71}$ | $1.1 \times 10^{-4}$ | $2.0 \times 10^{-3}$ | $2.0 \times 10^{-2}$ | $6.8 \times 10^{-13}$ | $9.1 \times 10^3$ |
| 33 | $As^{76}$ | $3.6 \times 10^{-6}$ | $1.8 \times 10^{-4}$ | $1.8 \times 10^{-3}$ | $2.3 \times 10^{-15}$ | $2.8 \times 10^5$ |
| 37 | $Rb^{86}$ | $1.6 \times 10^{-6}$ | $2.0 \times 10^{-5}$ | $2.0 \times 10^{-4}$ | $2.0 \times 10^{-14}$ | $6.3 \times 10^5$ |
| 38 | $Sr^{89}$ | $1.6 \times 10^{-6}$ | $10^{-5}$ | $9.9 \times 10^{-5}$ | $5.7 \times 10^{-14}$ | $6.3 \times 10^5$ |
| 38 | $Sr^{90} + Y^{90}$ | $8.5 \times 10^{-7}$ | $1.9 \times 10^{-6}$ | $1.7 \times 10^{-5}$ | $4.3 \times 10^{-12}$ | $1.2 \times 10^6$ |
| 39 | $Y^{91}$ | $1.6 \times 10^{-6}$ | $9.1 \times 10^{-6}$ | $9.1 \times 10^{-5}$ | $6.4 \times 10^{-14}$ | $6.3 \times 10^5$ |
| 40 | $Zr^{95} + Nb^{95}$ | $4.9 \times 10^{-6}$ | $2.7 \times 10^{-5}$ | $2.7 \times 10^{-4}$ | $1.2 \times 10^{-13}$ | $2.0 \times 10^5$ |
| 41 | $Nb^{95}$ | $1.9 \times 10^{-5}$ | $1.6 \times 10^{-4}$ | $1.6 \times 10^{-3}$ | $8.9 \times 10^{-13}$ | $5.3 \times 10^4$ |
| 42 | $Mo^{99}$ | $1.4 \times 10^{-5}$ | $5.6 \times 10^{-4}$ | $5.6 \times 10^{-3}$ | $3.0 \times 10^{-14}$ | $7.1 \times 10^4$ |
| 43 | $Tc^{96}$ | $5.5 \times 10^{-5}$ | $1.9 \times 10^{-3}$ | $1.9 \times 10^{-2}$ | $1.7 \times 10^{-13}$ | $1.8 \times 10^4$ |
| 44 | $Ru^{106} + Rh^{106}$ | $6.3 \times 10^{-7}$ | $1.8 \times 10^{-6}$ | $1.7 \times 10^{-5}$ | $1.9 \times 10^{-13}$ | $1.6 \times 10^6$ |
| 45 | $Rh^{105}$ | $10^{-5}$ | $5.0 \times 10^{-4}$ | $5.0 \times 10^{-3}$ | $1.2 \times 10^{-14}$ | $10^5$ |
| 46 | $Pd^{103} + Rh^{103*}$ | $2.7 \times 10^{-5}$ | $3.8 \times 10^{-4}$ | $3.8 \times 10^{-3}$ | $3.6 \times 10^{-13}$ | $3.8 \times 10^4$ |
| 47 | $Ag^{105}$ | $9.0 \times 10^{-5}$ | $6.1 \times 10^{-4}$ | $6.1 \times 10^{-3}$ | $3.2 \times 10^{-12}$ | $1.1 \times 10^4$ |
| 47 | $Ag^{111}$ | $3.1 \times 10^{-6}$ | $7.7 \times 10^{-5}$ | $7.7 \times 10^{-4}$ | $2.0 \times 10^{-14}$ | $3.2 \times 10^5$ |
| 48 | $Cd^{109} + Ag^{109*}$ | $2.1 \times 10^{-4}$ | $5.7 \times 10^{-4}$ | $5.3 \times 10^{-3}$ | $8.2 \times 10^{-11}$ | $4.8 \times 10^3$ |
| 50 | $Sn^{113}$ | $4.4 \times 10^{-4}$ | $1.8 \times 10^{-3}$ | $1.7 \times 10^{-2}$ | $4.2 \times 10^{-11}$ | $2.3 \times 10^3$ |

* The daughter products in these cases are isomers in an excited state.

**Table III. Single Exposure Values for Injection (or Puncture Wounds) when Insoluble Radioactive Material Is Deposited in the Wound — *Continued***

| | (1) | (2) | (3) | (4) | (5) | (6) |
|---|---|---|---|---|---|---|
| | | *μc injected that will result in a dose of:* | | | | |
| *Z* | *Isotope* | *0.3 rem in 1 wk* | *15.7 rem in 1 yr* | *150 rem in 70 yrs* | *mg corresponding to smallest value in columns 2, 3, or 4* | *Relative hazard H* |
| 52 | $Te^{127}$ | $3.2 \times 10^{-6}$ | $1.3 \times 10^{-5}$ | $1.3 \times 10^{-4}$ | $3.6 \times 10^{-13}$ | $3.1 \times 10^{8}$ |
| 52 | $Te^{129}$ | $1.4 \times 10^{-6}$ | $1.2 \times 10^{-5}$ | $1.2 \times 10^{-4}$ | $4.6 \times 10^{-14}$ | $7.1 \times 10^{8}$ |
| 53 | $I^{131}$ | $6.3 \times 10^{-6}$ | $1.5 \times 10^{-4}$ | $1.5 \times 10^{-3}$ | $5.0 \times 10^{-14}$ | $1.6 \times 10^{8}$ |
| 55 | $Cs^{137} + Ba^{137}$ | $4.7 \times 10^{-6}$ | $1.1 \times 10^{-5}$ | $9.5 \times 10^{-5}$ | $5.9 \times 10^{-11}$ | $2.1 \times 10^{5}$ |
| 56 | $Ba^{140} + La^{140}$ | $1.3 \times 10^{-6}$ | $2.1 \times 10^{-5}$ | $2.1 \times 10^{-4}$ | $1.8 \times 10^{-14}$ | $7.7 \times 10^{8}$ |
| 57 | $La^{140}$ | $4.8 \times 10^{-6}$ | $2.2 \times 10^{-4}$ | $2.2 \times 10^{-3}$ | $8.5 \times 10^{-15}$ | $2.1 \times 10^{8}$ |
| 58 | $Ce^{144} + Pr^{144}$ | $6.9 \times 10^{-7}$ | $2.0 \times 10^{-6}$ | $1.9 \times 10^{-5}$ | $2.2 \times 10^{-13}$ | $1.4 \times 10^{6}$ |
| 59 | $Pr^{143}$ | $3.3 \times 10^{-6}$ | $5.4 \times 10^{-5}$ | $5.4 \times 10^{-4}$ | $4.9 \times 10^{-14}$ | $3.0 \times 10^{8}$ |
| 61 | $Pm^{147}$ | $1.3 \times 10^{-5}$ | $3.1 \times 10^{-5}$ | $2.8 \times 10^{-4}$ | $1.4 \times 10^{-11}$ | $7.7 \times 10^{4}$ |
| 62 | $Sm^{151}$ | $4.3 \times 10^{-5}$ | $9.6 \times 10^{-5}$ | $8.4 \times 10^{-4}$ | $1.8 \times 10^{-9}$ | $2.3 \times 10^{4}$ |
| 63 | $Eu^{154}$ | $4.1 \times 10^{-6}$ | $9.2 \times 10^{-6}$ | $8.2 \times 10^{-5}$ | $2.8 \times 10^{-11}$ | $2.4 \times 10^{5}$ |
| 67 | $Ho^{166}$ | $5.7 \times 10^{-6}$ | $2.8 \times 10^{-4}$ | $2.8 \times 10^{-3}$ | $8.2 \times 10^{-15}$ | $1.8 \times 10^{8}$ |
| 69 | $Tm^{170}$ | $2.7 \times 10^{-6}$ | $10^{-5}$ | $10^{-4}$ | $4.4 \times 10^{-13}$ | $3.7 \times 10^{5}$ |
| 71 | $Lu^{177}$ | $9.2 \times 10^{-6}$ | $2.4 \times 10^{-4}$ | $2.4 \times 10^{-3}$ | $8.4 \times 10^{-14}$ | $1.1 \times 10^{8}$ |
| 73 | $Ta^{182}$ | $5.1 \times 10^{-6}$ | $2.1 \times 10^{-5}$ | $2.1 \times 10^{-4}$ | $7.8 \times 10^{-13}$ | $2.0 \times 10^{5}$ |
| 74 | $W^{181}$ | $2.9 \times 10^{-5}$ | $1.1 \times 10^{-4}$ | $10^{-3}$ | $5.6 \times 10^{-12}$ | $3.4 \times 10^{4}$ |
| 75 | $Re^{183}$ | $2.9 \times 10^{-4}$ | $1.1 \times 10^{-3}$ | $1.1 \times 10^{-2}$ | $4.8 \times 10^{-11}$ | $3.4 \times 10^{3}$ |
| 77 | $Ir^{190}$ | $3.1 \times 10^{-5}$ | $6.1 \times 10^{-4}$ | $6.1 \times 10^{-3}$ | $4.8 \times 10^{-13}$ | $3.2 \times 10^{4}$ |
| 77 | $Ir^{192}$ | $4.0 \times 10^{-6}$ | $2.0 \times 10^{-5}$ | $2.0 \times 10^{-4}$ | $4.3 \times 10^{-13}$ | $2.5 \times 10^{5}$ |
| 78 | $Pt^{191}$ | $2.9 \times 10^{-4}$ | $1.2 \times 10^{-2}$ | $1.2 \times 10^{-1}$ | $1.3 \times 10^{-12}$ | $3.4 \times 10^{3}$ |
| 78 | $Pt^{193}$ | $2.8 \times 10^{-4}$ | $9.6 \times 10^{-3}$ | $9.6 \times 10^{-2}$ | $1.8 \times 10^{-12}$ | $3.6 \times 10^{3}$ |
| 79 | $Au^{196}$ | $2.6 \times 10^{-4}$ | $7.6 \times 10^{-3}$ | $7.6 \times 10^{-2}$ | $2.1 \times 10^{-13}$ | $3.8 \times 10^{3}$ |
| 79 | $Au^{198}$ | $5.8 \times 10^{-6}$ | $2.4 \times 10^{-4}$ | $2.4 \times 10^{-3}$ | $2.3 \times 10^{-14}$ | $1.7 \times 10^{5}$ |
| 79 | $Au^{199}$ | $1.6 \times 10^{-5}$ | $6.3 \times 10^{-4}$ | $6.3 \times 10^{-3}$ | $8.0 \times 10^{-14}$ | $6.3 \times 10^{4}$ |
| 81 | $Tl^{200}$ | $3.3 \times 10^{-5}$ | $1.6 \times 10^{-3}$ | $1.6 \times 10^{-2}$ | $5.7 \times 10^{-14}$ | $3.0 \times 10^{4}$ |
| 81 | $Tl^{201}$ | $2.9 \times 10^{-3}$ | $1.2 \times 10^{-1}$ | 1.2 | $1.3 \times 10^{-11}$ | $3.4 \times 10^{2}$ |
| 81 | $Tl^{202}$ | $1.0 \times 10^{-4}$ | $1.8 \times 10^{-3}$ | $1.8 \times 10^{-2}$ | $1.8 \times 10^{-12}$ | $10^{4}$ |
| 81 | $Tl^{204}$ | $3.4 \times 10^{-6}$ | $8.1 \times 10^{-6}$ | $7.3 \times 10^{-5}$ | $7.9 \times 10^{-12}$ | $2.9 \times 10^{5}$ |
| 82 | $Pb^{203}$ | $2.1 \times 10^{-4}$ | $9.5 \times 10^{-3}$ | $9.5 \times 10^{-2}$ | $7.0 \times 10^{-13}$ | $4.8 \times 10^{3}$ |
| 82 | $Pb^{210}$ + dr | $1.5 \times 10^{-8}$ | $3.5 \times 10^{-8}$ | $3.1 \times 10^{-7}$ | $1.9 \times 10^{-13}$ | $6.7 \times 10^{7}$ |
| 84 | $Po^{210}$ | $1.6 \times 10^{-8}$ | $5.9 \times 10^{-8}$ | $5.8 \times 10^{-7}$ | $3.5 \times 10^{-15}$ | $6.3 \times 10^{7}$ |
| 85 | $At^{211}$ | $1.8 \times 10^{-7}$ | $9.2 \times 10^{-6}$ | $9.2 \times 10^{-5}$ | $8.9 \times 10^{-17}$ | $5.6 \times 10^{6}$ |
| 88 | $Ra^{226}$ + dr | $5.3 \times 10^{-9}$ | $1.2 \times 10^{-8}$ | $1.0 \times 10^{-7}$ | $5.4 \times 10^{-12}$ | $1.9 \times 10^{6}$ |
| 89 | $Ac^{227}$ + dr | $2.5 \times 10^{-9}$ | $5.8 \times 10^{-9}$ | $5.1 \times 10^{-8}$ | $3.5 \times 10^{-14}$ | $4.0 \times 10^{8}$ |
| 90 | $Th^{nat}$ | $2.3 \times 10^{-9}$ | $5.1 \times 10^{-9}$ | $4.5 \times 10^{-8}$ | $2.0 \times 10^{-5}$ | $5.0 \times 10^{-1}$ |
| 90 | $Th^{234} + Pa^{234}$ | $1.1 \times 10^{-6}$ | $1.2 \times 10^{-5}$ | $1.2 \times 10^{-4}$ | $4.7 \times 10^{-14}$ | $9.1 \times 10^{5}$ |
| 92 | $U^{nat}$ | $9.1 \times 10^{-9}$ | $2.0 \times 10^{-8}$ | $1.8 \times 10^{-7}$ | $2.7 \times 10^{-5}$ | $3.7 \times 10^{-1}$ |
| 92 | $U^{238}$ | $1.7 \times 10^{-8}$ | $3.8 \times 10^{-8}$ | $3.4 \times 10^{-7}$ | $1.8 \times 10^{-9}$ | $5.9 \times 10^{3}$ |
| 94 | $Pu^{239}$ | $1.6 \times 10^{-8}$ | $3.6 \times 10^{-8}$ | $3.2 \times 10^{-7}$ | $2.6 \times 10^{-10}$ | $3.8 \times 10^{4}$ |
| 95 | $Am^{241}$ | $1.5 \times 10^{-8}$ | $3.4 \times 10^{-8}$ | $3.0 \times 10^{-7}$ | $4.9 \times 10^{-12}$ | $2.0 \times 10^{6}$ |
| 96 | $Cm^{242}$ | $1.4 \times 10^{-8}$ | $4.8 \times 10^{-8}$ | $4.7 \times 10^{-7}$ | $4.2 \times 10^{-15}$ | $7.1 \times 10^{7}$ |

**Table IV. Single Exposure Values for Inhalation of Insoluble Radioactive Materials when the Lung Is the Critical Body Organ**

| | (1) | (2) | (3) | (4) | (5) | (6) |
|---|---|---|---|---|---|---|
| | | *MPI† or μc inhaled in 8 hours that will result in a dose of:* | | | | |
| *Z* | *Isotope* | *0.3 rem in 1 wk* | *15.7 rem in 1 yr* | *150 rem in 70 yrs* | *mg corresponding to smallest value in columns 2, 3, or 4* | *Relative hazard H* |
| 1 | $H^3$ (HTO or $H^3_2O$) | $1.1 \times 10^3$ | $2.7 \times 10^3$ | $2.3 \times 10^4$ | $1.2 \times 10^{-4}$ | $9.0 \times 10^{-4}$ |
| 4 | $Be^7$ | $1.9 \times 10^2$ | $1.2 \times 10^3$ | $1.2 \times 10^4$ | $5.7 \times 10^{-7}$ | $5.1 \times 10^{-3}$ |
| 6 | $C^{14}$ ($CO_2$) | $1.3 \times 10^2$ | $3.0 \times 10^2$ | $2.6 \times 10^3$ | $2.9 \times 10^{-2}$ | $3.6 \times 10^{-3}$ |
| 9 | $F^{18}$ | $5.3 \times 10^3$ | $2.8 \times 10^5$ | $2.7 \times 10^6$ | $5.7 \times 10^{-8}$ | $1.9 \times 10^{-4}$ |
| 11 | $Na^{24}$ | 23 | $1.2 \times 10^3$ | $1.1 \times 10^4$ | $2.6 \times 10^{-9}$ | $4.5 \times 10^{-2}$ |
| 15 | $P^{32}$ | 12 | $1.9 \times 10^2$ | $1.8 \times 10^3$ | $4.3 \times 10^{-8}$ | $8.4 \times 10^{-2}$ |
| 16 | $S^{35}$ | $1.3 \times 10^2$ | $6.3 \times 10^2$ | $6.0 \times 10^3$ | $2.9 \times 10^{-6}$ | $7.8 \times 10^{-3}$ |
| 17 | $Cl^{36}$ | 26 | 63 | $5.3 \times 10^2$ | 1.1 | $9.3 \times 10^{-5}$ |
| 19 | $K^{42}$ | 50 | $2.5 \times 10^3$ | $2.4 \times 10^4$ | $8.3 \times 10^{-9}$ | $2.0 \times 10^{-2}$ |
| 20 | $Ca^{45}$ | 80 | $3.0 \times 10^2$ | $2.8 \times 10^3$ | $4.3 \times 10^{-6}$ | $1.3 \times 10^{-2}$ |
| 21 | $Sc^{46}$ | 5.7 | 26 | $2.5 \times 10^2$ | $1.7 \times 10^{-7}$ | 0.18 |
| 21 | $Sc^{47}$ | 63 | $2.5 \times 10^3$ | $2.4 \times 10^4$ | $7.7 \times 10^{-8}$ | $1.6 \times 10^{-2}$ |
| 21 | $Sc^{48}$ | 13 | $6.3 \times 10^2$ | $3.7 \times 10^3$ | $8.7 \times 10^{-9}$ | $7.8 \times 10^{-2}$ |
| 23 | $V^{48}$ | 4.7 | 67 | $6.7 \times 10^2$ | $2.7 \times 10^{-8}$ | 0.21 |
| 24 | $Cr^{51}$ | $6.7 \times 10^2$ | $6.7 \times 10^3$ | $6.3 \times 10^4$ | $7.3 \times 10^{-6}$ | $1.5 \times 10^{-3}$ |
| 25 | $Mn^{56}$ | $4 \times 10^2$ | $2.1 \times 10^4$ | $2.0 \times 10^5$ | $1.8 \times 10^{-8}$ | $2.5 \times 10^{-3}$ |
| 26 | $Fe^{55}$ | $1.1 \times 10^3$ | $2.9 \times 10^3$ | $2.5 \times 10^4$ | $5.3 \times 10^{-4}$ | $8.7 \times 10^{-4}$ |
| 26 | $Fe^{59}$ | 9 | 63 | $6.0 \times 10^2$ | $1.8 \times 10^{-7}$ | 0.11 |
| 27 | $Co^{60}$ | 4.7 | 11 | 97 | $4.0 \times 10^{-6}$ | 0.21 |
| 28 | $Ni^{59}$ | $1.4 \times 10^2$ | $3.2 \times 10^2$ | $2.7 \times 10^3$ | 1.8 | $5.7 \times 10^{-5}$ |
| 29 | $Cu^{64}$ | $3.7 \times 10^2$ | $1.8 \times 10^4$ | $1.8 \times 10^5$ | $9.7 \times 10^{-8}$ | $2.7 \times 10^{-3}$ |
| 30 | $Zn^{65}$ | 21 | 67 | $6.0 \times 10^2$ | $2.5 \times 10^{-6}$ | $4.8 \times 10^{-2}$ |
| 31 | $Ga^{72}$ | 33 | $1.7 \times 10^3$ | $1.7 \times 10^4$ | $1.1 \times 10^{-8}$ | $3.0 \times 10^{-2}$ |
| 32 | $Ge^{71}$ | $8.7 \times 10^2$ | $1.7 \times 10^4$ | $1.6 \times 10^5$ | $5.3 \times 10^{-6}$ | $1.1 \times 10^{-3}$ |
| 33 | $As^{76}$ | 25 | $1.3 \times 10^3$ | $1.3 \times 10^4$ | $1.6 \times 10^{-8}$ | $3.9 \times 10^{-2}$ |
| 37 | $Rb^{86}$ | 11 | $1.4 \times 10^2$ | $1.3 \times 10^3$ | $1.4 \times 10^{-7}$ | $9.3 \times 10^{-2}$ |
| 38 | $Sr^{89}$ | 13 | 83 | $8.0 \times 10^2$ | $4.7 \times 10^{-7}$ | $7.8 \times 10^{-2}$ |
| 38 | $Sr^{90} + Y^{90}$ | 7 | 16 | $1.4 \times 10^2$ | $3.3 \times 10^{-5}$ | 0.14 |
| 39 | $Y^{91}$ | 13 | 77 | $7.3 \times 10^2$ | $5.0 \times 10^{-7}$ | $7.8 \times 10^{-2}$ |
| 40 | $Zr^{95} + Nb^{95}$ | 7.3 | 43 | $4.0 \times 10^2$ | $3.3 \times 10^{-7}$ | 0.14 |
| 41 | $Nb^{95}$ | 14 | $1.2 \times 10^2$ | $1.2 \times 10^3$ | $3.7 \times 10^{-7}$ | $6.9 \times 10^{-2}$ |
| 42 | $Mo^{99}$ | 28 | $1.2 \times 10^3$ | $1.2 \times 10^4$ | $6.0 \times 10^{-8}$ | $3.6 \times 10^{-2}$ |
| 43 | $Tc^{96}$ | 8 | $2.8 \times 10^2$ | $2.7 \times 10^3$ | $2.5 \times 10^{-8}$ | 0.13 |
| 44 | $Ru^{106} + Rh^{106}$ | 4.7 | 13 | $1.2 \times 10^2$ | $1.4 \times 10^{-6}$ | 0.21 |
| 45 | $Rh^{105}$ | 53 | $2.6 \times 10^3$ | $2.5 \times 10^4$ | $6.3 \times 10^{-8}$ | $1.9 \times 10^{-2}$ |
| 46 | $Pd^{103} + Rh^{103*}$ | $1.1 \times 10^2$ | $1.5 \times 10^3$ | $1.5 \times 10^4$ | $1.4 \times 10^{-6}$ | $9.3 \times 10^{-3}$ |
| 47 | $Ag^{105}$ | 4.3 | 30 | $2.9 \times 10^2$ | $1.6 \times 10^{-7}$ | 0.23 |
| 47 | $Ag^{111}$ | 26 | $6.7 \times 10^2$ | $6.3 \times 10^3$ | $1.7 \times 10^{-7}$ | $3.9 \times 10^{-2}$ |
| 48 | $Cd^{109} + Ag^{109*}$ | 97 | $2.7 \times 10^2$ | $2.4 \times 10^3$ | $4.0 \times 10^{-5}$ | $1.0 \times 10^{-2}$ |

* The daughter products in these cases are isomers in an excited state.

† The MPI as used here is the μc present initially in a volume of air to be breathed during the succeeding 8 hours and which then results in the indicated dose.

Table IV. Single Exposure Values for Inhalation of Insoluble Radioactive Materials when the Lung Is the Critical Body Organ — *Continued*

| | (1) | (2) | (3) | (4) | (5) | (6) |
|---|---|---|---|---|---|---|
| | | *MPI† or μc inhaled in 8 hours that will result in a dose of:* | | | | |
| *Z* | *Isotope* | *0.3 rem in 1 wk* | *15.7 rem in 1 yr* | *150 rem in 70 yrs* | *mg corresponding to smallest value in columns 2, 3, or 4* | *Relative hazard H* |
| 50 | $Sn^{113}$ | 23 | 97 | $9.0 \times 10^{2}$ | $2.2 \times 10^{-6}$ | $4.5 \times 10^{-2}$ |
| 52 | $Te^{127}$ | 22 | 90 | $8.7 \times 10^{2}$ | $2.4 \times 10^{-6}$ | $4.5 \times 10^{-2}$ |
| 52 | $Te^{129}$ | 5.3 | 47 | $4.3 \times 10^{2}$ | $1.8 \times 10^{-7}$ | 0.19 |
| 53 | $I^{131}$ | 22 | $5.3 \times 10^{2}$ | $5.0 \times 10^{3}$ | $1.7 \times 10^{-7}$ | $4.5 \times 10^{-2}$ |
| 55 | $Cs^{137} + Ba^{137*}$ | 12 | 29 | $2.4 \times 10^{2}$ | $1.5 \times 10^{-4}$ | $8.4 \times 10^{-2}$ |
| 56 | $Ba^{140} + La^{140}$ | 4.3 | 77 | $7.3 \times 10^{2}$ | $6.0 \times 10^{-8}$ | 0.23 |
| 57 | $La^{140}$ | 15 | $7.0 \times 10^{2}$ | $6.7 \times 10^{3}$ | $2.6 \times 10^{-8}$ | $6.9 \times 10^{-2}$ |
| 58 | $Ce^{144} + Pr^{144}$ | 5 | 15 | $1.4 \times 10^{2}$ | $1.6 \times 10^{-6}$ | 0.20 |
| 59 | $Pr^{143}$ | 27 | $4.7 \times 10^{2}$ | $4.3 \times 10^{3}$ | $4.0 \times 10^{-7}$ | $3.6 \times 10^{-2}$ |
| 61 | $Pm^{147}$ | $1.0 \times 10^{2}$ | $2.6 \times 10^{2}$ | $2.3 \times 10^{3}$ | $1.1 \times 10^{-4}$ | $9.6 \times 10^{-3}$ |
| 62 | $Sm^{151}$ | $3.3 \times 10^{2}$ | $8.0 \times 10^{2}$ | $6.7 \times 10^{3}$ | $1.4 \times 10^{-2}$ | $3 \times 10^{-3}$ |
| 63 | $Eu^{154}$ | 8.3 | 20 | $1.7 \times 10^{2}$ | $5.7 \times 10^{-5}$ | 0.12 |
| 67 | $Ho^{166}$ | 47 | $2.4 \times 10^{3}$ | $2.3 \times 10^{4}$ | $6.7 \times 10^{-8}$ | $2.1 \times 10^{-2}$ |
| 69 | $Tm^{170}$ | 22 | 87 | $8.3 \times 10^{2}$ | $3.7 \times 10^{-6}$ | $4.5 \times 10^{-2}$ |
| 71 | $Lu^{177}$ | 57 | $1.6 \times 10^{3}$ | $1.5 \times 10^{4}$ | $5.3 \times 10^{-7}$ | $1.8 \times 10^{-2}$ |
| 73 | $Ta^{182}$ | 8.7 | 37 | $3.3 \times 10^{2}$ | $1.3 \times 10^{-6}$ | 0.11 |
| 74 | $W^{181}$ | 6.3 | 25 | $2.3 \times 10^{2}$ | $1.2 \times 10^{-6}$ | 0.16 |
| 75 | $Re^{183}$ | 11 | 47 | $4.3 \times 10^{2}$ | $1.8 \times 10^{-6}$ | $9.0 \times 10^{-2}$ |
| 77 | $Ir^{190}$ | 43 | $8.7 \times 10^{2}$ | $8.0 \times 10^{3}$ | $6.7 \times 10^{-7}$ | $2.3 \times 10^{-2}$ |
| 77 | $Ir^{192}$ | 7.3 | 37 | $3.7 \times 10^{2}$ | $8.0 \times 10^{-7}$ | 0.14 |
| 78 | $Pt^{191}$ | 12 | $5.0 \times 10^{2}$ | $4.7 \times 10^{3}$ | $5.3 \times 10^{-8}$ | $8.4 \times 10^{-2}$ |
| 78 | $Pt^{193}$ | 12 | $4.0 \times 10^{2}$ | $4.0 \times 10^{3}$ | $7.3 \times 10^{-8}$ | $8.7 \times 10^{-2}$ |
| 79 | $Au^{196}$ | 28 | $8.7 \times 10^{2}$ | $8.3 \times 10^{3}$ | $2.3 \times 10^{-7}$ | $3.6 \times 10^{-2}$ |
| 79 | $Au^{198}$ | 27 | $1.2 \times 10^{3}$ | $1.1 \times 10^{4}$ | $1.1 \times 10^{-7}$ | $3.6 \times 10^{-2}$ |
| 79 | $Au^{199}$ | 63 | $2.5 \times 10^{3}$ | $2.4 \times 10^{4}$ | $3.2 \times 10^{-7}$ | $1.7 \times 10^{-2}$ |
| 81 | $Tl^{200}$ | 26 | $1.4 \times 10^{3}$ | $1.3 \times 10^{4}$ | $4.7 \times 10^{-8}$ | $3.9 \times 10^{-2}$ |
| 81 | $Tl^{201}$ | 90 | $3.7 \times 10^{3}$ | $3.7 \times 10^{4}$ | $4.0 \times 10^{-7}$ | $1.1 \times 10^{-2}$ |
| 81 | $Tl^{202}$ | 40 | $7.3 \times 10^{2}$ | $7.0 \times 10^{3}$ | $7.3 \times 10^{-7}$ | $2.5 \times 10^{-2}$ |
| 81 | $Tl^{204}$ | 27 | 67 | $6.0 \times 10^{2}$ | $6.3 \times 10^{-5}$ | $3.6 \times 10^{-2}$ |
| 82 | $Pb^{203}$ | 37 | $1.7 \times 10^{3}$ | $1.6 \times 10^{4}$ | $1.2 \times 10^{-7}$ | $2.7 \times 10^{-2}$ |
| 82 | $Pb^{210}$ | 0.12 | 0.29 | 2.5 | $1.6 \times 10^{-6}$ | 8.1 |
| 84 | $Po^{210}$ | 0.12 | 0.47 | 4.7 | $2.7 \times 10^{-8}$ | 8.1 |
| 85 | $At^{211}$ | 3.1 | $1.1 \times 10^{2}$ | $1.0 \times 10^{3}$ | $1.0 \times 10^{-9}$ | 0.48 |
| 88 | $Ra^{226} + dr$ | $2.8 \times 10^{-2}$ | $6.3 \times 10^{-2}$ | 0.53 | $2.8 \times 10^{-5}$ | 3.6 |
| 89 | $Ac^{227} + dr$ | $2.1 \times 10^{-2}$ | $4.7 \times 10^{-2}$ | 0.40 | $2.9 \times 10^{-7}$ | 48 |
| 90 | $Th^{nat}$ | $1.9 \times 10^{-2}$ | $4.3 \times 10^{-2}$ | 0.37 | $1.6 \times 10^{2}$ | $6.3 \times 10^{-7}$ |
| 90 | $Th^{234} + Pa^{234}$ | $8.3 \times 10^{-2}$ | 97 | $9.0 \times 10^{2}$ | $3.7 \times 10^{-9}$ | 0.12 |
| 92 | $U^{nat}$ | $7.3 \times 10^{-2}$ | 0.17 | 1.4 | $2.2 \times 10^{2}$ | $4.5 \times 10^{-7}$ |
| 92 | $U^{233}$ | 0.14 | 0.32 | 2.7 | $1.5 \times 10^{-2}$ | $6.9 \times 10^{-3}$ |
| 94 | $Pu^{239}$ | 0.13 | 0.30 | 2.5 | $2.1 \times 10^{-3}$ | $4.8 \times 10^{-2}$ |
| 95 | $Am^{241}$ | 0.12 | 0.29 | 2.4 | $4.0 \times 10^{-5}$ | 2.5 |
| 96 | $Cm^{242}$ | 0.11 | 0.40 | 3.7 | $3.2 \times 10^{-8}$ | $9.3 \times 10^{-2}$ |

## Table V. Single Exposure Values for Inhalation of Insoluble Radioactive Material when the GI Tract Is the Critical Body Organ

| (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) |
|---|---|---|---|---|---|---|---|---|
| | | | MPI† or μc inhaled in 8 hours that will deliver a dose of 0.3 rem during the following week to the organs cited: Equations 26, 28 | | | | | |
| Z | Isotope | mg/100 mc | Stomach | Small intestine | Upper large intestine | Lower large intestine | mg corresponding to smallest value in columns 4, 5, 6, or 7 | Relative hazard H |
| 1 | $H^3$ (HTO or $H^3_2O$) | $10^{-2}$ | $1.9 \times 10^4$ | $2.1 \times 10^4$ | $2.5 \times 10^4$ | *$1.2 \times 10^4$* | $1.2 \times 10^{-3}$ | $8.3 \times 10^{-5}$ |
| 4 | $Be^7$ | $2.9 \times 10^{-4}$ | $7.0 \times 10^3$ | $3.3 \times 10^3$ | $8.5 \times 10^2$ | *$4.2 \times 10^2$* | $1.2 \times 10^{-6}$ | $2.4 \times 10^{-3}$ |
| 6 | $C^{14}$ ($CO_2$) | 22 | $2.1 \times 10^3$ | $2.3 \times 10^3$ | $2.9 \times 10^3$ | *$1.4 \times 10^3$* | 0.31 | $3.2 \times 10^{-4}$ |
| 9 | $F^{18}$ | $1.1 \times 10^{-6}$ | *$1.7 \times 10^3$* | $2.3 \times 10^3$ | $1.1 \times 10^{15}$ | $10^{16}$ | $1.9 \times 10^{-8}$ | $5.8 \times 10^{-4}$ |
| 11 | $Na^{24}$ | $1.2 \times 10^{-5}$ | 92 | *57* | $8.7 \times 10^2$ | $6.2 \times 10^2$ | $6.6 \times 10^{-9}$ | $1.8 \times 10^{-2}$ |
| 15 | $P^{32}$ | $3.5 \times 10^{-4}$ | $1.7 \times 10^2$ | $1.8 \times 10^2$ | 38 | *19* | $6.6 \times 10^{-8}$ | $5.3 \times 10^{-2}$ |
| 16 | $S^{35}$ | $2.3 \times 10^{-3}$ | $2.0 \times 10^3$ | $2.2 \times 10^3$ | $3.1 \times 10^2$ | *$1.6 \times 10^2$* | $3.7 \times 10^{-6}$ | $6.3 \times 10^{-3}$ |
| 17 | $Cl^{36}$ | $4.1 \times 10^3$ | $4.3 \times 10^2$ | $4.8 \times 10^2$ | $5.8 \times 10^2$ | *$2.9 \times 10^2$* | 12 | $8.4 \times 10^{-6}$ |
| 19 | $K^{42}$ | $1.7 \times 10^{-5}$ | *98* | 102 | $1.1 \times 10^3$ | $8.5 \times 10^2$ | $1.6 \times 10^{-8}$ | $10^{-2}$ |
| 20 | $Ca^{45}$ | $5.6 \times 10^{-3}$ | $1.3 \times 10^3$ | $1.5 \times 10^3$ | $9.0 \times 10^2$ | *$4.4 \times 10^2$* | $2.5 \times 10^{-5}$ | $2.3 \times 10^{-3}$ |
| 21 | $Sc^{46}$ | $3.0 \times 10^{-3}$ | $2.2 \times 10^2$ | 95 | 19 | *9.2* | $2.7 \times 10^{-7}$ | 0.11 |
| 21 | $Sc^{47}$ | $1.2 \times 10^{-4}$ | $5.6 \times 10^2$ | $6.1 \times 10^2$ | 39 | *21* | $2.6 \times 10^{-8}$ | $4.8 \times 10^{-2}$ |
| 21 | $Sc^{48}$ | $6.7 \times 10^{-5}$ | $1.5 \times 10^2$ | 84 | 16 | *9.1* | $6.1 \times 10^{-9}$ | 0.11 |
| 23 | $V^{48}$ | $5.8 \times 10^{-4}$ | $1.3 \times 10^2$ | 69 | 14 | *7.1* | $4.1 \times 10^{-8}$ | 0.14 |
| 24 | $Cr^{51}$ | $1.1 \times 10^{-2}$ | $1.4 \times 10^4$ | $1.1 \times 10^4$ | $10^3$ | *$5.0 \times 10^2$* | $5.4 \times 10^{-6}$ | $2.0 \times 10^{-3}$ |
| 25 | $Mn^{56}$ | $4.6 \times 10^{-6}$ | $2.9 \times 10^2$ | $2.2 \times 10^2$ | *80* | $3.4 \times 10^2$ | $3.6 \times 10^{-9}$ | $1.2 \times 10^{-2}$ |
| 26 | $Fe^{55}$ | $4.6 \times 10^{-2}$ | $1.9 \times 10^4$ | $2.1 \times 10^4$ | $6.3 \times 10^3$ | *$3.1 \times 10^3$* | $1.4 \times 10^{-3}$ | $3.2 \times 10^{-4}$ |
| 26 | $Fe^{59}$ | $2.0 \times 10^{-3}$ | $2.7 \times 10^2$ | $1.5 \times 10^2$ | $1.4 \times 10^2$ | *68* | $1.4 \times 10^{-6}$ | $1.5 \times 10^{-2}$ |
| 27 | $Co^{60}$ | $8.7 \times 10^{-2}$ | $1.6 \times 10^2$ | 82 | 22 | *11* | $9.5 \times 10^{-6}$ | $9.1 \times 10^{-2}$ |
| 28 | $Ni^{59}$ | 1.3 | $2.2 \times 10^3$ | $2.5 \times 10^3$ | $1.9 \times 10^2$ | *94* | 1.2 | $8.2 \times 10^{-5}$ |
| 29 | $Cu^{64}$ | $2.6 \times 10^{-5}$ | $9.5 \times 10^2$ | $7.6 \times 10^2$ | $1.4 \times 10^2$ | *$1.1 \times 10^2$* | $2.8 \times 10^{-8}$ | $9.8 \times 10^{-3}$ |
| 30 | $Zn^{65}$ | $1.2 \times 10^{-2}$ | $7.5 \times 10^2$ | $3.7 \times 10^2$ | 99 | *49* | $5.9 \times 10^{-6}$ | $2.0 \times 10^{-2}$ |
| 31 | $Ga^{72}$ | $3.2 \times 10^{-5}$ | $1.2 \times 10^2$ | 79 | 15 | *11* | $3.5 \times 10^{-9}$ | $9.1 \times 10^{-2}$ |
| 32 | $Ge^{71}$ | $6.2 \times 10^{-4}$ | $1.1 \times 10^4$ | $1.2 \times 10^4$ | $7.8 \times 10^2$ | *$3.9 \times 10^2$* | $2.4 \times 10^{-6}$ | $2.6 \times 10^{-3}$ |
| 33 | $As^{76}$ | $6.4 \times 10^{-5}$ | $1.1 \times 10^2$ | $1.1 \times 10^2$ | 9.0 | *5.4* | $3.4 \times 10^{-9}$ | 0.18 |
| 37 | $Rb^{86}$ | $1.3 \times 10^{-3}$ | $1.7 \times 10^2$ | $1.7 \times 10^2$ | $2.5 \times 10^2$ | *$1.3 \times 10^2$* | $1.7 \times 10^{-6}$ | $7.7 \times 10^{-3}$ |
| 38 | $Sr^{89}$ | $3.6 \times 10^{-3}$ | $2.0 \times 10^2$ | $2.3 \times 10^2$ | 35 | *17* | $6.1 \times 10^{-7}$ | $5.9 \times 10^{-2}$ |
| 39 | $Y^{90}$ | $1.7 \times 10^{-4}$ | $1.3 \times 10^2$ | $1.5 \times 10^2$ | 9.6 | *5.2* | $9.1 \times 10^{-9}$ | 0.19 |
| 38 | $Sr^{90} + Y^{90}$ | 0.5 | $6.1 \times 10^2$ | $6.5 \times 10^2$ | 58 | *21* | $1.0 \times 10^{-4}$ | $4.8 \times 10^{-2}$ |
| 39 | $Y^{91}$ | $4.0 \times 10^{-3}$ | $2.0 \times 10^2$ | $2.2 \times 10^2$ | 13 | *6.6* | $2.6 \times 10^{-7}$ | 0.15 |
| 41 | $Nb^{95}$ | $2.5 \times 10^{-3}$ | $4.5 \times 10^2$ | $2.4 \times 10^2$ | 93 | *46* | $1.2 \times 10^{-6}$ | $2.2 \times 10^{-2}$ |
| 40 | $Zr^{95} + Nb^{95}$ | $4.7 \times 10^{-3}$ | $3.5 \times 10^2$ | $2.2 \times 10^2$ | 33 | *16* | $7.5 \times 10^{-7}$ | $6.2 \times 10^{-2}$ |
| 42 | $Mo^{99}$ | $2.2 \times 10^{-4}$ | $4.0 \times 10^2$ | $2.5 \times 10^2$ | $1.4 \times 10^2$ | *72* | $1.6 \times 10^{-7}$ | $1.3 \times 10^{-2}$ |
| 43 | $Tc^{96}$ | $3.1 \times 10^{-4}$ | $1.7 \times 10^2$ | 85 | 44 | *24* | $7.2 \times 10^{-8}$ | $4.2 \times 10^{-2}$ |
| 44 | $Ru^{106} + Rh^{106}$ | $2.9 \times 10^{-2}$ | 80 | 82 | 5.4 | *2.7* | $7.9 \times 10^{-7}$ | 0.37 |
| 45 | $Rh^{105}$ | $1.2 \times 10^{-4}$ | $3.5 \times 10^2$ | $2.9 \times 10^2$ | 38 | *22* | $2.6 \times 10^{-8}$ | $4.6 \times 10^{-2}$ |
| 45 | $Rh^{103*}$ | $3.0 \times 10^{-5}$ | *$2.3 \times 10^4$* | $2.8 \times 10^4$ | $3.7 \times 10^5$ | $6.4 \times 10^7$ | $7.0 \times 10^{-6}$ | $4.3 \times 10^{-5}$ |
| 46 | $Pd^{103} + Rh^{103*}$ | $1.3 \times 10^{-3}$ | $6.4 \times 10^3$ | $2.4 \times 10^3$ | $2.4 \times 10^2$ | *$1.2 \times 10^2$* | $1.6 \times 10^{-6}$ | $8.3 \times 10^{-3}$ |
| 47 | $Ag^{105}$ | $3.6 \times 10^{-3}$ | $1.5 \times 10^2$ | *73* | 19 | *9.4* | $3.4 \times 10^{-7}$ | 0.11 |
| 47 | $Ag^{111}$ | $6.3 \times 10^{-4}$ | $3.0 \times 10^2$ | $3.4 \times 10^2$ | 21 | *11* | $7.0 \times 10^{-8}$ | $9.1 \times 10^{-2}$ |
| 48 | $Cd^{109} + Ag^{109}$ | $3.9 \times 10^{-2}$ | $2.8 \times 10^3$ | $1.8 \times 10^3$ | $2.5 \times 10^2$ | *$1.3 \times 10^2$* | $5.1 \times 10^{-5}$ | $7.7 \times 10^{-3}$ |
| 50 | $Sn^{113}$ | $9.6 \times 10^{-3}$ | $7.5 \times 10^2$ | $4.0 \times 10^2$ | 88 | *44* | $4.2 \times 10^{-6}$ | $2.3 \times 10^{-2}$ |
| 52 | $Te^{127}$ | $1.1 \times 10^{-2}$ | $3.9 \times 10^2$ | $3.9 \times 10^2$ | 36 | *18* | $2.0 \times 10^{-6}$ | $5.5 \times 10^{-2}$ |
| 52 | $Te^{129}$ | $3.3 \times 10^{-3}$ | $1.1 \times 10^2$ | 88 | 12 | *6.1* | $2.0 \times 10^{-7}$ | 0.16 |
| 53 | $I^{131}$ | $8.0 \times 10^{-4}$ | $3.8 \times 10^2$ | *$2.9 \times 10^2$* | $7.0 \times 10^2$ | $3.5 \times 10^2$ | $2.3 \times 10^{-6}$ | $3.4 \times 10^{-3}$ |
| 55 | $Cs^{137} + Ba^{137*}$ | 1.3 | $3.2 \times 10^2$ | *$2.2 \times 10^2$* | $5.6 \times 10^2$ | $2.8 \times 10^2$ | $2.7 \times 10^{-3}$ | $4.5 \times 10^{-3}$ |
| 57 | $La^{140}$ | $1.8 \times 10^{-4}$ | $1.3 \times 10^2$ | 90 | 12 | *6.6* | $1.2 \times 10^{-8}$ | 0.15 |
| 56 | $Ba^{140} + La^{140}$ | $1.4 \times 10^{-3}$ | $3.3 \times 10^2$ | $2.6 \times 10^2$ | 19 | *7* | $9.5 \times 10^{-8}$ | 0.14 |

* The daughter products in these cases are isomers in an excited state.

† The MPI as used here is the μc present initially in a volume of air to be breathed during the succeeding 8 hours and which then results in the indicated dose.

**Table V. Single Exposure Values for Inhalation of Insoluble Radioactive Material when the GI Tract Is the Critical Body Organ — *Continued***

| (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) |
|---|---|---|---|---|---|---|---|---|
| | | | *MPI or μc inhaled in 8 hours that will deliver a dose of 0.3 rem during the following week to the organs cited: Equations 26, 28* | | | | | |
| *Z* | *Isotope* | *mg/100 mc* | *Stomach* | *Small intestine* | *Upper large intestine* | *Lower large intestine* | *mg corresponding to smallest value in columns 4, 5, 6, or 7.* | *Relative hazard H* |
| 59 | $Pr^{144}$ | $1.3 \times 10^{-6}$ | *$4.5 \times 10^{3}$* | $1.9 \times 10^{4}$ | $2.8 \times 10^{7}$ | $2.5 \times 10^{15}$ | $6.1 \times 10^{-8}$ | $2.2 \times 10^{-4}$ |
| 58 | $Ce^{144} + Pr^{144}$ | $3.2 \times 10^{-2}$ | $1.3 \times 10^{2}$ | 91 | *5.9* | *2.9* | $9.2 \times 10^{-7}$ | 0.34 |
| 59 | $Pr^{143}$ | $1.5 \times 10^{-3}$ | $3.6 \times 10^{2}$ | $4.0 \times 10^{2}$ | 25 | *13* | $2.0 \times 10^{-7}$ | $7.7 \times 10^{-2}$ |
| 61 | $Pm^{147}$ | 0.11 | $1.7 \times 10^{3}$ | $1.8 \times 10^{3}$ | $1.1 \times 10^{2}$ | *56* | $5.9 \times 10^{-5}$ | $1.8 \times 10^{-2}$ |
| 62 | $Sm^{151}$ | 4.2 | $5.6 \times 10^{3}$ | $6.2 \times 10^{3}$ | $3.8 \times 10^{2}$ | *$1.9 \times 10^{2}$* | $8.0 \times 10^{-3}$ | $5.3 \times 10^{-3}$ |
| 63 | $Eu^{154}$ | 0.68 | $2.4 \times 10^{2}$ | $1.5 \times 10^{2}$ | 22 | *11* | $7.5 \times 10^{-5}$ | $9.1 \times 10^{-2}$ |
| 67 | $Ho^{166}$ | $1.4 \times 10^{-4}$ | $1.9 \times 10^{2}$ | $2.1 \times 10^{2}$ | 17 | *10* | $1.4 \times 10^{-8}$ | 0.10 |
| 69 | $Tm^{170}$ | $1.6 \times 10^{-2}$ | $3.5 \times 10^{2}$ | $3.9 \times 10^{2}$ | 24 | *12* | $2.0 \times 10^{-6}$ | $8.3 \times 10^{-2}$ |
| 71 | $Lu^{177}$ | $9.2 \times 10^{-4}$ | $8.1 \times 10^{2}$ | $7.3 \times 10^{2}$ | 55 | *28* | $2.6 \times 10^{-7}$ | $3.6 \times 10^{-2}$ |
| 73 | $Ta^{182}$ | $1.5 \times 10^{-2}$ | $2.6 \times 10^{2}$ | $1.5 \times 10^{2}$ | 25 | *12* | $1.8 \times 10^{-6}$ | $8.4 \times 10^{-2}$ |
| 74 | $W^{181}$ | $1.9 \times 10^{-2}$ | $2.4 \times 10^{2}$ | $1.1 \times 10^{2}$ | 32 | *16* | $3.1 \times 10^{-6}$ | $6.3 \times 10^{-2}$ |
| 75 | $Re^{183}$ | $1.7 \times 10^{-2}$ | $4.0 \times 10^{2}$ | $2.0 \times 10^{2}$ | $10^{2}$ | *50* | $8.3 \times 10^{-6}$ | $2.0 \times 10^{-2}$ |
| 77 | $Ir^{190}$ | $1.6 \times 10^{-3}$ | $1.2 \times 10^{3}$ | $6.2 \times 10^{2}$ | $1.6 \times 10^{2}$ | *81* | $1.3 \times 10^{-6}$ | $1.2 \times 10^{-2}$ |
| 77 | $Ir^{192}$ | $1.1 \times 10^{-2}$ | $2.0 \times 10^{2}$ | $1.3 \times 10^{2}$ | 24 | *12* | $1.3 \times 10^{-6}$ | $8.4 \times 10^{-2}$ |
| 78 | $Pt^{191}$ | $4.4 \times 10^{-4}$ | $2.2 \times 10^{2}$ | $10^{2}$ | 33 | *18* | $7.7 \times 10^{-8}$ | $5.7 \times 10^{-2}$ |
| 78 | $Pt^{193}$ | $6.3 \times 10^{-4}$ | $2.4 \times 10^{2}$ | $1.2 \times 10^{2}$ | 40 | *21* | $1.3 \times 10^{-7}$ | $4.8 \times 10^{-2}$ |
| 79 | $Au^{196}$ | $8.3 \times 10^{-4}$ | $6.7 \times 10^{2}$ | $3.4 \times 10^{2}$ | 98 | *51* | $4.2 \times 10^{-7}$ | $2.0 \times 10^{-2}$ |
| 79 | $Au^{198}$ | $4.1 \times 10^{-4}$ | $2.7 \times 10^{2}$ | $2.3 \times 10^{2}$ | 25 | *14* | $5.5 \times 10^{-8}$ | $7.4 \times 10^{-2}$ |
| 79 | $Au^{199}$ | $5.0 \times 10^{-4}$ | $7.4 \times 10^{2}$ | $5.9 \times 10^{2}$ | 77 | *40* | $2.0 \times 10^{-7}$ | $2.5 \times 10^{-2}$ |
| 81 | $Tl^{200}$ | $1.7 \times 10^{-4}$ | $2.1 \times 10^{2}$ | $1.1 \times 10^{2}$ | 51 | *31* | $5.3 \times 10^{-8}$ | $3.3 \times 10^{-2}$ |
| 81 | $Tl^{201}$ | $4.6 \times 10^{-4}$ | $1.7 \times 10^{3}$ | $8.1 \times 10^{2}$ | $4.1 \times 10^{2}$ | *$2.2 \times 10^{2}$* | $10^{-6}$ | $4.6 \times 10^{-3}$ |
| 81 | $Tl^{202}$ | $1.8 \times 10^{-3}$ | $1.1 \times 10^{3}$ | $5.9 \times 10^{2}$ | $2.4 \times 10^{2}$ | *$1.2 \times 10^{2}$* | $2.2 \times 10^{-6}$ | $8.4 \times 10^{-3}$ |
| 81 | $Tl^{204}$ | 0.23 | $4.5 \times 10^{2}$ | $5.0 \times 10^{2}$ | 55 | *27* | $6.3 \times 10^{-5}$ | $3.7 \times 10^{-2}$ |
| 82 | $Pb^{203}$ | $3.4 \times 10^{-4}$ | $5.4 \times 10^{2}$ | $2.6 \times 10^{2}$ | 84 | *46* | $1.6 \times 10^{-7}$ | $2.2 \times 10^{2}$ |
| 84 | $Po^{210}$ | $2.2 \times 10^{-2}$ | 2.0 | 2.2 | 0.16 | *$6.4 \times 10^{-2}$* | $1.4 \times 10^{-8}$ | 16 |
| 83 | $Bi^{210} + Po^{210}$ | $8.0 \times 10^{-4}$ | $2.9 \times 10^{2}$ | $2.8 \times 10^{2}$ | 15 | *5.8* | $4.6 \times 10^{-8}$ | 0.17 |
| 82 | $Pb^{210} \rightarrow Po^{210}$‡ | 1.3 | $4.5 \times 10^{3}$ | $2.6 \times 10^{3}$ | $1.7 \times 10^{2}$ | *39* | $5.0 \times 10^{-4}$ | $2.6 \times 10^{-2}$ |
| 85 | $At^{211} \rightarrow Pb^{207}$ | $5.0 \times 10^{-5}$ | *2.4* | 2.7 | $3.1 \times 10^{2}$ | $3.3 \times 10^{2}$ | $1.2 \times 10^{-9}$ | 0.42 |
| 83 | $Bi^{214} \rightarrow Po^{210}$ | $2.2 \times 10^{-6}$ | *56* | $2.2 \times 10^{2}$ | $5.9 \times 10^{4}$ | $6.7 \times 10^{6}$ | $1.2 \times 10^{-9}$ | $1.8 \times 10^{-2}$ |
| 82 | $Pb^{214} \rightarrow Po^{210}$ | $3.0 \times 10^{-6}$ | *50* | 55 | $1.1 \times 10^{4}$ | $4.3 \times 10^{6}$ | $1.5 \times 10^{-9}$ | $2.0 \times 10^{-2}$ |
| 86 | $Rn^{222} \rightarrow Po^{210}$ | $6.5 \times 10^{-4}$ | 0.81 | 0.76 | $4.0 \times 10^{-2}$ | *$2.1 \times 10^{-2}$* | $1.4 \times 10^{-2}$ | |
| 88 | $Ra^{226} \rightarrow Po^{210}$ | $10^{2}$ | 2.3 | 2.5 | 0.14 | *$5.1 \times 10^{-2}$* | $5.2 \times 10^{-5}$ | 19 |
| 82 | $Pb^{211}$ | $4.0 \times 10^{-6}$ | *25* | 52 | $2.0 \times 10^{4}$ | $10^{8}$ | $10^{-9}$ | $4.0 \times 10^{-2}$ |
| 88 | $Ra^{223} \rightarrow Pb^{211}$ | $1.9 \times 10^{-3}$ | 0.48 | 0.49 | $3.5 \times 10^{-2}$ | *$1.8 \times 10^{-2}$* | $3.4 \times 10^{-10}$ | 56 |
| 90 | $Th^{227} \rightarrow Pb^{211}$ | $3.2 \times 10^{-3}$ | 1.8 | 1.9 | 0.11 | *$4.7 \times 10^{-2}$* | $1.5 \times 10^{-9}$ | 21 |
| 89 | $Ac^{227} \rightarrow Pb^{211}$ | 1.4 | $2.1 \times 10^{3}$ | $2.5 \times 10^{2}$ | 5.7 | *1.1* | $1.5 \times 10^{-5}$ | 0.91 |
| 83 | $Bi^{212}$ | $6.8 \times 10^{-6}$ | *10* | 17 | 16 | $1.9 \times 10^{3}$ | $7.2 \times 10^{-10}$ | $9.6 \times 10^{-2}$ |
| 82 | $Pb^{212} + Bi^{212}$ | $7.1 \times 10^{-5}$ | 6.4 | 2.6 | 0.18 | *0.15* | $1.1 \times 10^{-10}$ | 6.5 |
| 88 | $Ra^{224} \rightarrow Bi^{212}$ | $6.2 \times 10^{-4}$ | 0.58 | 0.64 | $4.4 \times 10^{-2}$ | *$2.2 \times 10^{-2}$* | $1.4 \times 10^{-10}$ | 45 |
| 90 | $Th^{228} \rightarrow Bi^{212}$ | 0.12 | 2.0 | 2.0 | 0.10 | *$3.5 \times 10^{-2}$* | $4.2 \times 10^{-8}$ | 29 |
| 89 | $Ac^{228} \rightarrow Bi^{212}$ | $4.4 \times 10^{-5}$ | $2.3 \times 10^{2}$ | $2.0 \times 10^{2}$ | *30* | 33 | $1.3 \times 10^{-8}$ | $3.3 \times 10^{-2}$ |
| 88 | $Ra^{228} \rightarrow Bi^{212}$ | 0.43 | $2.0 \times 10^{3}$ | $2.8 \times 10^{2}$ | 17 | *6.4* | $2.7 \times 10^{-5}$ | 0.16 |
| 90 | $Th^{232} \rightarrow Bi^{212}$ | $8.8 \times 10^{6}$ | 2.7 | 3.0 | 0.19 | *$9.2 \times 10^{-2}$* | $8.1 \times 10^{2}$ | $1.2 \times 10^{-7}$ |
| 90 | $Th^{234} + Pa^{234}$ | $4.3 \times 10^{-3}$ | $1.3 \times 10^{2}$ | $1.4 \times 10^{2}$ | 8.8 | *4.4* | $1.9 \times 10^{-7}$ | 0.23 |
| 92 | $U^{238}$ | $3.0 \times 10^{6}$ | 2.6 | 2.9 | 0.18 | *0.10* | $7.7 \times 10^{2}$ | $1.3 \times 10^{-8}$ |
| 92 | $U^{234}$ | $1.6 \times 10^{4}$ | 2.3 | 2.5 | 0.15 | *$7.7 \times 10^{-2}$* | $1.2 \times 10^{-2}$ | $8.1 \times 10^{-3}$ |
| 92 | $U^{235}$ | $4.6 \times 10^{7}$ | 2.5 | 2.7 | 0.17 | *0.08* | $1.2 \times 10^{2}$ | $8.6 \times 10^{-6}$ |
| 92 | $U^{233}$ | $1.1 \times 10^{4}$ | 2.2 | 2.5 | 0.15 | *$7.5 \times 10^{-2}$* | $7.9 \times 10^{-3}$ | $1.3 \times 10^{-2}$ |
| 94 | $Pu^{239}$ | $1.6 \times 10^{3}$ | 2.1 | 2.3 | 0.14 | *$7.1 \times 10^{-2}$* | $1.1 \times 10^{-3}$ | $8.8 \times 10^{-2}$ |
| 95 | $Am^{241}$ | 33 | 2.0 | 2.2 | 0.14 | *$6.7 \times 10^{-2}$* | $2.2 \times 10^{-5}$ | 4.6 |
| 96 | $Cm^{242}$ | $3.0 \times 10^{-2}$ | 1.8 | 2.0 | 0.12 | *$6.0 \times 10^{-2}$* | $1.8 \times 10^{-8}$ | 17 |

‡ The arrow (→) means the values given include the energies emitted by all the daughter products down to the stable isotope.

small amount of radioactive material on a mass basis that corresponds to the limiting MPI value. It is apparent that in many cases this quantity is so small that it could be inhaled on a single dust particle or could be injected unnoticed into the body as the result of a puncture wound. It is of interest to observe that, in spite of the low values of MPI in $\mu$c for natural thorium and natural uranium, the mg values are very large relative to the mg values for radionuclides such as $Ra^{226}$, and $Sr^{90} + Y^{90}$. This suggests that the radiation hazard may be proportional to the product of the probability of inhaling or injecting a given mass of the radioactive material and of the inverse of the minimum MPI value. Drinker and Hatch[7] have indicated that the average city and industrial air has a dust loading of from about $10^{-4}$ to $10^{-3}$ grams per cubic meter. This corresponds to an inhalation of 1 to 10 mg of dust in 8 hours. Therefore, in case of an accident with radioactive material it is not likely one would inhale more than 10 mg in an 8-hour period. There were 52,259 domestic shipments of radioactive material from our Laboratory[8] (ORNL) in 1954, and the average activity per shipment was 100 mc. These figures are exclusive of $Co^{60}$, which in many cases was shipped as large solid sources to be used as radiation therapy units. Therefore, for this comparison of radiation hazards, it is assumed that the amount involved in the accident is 100 mc in each case. Values of mg/100 mc are given in column 3 of Table V for convenient reference. The relative hazard in case of a spill or accident with one of these shipments of 100 mc of radioactive material is given by the equation,

$$H = \frac{10}{(\mathrm{MPI})(\mathrm{mg}/100\ \mathrm{mc})} \qquad (1)$$

In this equation, 10/(mg/100 mc) is the probability of inhalation of 10 mg of radioactive material. This term can have values $\leq 1$ in Equation 1, but when 10/(mg/100 mc) becomes $> 1$, it is assumed that all the radioactive material can be inhaled in 8 hours, and in this case the hazard is given by $H = 1/(\mathrm{MPI})$. The MPI to be used in Equation 1 is the smallest value given for the radioisotope in each table. In the case of a contaminated wound, the term, 10/(mg/100 mc), in Equation 1 is reduced to 1/(mg/100 mc) and this has a maximum value of 1. The values of $H$ in Tables II and III relate to the probability of getting radioactive contamination of 1 mg or less into a wound.

It is apparent that there are other factors which may affect the relative hazard in case of an accidental spill of radioactive material. One of the most important of these factors is the form—physical and chemical—of the material at the time of the accident. Space will not permit an evaluation of all these factors, but Table VI, prepared by G. R. Patterson,[9] gives information on the compounds shipped from our Laboratory (ORNL), the specific activity, the physical form, and an estimate of the probable fraction that would be available for intake by inhalation in case of an accident. These data and similar information for other chemical and physical forms and isotopic dilutions can be used to modify the basic data given for the single unadulterated radioisotopes in Tables I, II, III, IV, and V. Table VII is a summary of the 20 more dangerous radionuclides listed in the order of decreasing hazard.

### Equations Used in the Calculations

See Appendix for details of derivation and section on nomenclature for definition.

**Case 1 Table I:**

$$(\mathrm{MPI}) = \frac{3.12 \times 10^{-3} Dm}{fT\Sigma(bE)(\mathrm{RBE})N[T_r(1 - e^{-0.231/T_r}) + T_b e^{-0.693\ t_z/T}(1 - e^{0.231/T_b})]} \qquad (2)$$

**Case 2 Table II:**

$$(\mathrm{MPI}) = \frac{1.35 \times 10^{-2} Dm}{f_2' T\Sigma(bE)(\mathrm{RBE})N\ [1 - e^{-0.693\ t_z/T}]} \qquad (3)$$

**Case 3 Table III:**

$$(\mathrm{MPI}) = \frac{1.35 \times 10^{-5} D}{T\Sigma(bE)(\mathrm{RBE})N\ [1 - e^{-0.693\ t_z/T}]} \qquad (4)$$

**Case 4 Table IV:**

$$(\mathrm{MPI}) = \frac{25D}{T\Sigma(bE)(\mathrm{RBE})N\ [T_r(1 - e^{-0.231/T_r}) - 0.231\ e^{-0.693\ t_z/T}]} \qquad (5)$$

**Case 5 Table V:**

$$(\mathrm{MPI})_{\mathrm{GI}} = \mathrm{Min}\ [(\mathrm{MPI})_{stomach}, (\mathrm{MPI})_{intestine}] \qquad (6)$$

The solution of this equation for the GI tract is rather involved (See Appendix), and numerical values were found by use of the ORNL digital computer.

## CONCLUSIONS

In case a person receives a single exposure to relatively insoluble radioactive material, the localized dose is likely to be much greater if the material contaminates an open wound than if it enters the body by ingestion or inhalation. This may lend support to the common medical practice of excising contaminated tissue from such a wound. Table VII, which is a comparative summary of the 20 more dangerous radionuclides, indicates that many of the radionuclides are in the very hazardous group regardless of whether they are inhaled or injected; whether they are soluble or insoluble; and whether the wound site, the lung, the GI tract or some other organ is the critical body organ. For example, $Ac^{227}$, $At^{211}$, $Po^{210}$, $Am^{241}$, $Pb^{210}$, and $Sr^{90} + Y^{90}$ are generally among the more hazardous isotopes, the first four being among the 12 more hazardous in every case. For inhalation, the lung or GI tract is usually the critical body organ. In case of

accident, the ingestion hazard is considered to be negligible compared with the inhalation or injection hazard. The absorbed dose from each of the radionuclides is calculated by means of the digital computer at 31 points along the GI tract. The lower large intestine is usually the critical portion of the GI tract but, where the effective half-life is short, other portions, i.e., stomach, small intestine, and upper large intestine, may become the critical tissue. Five of the radionuclides—$Sr^{90}$ + $Y^{90}$, $Sm^{151}$, $Pu^{239}$, $Ra^{226}$, and $Th^{232}$—have very long biological half-lives so that the limiting restriction leading to the smallest MPI is the requirement that the integrated dose as a result of a single body intake must not exceed 150 rem in a lifetime (during the following 70 years). For the rest of the radionuclides, the smallest MPI is obtained when the integrated dose is limited to 0.3 rem during the week following the exposure.


## ACKNOWLEDGEMENTS

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## APPENDIX

### Derivation of General Equations of Dose Resulting From Single Exposure to a Radioisotope

#### Case 1

In this case it is assumed that a certain fraction, $f$, of the radioisotope of concentration MPC ($\mu c/cm^3$) is inhaled at a rate $V$ ($cm^3$/day) or at a rate of MPC ($I'\mu c$/day) and that it arrives in the critical body organ where it is eliminated exponentially both by radioactive and biological decay (Fig. 1). The dose is given by the equation,

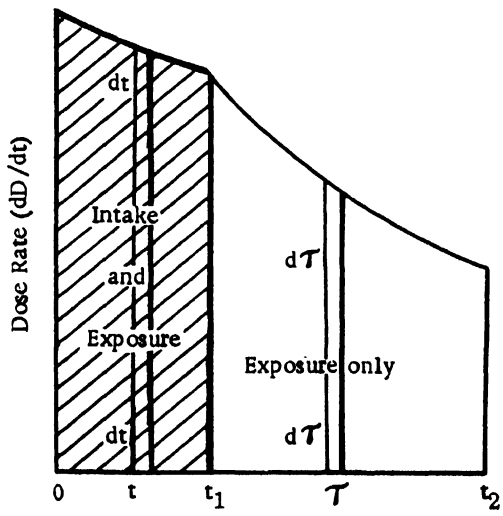



Figure 1

$$D = C \int_0^{t_1} \int_t^{t_2} \underbrace{f(\mathrm{MPC})V}_{\substack{\text{amount}\\ \text{arriving}\\ \text{in critical}\\ \text{organ}}} e^{\lambda_r t}\, e^{-\lambda(\tau - t)}\, dt\, d\tau \qquad (7)$$

$$D = C \int_0^{t_1} \int_t^{t_2} f(\mathrm{MPC})V e^{-\lambda_b t}\, e^{-\lambda \tau}\, dt\, d\tau \qquad (8)$$

in which $D$ is the dose in rem following a spill such that MPI $\mu c$ are initially available for body intake for time, $t_1$ and exposure to time, $\tau - t$. The terms $\lambda$, $\lambda_r$ and $\lambda_b$ are the coefficients of the total elimination, radioactive decay, and biological decay, respectively. The term MPI $= (\mathrm{MPC})Vt_1$ and

$$D = C \int_0^{t_1} \frac{f(\mathrm{MPC})I'e^{\lambda_b t}}{\lambda}(e^{-\lambda t} - e^{-\lambda t_2})dt$$

$$= C \int_0^{t_1} \frac{f(\mathrm{MPC})I'}{\lambda}(e^{\lambda_r t} - e^{\lambda t_2} e^{\lambda_b t})dt \qquad (9)$$

$$D = \frac{Cf(\mathrm{MPI})}{t_1 \lambda}\left[\frac{1 - e^{-\lambda_r t_1}}{\lambda_r} + \frac{e^{\lambda t_2}(1 - e^{\lambda_b t_1})}{\lambda_b}\right] \qquad (10)$$

$$C = 3.7 \times 10^4\ (\mathrm{dps}/\mu c)\ 3600 \times 24 \frac{(\mathrm{sec})}{(\mathrm{day})}$$
$$\times\ \Sigma(bE)(\mathrm{RBE})N(\mathrm{Mev/dis})\ 1.602 \times 10^{-6}$$
$$(\mathrm{ergs/Mev})\ 1/m\ (1/\mathrm{gm})\ 1/100\ (\mathrm{gm\text{-}rad/ergs})$$

$$Cf = \frac{51.2\, f\, \Sigma(bE)(\mathrm{RBE})N}{m}\ \text{rem/day to the critical organ of mass, } m\text{, due to 1 } \mu c \text{ taken in at time, } t = 0 \qquad (11)$$

$$\mathrm{MPI} = D\lambda m t_1 \Big/ \left(51.2\, f\Sigma(bE)(\mathrm{RBE})N \times \left[\frac{1 - e^{-\lambda_r/3}}{\lambda_r} + \frac{e^{-\lambda t_2}(1 - e^{\lambda_b/3})}{\lambda_b}\right]\right) \qquad (12)$$

$$\mathrm{MPI} = \frac{3.12 \times 10^{-3} Dm}{fT\Sigma(bE)(\mathrm{RBE})N\ [T_r(1 - e^{-0.231/T_r}) + T_b\, e^{-0.693\, t_2/T}(1 - e^{-0.231/T_b})]} \qquad (13)$$

#### Case 2

In this case it is assumed the MPI of soluble radioactive material enters the body by way of a wound at time $t = 0$ and that fraction $f_2$ arrives at the critical body organ by way of the blood. Here it remains for a time $t_2$ and delivers a dose $D$ as indicated by the equation,

$$D = C \int_0^{t_2} f_2'(\mathrm{MPI})e^{-\lambda t}\, dt = \frac{f_2' C(\mathrm{MPI})}{\lambda}(1 - e^{-\lambda t_2}) \qquad (14)$$

$$\mathrm{MPI} = \frac{1.35 \times 10^{-2} Dm}{f_2' T\Sigma(bE)(\mathrm{RBE})N(1 - e^{-0.693 t_2/T})} \qquad (15)$$

**Table VI. General Information Relative to Shipments of Radioactive Material from Oak Ridge National Laboratory[a]**

| (1) | (2) | (3) | (4) | (5) |
|---|---|---|---|---|
| *Radioisotope** | *Compound as shipped* | *Specific activity* | *Form and physical character* | *Estimate of probable fraction available for inhalation (%)* |
| $H^3$ (P) | H + $He^3$ Gas | ~2.63 c/cm³ at NTP (CF) | Gas | 100 |
| $Be^7$ (P) | $BeCl_2$ | >0.5 mc/ml (CF) | In HCl solution | 1 (fumes) |
| $C^{14}$ (P)1 | $BaCO_3$ | 0.25–1.5 c/gm C | Powder | 1 (dust) |
| $Na^{22}$ (P) | NaCl | >1 c/gm Na (CF) | In weak HCl solution | 0.1 |
| $Na^{24}$ (I) | $Na_2CO_3$ | 90 mc/gm Na | Powder | 1 |
| $Na^{24}$ (P) | NaCl | ~1 c/gm Na | In $H_2O$ solution | 0.1 |
| $P^{32}$ (I) | $KH_2PO_4$ | 30 mc/gm P | Crystalline-fine | 0.1 |
| $P^{32}$ (P)1 | Phosphate | ~0.025 mg P/mc | In weak HCl solution | 0.1 |
| $P^{32}$ (P)2 | Phosphate | >0.5 mc/ml (CF) | In weak HCl solution | 0.1 |
| $S^{35}$ (I) | S (elemental) | 0.3 mc/gm S | Sulfur sublimed | 1 |
| $S^{35}$ (P)1 | Sulfate | >1 mc/ml (CF) | In weak HCl solution | 0.1 |
| $S^{35}$ (P)2 | Ba sulfide | >10 c/gm S | In $Ba(OH)_2$ solution | 1 |
| $S^{35}$ (P)3 | S (elemental) | >1 c/gm S | In benzene solution | 1 |
| $Cl^{36}$ (I) | KCl | 0.0006 mc/gm Cl | Crystalline (very fine) | 0.1 |
| $Cl^{36}$ (P) | HCl | 0.01–0.2 mc/gm Cl | HCl solution | 1 |
| $A^{37}$ (P) | Argon and helium | 0.01–0.1 mc/ml (CF) | Gas | 100 |
| $K^{42}$ (I) | $K_2CO_3$ | 20 mc/gm K | Anhydrous powder | 1 |
| $Ca^{45}$ (I) | $CaCO_3$ | 0.4 mc/gm Ca | Powder | 1 (dust) |
| $Ca^{45}$ (P)1 | $CaCl_2$ | 0.2–0.4 mc/gm Ca | In HCl solution | Slight |
| $Ca^{45}$ (P)2 | $CaCl_2$ | 5–60 mc/gm Ca | In HCl solution | Slight |
| $Ca^{45}$ (P)3 | $CaCl_2$ | >1000 mc/gm | In HCl solution | Slight |
| $Ca^{45}$ (P)4 | Chloride | >0.1 μc/ml (CF) | In HCl solution | Slight |
| $Sc^{46}$ (I) | $Sc_2O_3$ | 750 mc/gm Sc | Powder | 0.2 |
| $Sc^{46}$ (P) | $ScCl_3$ | >5 c/gm Sc | In HCl solution | 0.1 |
| $Cr^{51}$ (I) | Cr Metal | 50 mc/gm Cr | Metal powder | Slight |
| $Cr^{51}$ (P) | $CrCl_3$ | 100–400 mc/gm Cr | In HCl solution | 0.1 |
| $Mn^{52-54}$ (P) | $MnCl_2$ | >0.05 mc/ml (CF) | In HCl solution | 0.1 |
| $Fe^{55-59}$ (I) | Fe metal | 0.7, 0.4 mc/gm Fe | Metal Powder | Slight |
| $Fe^{55-59}$ (P) | $FeCl_3$ | 1–4 mc $Fe^{59}$/gm Fe | In HCl solution | 0.1 |
| $Fe^{55}$ (P) | $FeCl_3$ | 0.5–1.5 c/gm Fe | In HCl solution | 0.1 |
| $Fe^{59}$ (P) | $FeCl_3$ | 0.5–1.5 c/gm Fe | In HCl solution | 0.1 |
| $Co^{57-58}$ (P) | $CoCl_2$ | <0.01 mc/ml | In HCl solution | 0.1 |
| $Co^{60}$ (I) | $Co_2O_3$ | 34 mc/gm Co | Powder and/or wire | Slight |
| $Co^{60}$ (P) | $CoCl_2$ | 0.5–5 c/gm Co | In HCl solution | 1 |
| $Ni^{63}$ (I) | Ni metal | 0.35 mc/gm Ni | Metal powder | Slight |
| $Ni^{63}$ (P) | $NiCl_2$ | 5–50 mc/gm Ni | In HCl solution | 0.1 |
| $Cu^{64}$ (I) | Cu metal | 300 mc/gm Cu | Metal turnings | Slight |
| $Zn^{65}$ (I) | Zn metal | 1.9 mc/gm Zn | Mossy metal | Slight |
| $Zn^{65}$ (P)1 | $ZnCl_2$ | 75–300 mc/gm Zn | In HCl solution | 0.1 |
| $Zn^{65}$ (P)2 | $ZnCl_2$ | ~5 c/g Zn (CF) | In HCl solution | 0.1 |
| $Ga^{72}$ (I) | $Ga_2O_3$ | 235 mc/gm Ga | Powder | Slight |
| $As^{73-74}$ (P) | Chloride or oxychloride | >0.05 mc/ml (CF) | In HCl solution | 1 (fumes) |
| $As^{76}$ (I) | $As_2O_3$ | 550 mc/gm As | Powder | 0.1 |
| $As^{77}$ (I) | $GeO_2$ | (CF) | Powder | 0.1 |
| $Se^{75}$ (I) | Se metal | 3.3 mc/gm Se | Powder | 1 |
| $Se^{75}$ (P) | Chloride | 50–100 mc/gm Se | In HCl solution | 1 |
| $Br^{82}$ (I) | KBr | 120 mc/gm Br | Crystalline | Slight |
| $Rb^{86}$ (I) | $Rb_2CO_3$ | 45 mc/gm Rb | Powder | 1 |
| $Sr^{89}$ (I) | $Sr(NO_3)_2$ | 0.11 mc/gm Sr | Crystalline (fine) | 0.1 |
| $Sr^{89}$ (P) | $SrCl_2$ | >1 mc/ml (CF) | In HCl solution | 0.1 |
| $SrY^{90}$ (P) | Chlorides | >1 mc/ml (CF) | In HCl solution | 0.1 |
| $Y^{90}$ (I) | $Y_2O_3$ | 115 mc/gm Y | Powder | 0.1 |
| $Y^{91}$ (P) | $YCl_3$ | >1 mc/ml (CF) | In HCl solution | 0.1 |
| $Zr^{95}$ (I) | $Zr(OH)_4$ or $ZO_2$ | 0.05 mc/gm Zr | Powder | Slight |
| Zr-$Nb^{95}$ (P) | Oxelate complex | >1 mc/ml (CF) | In oxalic acid solution | 0.1 |
| $Nb^{95}$ (P) | Nb complex | >1 mc/ml (CF) | In oxalic acid solution | 0.1 |
| $Mo^{99}$ (I) | $MoO_3$ | 7 mc/gm Mo | Powder | 0.1 |

* Symbols: (I) Irradiated; (P) Processed; and (CF) Carrier free.

Table VI. General Information Relative to Shipments of Radioactive Material from Oak Ridge National Laboratory[8] —*Continued*

| (1) | (2) | (3) | (4) | (5) |
|---|---|---|---|---|
| *Radioisotope** | *Compound as shipped* | *Specific activity* | *Form and physical character* | *Estimate of probable fraction available for inhalation (%)* |
| $Tc^{99}$ (I) | $MoO_3$ | (CF) | Powder | 0.1 |
| $Tc^{99}$ (P) | $NH_4TcO_4$ | 1 mg $Tc^{99} \approx 20\mu c$ $Tc^{99}$ | In weak $NH_4OH$ | 0.1 |
| $Ru^{97}$ (I) | $Ru_2O_3$ | 0.03 mc/gm Ru | Powder | 0.1 |
| $Ru^{103}$ (P) | Chloride | >1 mc/ml (CF) | In HCl solution | 0.1 |
| Ru-$Rh^{106}$ (P) | Chlorides | >2 c/g Ru (CF) | In HCl solution | 0.1 |
| $Pd^{109}$ (I) | Pd metal | 220 mc/gm Pd | Metal foil | Slight |
| $Ag^{110}$ (I) | $AgNO_3$ | 5 mc/gm Ag | Crystalline | Slight |
| $Ag^{110}$ (P) | $AgNO_3$ | 100–300 mc/gm Ag | In$NHO_3$ solution | 0.1 |
| $Ag^{111}$ (I) | Pd metal | (CF) | Metal foil (5 mil) | Slight |
| $Cd^{115}$ (I)1 | Cd metal | 20 mc/gm Cd | Metal, mossy | Slight |
| $Cd^{115}$ (I)2 | Cd metal | 1 mc/gm Cd | Metal, mossy | Slight |
| $Cd^{115}$ (P) | Cd nitrate | 5–20 mc/gm Cd | In $NHO_3$ solution | 1 (fumes) |
| $In^{114}$ (I) | In metal | 70 mc/gm In | Metal foil | Slight |
| $In^{114}$ (P) | Chloride | 50–150 mc/gm In | In HCl solution | 1 |
| $Sn^{113}$ (P) | Chloride | 0.1–1 mc/gm Sn | In HCl solution | 0.1 |
| $Sn^{113}$ (I) | Sn metal | | Mossy metal | Slight |
| $Sb^{122}$ (I) | Sb metal | 250 mc/gm Sb | Metal chunks | Slight |
| $Sb^{124}$ (I) | Sb metal | 12 mc/gm Sb | Metal powder | Slight |
| $Sb^{124}$ (P) | Sb chloride | 500–1500 mc/gm Sb | In HCl solution | 1 (fumes) |
| $Sb^{125}$ (I) | Sn + Sb metal | (CF) | Mossy metal chunks | Slight |
| $Sb^{125}$ (P) | Sb chloride | ~0.01 mc/ml (CF) | In HCl solution | 1 (fumes) |
| $I^{131}$ (I) | Te metal | (CF) | Powder | 1 |
| $I^{131}$ (P) | NaI in $Na_2SO_3$ | 5–20 mc/ml (CF) | In $Na_2SO_3$ solution | 0.1 |
| $Cs^{134}$ (I) | $Cs_2CO_3$ | 60 mc/gm Cs | Powder | 1 or less |
| $Cs^{134}$ (P) | CsCl | 1–3 c/gm Cs | In HCl solution | 0.1 |
| Cs-$Ba^{137}$ (P) | CsCl | 1–10 mc/ml (CF) | In HCl solution | 0.1 |
| $Ba^{131}$ (I) | $Ba(NO_3)_2$ | 0.25 mc/gm Ba | Crystalline and/or powder | 0.1 |
| Ba-$La^{140}$ (P) | Chlorides | >1 mc/ml (CF) | In HCl solution | 1 (fumes) |
| $La^{140}$ (I) | $La_2O_3$ | 525 mc/gm La | Powder | 0.1 |
| $Ce^{141}$ (I) | $CeO_2$ | 7.5 mc/gm Ce | Powder | 0.1 (dust) |
| $Ce^{141}$ (P) | $CeCl_3$ | >1 mc/ml (CF) | In HCl solution | 0.1 (fumes) |
| Ce-$Pr^{144}$ (P) | $CeCl_3$ | >1 m/ml (CF) | In HCl solution | 0.1 (fumes) |
| $Pr^{142}$ (I) | $Pr_2O_3$ | 550 mc/gm Pr | Powder | 0.1 |
| $Pr^{143}$ (P) | $PrCl_3$ | >0.1 mc/ml (CF) | In HCl solution | 0.1 |
| Nd-$Pm^{147}$ (P) | $NdCl_3$ | >0.1 mc/ml (CF) | In HCl solution | 0.1 |
| $Pm^{147}$ (I) | $Nd_2O_3$ | (CF) | Powder | 0.1 |
| $Pm^{147}$ (P) | $PmCl_3$ | >1 mc/ml (CF) | In HCl solution | 0.1 |
| $Sm^{153}$ (I) | $Sm_2O_3$ | 1000 mc/gm Sm | Powder | 0.1 |
| $Eu^{152-154}$ (I) | $Eu_2O_3$ | 72 mc/gm Eu | Powder | 0.1 |
| $Hf^{181}$ (I) | $HfO_2$ | 60 mc/gm Hf | Powder | 0.1 |
| $Ta^{181}$ (I) | Ta Metal | 105 mc/gm Ta | Metal Powder | Slight |
| $Ta^{181}$ (P) | Tantalate | 0.5–1.5 c/gm Ta | In KOH solution | 0.1 |
| $W^{185}$ (I) | $WO_3$ | 7 mc/gm W | Powder | 0.1 |
| $W^{185}$ (P) | Tungstate | 100–400 mc/gm W | In KOH solution | 0.2 |
| $W^{187}$ (I) | $WO_3$ | 350 mc/gm W | Powder | 0.1 |
| $Re^{186}$ (I) | Re | 1230 mc/gm Re | Metal powder | 1 |
| $Os^{191}$ (I) | Os metal | >3 mc/gm Os | Metal powder | 0.1 |
| $Ir^{192}$ (I) | Ir metal | 2.1 c/gm Ir | Metal powder | Slight |
| $Ir^{192}$ (P) | K or Na chloroiridiate | 1–5 c/gm Ir | In HCl solution | 0.1 |
| $Ir^{194}$ (I) | Ir metal | 1700 mc/gm Ir | Metal powder | Slight |
| $Au^{198}$ (I) | Au metal | 3 c/gm Au | Metal foil (1 mil) | Slight |
| $Au^{199}$ (I) | Au and Pt metal | (CF) | Metal foil (5 mil) | Slight |
| $Hg^{197}$ (I) | HgO | 200 mc/gm Hg | Powder | Slight |
| $Hg^{203}$ (I) | HgO | 72 mc/gm Hg | Powder | Slight |
| $Hg^{203}$ (P) | Mercury nitrate | 50–100 mc/gm Hg | In $HNO_3$ solution | 0.1 |
| $Tl^{204}$ (I) | $TlNO_3$ | 1.3 mc/gm Tl | Crystalline (fine) | 0.1 |
| $Tl^{204}$ (P) | Nitrate | 50–300 mc/gm Tl | In $HNO_3$ solution | 0.1 |
| $Bi^{210}$ (I) | Bi metal | 0.4 mc/gm Bi | Granular metal | Slight |
| FP (P)1 | Mixed fission products | 1–25 mc/ml | Strong $HNO_3$ solution | 1 |

**Table VII. Summary of the Twenty More Dangerous Radionuclides Listed in Order of Decreasing Hazard**

| *Inhalation* | | | *Injection* | | | *Inhalation* | |
|---|---|---|---|---|---|---|---|
| *Soluble radioactive material* | | | | | | *Insoluble radioactive material* | |
| *The indicated organ† as the critical body organ* | | | *Wound site the critical tissue* | | | *Lung the critical body organ* | *GI tract the critical body organ* |
| *Relative hazard from Table I* | | | *Relative hazard from Table II* | | | *Relative hazard from Table III* | *Relative hazard from Table IV* | *Relative hazard from Table V* |
| 210 $Ac^{227}$ + dr | *B* | 60 | $Ac^{227}$ + dr | *B* | $4 \times 10^8$ $Ac^{227}$ + dr | 48 $Ac^{227}$ + dr | 56 $Ra^{223} \rightarrow Pb^{211}$‡ |
| 9.6 $At^{211}$ + dr | *T* | 19 | $At^{211}$ + dr | *T* | $7.1 \times 10^7$ $Cm^{242}$ | 8.1 $Pb^{210}$ + dr | 45 $Ra^{224} \rightarrow Bi^{212}$ |
| 4.8 $Pb^{210}$ + dr | *B* | 13 | $Cm^{242}$ | *B* | $6.7 \times 10^7$ $Pb^{210}$ + dr | 8.1 $Po^{210}$ | 29 $Th^{228} \rightarrow Bi^{212}$ |
| 3.3 $Cm^{242}$ | *B* | 11 | $Pb^{210}$ + dr | *B* | $6.3 \times 10^7$ $Po^{210}$ | 3.6 $Ra^{226}$ + dr | 21 $Th^{227} \rightarrow Pb^{211}$ |
| 1.6 $Po^{210}$ | *S* | 8.3 | $Po^{210}$ | *S* | $5.6 \times 10^6$ $At^{211}$ + dr | 2.4 $Am^{241}$ | 19 $Ra^{226} \rightarrow Po^{210}$ |
| 1.4 $I^{131}$ | *T* | 1.9 | $I^{131}$ | *T* | $2.0 \times 10^6$ $Am^{241}$ | 0.48 $At^{211}$ + dr | 17 $Cm^{242}$ |
| 0.9 $Am^{241}$ | *B* | 0.53 | $Th^{234}$ + $Pa^{234}$ | *B* | $1.9 \times 10^6$ $Ra^{226}$ + dr | 0.23 $Ba^{140}$ + $La^{140}$ | 16 $Po^{210}$ |
| 0.3 $Ra^{226}$ + dr | *B* | 0.43 | $Ce^{144}$ + $Pr^{144}$ | *B* | $1.6 \times 10^6$ $Ru^{106}$ + $Rh^{106}$ | 0.23 $Ag^{105}$ | 6.5 $Pb^{212}$ + $Bi^{212}$ |
| 0.3 $Pu^{239}$ | *B* | 0.42 | $Ba^{140}$ + $La^{140}$ | *B* | $1.4 \times 10^6$ $Ce^{144}$ + $Pr^{144}$ | 0.21 $Ru^{106}$ + $Rh^{106}$ | 4.6 $Am^{241}$ |
| 0.2 $Sr^{90}$ + $Y^{90}$ | *B* | 0.36 | $Am^{241}$ | *B* | $1.2 \times 10^6$ $Sr^{90}$ + $Y^{90}$ | 0.21 $Co^{60}$ | 0.91 $Ac^{227} \rightarrow Pb^{211}$ |
| 0.13 $Th^{234}$ + $Pa^{234}$ | *B* | 0.34 | $Sr^{90}$ + $Y^{90}$ | *B* | $9.1 \times 10^5$ $Th^{234}$ + $Pa^{234}$ | 0.21 $V^{48}$ | 0.42 $At^{211} \rightarrow Pb^{207}$ |
| 0.11 $Ba^{140}$ + $La^{140}$ | *B* | 0.28 | $Ir^{192}$ | *S* | $7.7 \times 10^5$ $Ba^{140}$ + $La^{140}$ | 0.20 $Ce^{144}$ + $Pr^{144}$ | 0.37 $Ru^{106}$ + $Rh^{106}$ |
| 0.11 $Ce^{144}$ + $Pr^{144}$ | *B* | 0.25 | $Y^{91}$ | *B* | $7.1 \times 10^5$ $Te^{129}$ | 0.19 $Te^{129}$ | 0.34 $Ce^{144}$ + $Pr^{144}$ |
| 0.1 $Sr^{89}$ | *B* | 0.20 | $Ru^{106}$ + $Rh^{106}$ | *K* | $6.7 \times 10^5$ $P^{32}$ | 0.18 $Sc^{46}$ | 0.23 $Th^{234}$ + $Pa^{234}$ |
| 0.072 $Fe^{59}$ | *Bl* | 0.19 | $Tm^{170}$ | *B* | $6.3 \times 10^5$ $Sr^{89}$ | 0.16 $W^{181}$ | 0.19 $Y^{90}$ |
| 0.063 $Y^{91}$ | *B* | 0.18 | $Sr^{89}$ | *B* | $6.3 \times 10^5$ $Rb^{86}$ | 0.14 $Sr^{90}$ + $Y^{90}$ | 0.18 $As^{76}$ |
| 0.054 $Te^{129}$ | *K* | 0.17 | $Te^{129}$ | *K* | $6.3 \times 10^5$ $Y^{91}$ | 0.14 $Ir^{192}$ | 0.17 $Bi^{210}$ + $Po^{210}$ |
| 0.048 $Tm^{170}$ | *B* | 0.17 | $Au^{198}$ | *K* | $3.7 \times 10^5$ $Tm^{170}$ | 0.14 $Zr^{95}$ + $Nb^{95}$ | 0.16 $Te^{129}$ |
| 0.048 $Ru^{106}$ + $Rh^{106}$ | *K* | 0.12 | $Pu^{239}$ | *B* | $3.2 \times 10^5$ $Ag^{111}$ | 0.13 $Tc^{96}$ | 0.16 $Ra^{228} \rightarrow Bi^{212}$ |
| 0.048 $Au^{198}$ | *K* | 0.11 | $Fe^{59}$ | *Bl* | $3.1 \times 10^5$ $Te^{127}$ | 0.12 $Th^{234}$ + $Pa^{234}$ | 0.15 $La^{140}$ |

* Parent radionuclides with radioactive half-lives less than one-half hour have been excluded in this comparison.

† Symbols: *B*—Bone; *T*—Thyroid; *S*—Spleen; *Bl*—Blood; and *K*—Kidneys.

‡ The arrow (→) means the values given include the energies emitted by all the daughter products down to the stable isotope.

### Case 3

In this case it is assumed the MPI of insoluble radioactive material enters the body by way of a wound at time $t = 0$; that all the material remains at the site of the wound where it irradiates the critical tissue of mass $m = 10^{-3}$ gm, and that it is eliminated from the wound with a biological half-life, $T_b$, of 120 days. In this case Equation 15 becomes,

$$\mathrm{MPI} = \frac{1.35 \times 10^{-5} D}{T \Sigma (bE)(\mathrm{RBE}) N (1 - e^{-0.693 t_s / T})} \tag{16}$$

### Case 4

This case is essentially the same as Case 1 except the lung of mass $m = 10^3$ gm is always taken as the critical organ, the fraction retained in the lungs, $f = 0.125$, and the biological half-life $T_b$ is 120 days. Placing these constants in Equation 13,

$$\mathrm{MPI} = \frac{25D}{T \Sigma (bE)(\mathrm{RBE}) N \left[ T_r (1 - e^{-0.231 / T_r}) - 0.231\, e^{-0.693\, t_s / T} \right]} \tag{17}$$

### Case 5

The damage to a portion of the GI tract due to the inhalation of a radioactive isotope is influenced by the radioactive decay rate of the isotope and of the daughters which may replace it, and also by the partial or complete elimination of the isotope and daughters from the GI tract. In most cases the elimination from the GI tract by absorption into the blood stream occurs predominantly while the isotope passes through the small intestine. In this report we compute the damage to the GI tract, taking account of the effect of biological elimination.

We consider the GI tract as composed of four organs—the stomach, small intestine, upper large intestine, and lower large intestine. We assume one atom of a certain isotope is ingested and that this atom, or the daughters which may replace it, moves through the GI tract spending one hour in the stomach, 4 hours in the small intestine, 8 hours in the upper large intestine, and 18 hours in the lower large intestine. We assume the atom moves at a uniform rate along the intestinal tract. Finally we assume that the elimination from the GI tract occurs uniformly, i.e., at a constant rate during the 4-hour period in the small intestine, and that there is no absorption into the blood stream during the remaining periods of time.

Under these assumptions the atom has a definite position at any time, $t$, and thus the damage to a specified portion of the GI tract is easily calculated when the number of atoms at any time, $t$, is known. Suppose the parent isotope has a chain of $k$ daughters. When the isotope or its daughters are passing along a section of the GI tract where no absorption into the blood stream occurs, the number of atoms of each kind is determined by the solution of the following differential equations:

$$\frac{dN_0(t)}{dt} = -\lambda_0{}^r N_0(t) \qquad (18)$$

$$\frac{dN_i(t)}{dt} = \lambda_{i-1}{}^r N_{i-1}(t) - \lambda_i{}^r N_i(t)$$
$$i = 1, 2, \ldots k \qquad (19)$$

In Equations 18 and 19, $N_i(t)$ is the number of atoms of the $i$th daughter which are in the GI tract at time, $t$, and $\lambda_i{}^r$ is the radioactive decay constant for the $i$th daughter. The subscript, zero, refers to the parent isotope. To completely determine the functions $N_i(t)$, the values of these functions are needed at the time when they entered the section of the GI tract in question. Thus, for the stomach we may take $t = 0$ as the time of entry and then $N_0(0) = 1$, $N_i(0) = 0$ for $i \geqq 1$. The solution of Equations 18 and 19 at any time within the first hour is then given by,[10]

$$N_0(t) = e^{-\lambda_0{}^r t} \qquad (20)$$

$$N_i(t) = \left(\prod_{j=0}^{i-1} \lambda_j{}^r\right) \sum_{n=0}^{i} \frac{e^{-\lambda_n{}^r t}}{\prod_{\substack{p=0 \\ p \neq n}}^{i} (\lambda_p{}^r - \lambda_n{}^r)}$$
$$i = 1, 2, \ldots k$$

$\prod_{j=0}^{i-1} \lambda_j{}^r$ means the product of the $\lambda_j{}^r$ indicated (21)

The solutions of Equations 20 and 21 can be extended for arbitrary numbers of atoms of each daughter entering by simply regarding each daughter atom that enters the GI tract as the parent of a somewhat shorter chain of daughters and applying the same formulae (20 and 21) with the corresponding values for $k$ and the $\lambda$'s. We shall not write here the most general solution which is a rather cumbersome expression. It is, however, easy to write the appropriate solution in any case where $k$ is not very large.

If the atoms are passing through the small intestine, Equations 18 and 19 need to be modified since they neglect absorption into the blood stream. We have assumed that the absorption takes place at a constant rate. Thus, of $N_i(t)$ atoms of the $i$th daughter which exist at time, $t$, in the small intestine, i.e., if time is taken in days $1/24 < t < 5/24$ a certain fraction is absorbed per unit time, and we denote this fraction by $\lambda_i{}^b$.

The constant, $\lambda_i{}^b$, plays a role quite analogous to that of the radioactive decay constant, $\lambda_i{}^r$, and we term $\lambda_i{}^b$ the biological elimination constant. We put $\lambda_i = \lambda_i{}^r + \lambda_i{}^b$ so that if $N_i(t)$ atoms exist at time, $t$, the number eliminated in time, $dt$, either through radioactive decay or through absorption into the blood stream, is $N_i(t)\lambda_i dt$. Thus, for $1/24 < t < 5/24$ Equations 18 and 19 become,

$$\frac{dN_0(t)}{dt} = -\lambda_0 N_0(t) \qquad (18')$$

$$\frac{dN_i(t)}{dt} = \lambda_{i-1}{}^r N_{i-1}(t) - \lambda_i N_i(t) \qquad i = 1, 2, \ldots k \qquad (19')$$

and the appropriate boundary conditions are the values of the functions $N_i$ (1/24) determined by Equations 20 and 21 for $t = 1/24$. It is easily verified that,

$$N^0(t) = e^{-\lambda_0(t-1)} \qquad (20')$$

$$N_i(t) = \left(\prod_{j=0}^{i-1} \lambda_j{}^r\right) \sum_{n=0}^{i} \frac{e^{-\lambda_n(t-1)}}{\prod_{\substack{p=0 \\ p \neq n}}^{i} (\lambda_p - \lambda_n)}$$
$$i = 1, 2, \ldots k$$

is the solution of Equations 18′ and 19′ with the boundary conditions $N_0$ (1/24) = 1, $N_i$ (1/24) = 0, $i = 1 \ldots$ k. This is easily verified by direct substitution and comparison with Equations 20 and 21. Thus, the solution which has the same values at $t = 1/24$ as Equations 20 and 21 is,

$$N_0(t) = N_0(5/24) e^{-\lambda_0{}^r (t - 5/24)} \qquad (20'')$$

$$N_i(t) = \sum_{q=0}^{i} N_q(1/24) \left(\prod_{j=q}^{i-1} \lambda_j{}^r\right) \sum_{n=q}^{i} \frac{e^{-\lambda_n(t-1/24)}}{\prod_{\substack{p=q \\ p \neq n}}^{i} (\lambda_p - \lambda_n)}$$
$$i = 1, 2, \ldots k \qquad (21'')$$

with $N_q(1/24)$ given by Equations 20 and 21 for $t = 1/24$.

For $5/24 < t < 31/24$ Equations 18 and 19 hold, and thus for $t$ in this range, we have,

$$N_0(t) = N_0(5/24) e^{-\lambda_0{}^r (t - 5/24)} \qquad (20''')$$

$$N_i(t) = \sum_{q=0}^{i} N_q(5/24) \left(\prod_{j=q}^{i-1} \lambda_j{}^r\right) \sum_{n=q}^{i} \frac{e^{-\lambda_n{}^r (t - 5/24)}}{\prod_{\substack{p=q \\ p \neq n}}^{i} (\lambda_p{}^r - \lambda_n{}^r)}$$
$$i = 1, 2, \ldots k \qquad (21''')$$

The value of $\lambda_i{}^b$ is based on experimental values which determine the amount of an isotope absorbed into the blood stream. Let $f_i$ be the fractional amount of an isotope, $i$, absorbed into the blood stream. Then $\lambda_i{}^b$ is found by solving

$$f_i = e^{-\lambda_i{}^r} \int_0^{4/24} \lambda_i{}^b e^{-\lambda_i t} dt = \frac{\lambda_i{}^b}{\lambda_i} e^{-\lambda_i{}^r} (1 - e^{-\lambda_i 4/24})$$
$$= \left(1 - \frac{\lambda_i{}^r}{\lambda_i}\right) e^{-\lambda_i{}^r} (1 - e^{-4/24\ \lambda_i}) \qquad (22)$$

or,

$$1 - \frac{\lambda_i{}^r}{\lambda_i}\left(1 - e^{-4/24\ \lambda_i}\right) = e^{\lambda_i{}^r} f_i \text{ for } \lambda_i = \lambda_i{}^r + \lambda_i{}^b$$

Because of uncertainty in the experimental values of $f_i$, and since our assumptions are not precisely fulfilled, it may happen that

$$e^{\lambda_i{}^r} f_i > 1$$

and the equation does not apply. When $e^{\lambda_i{}^r} f_i$ exceeded 0.95, the latter value was used in place of

$$e^{\lambda_i{}^r} f_i$$

in Equation 22.

The MPI to give a total damage of 0.3 rem to any given portion of the GI tract can now be readily calculated. The expression,

$$f \sum_{i=0}^{k} N_i(t)\lambda_i^r \Sigma(bE)_i(\mathrm{RBE})_i\, dt \qquad (23)$$

represents the total effective energy released in the GI tract in time, $dt$, due to the inhalation of one atom of parent. The fraction, $f$, of inhaled atoms arriving in the stomach is taken as 0.625. During the first hour, the radioisotopes are assumed to be in the stomach and so the rem dose to the stomach per atom of parent inhaled is given by,

$$\text{Dose to stomach/atom} = \frac{1.602 \times 10^{-8} f}{2\,m} \int_0^{1/24} \sum_{i=0}^{k} N_i(t)\, \lambda_i^r (bE)_i(\mathrm{RBE})_i\, dt \qquad (24)$$

A factor of 1/2 has been used since the stomach wall is only irradiated from one side. Then the MPI in $\mu$c is given by the equation,

$$\mathrm{MPI}_{stomach} = \frac{0.3\, m\lambda_0^r}{25.6\, f \int_0^{1/24} \sum_{i=0}^{k} N_i(t)\, \lambda_i^r (bE)_i(\mathrm{RBE})_i\, dt} \times \frac{\lambda_0^r\, t_1}{(1 - e^{-\lambda_0^r t_1})} \qquad (25)$$

$$\mathrm{MPI}_{stomach} = \frac{1.17 \times 10^{-2}\, m}{f T_0^r \int_0^{1/24} \sum_{i=0}^{k} \frac{N_i(t)\,(bE)_i(\mathrm{RBE})_i}{T_i^r}\, dt} \times \frac{0.231}{T_0^r(1 - e^{-0.231/T_0^r})} \qquad (26)$$

The fraction on the left of (25), (26) and (28) gives the number of $\mu$c actually inhaled and the fraction on the right corrects for radioactive decay before entering the body. Thus MPI is the equivalent amount at time $t = 0$. For other portions of the GI tract, we return to Equation 23 and assume that the energy released in time, $dt$, is absorbed in the section of the GI tract through which the isotope moved during time, $dt$. If $\tau$ is the total time in the section of the GI tract considered, the energy released in time, $dt$, is absorbed in a mass of $mdt/\tau$ and the rem dose per atom of parent at the site in the intestine is given by,

$$\text{Dose to intestine/atom} = \frac{1.602 \times 10^{-8} f\tau}{2\,m} \times \sum_{i=0}^{k} N_i(t)\, \lambda_i^r (bE)_i(\mathrm{RBE})_i \qquad (27)$$

The number of $\mu$c of parent which deliver 0.3 rem to the site in the intestine is given by,

$$\mathrm{MPI}_{intestine} = \frac{1.17 \times 10^{-2}\, m}{f\tau T_0^r \sum_{i=0}^{k} \frac{N_i(t)\,(bE)_i(RBE)_i}{T_0^r}} \times \frac{0.231}{T_0^r(1 - e^{-0.231/T_0^r})} \qquad (28)$$

The $\mu$c of parent which may be inhaled without exceeding the dose of 0.3 rem in any given portion of the GI tract is then the minimum of Equations 26 and 28.

$$\mathrm{MPI}_{GI} = \text{minimum } [\mathrm{MPI}_{stomach}, \mathrm{MPI}_{intestine}]$$

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# Health Protection of Workers Exposed to Ionizing Radiations

**By A. A. Letavet, USSR**

The health protection of people who may be subjected to the action of ionizing radiations, while engaged in work devoted to the use of atomic energy for peaceful purposes, is ensured in the USSR by the following system of national measures:

1. Legislation regarding the maximum permissible levels of irradiation and concentration of radioactive isotope content in the air and water.
2. Legislation regarding the duration of the working day and vacations.
3. Publication of compulsory hygienic rules for all kinds of work performed under conditions of ionizing radiations.
4. The setting up of state control over the observance of laws promulgated in this realm.
5. Organization of medical service aimed at systematically supervising the state of health of the workers in question.

The hygienic standards of the maximum permissible levels of external irradiation, as well as of concentrations of radioactive isotopes in the air and water are issued by the Ministry of Health of the USSR. The established standards have the force of law to be abided by every institution and enterprise.

The Ministry of Health of the USSR has ruled that the dose of 0.05 roentgen (or 0.05 of physical equivalent of roentgen for beta-irradiation) in a working day is the maximum permissible dose of exterior gamma-radiation. This standard conforms with the recommendation of the International Radiological Congress held in London in 1950.

However, this international recommendation, as well as recommendations of various scientific organizations and committees in some countries (the USA, Britain, and others) accepts a dose of 0.3 roentgen a week (or a physical equivalent of a roentgen) as an equivalent standard. We believe it is preferable to accept the daily dose of 0.05 roentgen in a working day as the basic standard. Here we proceed from the general concept drawn from the findings of many investigations carried out in the USSR and abroad that finely fractioned doses of irradiation lead to better restoration and, consequently, are of less danger than concentrated doses. In line with this thesis accepted in the USSR, all preventive measures (protection, the fixing of a permissible working day for certain conditions, etc.) are designed so as not to exceed the daily dose of 0.05 roentgen. A summary weekly dose of 0.3 roentgen is used only as an exception, in rare cases, when the conditions or nature of the work do not permit the observance of the fixed daily dose.

Table I shows the coefficients accepted in the USSR of the relative biological effectiveness of some kinds of radiations and the corresponding admissible daily doses of irradiations in physical equivalents of a roentgen.

In order to prevent the harmful effect of radioactive substances penetrating into the body, maximum permissible levels of radioactive isotopes in the air and water have been established. Such levels of concentration have been fixed by the Ministry of Health for all widely used radioactive isotopes. The list of maximum permissible levels of concentrations is enlarged and perfected by the Ministry of Health as the application of these isotopes extends to new fields, and as more scientific data is accumulated concerning their action.

The maximum permissible concentration of radioactive substances in the air and water have been computed on the basis of: (1) experiments on animals for the absorption, distribution, removal and action on the body of individual radioactive isotopes; and (2) data on the physical properties of isotopes—the type of disintegration, the energy of ionizing particles, the half-life, as well as the solubility of compounds and their dispersions.

In experiments on animals particular attention was given to the resorption of radioactive isotopes from the gastrointestinal tract and the lungs, their distribution among the organs, the rate of elimination, and the setting up of an equilibrium between the intake and elimination. Account was also taken of the natural intake, the content and exchange of stable bioelements in the body of man and animals,

Table I

| *Type of radiation* | *Coefficient of relative biological effectiveness* | *Maximum permissible daily doses of irradiation in physical equivalents of roentgen* |
|---|---|---|
| Gamma and roentgen rays | 1.0 | 0.05 |
| Beta-radiation | 1.0 | 0.05 |
| Alpha-radiation | 10.0 | 0.005 |
| Thermal neutrons | 5.0 | 0.01 |
| Fast neutrons (up to 20 Mev) | 10.0 | 0.005 |
| Fast neutrons (over 20 Mev) | 20.0 | 0.0025 |

Original language: Russian.

i.e., the natural mineral exchange. In the case of radioactive substances and their compounds which are with difficulty resorbed from the gastrointestinal tract, account was taken of the specific action of radiations on the intestinal tract itself in the presence of radioactive substances, with due regard to the peculiarities of the neuro-receptive apparatus of the digestive organs.

In the application of the experiments on animals to the solution of the problem under consideration, various factors were taken into account such as the method of injecting radioactive substances into the body, the duration of the experiments, the intensity of metabolism in various kinds of animals, their radiosensitivity, and longevity, as well as the difference of content of chemical elements in the tissues of man and experimental animals.

With the same object in view, experiments were conducted on long-term effects of small doses of radioactive substances.

With all these data available, a quantitative calculation of the maximum permissible concentrations was made, with the provision that with the maximum concentration of the radioactive element (equilibrium condition), the daily dose of irradiation should not exceed 0.05 of the biological equivalent of a roentgen in any part of the tissue of the human body as well as the gastrointestinal tract.

The computation of maximum permissible concentrations for radioactive elements whose stable isotopes are bioelements of the human body can be made on the basis of the maximum concentration of stable isotopes in the human tissues and their daily intake with food.

A comparison of maximum permissible levels of concentrations of radioactive isotopes in the air and water required in the USSR with the standard of international recommendations shows that they do not differ considerably. This can be seen from Table II illustrating the maximum permissible concentrations in water for several isotopes.

The next table shows the maximum permissible concentrations of some radioactive isotopes in the air and water, required in the USSR (Table III).

Higher hygienic requirements are in force with regard to drinking water of centralized water supplies, as well as with regard to atmospheric air of residential sections of towns and workers' settlements.

Table II

| *Element* | *USSR: maximum permissible concentration for water, in curies per liter* | *Standards recommended by the Radiological Congress in London in 1950, in curies per liter* |
|---|---|---|
| $Ra^{226}$ | $5 \times 10^{-11}$ | $4 \times 10^{-11}$ |
| $I^{131}$ | $0.5 \times 10^{-8}$ | $3 \times 10^{-8}$ |
| $Co^{60}$ | $5 \times 10^{-8}$ | $1 \times 10^{-8}$ |
| $P^{32}$ | $1 \times 10^{-7}$ | $2 \times 10^{-7}$ |
| $Na^{24}$ | $1 \times 10^{-5}$ | $0.8 \times 10^{-5}$ |
| $Sr^{90} \rightarrow Y^{90}$ | $1 \times 10^{-9}$ | $0.8 \times 10^{-9}$ |

Table III*

| *Element* | *Maximum permissible concentrations in curies per liter* | |
|---|---|---|
| | *Air* | *Water* |
| $Ra^{226}$ | $1 \times 10^{-14}$ | $5 \times 10^{-11}$ |
| $Rn^{222}$ | $1 \times 10^{-11}$ | $1 \times 10^{-9}$ |
| $Sr^{90} \rightarrow Y^{90}$ | $1 \times 10^{-12}$ | $1 \times 10^{-9}$ |
| $I^{131}$ | $5 \times 10^{-12}$ | $5 \times 10^{-9}$ |
| $Sr^{89}$ | $1 \times 10^{-11}$ | $1 \times 10^{-8}$ |
| $Ca^{45}$ | $5 \times 10^{-11}$ | $5 \times 10^{-8}$ |
| $Ba^{140} \rightarrow La^{140}$ | $5 \times 10^{-11}$ | $5 \times 10^{-8}$ |
| $P^{32}$ | $1 \times 10^{-10}$ | $1 \times 10^{-7}$ |
| $Cs^{137} \rightarrow Ba^{137}$ | $5 \times 10^{-10}$ | $5 \times 10^{-7}$ |
| $Zr^{95} \rightarrow Nb^{95}$ | $5 \times 10^{-10}$ | $5 \times 10^{-7}$ |
| $Ag^{110}$ | $5 \times 10^{-10}$ | $1 \times 10^{-6}$ |
| $C^{14}$ | $5 \times 10^{-9}$ | $1 \times 10^{-6}$ |
| $Mn^{56}$ | $1 \times 10^{-9}$ | $1 \times 10^{-6}$ |
| $S^{35}$ | $5 \times 10^{-9}$ | $1 \times 10^{-5}$ |
| $Na^{24}$ | $5 \times 10^{-9}$ | $1 \times 10^{-5}$ |

* Maximum permissible concentrations of radioactive substances are for the air of working premises in the form of gases and aerosols and for the water of open pools used for non-centralized water supplies.

Soviet legislation provides for special privileges, such as a longer paid holiday and a shorter working day for kinds of work involving exposure to ionizing radiation of higher intensity, though still below the maximum permissible dose.

In assessing 0.05 roentgen or physical equivalent of roentgen as the maximum permissible dose of irradiation both for the USSR and as internationally recommended, one should bear in mind that this dose still exceeds the natural radiation background more than a hundredfold; this follows from the assumption that the most likely daily dose of the background amounts to 0.0004 of the physical equivalent of a roentgen.

Hence, from the viewpoint of maximum concern for the health of people subjected to the action of ionizing radiations, it is advisable to resort to a reduced working day and longer annual paid vacations, along with technical measures aimed at the further reduction of the dose of radiation at the place of work. The object of these measures is to improve and to ensure a more complete process of recovery, which is of signal importance for the preservation of health.

Here are some instances from Soviet legislation dealing with the working day and longer paid vacations for some categories of workers subject to the action of ionizing radiations.

A six-hour working day has been fixed for every kind of work at nuclear reactors, at cyclotrons, betatrons and other accelerating units; for work connected with the preparation and operation of neutron sources; for work on the gamma defectoscopy of metals and various articles (at both portable and stationary units); for operations dealing with the use of artificial radioactive isotopes in an open form, possessing either a considerable half-life or unfavorable conditions of distribution in the body, when

present in quantities over 1 millicurie per working place (when using other isotopes—in quantities over 10 millicuries); in the case of industrial use of radioactive luminous substances of constant action; and when preparing radon baths, etc. The working day is reduced to 5 hours under specific conditions of treating patients with gamma therapy at medical establishments. It is worth mentioning that a shorter working day does not involve a reduced salary.

A longer paid vacation of 24 days is granted to everyone engaged in work dealing with radioactive isotopes in quantities over 1 millicurie at the working place, servicing stationary units for gamma defectoscopy, or engaged in the preparation of radon baths. A 30-day paid vacation is granted to those working at nuclear reactors, preparing neutron sources, operating accelerating units, cyclotrons, betatrons, etc.; working at portable units for gamma defectoscopy or at places where luminous substances of constant action are used; and to medical staff treating patients with gamma therapy, and others.

It is expedient to grant facilities, such as a shorter working day or a longer annual vacation, in cases when the average total dose of radiation for some reason or other exceeds approximately tenfold the natural background of ionizing radiations, despite the application of modern protective techniques.

In the case of widespread kinds of work, when radioactive products are used in industry, in scientific, medical and other establishments, the State Sanitary Inspection of the Ministry of Health of the USSR issues special hygienic rules aimed at fully preventing the harmful action of radioactive products on the human body. To date such rules have been issued for: (1) work dealing with radioactive isotopes; (2) industrial gamma defectoscopy; (3) work connected with radioactive luminous substances; and (4) medical establishments.

The rules contain detailed instructions concerning the storage and handling of radioactive products, the building and equipment of premises where work with radioactive substances is performed, protection from radiation, the supply of clean air, measures dealing with personal hygiene, etc.

The rules are compulsory for every chief of enterprises and offices where radioactive products are used. The observance of these rules and standards is checked up by the authorities of state industrial and sanitary supervision. In case of violation of the rules or standards, the representative of the supervisory authorities is empowered to apply sanctions depending on the nature of the violation: (1) to impose a fine on the management of the enterprise or office; (2) to have the department or laboratory closed down; and (3) in cases of gross violations, to sue in court.

In addition to current supervision of the observance of the required hygienic rules and standards at operating enterprises, the Soviet State Sanitary Inspection also effects so-called preventive Sanitary Supervision, i.e., passes judgment on designs of factories, new or reconstructed, as far as hygienic requirements and standards are concerned. Hence, no industrial enterprise or laboratory using radioactive substances can be commissioned without the approval by a competent committee of physicians and physicists on the observance of safety rules.

Properly designed protection from gamma and neutron radiation is of uppermost importance in ensuring safe conditions of work at various enterprises and laboratories using radioactive products.

Up to recently tables have been recommended in Soviet and foreign publications for protection from gamma radiations comprising the following quantities: the activity of the source and the energy of gamma rays (for instance, the well-known Hammertsfelder table). The general shortcoming of these tables was their lack of accuracy since the multiple scattering of gamma rays in the protective facilities was not taken into consideration, as well as the limited sphere of their application.

In the USSR "Universal tables for calculating protection from gamma rays" have been prepared, which are to a considerable extent devoid of the above shortcomings. The basic quantities in these tables are factor of reduction and the energy of gamma rays. The tables are in accordance with the theory of gamma ray reduction in a wide beam for major building materials: lead, iron, concrete, and water. An experimental check up of the tables has produced quite satisfactory results in the case of both monochromatic sources and those with a complex spectrum of gamma rays. The table is universal in that it helps to calculate promptly and easily the protection from gamma rays within 0.1 to 10 Mev of energy under any conditions which may be encountered in practice; for instance, to ensure a physical dose of any magnitude, for any distance from the source, for sources of various shapes and various specific activities, for parallel, slanting and radial beams, etc. The tables may be used to calculate protection from gamma rays from monochromatic sources as well as sources spreading a complex spectrum of gamma rays.

By way of illustration, universal tables (Tables IV and V) for calculating protection from gamma rays for lead are attached to this report.

Convenient nomograms for rapidly calculating protection have also been prepared for separate isotopes and their mixtures. A sample of such a nomogram (Fig. 4) for calculating protection from gamma radiation of radioactive cobalt is given in the addenda. The latter comprises a table of conversion coefficients, which makes it possible to use the nomogram for radioactive cobalt for finding the degree of protection from gamma rays of any other isotopes within a range of energy from 0.1 to 10 Mev.

Great significance is attached to ventilation facilities so as to ensure purity of the air. It should, however, be pointed out that the high degree of purity

**Table IV. Protection in Terms of Reduction Factor Using Lead (Wide Parallel Beam). Thickness, x, in Terms of mm of Lead of 11.34 gm/cm³ Density**

| *Reduction factor, K* | *Energy of gamma rays in Mev* | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | *0.1* | *0.2* | *0.3* | *0.4* | *0.5* | *0.6* | *0.7* | *0.8* | *0.9* | *1* | *1.25* | *1.5* | *1.75* | *2.0* | *2.2* | *3* | *4* | *6* | *8* | *10* |
| 1.5 | 0.5 | 1 | 1.5 | 2 | 2 | 3 | 4 | 6 | 7 | 8 | 9 | 11 | 12 | 12 | 12 | 13 | 12 | 10 | 9 | 9 |
| 2 | 1 | 2 | 3 | 4 | 5 | 7 | 8.5 | 10 | 11 | 13 | 15 | 17 | 18 | 20 | 20 | 21 | 20 | 16 | 15 | 14 |
| 5 | 2 | 4 | 6 | 9 | 11 | 15 | 19 | 22 | 25 | 28 | 34 | 38 | 41 | 43 | 44 | 46 | 45 | 38 | 33 | 30 |
| 8 | 2 | 5 | 8 | 11 | 15 | 19 | 23 | 28 | 32 | 35 | 42 | 48 | 52 | 55 | 57 | 59 | 58 | 50 | 43 | 38 |
| 10 | 3 | 5.5 | 9 | 13 | 16 | 21 | 26 | 30 | 35 | 38 | 45 | 51 | 56 | 59 | 61 | 65 | 64 | 55 | 49 | 42 |
| 20 | 3 | 6 | 11 | 15 | 20 | 26 | 32 | 38.5 | 44 | 49 | 58 | 66 | 72 | 76 | 78 | 83 | 82 | 71 | 63 | 56 |
| 30 | 3.5 | 7 | 11.5 | 17 | 23 | 30 | 36 | 43 | 50 | 55 | 65 | 73 | 80 | 85 | 88 | 93 | 92 | 80 | 72 | 63 |
| 40 | 4 | 8 | 13 | 18 | 24 | 31 | 38 | 45 | 52 | 58 | 69 | 78 | 86 | 91 | 94 | 100 | 99 | 87 | 78 | 68 |
| 50 | 4 | 8.5 | 14 | 19 | 26 | 32 | 39 | 46 | 53 | 60 | 72 | 82 | 90 | 96 | 100 | 106 | 110 | 92 | 83 | 73 |
| 60 | 4.5 | 9 | 14.5 | 20 | 27 | 34 | 42 | 49 | 56 | 63 | 75 | 86 | 95 | 101 | 104 | 110 | 109 | 97 | 87 | 77 |
| 80 | 4.5 | 10 | 15.5 | 21 | 28 | 37 | 45 | 53 | 60 | 67 | 80 | 92 | 101 | 107 | 111 | 117 | 116 | 104 | 94 | 82 |
| 100 | 5 | 10 | 16 | 23 | 30 | 38 | 47 | 55 | 63 | 70 | 84 | 96 | 106 | 113 | 117 | 122 | 121 | 109 | 99 | 87 |
| 200 | 6 | 12.5 | 19 | 26 | 34 | 44 | 53 | 63 | 72 | 80 | 96 | 111 | 122 | 129 | 133 | 140 | 138 | 126 | 114 | 102 |
| 500 | 6.5 | 14 | 22 | 31 | 40 | 51 | 61 | 72 | 82 | 92 | 113 | 129 | 142 | 150 | 154 | 163 | 161 | 149 | 133 | 119 |
| 1000 | 7 | 15 | 24 | 33 | 44 | 57 | 69 | 81 | 92 | 102 | 123 | 141 | 155 | 165 | 170 | 180 | 178 | 165 | 151 | 133 |
| 2000 | 8.5 | 17 | 27 | 38 | 50 | 63 | 76 | 88 | 100 | 111 | 135 | 154 | 168 | 179 | 185 | 197 | 195 | 181 | 166 | 148 |
| 5000 | 9 | 19 | 30 | 42 | 55 | 70 | 85 | 99 | 112 | 124 | 149 | 170 | 186 | 198 | 205 | 219 | 217 | 203 | 185 | 166 |
| 8000 | 10 | 20 | 31.5 | 44 | 57 | 73 | 90 | 104 | 118 | 130 | 158 | 180 | 196 | 208 | 215 | 130 | 229 | 215 | 196 | 175 |
| $10^4$ | 10.5 | 21 | 33 | 46 | 59 | 75 | 91 | 106 | 120 | 133 | 161 | 183 | 201 | 213 | 221 | 235 | 234 | 220 | 201 | 180 |
| $2 \times 10^4$ | 11 | 22 | 35 | 48 | 63 | 80 | 97 | 113 | 128 | 142 | 172 | 195 | 213 | 227 | 235 | 251 | 250 | 236 | 217 | 195 |
| $5 \times 10^4$ | 11.5 | 23.5 | 37 | 52 | 69 | 87 | 105 | 123 | 140 | 156 | 188 | 214 | 233 | 247 | 255 | 273 | 272 | 258 | 237 | 215 |
| $1 \times 10^5$ | 11.5 | 24 | 38 | 54 | 72 | 92 | 111 | 130 | 148 | 165 | 201 | 227 | 247 | 262 | 270 | 289 | 289 | 275 | 253 | 229 |
| Correction for slant rays* $\epsilon =$ | 0.17 | 0.85 | 2.10 | 3.98 | 5.80 | 7.30 | 8.9 | 10.1 | 11.6 | 12.6 | 15.3 | 17.3 | 18.7 | 19.6 | 20.0 | 21.8 | 21.2 | 19.6 | 18.2 | 16.7 |

* The correction for "slant rays" is introduced only for very remote sources seen at angle $2\psi$ or from point sources but measured by an extended detector. This correction should be deducted from quantity $x$ obtained from this table. The magnitude of the correction equals $\Delta x_0 = d_0/\mu_0 = \epsilon\, d_0$, where $\epsilon$ is given on the last line of this table and $d_0$ in Table V.

Table V*

| $K$ | $\psi = 15°$ $d_0$ | $\psi = 30°$ $d_0$ | $\psi = 45°$ $d_0$ | $\psi = 60°$ $d_0$ | $\psi = 75°$ $d_0$ | $\psi = 90°$ $d_0$ |
|---|---|---|---|---|---|---|
| 1.5 | 0.012 | 0.03 | 0.06 | 0.10 | 0.17 | 0.27 |
| 2 | 0.016 | 0.05 | 0.11 | 0.18 | 0.29 | 0.42 |
| 5 | 0.036 | 0.11 | 0.24 | 0.41 | 0.62 | 0.80 |
| 8 | 0.050 | 0.14 | 0.30 | 0.50 | 0.75 | 0.95 |
| 10 | 0.057 | 0.15 | 0.33 | 0.55 | 0.80 | 1.02 |
| 20 | 0.071 | 0.20 | 0.44 | 0.70 | 0.99 | 1.23 |
| 30 | 0.083 | 0.23 | 0.49 | 0.79 | 1.08 | 1.31 |
| 40 | 0.092 | 0.24 | 0.52 | 0.83 | 1.13 | 1.37 |
| 50 | 0.096 | 0.25 | 0.54 | 0.87 | 1.17 | 1.42 |
| 60 | 0.105 | 0.27 | 0.57 | 0.91 | 1.22 | 1.47 |
| 80 | 0.114 | 0.28 | 0.60 | 0.97 | 1.28 | 1.52 |
| 100 | 0.118 | 0.29 | 0.63 | 1.00 | 1.32 | 1.56 |
| 200 | 0.128 | 0.34 | 0.71 | 1.10 | 1.43 | 1.68 |
| 500 | 0.162 | 0.41 | 0.82 | 1.25 | 1.56 | 1.82 |
| 1000 | 0.176 | 0.45 | 0.89 | 1.32 | 1.66 | 1.92 |
| 2000 | 0.186 | 0.48 | 0.94 | 1.39 | 1.72 | 1.99 |
| 5000 | 0.215 | 0.54 | 1.04 | 1.49 | 1.83 | 2.10 |
| 8000 | 0.231 | 0.58 | 1.09 | 1.55 | 1.89 | 2.17 |
| $10^4$ | 0.240 | 0.59 | 1.10 | 1.57 | 1.91 | 2.19 |
| $2 \times 10^4$ | 0.254 | 0.61 | 1.15 | 1.62 | 1.97 | 2.26 |
| $5 \times 10^4$ | 0.266 | 0.66 | 1.23 | 1.71 | 2.06 | 2.34 |
| $10^5$ | 0.279 | 0.69 | 1.28 | 1.76 | 2.12 | 2.40 |

* This table is intended to determine the corrections for the effect of "slant rays". The table comprises the values $\Delta x_0 = d_0/\mu_0 = ed_0$, where $\mu_0$ is the coefficient of attenuation for a narrow beam, and $\Delta x_0$ is the magnitude of the correction. The correction is always deducted from the basic value of thickness $x$ obtained from Table IV. In this table, $K$ is reduction factor; $\psi$ is half the angle of visibility of the source from the observation point or of detector from the point source as given in Table IV.

of the air required in compliance with the standards of maximum permissible levels of radioactive substances in the air cannot be achieved unless the ventilation facilities are planned as a unit with hermetically sealed equipment, convenient for operation, and well planned premises. Unless the latter factor is observed, the ventilation system alone is incapable of providing the required cleanliness of the air.

Hermetic cubicles with manipulating appliances or rubber gloves built in the front walls are widely used for the protection of the air. The cubicles are ventilated in a closed system: the incoming air is fed through a separate main of air ducts, while the exhausted contaminated air is purified by an individual filter in the cubicle. It is such purification by individual filters which prevents the system of exhaust air ducts and ventilation pipes from being contaminated.

To prevent leakage, a vacuum of 5 to 50 mm of $H_2O$ is maintained in the cubicles.

When working with small quantities of radioactive substances, the area of apertures in the fume hood of the non-hermetic type (chemical) is reduced to the minimum. As in the hermetically sealed cubicles, rubber gloves for the operators are mounted into the front walls. High air velocity—from 1.0 to 2.0 meters a second, depending on the nature of radioactive substances and the kind of work, are provided for in the apertures to prevent the flow of radioactive substances into the air of the workers' premises.

Special individual table-vacuum-stands provided with a shelter and local ventilation exhausts are used for the work of applying radioactive luminous paints, which fully guarantees against the pollution of the air of the premises by radon.

The incoming air, preliminarily cleaned at the filters, is supplied primarily to the working zone.

Particular attention is given to the protection of the atmospheric air. The air removed through ventilation systems is purified at various dry and moist filters and then released through stacks high into the atmosphere.

Along with general prophylactic measures for work connected with radioactive substances, providing for the necessary equipment of the premises, protecting arrangements, ventilation, etc., much attention is paid in the USSR to the problems of individual protection of the operators, and notably to protective garments.

For all kinds of work connected with radioactive substances, the personnel is provided free of charge with all the necessary protective garments and other protective means (overalls, smocks, gloves, armlets, aprons, special pneumosuits, respirators, etc.).

For work connected with small quantities of radioactive substances, smocks, overalls and short overalls made of white cotton fabric with a maximum smooth surface, like moleskin and sateen, are used as protective garments. Besides, utmost attention is given to simplifying the design of the garments so as to facilitate the subsequent cleaning from radioactive contamination.

For the protection of the arms and hands, rubber or plastic gloves are used, as well as armlets made of plastic film.

Special short overalls or aprons with armlets made of various plastic materials of Soviet make are put on on top of the cotton garments when the work is connected with radioactive substances in greater quantities. The designs (Figs. 1–2) provide fuller protection of the surface of the body from radioactive substances, as well as from acids and alkali. The seams made by means of high frequency welding lend uniformity to the plastic material and ensure hermetic sealing; and at the same time make possible the use of effective methods of decontamination by means of acids and alkalis.



Figure 1

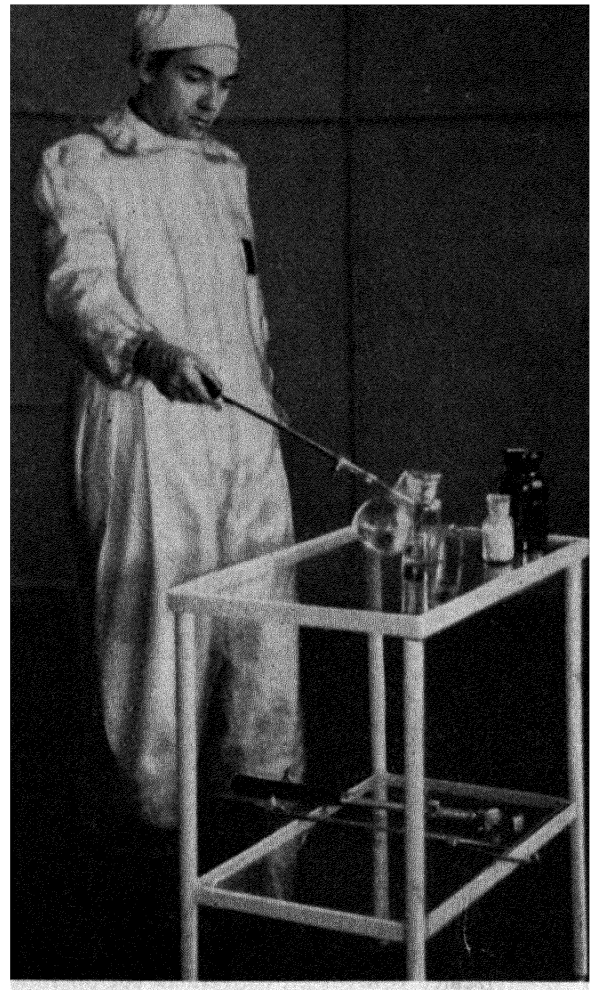


Figure 2

Special air-pumped protective suits are used for repair work and in cases when for some reason or other the air of the building has been contaminated with radioactive substances.

These so-called pneumatic suits are fabricated of various Soviet-made plastic materials. Figure 3 presents a general view of the pneumatic suit.

The pneumatic suit consists of three main parts: (1) the headgear—light transparent helmet of clear plastic; (2) overalls made of plastic films of special design; and (3) air supplying parts—hoses and forcing fans, portable or stationary.

The helmet we use has the following advantages over the various types of gas-masks and respirators: the respiratory organs of the worker are altogether isolated from the surrounding air containing radioactive impurities, good visibility is ensured and the field of vision is not limited; there is no squeezing of the head or irritation of the skin on the face. The helmet is so designed that air may also be delivered under the overalls, which makes for the normal thermo-regulation through evaporation.

The design of the film overalls ensures full protection of the surface of the body from radioactive impurities. At the same time the plastic material of

the overalls, as well as the helmets, are easily cleansed from any radioactive material which may contaminate them outside.

For the most favorable conditions of work in a pneumatic suit, 150–200 liters of air per minute are required. Then the operator does not feel the lowered heat yield caused by evaporation, and the incoming air does not excessively blow the suit. The amount of incoming air is quite sufficient to maintain a certain counterpressure (25–30 mm $H_2O$) so as to prevent the penetration under the suit of air contaminated by radioactive substances.

Examinations made of physiological alterations in the body of an operator working in a pneumatic suit have shown that quite a favorable microclimate of its own is created in the pneumatic suit; this enables the operator to remain for a long time in contaminated atmosphere without injuring his health.

In some cases filtering dust respirators furnished with high efficiency filters designed for high-dispersion aerosols are used for work of short duration under conditions of possible air contamination with radioactive aerosols.

Use is made of various conventional types of tongs, holders, clamps and similar tools aimed at increasing the distance between the radioactive substance and the operator when the latter deals with small quantities of radioactive substances.



Figure 3

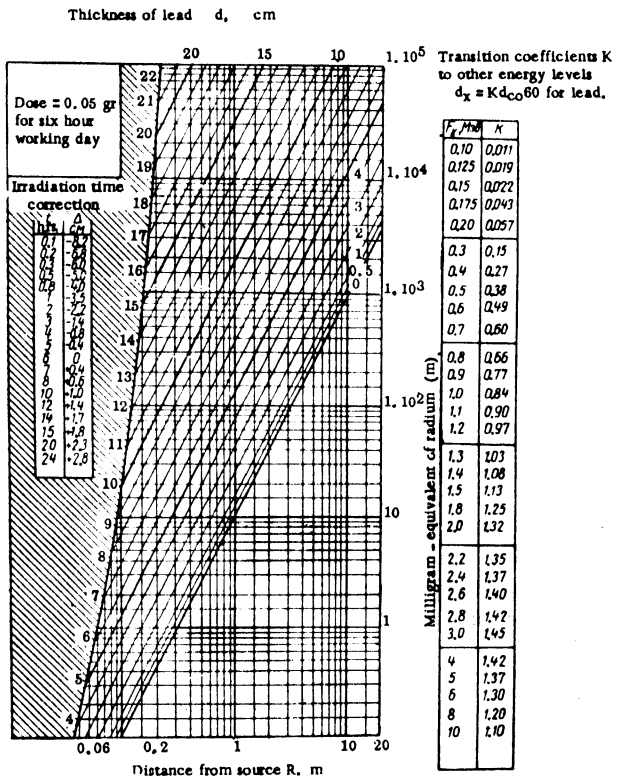



Figure 4. Nomogram: lead shielding from gamma-radiation

Manipulators varying in complexity are used for large quantities of radioactive substances. The design of such appliances is technically outside the medical and biological sphere and therefore there is no need to dwell on it.

Mention should be made of work performed with the help of very simple manipulators under water. This method is applied when packing radioactive cobalt and it provides quite a safe protection for the operator.

The reactors are operated automatically. Parts are replaced with the help of remote control by operators completely protected from radiation by thick walls. Various optical appliances are used to supervise the operations.

Dosimetric control is in use at enterprises and institutions where radioactive substances are used. A special dosimetric service headed by the chief engineer is instituted at enterprises dealing with considerable quantities of radioactive substances; at enterprises with a small volume of such work, dosimetric control is effected by safety inspectors.

General control over labor conditions, including radiation safety, is, as has been pointed out, effected by state industrial-sanitary supervisory organs.

The task of dosimetric control is to keep a record of working conditions and the dangerous action of radioactive radiations on the body.

Dosimetric control is carried out by means of numerous dosimetric instruments produced by Soviet

industry. Measurements with the use of these instruments are taken according to uniform methods and instructions.

Dosimetric measurements are effected by means of portable as well as stationary units including those automatically operated. The range of work of the dosimetric instruments is as follows: (1) control over gamma fields at the place of work; (2) determination of air contamination by active gases or aerosols; (3) analyzing the degree of contamination of working surfaces, the operator's body, clothing and footwear; and (4) determining pollution of water in the sewers and water pools.

Individual dosimetric control with the use of photographic films and pocket electric counters with indicator scales is employed to determine the total dose of radiation to which individual workers are subjected in a definite period of time.

Various types of radiotechnical apparatus have been designed in the USSR to solve the numerous problems of dosimetry and radiometry of ionizing radiation.

Representatives of dosimetric service or competent safety inspectors carry out systematic supervision of every kind of work where the operator may be subjected to the action of ionizing radiations. If repair or emergency work is to be performed in conditions of an intensified gamma field or air contamination, the representative of dosimetric service or an authorized representative issues to the operators a special permit indicating the period of time during which the latter may remain in the dangerous zone without being overexposed. The permit also specifies the proper outfit required for this kind of work. The dosimetric service inspector is authorized to stop the work if these rules, as well as other safety requirements, are violated. The most important kinds of work are performed under the supervision and control of a dosimetric inspector.

Soviet medical service devotes much attention to regular inspection of the health of every worker who may be subjected to the action of ionizing radiation in his pursuits connected with the peaceful use of atomic energy.

Systematic compulsory examinations carried out by medical establishments every 6 or 12 months form a major part of the system of public health service. The timing of these examinations, depending on the amount of the radioactive substances used and the nature of work, is approved by the Ministry of Health and is obligatory for all those concerned.

The aim of periodical medical examinations is to detect in time the beginning of occupational diseases, and the development of other diseases incompatible with work in conditions of ionizing radiations.

At important enterprises periodical medical inspections are made by medical centers functioning there and called medico-sanitary departments. At small enterprises, as well as at scientific research institutions and laboratories not provided with special medico-sanitary service, such examinations are made by the nearest medical-prophylactic establishments to which the given enterprise is attached for medical service.

The medical-prophylactic institution making medical examinations keeps a special record of people who have to deal with radioactive substances.

The following specialized physicians take part in the medical examinations: a therapist, a neuropathologist, a gynecologist; when necessary a dermatologist or ophthalmologist is consulted.

In every case without exception a complete blood test is taken: the number of erythrocytes, leukocytes and haemoglobin, the leukocyte formula, the number of thrombocytes and reticulocytes, determination of blood coagulation and the duration of bleeding. In the morphological blood test much attention is also paid to the qualitative characteristics of the blood cells: the size of erythrocytes, the change in their form, neutrophil fragments, cytolysis, etc.

Persons inhaling radioactive gas (radon) undergo roentgenography of the lungs not less than once a year besides general clinical examinations.

Particular attention is paid to neurological examinations, since, according to many Soviet investigators, the nervous system responds particularly early to the action of ionizing radiation.

After every medical examination the physician of the establishment responsible for making inspections jointly with the members of the medical commissions, formulates his opinion on the condition of the person under examination, specifying the necessary medical and prophylactic measures. Their findings are entered into a special individual ledger kept in the institution, or are sent to the respective medical establishment if the worker in question has changed his place of work or residence. This system of regular medical inspections over a number of years makes it possible to accumulate comprehensive data on the possible effect of minute doses of ionizing radiations, which individual experiments fail to yield.

Treatment at night sanatoriums is widely applied in the USSR, aimed at timely prevention of general and occupational diseases of the workers. A night sanatorium is a medical-prophylactic establishment where a worker, while pursuing his occupation, is provided after work with the necessary regimen and medical treatment (dietetic food, physio-therapeutic procedures, medicinal therapy) which would have been impossible under conditions of conventional outpatient treatment. Accommodation at a night sanatorium is free of charge. The period of treatment depending on the medical indications varies from two to four weeks.

Persons working under conditions of ionizing radiation are recommended to spend their annual vacation which, as has been mentioned, is prolonged, at sanatoriums and rest homes. Experience has shown that a visit to a sanatorium or a rest home is very effective in eliminating the changes which with

some probability might be ascribed to the action of ionizing radiations.

Sanatorium and rest home accommodations are provided at a moderate price (30% is paid by the worker, and 70% from the social insurance fund).

Research work dealing with problems of industrial hygiene under conditions of ionizing radiations is conducted in the USSR at scientific institutes of industrial hygiene and occupational diseases; such institutes, with corresponding laboratories and departments, function in various towns. They are engaged in experimental investigations aimed at substantiating the standard of maximum permissible concentrations of radioactive isotopes, in scientifically analyzing the findings of periodical medical examinations, and elaborating the required protective measures, safety rules and instructions for work with radioactive substances.

The scientific and research work in this realm is planned and generalized by the Central Institute of Industrial Hygiene and Occupational Diseases, which forms part of the Academy of Medical Sciences, the highest scientific medical institution in the USSR.

Experience accumulated over a number of years has shown that the system of prophylactic measures applied in the Soviet Union guarantees the preservation of the health of all those engaged in the peaceful employment of atomic energy.

# The Protection of Workers Against Ionising Radiations*

## By the International Labour Office

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* This paper in its complete form was made available to participants at Geneva as a printed booklet. The table of contents indicates which sections of the paper have been omitted from these Proceedings. The full text can be obtained from the International Labour Office, Geneva.

*The following is an abridged version of the paper submitted by the International Labour Office to the Conference. The chapters dealing with injuries caused by ionising radiations, standards of exposure, uses of these radiations and safety in the transport of radioactive substances, as well as the five appendices appearing in the full paper, are not reproduced in this text.*

## I. INTRODUCTION

At its 38th Session, held in June 1955, the International Labour Conference adopted a resolution in which, among other things, it requested the Governing Body of the International Labour Office, in the light of the information available as a result of the International Conference on the Peaceful Uses of Atomic Energy, to consider what part the ILO could play in advising and assisting governments and industry in promoting the highest possible standards of health, safety and welfare among workers in atomic plants and in other undertakings affected by the development of the industrial uses of atomic energy.

This resolution is the most recent manifestation of the interest of the International Labour Organisation in the protection of workers against injury and disease arising out of their employment.

Among the many questions with which the Organisation has been more particularly concerned in this field is that of the risks presented by ionising radiations and measures for the protection of the workers who are exposed to these risks.

As long ago as 1949, the International Labour Office showed that it was aware of the importance of ionising radiations in modern industry by convening a meeting of experts on dangerous radiations and devoting to this subject a whole chapter of the *Model Code of Safety Regulations for Industrial Establishments for the Guidance of Governments and Industry*. This chapter, which comprises over 200 provisions,[2] was in fact the first set of international safety and health standards for the industrial use of X-rays and radioactive substances.

Although the *Model Code* has in no sense the binding force of an international convention, it might be of interest to point out that several governments have used it as a basis for the preparation of their own regulations. For example, it has recently been used in this way by the British Government in the preparation of statutory regulations which will control the use of X- and gamma rays in factories and other industrial establishments.

The provisions on ionising radiations in the *Model Code* are, however, by no means the first manifestation of the interest of the ILO in the subject.

For instance, in the years 1932–34 the International Labour Office published in the encyclopaedia *Occupation and Health* three articles dealing respectively with the occupational pathology of roentgen ray operators, radium and radioactive substances, and uranium. The information given in these articles is obviously now out of date, but they are not devoid of interest and they make it easy to realise the immense progress accomplished during the last 25 years, both in the industrial and other uses of X-rays and radioactive substances and in the knowledge of the risks and the indispensable precautions that they require.

This progress is not less evident from the increasing number of references to X-rays and radioactive substances quoted since 1923 in two periodicals published by the International Labour Office, the *Bibliography of Industrial Hygiene*† and *Occupational Safety and Health*.‡

Further, the number of requests for information on the subject received by the occupational safety and health division of the International Labour Office is tangible evidence both of the development of the uses of ionising radiations in modern technology and of the importance of the precautions that they call for. These requests, which emanate from hospital services as well as from industry, are concerned with the nature of the hazards, protective measures in the strict sense, and provisions for limiting the duration of exposure or arranging working hours and holidays of the exposed personnel.

It should also be mentioned that pathological manifestations due to X-rays, radium and other radioactive substances are included among the compensable occupational diseases listed in the Convention concerning workmen's compensation for occupational diseases adopted in 1934 by the International Labour Conference, and ratified by 29 countries. These manifestations are now compensable as occupational accidents or diseases in a total of 49 countries.¶ It is, however, possible that the extension of the peaceful uses of atomic energy to countries that are relatively less advanced and whose legislation in this respect is inadequate will create certain difficulties.

Lastly, a meeting of occupational health experts held in Geneva in December 1954 under the auspices of the International Labour Organisation recommended, as one of the items to be included in the international list of occupational diseases to be notified to the labour inspectorate or other authority concerned with the protection of the health of workers in places of employment, diseases due to "ionising radiations such as X-rays and radiations from radium or other natural or artificial radioactive substances". Notification is of capital importance, if only because it facilitates the initiation of the necessary preventive and protective measures and ensures their effective application.

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† Published in 1948-50 as the *Bibliography of Occupational Medicine*.

‡ Formerly the *Industrial Safety Survey*.

¶ In three of these countries, compensation is awarded only if the disease has been contracted in the course of specified work. In some federal States such as Australia, Canada and the United States, compensation is not provided for in all the federated states or provinces.

The purpose of the present report is to provide a brief survey of the field of protection of workers against ionising radiations. It is designed to give general but practical information on the nature of injuries due to ionising radiations, the standards of exposure and the methods of protection appropriate to the various categories of use, with special emphasis on the industrial field. The report also briefly mentions some of the problems arising out of the mining and processing of radioactive ores and the transport of radioisotopes.

On the other hand, it intentionally ignores those aspects of protection against ionising radiations of concern to public health authorities as well as those safety problems—and they are many—which arise in establishments for the production of atomic energy and its by-products. In the international field the former are of more special concern to the World Health Organisation and the International Commission on Radiological Protection, while the latter, and more especially those related to the operation of reactors, will most probably form the subject of several national reports to the Conference. The International Labour Office is aware that these problems have been thoroughly studied in various countries that possess imposing resources and have accumulated in this field a fund of knowledge that is much more precise and extensive than would be possible in the case of an international organisation.

It is interesting to observe that up to the present the atomic energy industry is widely believed to be safer than other comparable branches of industry. That this is so is doubtless largely because it is still in the experimental stage, is progressing cautiously under government auspices and is controlled by highly specialised staff having the necessary background knowledge. This, moreover, is confirmation of a generally recognised fact, namely that the frequency of occupational accidents is often relatively low in industries where the risks are so serious and so widely known that special precautions are properly enforced and readily accepted.

The International Labour Office does not possess information that can be considered adequate on the situation in uranium mining and processing. However, it can be asserted that at the present time the safety and health conditions prevailing in undertakings where X-rays or radioactive substances are used for industrial or medical purposes are generally good and are adequately supervised, although protection of the personnel seems to be generally better in industrial undertakings than in hospitals.

This situation, however, might take an unfavourable turn with the extension of the uses of atomic energy to regions or countries with little industrial experience and underdeveloped inspection services; it might also deteriorate as man becomes familiar with risks that are not perceptible by any of his senses, and for this reason neglects or even despises the most elementary safety precautions. Lastly, competition to reduce the cost of power from nuclear energy, the cost of devices based on this energy or the cost of other uses of it may also tend to narrow the margin of safety hitherto observed.

Since one of the consequences of the development of the peaceful uses of atomic energy will be to make available to industry and other branches of economic activity a steadily increasing quantity of artificial radioisotopes, it is desirable that the subject of the protection of workers who are likely to come into contact with these substances should receive the close attention of the Conference.

In fact, while in nuclear research centres or atomic energy establishments the risks inherent in ionising radiations and the indispensable precautions that they call for are perfectly well known, this is not so in the numerous industrial and other establishments that are already using radioactive substances or will be using them in the near future. It is thus on this kind of use that preventive action, and hence training activities, should concentrate if workers are to be adequately protected.

In this connection it should be observed that for the International Labour Organisation, the term "workers" comprises all persons in employment. The Organisation thus has an equal interest in all classes of persons whose occupation may expose them to the action of ionising radiations, whether these persons are employed as manual workers or salaried employees in industry, in mining, in agriculture, in transport or in public or private services.

One of the basic requirements of labour protection consists in the application of codes of practice and statutory regulations wherever dangerous radiations are being used, and this application should begin as soon as possible because occupational safety and health must always be taken into consideration when planning new projects, and because also a long period may elapse between excessive radiation exposure and the manifestation of the first injurious effects resulting from it. It is vital that there should be no repetition of the tragic effects on the health of workers that have resulted in the past from the lack of clear standards and fully enforced precautions in various industries in so many industrialised countries.

In the legislative field much has been accomplished already, or is about to be undertaken, in countries such as Australia, Austria, Brazil, France, the Federal Republic of Germany, New Zealand, Norway, Sweden, Switzerland, Turkey, the United Kingdom and the United States, but much remains to be done. This question will no doubt also be dealt with in other reports submitted to the Conference.

The field of activity opened to international organisations by the development of the peaceful uses of atomic energy appears to be extremely wide and, while it is important that each organisation should be ready to play the part that its mission confers on it, it is no less important that in performing their respective tasks the various international organisa-

tions concerned should establish and maintain close contact with one another, and so far as possible avoid duplication and dispersal of effort.

It is in the field of labour protection and more particularly in matters of safety, health, welfare and workmen's compensation that the International Labour Organisation considers that it has a valuable contribution to make to the common cause.

It is less concerned with those aspects of the matter which relate mainly to the economic implications of the application of nuclear power on a large scale, or to public health, or to the purely scientific, medical and agricultural applications of atomic energy and its by-products.

Although the present report deals only with occupational safety and health questions, the large-scale development of the peaceful uses of atomic energy will doubtless also raise many problems of general social policy of varying importance. This has been the case in all great advances in technology. This is not the place to dwell on these problems which, moreover, have recently formed the subject of a preliminary general study by the International Labour Office.§ It will be for the International Labour Organisation to seek to facilitate their solution in keeping with the objectives defined in its Constitution.

## II. INJURIES CAUSED BY IONISING RADIATIONS

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## III. STANDARDS OF EXPOSURE TO IONISING RADIATIONS

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## IV. USES OF IONISING RADIATIONS

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## V. PROTECTIVE MEASURES

### Underlying Principles

These in essence are simple. The worker must be protected (1) from external sources of irradiation, and (2) from the deposition of radioactive materials internally in the body.

The first can be achieved by using the minimum X-ray tube strength and current or amount of radioactive material necessary for the work being undertaken, and using it for the minimum necessary time; together with such shielding or distance protection, or both, as will bring dosage rates at working positions down to, and preferably well below, the maximum permissible dosage rates set out in the third part of this report for the appropriate type of radiation.

A general indication of the amount of shielding necessary for X- and gamma rays, alpha and beta particles and neutrons has been given in the second part of this report, where the properties of these radiations were described. Nothing more detailed can be offered within the limits of this paper except to recall that X- and gamma rays may require shielding by considerable thicknesses of lead or concrete according to their energy and strength, alphas normally not at all or by negligibly small thicknesses of glass or other solid material, betas usually by a few millimetres of plastics or metals, and neutrons by materials containing hydrogen or other light elements (e.g., water, paraffin wax or concrete) to slow the neutrons down, followed by a material to absorb the resulting thermal neutrons (e.g., boron or cadmium) and a final material to absorb any resultant radiation produced (e.g., water for the alpha particles from boron or lead for the gamma rays from cadmium). Quantitative data on shielding against X-rays up to 1 million volts and radium gamma rays are included in an appendix to the *Model Code of Safety Regulations for Industrial Establishments for the Guidance of Governments and Industry,* while more detailed figures for X-rays up to 2 million volts may be found in the 1955 Recommendations of the International Commission on Radiological Protection, and on shielding against the other radiations in the United States National Bureau of Standards series of handbooks and many other national publications.

Though the shielding necessary will range from nothing or distance protection only up to massive thicknesses of concrete or lead, it should be remembered that distance protection itself is nearly always most valuable, particularly with gamma rays, because of the operation of the powerful inverse square law, and therefore the worker should be as far from the radiating sources as possible and, because of scatter, from material on which the radiation falls. Moreover, to economise in the amount of shielding necessary and in the interest of efficiency, shielding should always be provided as near the source as possible and should enclose it to the greatest extent practicable so as to minimise scattered radiation. Shielding may be permanent as in X- or gamma radiography rooms built of brick, concrete or lead; portable and movable, as in gamma radiography operating and storage containers; or temporary, as when interlocking lead bricks are used around "hot" sources being processed in radio laboratories.

Protection against the second main hazard—the deposition of radioactive materials within the body, whether from dusts, gases or solutions—is more difficult to achieve because permissible body burdens and permissible concentrations of radioisotopes in air and water are set so low. It can only be achieved by devoting scrupulous attention to the protection of all portals of entry to the body—nose and lungs, mouth and alimentary tract, and skin—by the installation in properly designed and equipped workrooms of adequate ventilation (local and general), by the adoption of measures to avoid spillage or the spread of contamination, by insisting that strict attention be paid to cleanliness of the person and of working equipment, by the prohibition of eating, drinking

§ See "Atomic Energy and Social Policy" in *International Labour Review* (Geneva, ILO), Vol. LXXII, No. 1, pp. 1-20, July 1955.

and smoking, etc., in the workrooms concerned, by the use of personal protective equipment and clothing by those at risk, and by the institution of regular monitoring procedures.

## General Precautions

### Planning and Equipment

Much can be achieved which would later be difficult if the planning, layout and equipment of X-ray installations and of workrooms and laboratories where radioisotopes are to be used as sealed or unsealed sources are carefully thought out and discussed in advance with the appropriate national authorities in the protection field or with a qualified expert.

Wherever possible there should be segregation of ionising radiation work from work not involving such radiation—in the case of X- and gamma radiography by the provision of special radiography rooms and in the case of the use of radioactive substances by the provision of rooms set apart for the purpose, so limiting to a minimum the number of workers liable to be exposed to radiation. On no account should radioactive laboratories and similar workrooms be overcrowded. The provision of ample floor space is vital, since it allows full play for the operation of the distance factor in protection and prevents proximity of workers leading to accidents such as the breakage or spillage of vessels containing radioactive materials with resultant inhalation, ingestion and contamination risks.

To counterbalance the potentially serious risks from radiation absorption, etc., everything should be done in the layout of workrooms to facilitate the maintenance of a high standard of the workers' general health. This implies that underground workrooms, unless specially air-conditioned, etc., are undesirable for this type of work, which should be carried out in rooms provided with ample natural lighting and means for the entry of sunshine and fresh air. Additional artificial lighting on a generous scale should also be provided on account of the extra safety afforded to those handling radioisotopes in a good light at all times.

The equipment provided to facilitate the use of ionising radiations and the shielding, permanent or temporary, which has to be used must always be adequate in amount and quality. It is false economy to try to make do with the bare minimum, because this means that radiation dosages to workers may be constantly at or near the maximum permissible rates and that as soon as some extension of the work involves the use of stronger sources—a thing that is constantly happening in industrial practice as attempts are made to reduce irradiation times and get through more work in the same time—the existing protective and other arrangements become inadequate and can frequently be altered only at considerable expense and after some delay.

In the planning of X-ray rooms housing powerful tubes, where the cost of protective barriers might be excessively high, it can often be arranged that the tube operates only in certain planes and directions for which ample shielding and distance protection is readily provided or already in existence; alternatively, limitations can be placed in the daily operating time of the tube.

To ensure that safe conditions exist before a new or modified radiological department is put into use, a thorough radiation survey should be made by the national protection organisation concerned or by some other qualified expert.

### Inhalation and Ingestion Precautions

For the reasons already given these precautions are supremely important and too much forethought and attention cannot be devoted to them. Extreme care must be used when working with radioisotopes of long half-life known to concentrate in particular organs of the body—examples are radium, $Ca^{45}$, $Fe^{55}$, $Co^{60}$, $Sr^{90}$, $Y^{91}$, $Zr^{95}$, $Ce^{144}$, $Pm^{147}$ and $Bi^{210}$. Care must also be taken with others known to be specifically absorbed or to have a long biological half-life, such as $I^{131}$, $C^{14}$, $P^{32}$, $Cs^{137}$ and $Au^{198}$.

Where there is any possibility at all of radioactive dusts, sprays, fumes or gases being produced or radioactive liquids handled, the utmost use should be made of complete enclosures around the operation concerned, and the enclosure should be put under adequate suction. Very frequently use can be made of small exhausted fume cupboards or of dry boxes fitted with rubber sleeves cemented to the box front, through which the operator, wearing rubber gloves, puts his hands to carry out the necessary manipulations. Such cabinets, which incidentally give their users a feeling of confidence, should normally exhaust their contents through filters so as to remove any radioactive dusts before the effluent is discharged outside the workroom, and great care needs to be taken in siting effluent discharges to ensure that no recycling of contaminated air occurs through neighbouring windows or plenum ventilation intakes.

The use of respirators will rarely be necessary if full use is made of the fume cupboard arrangements mentioned above, but these have a place and should be available for use, for example, by a worker engaged in clearing up any radioactive dust spillage.

Scrupulous attention must be paid to avoidance of the spread of radioactive contamination; this means careful and neat methods of working, the use of catch trays under all radioactive materials being handled, the immediate wiping up or clearing up of any spillage that may occur and the collection of all radioactive waste and rinse solutions in special containers to prevent discharge to sewers, unless special arrangements have been made with the authorities.

Working surfaces liable to be contaminated can be covered with disposable sheets of bitumenised paper, absorbent paper or polyvinyl chloride sheeting.

Contamination of hands can be minimised by rubbing them with suitable barrier creams or by wearing thin rubber or PVC surgeons' gloves, which should be washed, on the hands, before removal and the hands then washed before being monitored. Gloves should always be worn when handling unsealed sources. The wearing of suitable protective clothing, such as caps, overalls (preferably surgeons' gowns fastening at the back and without pockets), waterproof and washable aprons or bibs, and sometimes underclothing and footwear, as appropriate, is an essential means of assisting the worker in preventing bodily contamination by radioactive material. Such clothing, which should not be worn in non-active rooms, is best laundered on the premises and kept in special accommodation set apart for the purpose and immediately adjoining the workrooms where it is to be used.

Ample washing facilities must be available and frequent use made of them. Gentle but thorough washing of parts of the body with soap and warm water is normally sufficient to remove radioactive contamination, but in difficult cases the affected areas can be carefully rubbed with titanium dioxide paste, or with a strong solution of potassium permanganate followed by a 5% solution of sodium bisulphite.

The introduction of food and drink, smoking materials and cosmetics into workrooms where unsealed radioactive materials are handled must be strictly forbidden.

All cleaning of workrooms where inhalation and ingestion risks exist should be done by wet methods —the workrooms being designed and constructed to facilitate this—or by the use of vacuum cleaners.

### Limitation of Working Periods

There is no generally agreed figure for the number of hours per week which should be worked by those regularly liable to be exposed to ionising radiation. The ILO *Model Code* drawn up in 1949 recommended[4] a five-day working week of 35 hours with a minimum of four weeks' annual holiday, preferably taken consecutively; together with three months completely away from ionising radiations in some other employment once in every period of 15 months. The first part of this recommendation was that originally made by the British X-Ray and Radium Protection Committee.

Brazil, by an Act of November 1950, fixes the maximum working hours for personnel required to handle X-ray equipment and radioactive substances as low as 24 hours a week, and allows them 20 consecutive days' leave every six months. Some European countries have in recent years favoured a maximum of 48 hours a week (e.g. the present United Kingdom Luminising Regulations require this) but present opinion favours lower figures.

Linked with the question of permissible hours of work is the question of age and sex. Most countries feel that the employment of young persons under 18 in ionising radiation work is undesirable, since they are biologically at a stage of rapid development and in addition are apt to be less responsible in their attitude towards taking the necessary precautions and safeguards.

Other nations, such as the United States, have from time to time felt that both men and women over 45 years of age might be allowed a basic permissible dose of radiation (with certain limitations for specially critical body organs) double that for younger adults, since by that age the gonads have ceased to be the critical organ. This relaxation has not yet found international acceptance in the Recommendations of the International Commission on Radiological Protection, nor have any specific proposals as to special treatment for women of reproductive age. It is, however, widely felt that the protection of pregnant women against ionising radiation is most important, since human beings in the prenatal stage are known to be more than normally radiosensitive.

All such matters are at present subjects of discussion and inquiry by the International Commission and by national committees affiliated to it, and until its authoritative guidance can be given it would seem reasonable to limit the weekly exposure of workers to ionising radiation to a maximum of 40 hours in a five-day week, to allow them an annual holiday of three or four weeks—preferably taken consecutively—and to exclude young persons under 18 from such work.

It is at present considered unlikely that women under 45, who on account of their reproductive functions may be more radiosensitive than men, will suffer any ill effects from ionising radiations if the present international permissible limits for exposure are not exceeded. There is, however, firm evidence that luminisers, who happen to be practically always of the female sex, do benefit haematologically from a three months' change of occupation to non-radioactive work once in every period of 15 months.

The question often arises what should happen to workers of any age or sex who for some reason exceed the maximum permissible dose of any type of ionising radiation in any one week. The best advice available is that in such a case an average weekly value should be assessed for the irradiation of the worker during the 13 weeks prior to, and including, the week in which over-exposure occurred. If this average value exceeds the maximum permissible weekly value, the worker should be placed under medical supervision and his duties should be rearranged so as to involve considerably less exposure for a compensatory period. If the average weekly value is less than the maximum permissible value, the worker may continue with his normal duties.

### Monitoring

Apart from the personal monitoring equipment referred to in the next subsection, all establishments

in which there is occupational exposure to ionising radiations on any but a very minor scale should be equipped with suitable site-monitoring apparatus so that tests can be made at working positions to assess the adequacy and completeness of the protection provided when use is first made of irradiating apparatus or radioisotopes; thereafter regular routine checks can be carried out to ensure that protective equipment is being properly maintained; and lastly, routine surveys can be made for radioactive contamination. Records of all such tests should be kept.

The amount of equipment necessary will vary enormously according to the extent of the possible risks. In an atomic energy establishment there will be need for the full complement of instruments. These will include ionisation chamber radiation monitors to measure beta, gamma, proton and slow and fast neutron radiation levels in working areas; proportional counters for measuring neutrons and alpha particles—particularly alpha contamination on hands, feet and clothing or on bench surfaces; Geiger counters for sensitive survey work generally, especially for measuring radioactive contamination, and sometimes linked with alarms and mounted on laboratory doorways, to give warning of contaminated persons passing through; scintillation counters for alpha or gamma contamination monitoring and for testing urine samples for alpha emitting material; and special portable instruments for assessing radioactive dust or gas contents of working atmospheres (including those downwind of reactor chimney stacks or in the discharge ducts from filter systems) to enable efficiency of maintenance to be assessed.

In other industrial establishments using X-rays or radioisotopes an ionisation-type radiation monitor will frequently suffice for routine monitoring work. It is surprising how useful a single instrument can be, for example in a radium luminising department, to locate radioactive bench and floor contamination and spillages, to measure the build-up of luminous compound in ducting systems and to check that old stocks of luminised compass dials, hands, pointers and other radioactive materials are not left lying about on working benches or "forgotten" in cupboards or drawers, so contributing unnecessarily to the background radiation of the workroom.

There should, however, be set up in each country a central monitoring service available to all users of ionising radiations so that periodically, or for unusual types of radiation hazard, its services may be enlisted to make impartial and authoritative surveys. This body should have available the more elaborate and expensive types of monitoring equipment that are necessary to carry out accurate determinations of radioactive body burdens such as might result from long-continued or especially hazardous work with radioactive materials, e.g. in radium luminising, or from the accidental ingestion or inhalation of radioisotopes generally.

**Film or Dosemeter Tests of Radiation Exposure**

The measurement by film or dosemeter of the individual external radiation dosages received by a worker is of the greatest importance, for not only does it act as the basic check on his safety and health in so far as ionising radiations are concerned but it is also an excellent way of educating him in what he can and cannot do safely. The view of the International Commission on Radiological Protection is that if, as a result of the initial radiation survey, the person in charge of the department is satisfied that the doses received by his staff can never exceed the maximum permissible dose, periodic personnel monitoring should suffice. When, however, the safety of personnel depends upon adherence to a particular technique or proper functioning of equipment rather than upon low-level sources of radiation or provision of permanent protective barriers, continuous monitoring should be carried out.

It is very important that records of radiation exposures should be faithfully kept, as they form an invaluable guide for medical consideration if at a later date ill health possibly attributable to radiation absorption should occur.

Film badges, which may be worn in a breast pocket, pinned on an overall, strapped to a wrist or as a finger ring, as appropriate, and carried for a week or two before being developed, are readily available at nominal charge in all the major countries of the world. They can be used for the measurement of X- or gamma radiation or, if covered on both sides over half the film's area by 1 mm of lead, for the measurement of both beta and gamma radiation separately where both radiations are present, as in a radium luminising department.

For slow neutron measurement, films covered with a thin layer of cadmium can be used. The cadmium captures the slow neutrons and the resulting gamma ray emission registers on the film. Alternatively a lithium- or boron-loaded film emulsion can be used to capture the neutrons and to register the resulting alpha particle tracks, which can be counted.

Exposure to fast neutrons can be measured by film badges indirectly through the proton recoils caused in the hydrogen atoms present in unloaded emulsion, but there are complications and the accuracy is low.

A most useful alternative to the film badge—particularly when records of accumulated X- or gamma radiation exposures are required at daily or even shorter intervals—is the personal pocket dosemeter. In one form of this, small highly insulated ionisation chambers are charged and worn during the appropriate period. They discharge proportionately to the radiation received, and are read with an electrometer. Slow neutron measurement can be obtained by lining them with boron or filling them with boron fluoride gas. In another form, fountain-pen-type quartz-fibre electroscopes, charged before use, are worn for the convenient period, after which they are

read by holding them up to a good light and observing through an eyepiece the position of the fibre on a scale calibrated directly in milliroentgens.

Also available for use in cases where radiation intensities are unknown and likely to vary considerably, or in cases where the worker's attention may be fully concentrated on some operation, are small battery-operated pocket alarm instruments which give an audible alarm when a maximum permissible dose has been reached.

### Handling Equipment

On the question of handling equipment it is almost impossible to generalise, and each class of occupational radiation exposure has to be treated on its merits. The aim in each case will be to reduce radiation exposures or to prevent personal contamination by radioisotopes whether as solids, powders, liquids or gases.

Handling equipment can vary from a simple pair of forceps or tongs used to hold a test tube or beaker containing millicurie amounts or less of a gamma- or beta-emitting radioactive liquid to an elaborate power-driven and remote-controlled "master-slave" pair of mechanical hands for the manipulation of multicurie sources of fission products during processing in atomic energy establishments; or from a simple lead pot used for the handling of small quantities of radium luminous compound to the massive shielded operating and storage containers mounted on bogies for use with industrial radiographic cobalt-60 gamma-ray sources of between 10 and 100 curies—containers weighing nearly a ton, and operated from a distance pneumatically or by remote cable control.

Handling tongs, often made in stainless steel, are available in lengths of up to several feet and with interchangeable rubber-covered jaws activated by levers or other devices situated at the handle end; they are suitable for the manipulation of a wide variety of radiolaboratory equipment.

When the isotopes concerned are substantially beta emitters, use can be made of transparent plastic shields fitting closely around the equipment to allow of close handling with good visibility, and in this connection the protection of the eyes by goggles may also be of value.

Since radioactive solutions must never be pipetted by mouth, piston-operated types of pipette should be used instead; remotely controlled pipettes are normally used for handling gamma-emitting radioactive solutions behind lead bricks or when the operator has to keep a safe distance from an unshielded operation.

In radiochemical laboratories a wide variety of special gadgets have been perfected for the handling of radioactive liquids during such operations as filtering, precipitation, distribution, and titration, while to avoid all handling during various transfer operations, bottles containing liquids and powders, etc., are often moved within fume cupboards by small cranes or on small-scale railways or conveyors which may pass, through internal doors, between one fume cupboard and another. As the strengths of radioisotopes used industrially and medically increase, as they are constantly doing, more and more use will have to be made of handling and mechanical handling equipment, automatic sequence-time controls and remote controls if radiation hazards are to be avoided.

### Waste Disposal

No clear national or international policies have yet been formulated for the ultimate disposal of radioactive wastes from factories, hospitals or other establishments; and for the purpose of this paper waste disposal is referred to only in so far as it raises internal problems in the premises. The best policy is that largely adopted in the United States whereby waste radioactive material can be returned from each establishment to a central disposal authority, usually the Atomic Energy Commission, where it can be dealt with under the best and safest conditions possible. Unfortunately a solution of the problem along such lines is not entirely practicable as yet in most countries, and in addition individual batches of waste material may be very low in activity or exist in very dilute form. As a general principle active material should be precipitated where possible and disposed of as an insoluble residue.

Efforts should be made to keep liquid wastes as concentrated as possible since they are then more easily handled. For large establishments delay tank storage may be necessary; in smaller ones washings and active wastes can be accumulated in carboys, shielded if necessary and allowed to decay naturally. This is feasible with some of the shorter-lived isotopes such as $Na^{24}$, $P^{32}$ and $I^{131}$. For longer-lived beta emitters such as $Sr^{90}$ which tend to concentrate in specific body organs, and for all long-lived gamma emitters, the solutions, after being rendered alkaline and diluted by adding stable isotopes of the same element in the same chemical form, can be flushed down sewers in small quantities, the aim being to keep concentrations in the sewerage system below the drinking water tolerances for the isotopes concerned. For those mentioned this is likely to be achieved by limiting individual flushes to 5 $\mu$c and total quantities per week to 50 $\mu$c per establishment. Lower limits still may be set by national authorities for solutions containing alpha emitters such as radium. For the less hazardous beta-gamma emitters quantities disposed of in sewers may be greater, say not more than 100 $\mu$c per flush or 1 mc per week. Traps and sink fittings will need monitoring regularly to ensure that the waste is not accumulating by absorption in the drainage system and to keep the background radiation down to less than 10 mr per eight-hour day.

If larger quantities of liquid than can be so dealt with accumulate, they will have to be stored in a shielded container and special disposal arrangements will have to be made with the appropriate authority;

they may, for example, after evaporation or concentration, be incorporated with cement into concrete blocks and disposed of as solid waste.

Solid active waste, as it accumulates in workrooms, together with broken contaminated glassware, should be placed at once in closed, clearly labelled refuse bins fitted with moisture-proof disposal linings; and combustible waste such as contaminated wiping tissues, filter papers, catch tray absorbent linings, animal waste and so on should be segregated from non-combustible waste and subsequently concentrated by burning in an incinerator under carefully controlled conditions. Regular clearances of such bins must be made, at least daily. The resulting wastes may be held in a suitable store until their residual activity is very small, where this is possible, before being disposed of with ordinary rubbish. The activities per load should be limited to 5 $\mu$c for the more hazardous beta and gamma emitters and to 100 $\mu$c for the other beta and gamma isotopes.

Where longer-lived or more dangerous isotopes (especially the alpha emitters) are concerned, the best solution at present appears to be to form the dampened waste with cement into concrete blocks and to arrange for these to be buried at least 5 feet down in an approved rubbish dump or on some other marked and guarded site set apart for such a purpose by each national authority or by the isotope users themselves. Where possible, non-radioactive isotopes of the radioactive elements being disposed of in this way should be uniformly added to the cement mix to dilute the active component to a low specific activity.

Gaseous wastes from fume cupboards or incinerators should be filtered before being released into the atmosphere and the filters eventually treated as solid waste. The effluents will need to be released high enough and at a safe place so that they do not reenter buildings through windows or plenum intakes.

Waste disposal from atomic energy plants is a special subject which cannot be dealt with here.

### Supervision and Training

This is a most important part of the work of any radiological department, since workers liable to be exposed to ionising radiation need constant supervision with regard to the precautions they should take. They will only take them intelligently if they have been carefully instructed with regard to radiation hazards and the reasons which prompt the methods of control. In each department there should be a specially nominated officer responsible for the observance of radiation protection measures in the department and for the maintenance in good order of protective equipment and shielding. This safety officer should be scientifically or technically qualified where at all possible, and where not should at least have made some special study of, or have taken a course in, the subject of radiation hazards and their control. In the larger establishments—and this will certainly be so in the atomic energy establishments—there may well be justification for undertaking such safety and health work on a full-time basis.

The safety officer should make it his duty to carry out routine surveys of radiological departments with suitable monitoring apparatus to determine the adequacy of, or the need for improving, all protective measures, and he should pay particular attention to checking for the presence of radioactive contamination where this is a possibility.

He should also be responsible for seeing that film badges and personal pocket dosemeters are worn by the workers concerned, and that proper records of exposures so observed are kept and used as a guide to the efficacy of the precautions in force.

Equally the workers themselves have a duty to follow the recommendations and instructions of the safety officer or departmental head in the interests of radiation protection, to use all appliances and equipment provided for their health and safety, and to report promptly any defects or inadequacies in shielding or protective arrangements which they may observe. Workers should be reminded that neglect or carelessness on their part may harm or endanger others. Their training, which will need to be reinforced occasionally by refresher courses, is all the more necessary because of the insidious nature of the danger which is being guarded against. The workers will need to be constantly reminded that if there is no justification for fear in radiation work there is equally no room for complacency.

In the larger establishments the safety officer, in addition to carrying out the duties mentioned above, will be expected to give guidance on the siting, layout and equipment of radiological departments and laboratories, and to advise on the design and utilisation of ventilation, shielding, waste disposal systems, remote-control equipment and protective clothing. He will also be responsible for maintaining in proper calibration all monitoring instruments, for instructing workers in radiation protection measures, and for taking charge of and advising on protection measures when emergency operations and accidents bring with them sudden radiation hazards. Such officers should also be especially charged with securing the enforcement of any national statutory regulations or the application of codes of practice operating in their field.

The work of safety officers brings with it the need for special training courses, and these are now, or shortly will be, well provided for in the major countries working with atomic energy. The Harwell Isotope School, for example, since its foundation in 1951, has trained nearly 400 postgraduate students from some 30 different countries in courses of four weeks which include both lectures and laboratory work. In the United States since 1948 some 200 graduate fellows have additionally graduated in radiological physics after a nine months' theoretical

course and a three months' practical laboratory course controlled by the Oak Ridge Institute of Nuclear Studies.

### Medical Supervision

Medical supervision is no substitute for effective technical measures, but has its place in radiation protection work.

Pre-employment medical examination is necessary not only to help in preventing the employment of individuals unsuitable for the work or its conditions on account of significant skin, bone, gonad, blood, lung or other pathological abnormalities, but also to enable control sets of blood counts to be made and recorded for future reference. Further, persons regularly employed on work involving exposure to ionising radiation should be clinically examined yearly as a routine precaution.

There is much controversy and uncertainty at the present time as to the value of blood counts and the significance of blood changes in connection with radiation work, but it is generally considered that changes in the blood picture often afford the earliest detectable indication of biological damage from radiation absorption; and the general consensus of opinion as reflected by the 1955 Recommendations of the International Commission on Radiological Protection is that provided that radiation monitoring (both site and personnel monitoring) is carried out in all circumstances involving occupational exposure to ionising radiation (external or internal), then routine blood counts are (1) unnecessary in the case of workers who receive doses not exceeding one-third of the permissible doses; (2) optional in the case of workers who receive doses between one-third and two-thirds of the permissible doses; and (3) desirable in the case of workers who receive doses exceeding two-thirds of the permissible doses.

Blood counts, where done, are normally carried out at intervals of from three to six months.

It is further recommended that abnormal effects indicated by the haematological test should be discussed with a radiologist and a haematologist before any action is taken involving suspension or dismissal from radiological work.

Since the individual trend in blood counts may be of great importance, the technique employed must be standardised and all observations made under similar conditions. This suggests the desirability of each country setting up a centralised blood count service available to all radiation workers.

A medical record form substantially that proposed to be used in the United Kingdom and suitable for recording the clinical and haematological examinations of radiation workers in a standard manner, together with notes on haematological examinations and technique, are reproduced as Appendix III.** The form should be used to an extent to be determined by the medical officer in charge of each establishment.

** Not reproduced here.

In the intervals between periodical examinations the medical officer may need to ensure that adequate facilities exist for the decontamination by thorough irrigation of wounds contaminated with radioactive material, or for the combating of accidental over-ingestion of toxic amounts of radioactive substances by emetics, for example, or by the administration of non-radioactive competitors of similar chemical nature and bodily fate.

### Chemical and Biochemical Protection

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## Special Precautions

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### Factories

The main categories of risk in factories will be considered under separate heads.

### X-Radiography of Castings and Other Articles, and of Welds

X-ray tubes used for industrial purposes should be of the shock-proof self-protecting type in which the protection necessary according to the kilovoltage of the tube and the tube current is incorporated integrally in the design. Any continuously excited tube should be provided with a shutter affording adequate protection and with arrangements by which it can be opened and closed only from the control panel, which should be provided with signal lights to indicate the shutter's position.

It is most important that no person, in the course of his work, should be deliberately or inadvertently exposed to a direct beam of X-rays. To minimise this risk and to give protection from scattered radiation, X-radiography should, wherever practicable, be done only within a specially constructed and protected X-ray room, from which all persons are excluded while the X-ray apparatus is in operation. To achieve this effectively, efficient interlocks should be provided and carefully maintained on all entrances to prevent any person entering the X-ray room while any tube is energised. The construction of the X-ray room, which should be of ample area, should be such as to give adequate protection against direct and scattered radiation, under all operating conditions, to all persons outside the enclosure, whether they are on the same floor level or working in rooms above or below the X-ray room itself. The aim should be to reduce the free-air dosage rate on the outer wall and door, ceiling and floor surfaces, as necessary, to a figure not exceeding 7.5 mr/hr, and to ensure that there are no radiation leakages.

Assuming that there is no occupied room below, or that the flooring of the X-ray room affords adequate protection, it is generally desirable that the tube primary beam should be directed downwards so that protection will be needed only against scattered radiation. The scatter itself can be minimised by standing the work to be radiographed on a lead sheet. Means should be provided within the X-ray

room to enable persons accidentally shut in to leave the enclosure without delay, or safely to interrupt the main electrical supply which enables the X-ray tube to be energised. In this connection it is useful to provide a ramp-actuated electric warning bell preceding the ordinary door interlock system.

Where for valid reasons the construction of a proper X-ray enclosure is impracticable or uneconomic because of the infrequent use made of X-ray apparatus, the operator of an X-ray set should be shielded from direct and scattered X-radiation by the provision of a cabinet or screening affording adequate protection throughout. Other persons in the vicinity should either be similarly protected or be excluded from the area around the X-ray tube and work being radiographed within which the radiation dosage rate in free air exceeds 7.5 mr/hr. This area should be fenced off and placarded with a warning notice. In the case of mobile X-ray sets taken to objects too heavy or inconvenient to move to an X-ray room, the screening will commonly take the form of a lead-lined cabin built around the operator's control panel on the mobile truck carrying the X-ray tube. Protection of workers other than the set operator may be provided by excluding them from a suitably defined area, also fenced off and placarded with a warning notice. In addition, lead-lined cones should be provided for tubes used in open workshops in order to reduce the amount of unwanted scattered radiation as much as possible. The use of a suitable number of protective lead screens, cemented between plywood to prevent creep and mounted on wheels to facilitate easy movement, is most desirable for shielding closely as much as possible of the area around the tube and the work being radiographed. Whenever possible the direct beam should be pointed away from areas where people are at work, and in devising these various safeguards the possibility of overhead crane drivers entering the danger area should not be overlooked.

Identifying numbers or letters of lead, iron, etc., are often affixed by adhesive tape, magnetically or otherwise, to objects being radiographed. Should they become displaced during actual exposures it is absolutely essential that no attempt should be made to refix or replace them by hand before switching off the X-ray tube. Neglect of this advice has already resulted in one industrial radiographer receiving such severe X-ray burns to a hand that the forearm had subsequently to be amputated.

For both fixed and mobile installations, lights or audible signals, or both, should be provided for the purpose of warning persons in the vicinity of the control panel, X-ray tube and high-tension generating equipment, and so arranged that they operate automatically from the control panel immediately before and during the energisation of the high-tension circuit of the X-ray tube. Observation windows fitted into X-ray room enclosures or into the protective screens or cabinets around mobile sets should be of lead-glass or other material giving a standard of protection not less than that of the main protection provided.

### X-Ray Fluoroscopy

Although it is not desirable to work at screen illumination levels where eyestrain may develop, all fluoroscopic examinations should be done with the smallest practicable X-ray beam intensities and tube apertures, and wherever possible the fluorescent screen should be viewed indirectly by the use of inclined mirrors. The screen itself will need to be protected by a sufficient thickness of lead-glass. In all normal circumstances the X-ray apparatus used for the fluoroscopic examination should be completely enclosed in a protective cabinet lined throughout—with no gaps at joints—with sufficient lead or its equivalent in other materials to reduce the radiation dosage rate at its outer surface to a figure in free air not exceeding 7.5 mr/hr. Such a cabinet should be fitted with effective interlocking devices to ensure that when the fluorescent screen or any part of the cabinet such as an access door is opened for any purpose the X-ray tube electrical circuit is broken.

As far as possible articles for fluoroscopic examination should be placed in position before energising the X-ray tube. They should be inserted into, or oriented in, the beam by methods which preclude the possibility of the operative's hand or forearm or other part of the body getting into the beam itself or into heavy scatter. There should never be any need for operatives to have to wear leaded-rubber gloves or aprons, since the methods employed would include the use of conveyors, turntables, sliding trays, dial feeds, tongs or other mechanisms which can be operated from outside the enclosed cabinet. Where conveyor belts and similar arrangements are used to feed the articles for examination into and out of the cabinet, extension tunnels from the cabinet openings should be provided, protected at each end across their full width by hanging leaded-rubber flaps split into a number of strips of the order of 1 inch or so in width, so that the packages passing through lift the minimum possible number of strips each time. The tunnels should be of sufficient length to prevent a worker's arm being pushed far enough down to reach the direct beam of radiation, and to prevent the emission of any appreciable amount of scattered radiation. Where articles for fluoroscopic examination pass along the conveyor with gaps between them, this latter requirement is best achieved by having two leaded-rubber strip curtains in each tunnel fixed at such a distance apart that one curtain is always sealing the opening when the other is being raised by the article passing through.

Faulty articles disclosed by the fluoroscopic examination and needing to be identified should be marked in some safe way, as by the use of a pricker or a swab kept moistened with a dyestuff and operated from outside the cabinet on the exit side of the fluorescent screen.

### X-Ray Crystallography

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### Gamma Radiography of Castings and Other Articles and Welds

Gamma radiography sources are all hermetically sealed in metal capsules, and so are free of inhalation or ingestion risks and also of alpha and beta radiation hazards, but are necessarily such powerful emitters of gamma radiation that the greatest care is needed in their handling and use. Records should be kept of all gamma ray sources received at the factory and of their subsequent disposal when spent. Precautions must be based on restricting the exposure of workers to the minimum possible time, on shielding and on distance protection; and, as with X-rays, regard must be paid to protection against scattered radiation as well as against the direct rays.

Very little radium or radon is now used for gamma radiography in factories, the common sources being cobalt-60, iridium-192, tantalum-182, caesium-137 and thulium-170. These sources consist of standardised small metal cylinders mounted within standardised aluminium alloy capsules so constructed that long-handled tools and rods can be used to insert them in or withdraw them from their protective containers as required. These handling tools should always be used. The sources themselves must on no account be picked up in the hands, not even once. If radium or radon sources are in use they should be tested periodically for leakage—three-monthly intervals are suggested—and at any time when damage is suspected.

The safe storage of these strong gamma ray sources, whether they are for use in a factory or by contractors on site, is a matter of importance. A site well away from normal working areas is to be preferred, and a choice can be made between a conventional type of safe which may be constructed of lead, iron or concrete, etc., and a lined borehole of suitable depth provided with a padlocked cover.

The amount of protection provided should, in general, be such that the free-air gamma radiation dosage rate at any point on the outer surface of the safe is not in excess of 7.5 mr/hr. If more than one radioactive source is stored in any safe, each should be kept in a separate individually protected compartment so as to reduce to a minimum the high dosage rate to which a person withdrawing a source is otherwise momentarily liable to be exposed. Where the source can conveniently be left in its protective container (see below) it is advisable to place the whole unit in the safe if this is possible. Radioactive sources that are not in use, in transit or being tested should be stored under lock and key. If radium or radon sources are stored, provision of mechanical ventilation for the safe or borehole should be made in order to deal with the possible escape of radon from leaking capsules. Only authorised persons should have access to safes. It should be a principle that sources be withdrawn only for the minimum time necessary.

Sources should be conveyed between safe and working site in protective containers, labelled to show the nature and approximate radioactive strength of the capsules within. There is a variety of such containers commercially available, suitable for a wide range of source strengths and gamma ray energies. These containers are constructed of lead or of high-density tungsten alloys. On account of the conflicting considerations of weight and the need for portability, it would often be impracticable in the case of the stronger sources to design such containers so as to reduce the free-air radiation dosage rate at their outer surfaces to 7.5 mr/hr or less; and as a compromise most of them can be designed to give a maximum of 1000 mr/hr at the surface or of 7.5 mr/hr at a distance of 1 m from the container.

In the conveyance of sources, therefore, this should be borne in mind, and to obtain the necessary distance protection it is usually possible to make use of a small push trolley or to carry containers slung on a long rod between two persons, the rod being pushed through or under the carrying handle. An operator who uses the sources may at times, especially with very strong sources, be exposed for short periods to free-air gamma ray dosage rates in excess of 7.5 mr/hr. For such a worker the integrating-type pocket ionisation chamber dosemeters, which may also be worn strapped to the wrist, are particularly valuable in giving an "on the spot" indication of the dosage received up to any instant in the weekly cycle; this enables the subsequent work to be so adjusted as to ensure that the total weekly permissible radiation dosage is not exceeded. In making the preliminary trial set-ups of work to be radiographed, a dummy source should be used; the active source should only be brought from the safe when all adjustments have been completed.

Many firms set apart a room for gamma radiography, the articles for examination being brought to the room, and this arrangement is to be preferred. The room will normally be constructed of brick or concrete, and should be so designed that, allowing for the distance factor, a free-air dosage rate of 7.5 mr/hr is not exceeded on its outer wall and door surfaces. All workers (other than the operator while making necessary source adjustments) should be excluded from the room during exposures. Gamma radiography rooms within which exposures are in progress should always be locked when left unattended.

It is one of the advantages of the gamma radiographic method that the sources can easily be conveyed to parts of the factory or site for articles to be radiographed on the spot, and when this is done the radiography should, as far as practicable, be isolated from other work in the vicinity; the gamma ray beam should be directed away from other working areas and its cross-section limited to the minimum necessary for the work. If a pit can be made use of for the work so much the better. In such work

on site the danger area within which a gamma radiation dosage rate in free air of 7.5 mr/hr would be exceeded should be temporarily roped-off or otherwise delimited, warning notices should be affixed, and warning lights or audible signals provided and operated immediately before and during radiographic exposures. For example, the roped-off areas around 500 mc unshielded sources of radium or radon, cobalt-60, tantalum-182, caesium-137 and iridium-192 should have radii of not less than 25 ft, 31 ft, 21 ft, 15 ft and 14 ft respectively. Where some shielding can be provided, as where single exposures are made with the source in its protective container and with only the front aperture open, the distances can be correspondingly less. For panoramic exposures for which articles to be radiographed are placed in a ring around a central source and the source has necessarily to be brought out of its protective container, use can be made of Bowden cable arrangements for removing the source from and returning it to its protective container by a worker operating at a distance. Alternatively a clockwork time-controlled motor can be used to expose and return the source at predetermined times, so enabling the radiography, if desired, to be done automatically at night in the complete absence of workers.

The use of such remote-control methods becomes almost mandatory when sources much above one curie of cobalt-60, two curies of tantalum-182 or five curies of iridium-192 or caesium-137 are in constant use for exposures by the panoramic technique, since otherwise the permissible handling times for such sources at a distance of about one metre are reduced to only a few minutes per day. For the use of stronger sources still, a specially constructed and protected gamma radiography room should be designed, within which the exposures are made by remote-control gear operated from outside; this control gear should be so interlocked with all entrance doors that operators cannot enter the room while the source is exposed, i.e., the procedure should be very similar to that now followed with X-rays.

#### Use of Radioactive Static Eliminators

These devices function by producing intense local ionisation of the air in the vicinity of the parts of the machine on which they are installed. Close proximity of workers or close handling of the devices should be avoided as far as humanly possible.

Whenever possible, eliminators should be installed facing inwards on machines rather than outwards, and the safest procedure is to make a radiation survey around each installation with a suitable monitor, or to use film badges to see whether shielding and distance protection are needed and, if so, how much. In the case of the typical strontium-90 (+yttrium-90) and thallium-204 eliminators from which only beta radiation travelling several yards in air is emitted, any necessary protection for machine operators and neighbouring workers can easily be provided by fixed, hinged or movable shields of such materials as 9 mm (for strontium) or 3 mm (for thallium) sheet Perspex or similar material, 5 mm or 2 mm thick glass or 1 mm or 0.5 mm of almost any common metal or alloy such as copper, brass, iron or aluminium. In the case of eliminators based on radium, protection is more difficult on account of the penetrating gamma rays emitted. Depending on local circumstances and the strengths of sources used, it may be necessary to use some lead shielding if workers need to approach closely to the devices for appreciable periods of time.

Static eliminators are normally provided by their makers with metal guard bars or shields, thick enough when in position over the active surface to prevent the transmission of all beta and alpha radiation. These guard bars, for which an easy clip fixing is to be preferred, should always be affixed when the units are being installed on or removed from the machines, when they are in store (in boxes or cabinets under lock and key) off the machines, and when local maintenance adjustments other than momentary ones are being done on the machines in the vicinity of the eliminators. When major repairs on machines near eliminators have to be undertaken the eliminators with their guard bars affixed should be removed to store. It is often practicable to provide an interlocking arrangement so that opening up parts of the machine near the eliminator, or parts of any cabinet enclosing it, for the purpose of local maintenance and adjustment or for clearing product chokages, etc., automatically moves a guard bar over the active surface.

The design and construction of eliminators, as also of thickness gauges,†† should be such as to ensure that the radioactive materials are completely sealed in, as they are when firmly bonded in solid solution within noble metals in the foils, plaques and wires supplied by national atomic energy authorities or commissions, so that they cannot become dispersed into the surrounding workroom, either suddenly by accidental fracture or as the result of melting in a fire, or gradually as a result of the normal operation of the process, so as to cause ingestion, inhalation or contamination risks. The user will need to protect the devices from all rough and unnecessary handling and from the action of abrasive or corrosive chemicals. Any cleaning of the active surface of the foils, etc., should be done as quickly as possible with a soft cloth, brush or cotton wool swab moistened with a solvent; this should be done either mechanically or by a worker wearing rubber gloves and with the source facing away from him.

To draw the attention of workers to the potential danger from the radioactive material, suitable parts of the eliminator might well be painted a distinguishing colour; the housings should also bear the manufacturer's name and serial number and a per-

†† See following subsection.

manent marking giving a short form of warning. The warning might be worded—

Radioactive strontium-90 source
($x$ millicuries, and date).
Poisonous. Do not destroy.

—to indicate the nature of the units to operatives who, in ignorance, might otherwise handle them or work very near to them in an unshielded state.

### Use of Radioactive Thickness Gauges

Precautions are generally very similar to those referred to above for static eliminators, since the sources are largely used in the same form of radioactive foils, and are mostly beta emitters of 5–10 mc strength chosen, according to the thickness of material being gauged, from $Sr^{90}$ ($+Y^{90}$), $Tl^{204}$, $Ru^{106}$ ($+Rh^{106}$), $Ce^{144}$ ($+Pr^{144}$), $Pm^{147}$ and $S^{35}$. The foil sources are usually sealed over with a 0.001-in. thick aluminium foil and then further protected from mechanical damage by a stout metal gauze. In many types of gauge this unit is also protected by a metallic ray-proof housing fitted with a ray-proof shutter system, and the only risk normally to be considered from an installed instrument is that from any direct and scattered beta radiation. This is usually very small, for much of the radiation, emitted over a small solid angle, is intercepted by the material being gauged or by instrument or machine fittings.

Monitoring around the installed device will usually disclose that the maximum permissible radiation dose rate is rarely exceeded 18 in. from the measuring gap, and any shielding necessary can readily be provided by Perspex flanges above or below the gap. Since workers rarely work close to thickness gauges for long periods, the protection problem is reduced to ensuring that operatives do not risk getting skin burns by closely handling unshielded active discs or by doing maintenance work, etc., on machines in the immediate vicinity of the gauges for long periods. Clip-on, sliding or rotating cover shields for each active source allow the radiation to be cut off during installation or maintenance work, and can be arranged to operate automatically when there is no strip in the measuring gap or when there is a power failure.

Gamma ray thickness and level gauges and X-ray gauges can be considered on their merits, and appropriate shielding arrangements made, using lead, iron or other high-density materials.

### Other Less Common Uses of Radioactive Materials

Precautions for these can be deduced by analogy from those given under previous heads. Tracer uses require special precautions similar to those described below for the handling of unsealed sources.

### Hospitals

With regard to the medical use of X-rays, a full account of the general and special precautions required (1) for diagnostic departments (fluoroscopy, general, dental and mass radiography); (2) for therapeutic installations (deep, skin, short-distance and contact therapy); and (3) in the protection of patients, has been given in the report of International Subcommittee III on protection against X-rays generated at potentials of 5 kV up to 2 Mv, in the Recommendations of the International Commission on Radiological Protection (1955). Since this document is now readily available it would be pointless to repeat the special precautions here.

It is of some interest to note that medical users of ionising radiations, on the evidence of the United Kingdom Film Badge Service run by the National Physical Laboratory since November 1952, show a consistent tendency to absorb rather more radiation from occupational exposure than do industrial or research workers, though a marked improvement in both categories has taken place since the commencement of the Service. This is exemplified in the table below, compiled by Smith (1953) giving the latest available summarised film test results on over 6000 medical, industrial and research workers throughout the United Kingdom. Films are at present being issued at the rate of about 50,000 a year.

It is also of interest that in the experience of the United Kingdom Royal Cancer Hospital and others, as reported by Mayneord (1951), the medical workers receiving the largest doses of radiation are not those in charge of X-ray equipment (even at 2 Mev or 20 Mev), who receive only small doses, but those handling large numbers of discrete radioactive sources, particularly radium and radon. This is not surprising in view of the relative ease with which X-ray equipment can be operated from safe working positions in shielded control cubicles or from behind protective screens, while the radioisotopes, whose radiation never ceases to be emitted, have to be prepared and handled in ever increasing amounts. Moreover the safe nursing of patients with perhaps 100 mg of radium or several hundred millicuries of other radioisotopes in their bodies creates a difficult protection problem, as does also the accurate insertion of such sources into position on or in the patients' bodies by the medical practitioners concerned.

**Analysis of Results of National Physical Laboratory Radiation Monitoring Tests**

| *Dose received (in r per week)* | *Percentage of workers in radiation group* | | | |
|---|---|---|---|---|
| | *Medical* | | *Industrial and research* | |
| | *1943–50* | *1951–52* | *1943–50* | *1951–52* |
| Less than 0.05 | 53.0 | 66.4 | 67.7 | 71.7 |
| 0.05-0.1 | 18.6 | 16.0 | 15.2 | 12.8 |
| 0.1-0.2 | 12.6 | 10.7 | 7.3 | 8.0 |
| 0.2-0.3 | 6.5 | 3.4 | 2.3 | 4.5 |
| 0.3-0.4 | 2.8 | 1.3 | 1.4 | 1.2 |
| 0.4-0.5 | 1.3 | 1.2 | 1.1 | 0.9 |
| Greater than 0.5 | 5.2 | 1.0 | 5.0 | 0.9 |

The best hope of achieving radiation safety for nurses and hospital staff in this difficult field appears to lie in education and experience and the use of techniques deliberately designed with due regard to protective requirements. Nurses and others should certainly approach patients constituting a gamma-ray radioactive risk for only the minimum essential time, and should normally keep at a distance of at least two metres at other times, while the patients' beds should be labelled with warning notices.

The special precautions for sealed beta-gamma sources used in gamma-ray beam therapy departments will be essentially similar in principle to those already described for factories in the subsection on the gamma radiography of castings and other articles and welds.

In hospitals special risks also arise from the considerable amount of active material in the form of radium, radon and cobalt needles and tubes and radium and $Sr^{90}$ beta ray plaques which have to be manipulated. The beta ray risks need taking care of by the intelligent use of distance protection, handling tongs or Perspex shields interposed between work and worker during bench and similar manipulations, which should be carried out as quickly as possible. Similar requirements apply to radium, radon and cobalt sources, except that screenage by at least 2.5 cm of lead shielding will be necessary. Tubes and applicators should be made up in separate, ventilated rooms, the applicators preferably by temporary workers not engaged on such work for periods exceeding six months.

Radium containers will need to be tested for radon leakages at least once in every period of three months, and to be stored, as will radon seeds and gaseous radioactive materials, in ventilated safes. All gamma emitters will need to be stored in shielded safes provided with drawers individually protected. Access to safes should be limited to a very few key persons, and transport should be in long-handled shielded boxes mounted on trolleys so as to give useful distance protection. A strict account must be kept in record form of the use and whereabouts of all radioactive material. Dressings and excreta from patients being treated with sealed radioactive sources will need to be checked or held for subsequent checking to ensure that all sources are accounted for. Precautions for the disposal of active waste will follow from the rules and advice given above and for the handling of unsealed sources from the subsection on this subject below and from that on inhalation and ingestion precautions above.

There will also be need to adopt a few additional precautions peculiar to the nature of the work, such as the daily routine monitoring of the clothing or bedding of hospital patients being treated with unsealed radioisotopes, the cleansing of such contaminated articles by laundering on the premises, or their storage until they are safe for laundering at public laundries.

### Agriculture

The current uses of radioisotopes in agriculture are almost wholly in the tracer field, and accordingly the special precautions necessary are essentially those given in the subsection on inhalation and ingestion precautions[2] and in the succeeding subsection on the use of unsealed sources used in radiochemical research and process laboratories.

### Use of Unsealed Sources

The largest use of unsealed sources is made in factories (radioactive luminous compound) and in radiochemical research, hospital and process laboratories.

The inhalation, ingestion or absorption through wounds of radioactive materials, especially from alpha emitters, involve a great danger. The general precautions have been dealt with and the general and special precautions appropriate to the use of unsealed radioactive substances, including luminising, are also set forth in the *Model Code of Safety Regulations for Industrial Establishments*,[5] in so far as they could be formulated in 1949 when the *Code* was prepared. It remains to mention or emphasise various precautions which have been seen to have special value in the light of experience gained since then.

### Luminising

So far as luminising is concerned it is found that, owing partly to the steady and continuous nature of the luminisers' work, these operatives, as a class, work closer to the permissible radiation dosage limits than any other type of radiation worker in factories. Moreover, unless the most meticulous attention is paid to cleanliness in methods of working there is a steady tendency for the radiation background of a luminising room to increase because of the spread of contamination from hands, clothing, implements, splashes, and the accumulation of radioactive dust and radon decay products in exhaust hood ducting, and also because once there, the radioactive contamination is substantially there for good on account of the very long half-life of radium. Also, when used as a powder likely to involve most serious inhalation and ingestion hazards, radium and its decay products emit alpha, beta and gamma radiations, together with a radioactive gas—a combination of risks met with nowhere else in the factory usage of radioactive materials.

Regular monitoring and routine ultra-violet light surveys in luminising rooms are of the utmost importance, since they make possible the detection and immediate removal of any splashes or areas of contamination, together with "forgotten" sources of radiation, such as ductwork or accumulations of old luminised parts on benches and in cupboards and drawers. For the same reason the accumulation of piles of luminised work around luminisers' working positions must be firmly discouraged.

The utmost use should be made of sleeved dry

boxes for luminising, or of localised exhaust hoods enclosed to the greatest extent practicable. Associated ductwork should be so constructed that it can readily be dismantled for cleaning.

The recent developments of mechanical methods of luminising have also emphasised the need for regular and thoroughly effective wet methods of cleaning of machine parts—especially sumps—liable to become contaminated during working periods, and for special attention to be paid to the shielding of the gamma radiation from such sumps which otherwise would be apt on occasion to produce background radiation at working places almost up to the maximum permissible dosage rate.

It has also been found that some of the varnishes and binding media used to incorporate the radioactive luminous compound into a paste before application contain plastic bases which are largely insoluble in water, so that contaminated overalls are returned from the laundries still radioactively contaminated though ostensibly clean. This matter needs study in relation to each source of supply of the varnishes and media used, but since many of the synthetic bases are soluble in both white spirit and trichlorethylene, one answer to the problem is to dry clean such luminisers' overalls instead of laundering them with hot soapy water. The use of spongeable plastic overalls instead of porous cloth ones and of large spongeable and waterproof bibs will also have special value in this connection.

In view of the particularly hazardous nature of radium, more attention than at present should be paid to periodic assessments of the total body burdens of radium which luminisers may accumulate, and firm steps should also be taken by each country to prohibit outwork in this occupation, i.e., the practice of taking articles home and luminising them there, often in the most unsuitable and dangerous conditions—a matter which is linked with current labour shortages in certain countries, and of which instances still occasionally come to light.

### Radiochemical Research and Process Laboratories

Earlier sections in this report have partially dealt with the precautions and design considerations which should apply to these laboratories, and further special points are mentioned below.

Reference has already been made to the radioelements specially liable to cause harm if deposited within the body. Where the radiation received is much in excess of 10 mr per 8 hours the processes must be considered as involving a possible hazard, and in these circumstances the following quantities of the various isotopes should be considered hazardous:

1. Any beta-active isotope which is specifically absorbed in a particular organ and for which the half-life is greater than a few days, handled in amounts exceeding 0.1 mc. In this connection the following isotopes are especially hazardous: $C^{14}$, $Ca^{45}$, $Fe^{55}$, $Fe^{59}$, $Co^{60}$, $Sr^{89}$, $Sr^{90}$ + $Y^{90}$, $Y^{91}$, $Zr^{95}$ + $Cb^{95}$, $Nb^{95}$, $Ru^{106}$ + $Rh^{106}$, $Sb^{125}$, $Te^{129m}$ + $Te^{129}$, $I^{131}$, $Cs^{137}$ + $Ba^{137}$, $Ba^{140}$ + $La^{140}$, $Ce^{144}$ + $Pr^{144}$, $Pr^{143}$, $Pm^{147}$, $Eu^{154}$, $Tm^{170}$, $Ir^{192}$.

2. Any material emitting beta rays in amounts larger than 1 mc, regardless of half-life.

3. Any material emitting gamma rays in amounts greater than that equivalent to 0.1 mg of radium.

4. Any alpha-active isotopes, for the handling of which special precautions are necessary.

A laboratory set apart and preferably built specially for the purpose should be used for active work, and if at all feasible two such laboratories or distinct sections of the same laboratory should be used, one for high levels of activity and the other for inactive and low-level work.

Floor space of approximately 200 square feet per person should be allowed, and general ventilation should be so arranged that air movement is from rooms or areas of no activity to rooms of higher activity, without any recirculation

Brick and concrete buildings are preferable to those made of wood, plaster boarding and other light materials, not only on account of resistance to fires, which might release radioactive materials into the atmosphere, but also because of the better shielding provided. Walls, floors, ceilings and woodwork should be treated to give them a non-porous washable surface readily capable of being cleaned and decontaminated or replaced. Concrete floors should preferably be covered with rubber, waxed linoleum or asphalt tiles (all easily replaced if necessary) or, if traffic is too heavy for this, painted to seal the pores with chlorinated rubber paints.

Interior walls may be tiled, painted with hard glossy paints or coated with strippable plastic paints. Water, gas, electric, etc., service pipes should run under the floor or in specially enclosed channels. The whole aim in the design of a radiochemical laboratory should be to avoid unnecessary ledges, surfaces or corners on or in which dust can accumulate, or any fittings or fixtures which render cleaning difficult. For the same reason laboratory furniture, books and papers should be kept to a minimum, and reagent and apparatus storage space should be enclosed.

Bench and fume cupboard surfaces of stainless steel are very satisfactory for resisting the absorption of radioactive contamination, but many other materials such as laminated plastics, glass sheets and polished teak can be used. Stainless steel is probably the best available material for sinks and for catch trays, though for the latter disposable heat-welded trays of polythene sheet made up as required are excellent.

In all decontamination the technique should be such that the contamination is removed, and not merely spread or worked into the surface. Metal surfaces can usually be decontaminated by washing with a solution of detergent in hot water. If this is ineffective a one-normal nitric acid wash should be used, followed if necessary by the use of a proprietary sequestering agent. Wood and concrete surfaces, being

absorbent, are usually extremely difficult to decontaminate. Detergent wash and sequestering agents may be used, but in bad cases of contamination it will be necessary to remove the entire surface.

Open bench surfaces will rarely be necessary on a large scale in radiochemical laboratories, since practically all handling of and operations on unsealed sources will take place in individually exhausted and filtered fume cupboards and gloved boxes (fitted with side transfer boxes). The cupboards and boxes should be ample in number and the cupboards provided with sufficient draught to ensure that linear air speeds of up to about 120 ft/min are possible when the front windows are fully open, though this will rarely be necessary. Fume cupboard and gloved boxes will again need to be so constructed and designed as to allow of ready and effective decontamination, and to be shielded according to the levels of activity being handled.

A storage safe for active sources, typically constructed of 1.5 in. lead or 9 in. brickwork, will meet most laboratory requirements, and any additional shielding for individual sources can be provided around them by lead blocks, etc. An additional "delay storage" safe for apparatus contaminated with short-lived isotopes can be very useful.

Protective and ordinary clothing, footwear and washing facilities should be housed in a vestibule to the radiochemical laboratory on a clean and dirty side system with a barrier between, and the same room should house the necessary hand, foot and clothing monitoring equipment.

Persons with cuts, abrasions or body wounds should not be allowed to work with unsealed radioactive sources. Waste disposal should follow the arrangements already discussed and any spillages of radioactive material should be cleaned up immediately and thoroughly.

Enough has been said to indicate the type of special precautions necessary in radiochemical laboratories, but a full consideration would need a paper devoted specially to the subject, and many such authoritative papers have already been published.

## VI. PROBLEMS ARISING IN URANIUM MINING AND PROCESSING

### Inhalation of Radon and of Radioactive Dusts

Stimulated by the world-wide search for the basic material of atomic energy production, the mining and processing of uranium ore is proceeding on an increasingly large scale in many countries of the world, notably in Australia, the Belgian Congo, Canada, Czechoslovakia, France, the Union of South Africa, the United States and the USSR. Sources of high-grade ore are limited, but considerable deposits of medium-grade ore are known and the occurrence of workable low-grade ore is widespread. The past history of most branches of the world's mining industry shows all too clearly that the conditions of work may vitally affect the health and safety of the workers concerned. Uranium ore is frequently associated with siliceous rock structures which when comminuted give rise to dusts capable of causing pneumoconiosis, and in particular silicosis.

Uranium ore is itself a highly toxic mineral, and can give rise to toxic conditions in the kidneys in particular. It has recently been shown that fine uranium ore dust (less than 0.5 micron mean mass diameter), as produced by modern mechanical methods, is far more toxic than dust composed of larger particles (more than 2.5 microns mean mass diameter). Other toxic substances such as chromium, nickel and arsenic may also be found in conjunction with the uranium.

In addition to these health risks, which are serious enough, there is some evidence that uranium miners have contracted lung cancer, thought to be due to the inhalation of alpha-emitting radioactive substances associated with the uranium, for instance radon and its disintegration products. The latter are readily deposited on any dust particles present in the mine air, and can be present in considerable amount if the mine is insufficiently or poorly ventilated.

It is known that until quite recent years some of the world's uranium mines, whether worked dry by hand methods throughout or by machine drilling, have relied entirely on natural ventilation to keep down the dust, though this state of affairs is being rapidly changed as more intensive mechanical methods of ore getting and transport bring complementary advances in mechanical ventilation and dust suppression methods. Nevertheless it is very disquieting to note that owing to the secrecy enveloping so much of the mining of uranium-radium ores in recent years, very little information has been made available in the world's health literature concerning conditions in these mines and the health of the miners employed in them.

It is generally agreed that as far as the radioactive risks are concerned, those due to radon are greater than those due to the radioactive dusts associated with uranium mining, but it is recognised that the adsorption on dust particles of the daughter elements of radon involves a serious radioactive risk.

The history of industrial health teaches us conclusively that whenever there is long-continued occupational exposure to dusts and gases there is probably danger, first to the lungs and respiratory passages and secondly to the remainder of the body tissues, resulting from the lungs' capacity to absorb toxic materials and transfer them to the blood stream.

The whole subject of the possible and probable ill effects on the workers' health of radon and radioactive dusts in the amounts present in uranium mining is one on which much more work needs to be done, and the results should be made available for critical study.

Dust is produced in all mining operations, but more especially in drilling, blasting, loading, transport, caving and filling mined-out areas. When radioactive ores are mined there are thus many sources of

radiation inherent in the operation of the mine.

A risk peculiar to the mining of radioactive ores is presented by the mine water. The water supply of a uranium mine may be the seepage water of the mine itself. This water will contain a considerable amount of suspended matter, some of it radioactive, derived from the workings, and if fed to pneumatic drills and atomising sprays to damp down the spoil during mucking-out, tramming and ore transfer operations, will ultimately release its solid burden as airborne particles to add to breathing hazards. Such mine water also dissolves radon at places where the concentration is high and returns it to the atmosphere where the water sprays are in use, thus tending to maintain a mean equilibrium concentration in the galleries.

Finally, there is a possibility of contamination with radon from the spoil or ore heaps on the surface.

### Protective Measures

Although all desirable information is not yet available on the dangers of uranium mining, it is already clear that the hazards can be very greatly reduced by the methods currently employed to combat the risks presented by silicogenic dusts and, to a lesser extent, by firedamp.

Precautions against dust in mining were exhaustively studied by a meeting of experts from 16 countries convened by the International Labour Office in December 1952. The meeting examined some 90 papers on the subject and drew up a set of recommendations, in the nature of a summary code of practice, which have been circulated to all the States Members of the International Labour Organisation.‡‡ The importance of adequate ventilation cannot be overemphasized.

Basic precautions against firedamp are set out in the *Model Code of Safety Regulations for Underground Work in Coal Mines for the Guidance of Governments and of the Coal-Mining Industry,*¶¶ adopted by a tripartite technical conference convened by the International Labour Office in 1949. Some of these precautions are applicable to mines in which radon is present.

A special precaution required at these mines consists in installing downcast shafts clear of spoil or ore heaps on the surface, and providing them with brick or timber chimneys raising the intake well above ground level. Further, a radon-free water supply should be used for sprays and drills. Any mine water used for this purpose should be freed of dissolved radon and suspended radioactive material in a water treatment plant.

Though the hazards at ore crushing and treatment plants, such as those in the atomic energy factories where the uranium is extracted and purified, are not as severe as in the mines themselves, and external radiation plays only a small part, workers must be prevented from inhaling or ingesting the radioactive dusts containing uranium and radium. Numerous precautions that serve this purpose will be found in the *Model Code of Safety Regulations for Industrial Establishments for the Guidance of Governments and Industry*§§ already referred to, published by the International Labour Office in 1949.




**Figure 1. Symbol adopted by the International Labour Organisation for dangerous radiations**

Emphasis may be laid here on the need for an exceptional standard of plant cleanliness, and the use of enclosed and exhaust-ventilated plant. Arrangements for monitoring, medical supervision, segregation of processes, the use of suitable protective clothing, etc., follow the requirements already referred to in Part V.

As far as the International Labour Office is aware there are no national regulations in force dealing specially with radiation risks in uranium mines. In the Union of South Africa a code of practice concerning safety precautions in uranium extraction and processing plants is in preparation, and in the Federal Republic of Germany a conference of state labour inspection officials has recently agreed that protective regulations should be drawn up for the extraction of radioactive ores.

As already mentioned, there is a serious lack of information on radiation risks in the mining and processing of uranium and on the application of the necessary precautionary measures in the countries concerned. It is very desirable that full information on these two matters be made generally available.

## VII. SAFETY IN THE TRANSPORT OF RADIOACTIVE SUBSTANCES

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### Labelling

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The symbol proposed by the ILO for dangerous radiations has been adopted or is being considered for adoption by a number of organisations (see Fig-

‡‡ Extracts from the recommendations which may be applicable to uranium mines are seen in Appendix V (not reproduced here).

¶¶ Geneva, ILO, 1950.

§§ See in particular Chapter X (Dangerous and Obnoxious Substances).

ure 1.) Since there is general agreement at the international level on the necessity for securing the widest possible uniformity in systems used for marking dangerous materials in all branches of industrial activity, including the various branches of the transport industry, it would appear highly desirable that this symbol should be used on danger labels which are to be affixed to containers of radioactive materials destined for international exchange.

## REFERENCES

1. International Labour Office, *The Protection of Workers Against Ionising Radiations*, Geneva (1955). Price: 50 cents or 3/–.
2. ILO, *Op. cit.*, Appendix I (not reproduced here).
3. ILO, *Op. cit.*, Appendix I, pp. 55–57 (not reproduced here.
4. ILO, *Op. cit.*, Regulation 207 (2), Appendix I (not reproduced here).
5. ILO, *Op. cit.*, Regulation 209 (1–46), Appendix I (not reproduced here).

# Measurements of Low-Level Radioactivity, Particularly the $\gamma$-Radiation from Living Subjects

**By Rolf M. Sievert,* Sweden**

It is said that the peaceful use of atomic energy will probably not involve more hazards for those who work in its establishments than is the case in many other industrial works. Reactors and other plants handling large quantities of radioisotopes require extensive protective measures, but if these are adequately realized the risk of accidents will be reduced to a minimum. But atomic energy utilization and the progressive use of large quantities of radioisotopes involve another type of hazard, which in principle is new. In some areas or over the entire world we have to expect increasing radioactivity, occasionally of comparatively short duration but often consisting of long-lived elements which may progressively accumulate, adding steadily to the natural amount of ionizing radiations.

The biological action of small radiation quantities has thus grown to a problem of the greatest importance. Our knowledge, however, is very limited in this field, especially as to the genetic action. It is possible that those who speak in gloomy terms of what will happen in the future if the general level of ionizing radiation increases, are overestimating the genetical hazards, although none can assert that they are definitely wrong. If we ask the specialists in genetics, we get no definite answer. There seem to be a number of fundamental problems that ought to be solved before the question of the genetical hazards associated with small quantities of ionizing radiations can be answered, and some of them may be very difficult to treat. What, for instance, is the spontaneous mutation rate in man, and what fraction of this rate is due to the natural ionizing radiation? Are not the changes in equilibrium between the mutations produced and the selection forces of prime importance in the discussion? If so, must we not take into account the enormous role in selection which is played by development in medicine and technique?

We are all aware of the urgent need of research work on the genetical effects of ionizing radiations. But we must not be too optimistic when estimating the time required before we can hope to get definite results that will give us the answer to the main question: what radiation doses can be permitted from a genetical point of view? In the meantime we must keep check of the radiation level throughout the world. It is essential also to know the radiation conditions in man before and after starting atomic energy work if, in the future, results of genetical research are to be utilized without delay.

Before the advent of artificial radioactivity, man was exposed to an ionization caused in part by cosmic radiation, but mainly by radiation coming from the ground or from the body itself, due to radioactive materials. One has to consider $\gamma$-radiation from elements in the U-Ra and Th-series and from $K^{40}$ and corpuscular radiation from activities contained in the body. Of special importance is the radon and thoron, since it can cause an uptake of activity through inhalation.

For some years measurements of natural radioactivity have proceeded at the Institute of Radiophysics in order to elucidate the limits within which the background radiation and the content of radioactive material in air, water and living subjects vary in our country. For such investigations, recording instruments placed on the spot for a long time would be the best technical solution in many cases. But several investigations can appropriately be conducted with simpler methods, preferably when a great number of observations are to be made simultaneously, to secure a general view of the magnitude of different effects and the circumstances which are relevant for further research work. Simple instruments often have another advantage—they do not fail at the critical moment.

In our case it was essential to have a method suitable for many different purposes, such as measurement of radioactivity in air, waters, and solid samples, having a very broad range of readings and tolerating rough handling. The measurements we have made hitherto point to the necessity of making a number of determinations at or near the place where the samples are collected. It is often much easier to draw relevant conclusions if readings can be taken, as required, from different positions in a house, on a river or in the air, where radioactivity is observed. Consequently, we have equipped a mobile laboratory for such investigations.

Due to the increasing use of large quantities of radioactive isotopes, it is desirable to have facilities for rapid investigation on the spot if radioactive

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material is lost or if accidents of some other nature happen. The Institute of Radiophysics is responsible for the supervision of radiation protection in all work in Sweden which involves the use of radioactive substances and has therefore found it necessary to adapt the radiation measurement ambulance for radiation protection service too.

For the purpose described here a further development of the so-called condenser chamber method[1,2] has proved apposite. In this method, which is shown in Fig. 1, each ion chamber *A* forms a self-contained unit with a small charging battery *B*. After a suitable exposure, readings are taken by inserting a contact plug *C* in the frame *D* of the chamber. The ionization is measured by a reading instrument *E* which can accordingly be used for an arbitrary number of chambers. This is of special advantage where space is very limited, if it is difficult to prevent contamination of the ion chamber, or if a large number of exposures have to be made simultaneously. Different applications of the method will be seen from Fig. 1. The instruments placed in the ambulance are shown in Fig. 2. For determining very low-level radioactivity the condenser chambers are moved to the low-background laboratory that will be described later. Figure 3 shows the result of a determination of the radium concentration in the pipe water in one of our cities. A one-liter sample is evaporated to 50 ml volume, this quantity simply being placed inside the chamber in a container permitting the radon to diffuse

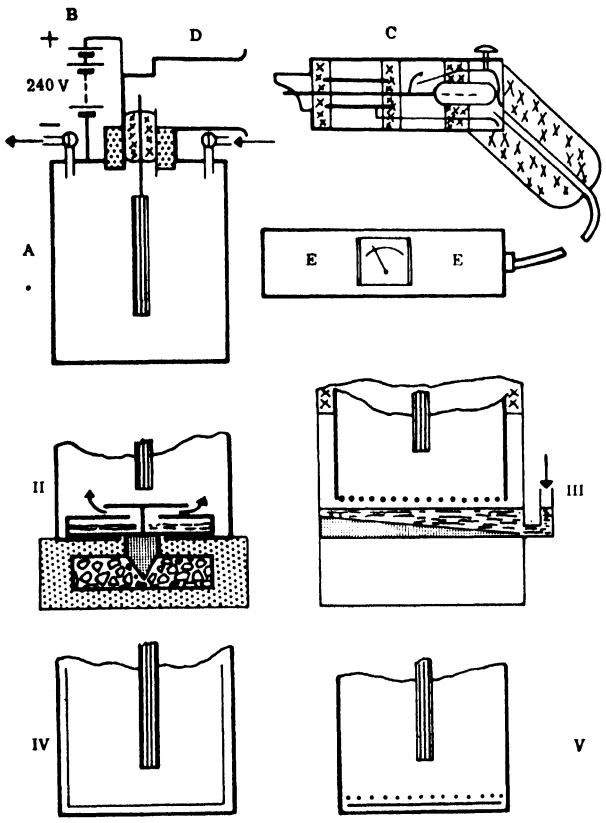



Figure 1. Different applications of the modified condenser chamber method

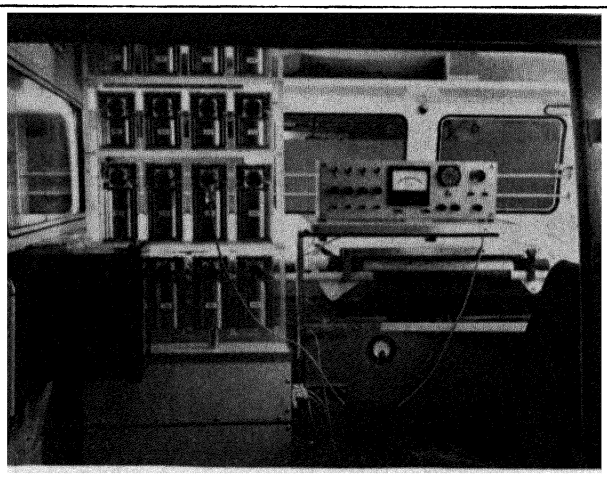


Figure 2. Inside view of the mobile laboratory

into the chamber. From the graph it is evident that concentrations down to about $10^{-16}$ gm Ra per ml may be detected.

Details of the methods of measurement and the pieces of apparatus used and a description of the ambulance for radiation measurements will be published later on.

For γ-background observations an 8-liter high-pressure ion chamber is used together with a direct-reading instrument permitting observations to be made in less than one minute. Investigations of the background γ-radiation have been made by Hultqvist[3] in 677 Swedish houses of three types: wooden, red brick, and concrete houses.

At the beginning we found some houses having comparatively high backgrounds and therefore thought that we should discover a sufficient number of such houses to provide us with material suitable for testing health conditions and making mutation tests for comparison of high- and low-level radiation effects. This has not been verified. As is shown in the graph in Fig. 4, there are, except in a few cases, only comparatively small differences in the radiation, indicating that such investigations would not be likely to give any significant results. The cosmic radiation is included in the figures.

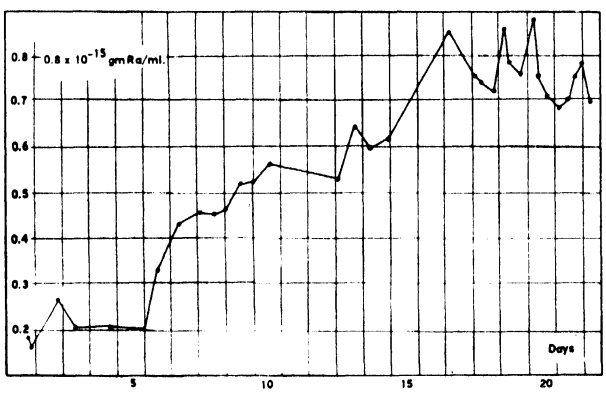



Figure 3. Record of determination of the radium concentration in a water sample

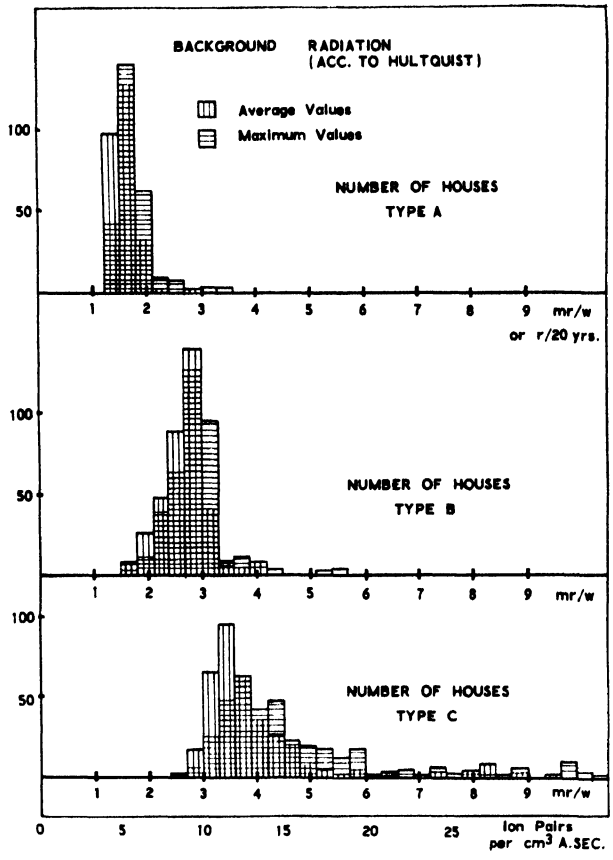



**Figure 4. Results of background measurements in houses. *A*, wooden; *B*, red brick; and *C*, concrete houses**

With the modified condenser chamber method mentioned, radon and thoron measurements have been made by Hultqvist[3] in different types of houses. The results show great variations, naturally depending very much on ventilation conditions. From Table I it is clear that very bad ventilation may lead to a high concentration of radon, slightly below that which the International Commission on Radiological Protection recommended as the highest permissible. It is worthy of note that the highest radon concentration was observed in a house having quite a normal γ-background. I shall return to this case later on.

The content of radioactive substances in human beings has been the subject of much discussion during the last few years. Krebs[4] found an average total radium content of about 10 nanogram (ng); Hursh and Gates,[5] however, only a tenth of a nanogram. Both these investigations were made on cremation ashes. Our first measurement in Stockholm indicated an average amount of less than 4 ng. Further measurements of the γ-radiation of living subjects suggest that the total-body content of radium contributes to the γ-radiation less than 10% of the total radiation which is again less than 1 ng and mainly due to $K^{40}$ (compare Burch and Spiers[6]). However, observations of the natural γ-radiation from human subjects involve some difficulties, and evaluation of the results obtained is not easy.

In 1948 at the Institute of Radiophysics we constructed a high-pressure ion chamber apparatus for the measurement of γ-radiation from living subjects down to the natural level. The instrument,[7] due to insufficient space, could not be adequately shielded against the γ-background radiation, and consequently a substantial correction had to be made for the absorption of this radiation. The variations of the cosmic radiation as well as a radon plant in the vicinity also caused disturbances. In 1951 a low-background laboratory, 8 by 4 meters in size, was therefore established in a solid rock just outside Stockholm. The laboratory is situated in a subway belonging to a sewage disposal plant and automobiles can be driven to a point close to its door, which has proved very suitable because of the high concentration of radon in the subway, although the ventilation is very good.

In the low-background laboratory we have now investigated more than 500 persons. During 1954 and 1955 the apparatus was running continuously without any changes in the technical arrangements. Most of the investigations during that time have referred to persons who have had no known contact with radioactive material other than natural sources. We thus have collected a normal material consisting of young persons from a school, middle-aged people from a large commercial enterprise, and old people from a home for the aged. Typical records are shown in Fig. 5.

The number of tested subjects, of varying sex, age, and weight, will be seen from Table II. The γ-radiation observed at various body weights, the standard errors of the various groups and the standard deviation of the single observations are seen in the graphs in Figs. 6–8.

In the graph in Fig. 9 the mean values for subjects in the age groups below 30 years, between 30 and 60 years, and above 60 years are plotted against the body weights. From the curves it is obvious that the males have a higher γ-radiation than the females and that the γ-radiation decreases with increasing age. This is easy to understand if we consider that most of the potassium in the human body is to be found in muscle

**Table I. Radon and Thoron Content of Air in Houses of Different Types* (in % of the number of flats investigated)**

| *Type of house* | *A* | | *B* | | *C* | |
|---|---|---|---|---|---|---|
| Number of flats | 55 | | 87 | | 83 | |
| Ventilation 10–20 min before investigation | yes | no | yes | no | yes | no |
| Radon content in $10^{-10}$ c/l air: | | | | | | |
| <0.5 | 71½ | 74 | 40½ | 26 | 20½ | 2 |
| 0.5–1.0 | 25 | 15 | 40½ | 32 | 20½ | 7½ |
| 1.0–2.0 | 3½ | 11 | 8 | 22 | 14 | 22 |
| 2.0–5.0 | – | – | 11 | 16 | 34½ | 33½ |
| 5.0–10.0 | – | – | – | 4 | 10½ | 27½ |
| 10.0–15.0 | – | – | – | – | – | 5½ |
| 15.0–30.0 | – | – | – | – | – | 2 |

* According to Hultqvist[3].

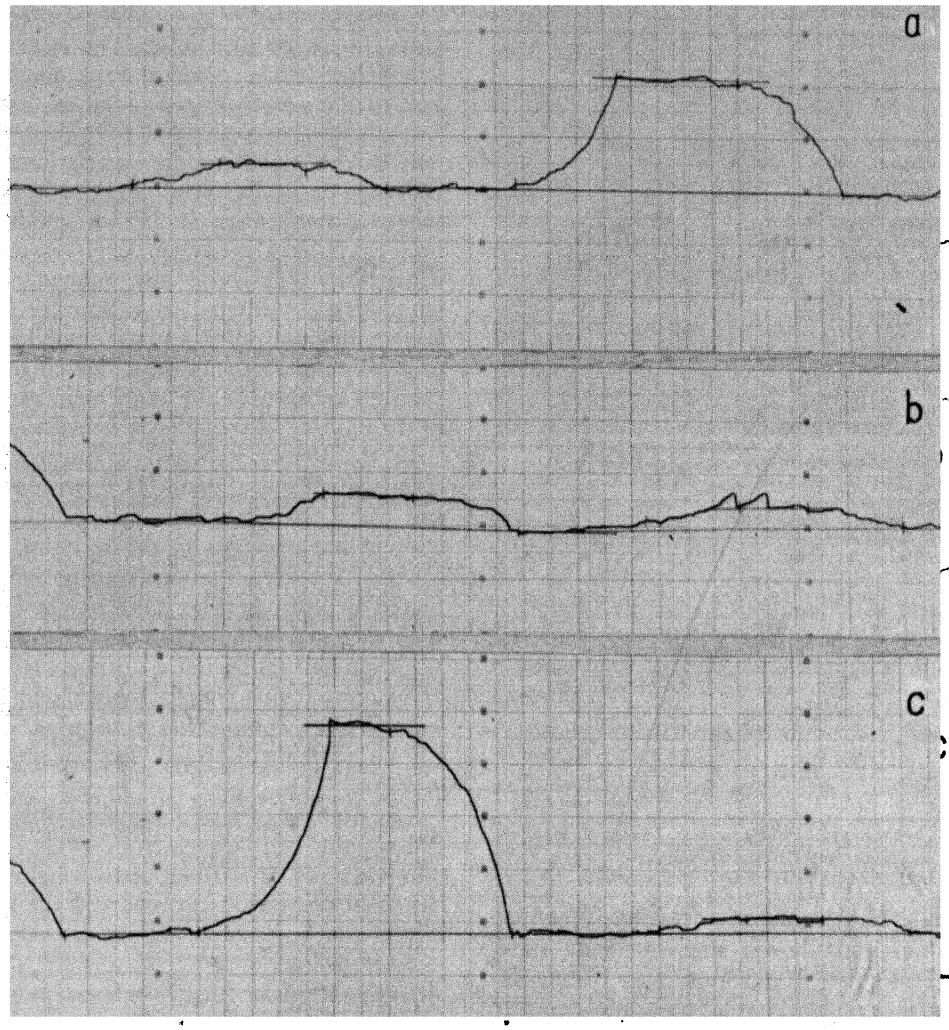



**Figure 5.** Records from the low-background laboratory. From the right (½ hour between two vertical lines): *a*, calibration with 0.02 μg radium, γ-radiation from boy weighing 34 kg; *b*, γ-radiation from young subject showing two cosmic ray showers, γ-radiation from adult woman; *c*, γ-radiation from girl weighing 24 kg, γ-radiation from contaminated person

Table II. Number of Subjects in the Normal Material (144 males, 162 females)

| *Weight class (kg)* | *Sex* | *Number of subjects* | | |
|---|---|---|---|---|
| | | *< 30 yrs* | *30–60 yrs* | *> 60 yrs* |
| <25.0 | M | – | – | – |
| | F | 6 | – | – |
| 25–35.0 | M | 18 | – | – |
| | F | 18 | – | – |
| 35–45.0 | M | 20 | – | – |
| | F | 17 | – | – |
| 45–55.0 | M | 11 | 4 | – |
| | F | 22 | 7 | 7 |
| 55–65.0 | M | 9 | 9 | 4 |
| | F | 16 | 6 | 15 |
| 65–75.0 | M | 11 | 10 | 14 |
| | F | 4 | 5 | 18 |
| 75–85.0 | M | 6 | 7 | 13 |
| | F | – | – | 15 |
| 85–95.0 | M | 2 | – | 6 |
| | F | – | 3 | 3 |
| Total | | 160 | 51 | 95 |

tissue and that the fat has a low potassium content. From old people with atrophied muscles and from heavy subjects having much subcutaneous fat absorbing the radiation, the γ-radiation yield is lower than from young and middle-aged subjects. The differences observed may consequently be attributable to the potasium content of the human beings and do not indicate any changes in radium content during life.

Calibrations made by means of the rubber dummies containing potassium showed for an average 70 kg man with a potassium content of 140 gm, a γ-radiation corresponding to 7.29 ng Ra-equivalent, and for a 40 kg body containing 80 gm potassium, 5.06 ng Ra-equivalent. Using these figures the potassium amounts can be calculated, assuming that the radiation is caused solely by potassium. In Table III the calculated potassium contents are compared with those obtained by other authors.

According to the results described, the total radium content of noncontaminated subjects is very likely to be less than a nanogram. This is also suggested by the

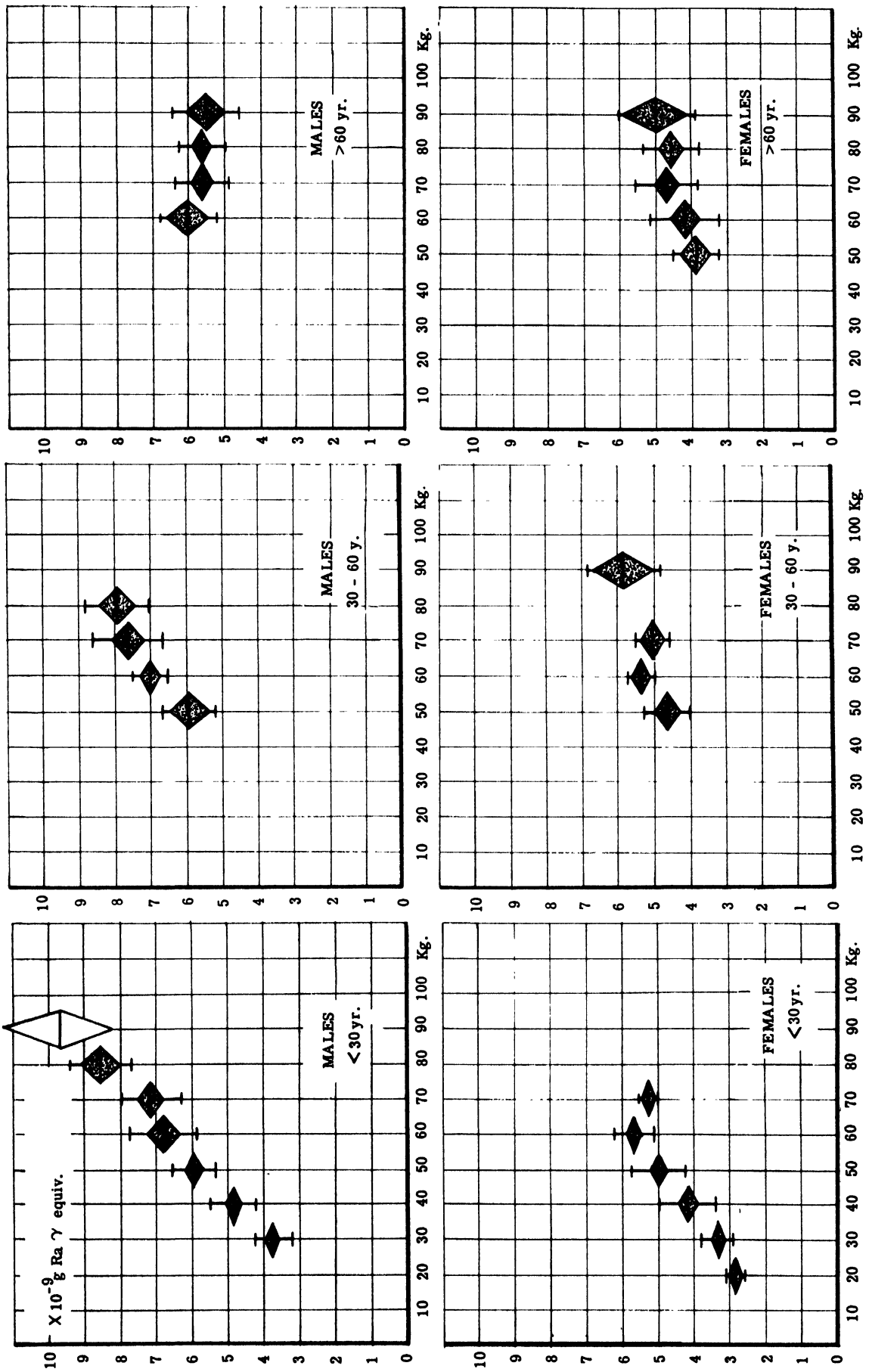



Figure 6.

Figure 7.

Figure 8.

The observed γ-radiations from people of both sexes, weight and age. The standard error of each weight group is represented by the height of the rhombi and the standard deviation of each single observation by the length of the vertical lines

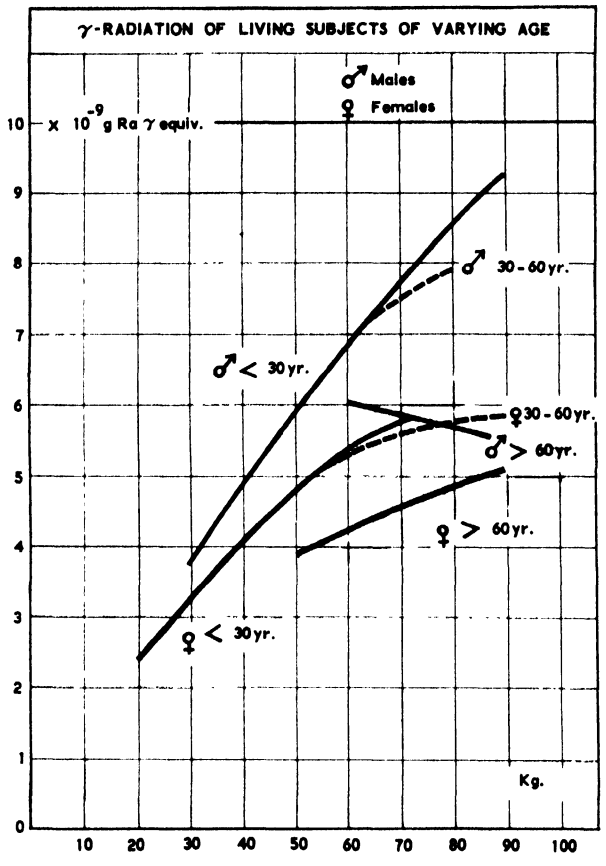



Figure 9. The average γ-radiation at varying ages as a function of weight

results obtained from subjects who have dwelt their whole lives in a city where the radium content of the water is at least 5 times that in Stockholm. As will be seen from Table IV, which contains preliminary results only, there may perhaps be a difference of the magnitude of 10% between the γ-radiation of subjects from Stockholm and those from city A, but not very much more. This would mean less than 0.2 ng total-body content in subjects from Stockholm if the radium originates entirely from the water consumed. Investigations of subjects from places with high radium contents in their potable waters are proceeding.

An interesting finding occurred during the investigations. The first record shown in Fig. 9, quite different from the normal curves, was obtained from one of the tested males. We measured the γ-radiation from his clothes and found that the slope of the curve was due mainly to decay products of radon (b and d of Fig. 10) and that the man himself also had a trifle higher value than would be normal (c and e of Fig. 10). An investigation of the air in his flat showed an extremely high radon content, very near the highest permissible concentration according to the recommendations of the International Commission on Radiological Protection. The γ-background, however, was not very high, about 4 mr/week. As far as we could see, the effect was due to bad ventilation. The case is the subject of further investigations.

About 100 persons working in the Swedish Atomic Energy Company have also been investigated. If we exclude some ten people who occasionally have shown a slight contamination, the mean values fit quite well into the normal curve.

The doses caused by the background γ-radiation in Swedish houses vary from 1 to 10 mr/week. Only in a very few houses does the dose exceed 5 mr/week. If, say 50 r, is considered to double the spontaneous mutation rate, then it seems scarcely worth while to seek any genetical differences between the populations living in houses of the different types tested. Further study of the radon amount in houses in combination with health tests would, however, be of interest.

In investigations of natural radiations of the kind dealt with in this paper, it may be said in general that a mobile laboratory has proved to be of great advantage, giving closer contact with the local problems. No doubt, however, a well-equipped ordinary laboratory for qualitative radiation measurements and more accurate determinations constitute a necessary additional facility.

As regards measurements of the γ-radiation from living subjects the following conclusions can be drawn on the basis of investigation of 306 persons who have lived most of their lives in Stockholm, where the radium content of the pipe water is less than $0.2 \times 10^{-12}$ curie per liter.

It has been found appropriate to have instruments for measuring γ-radiation from human subjects which are designed for continuous running requiring a minimum of technical service and so simple that an unskilled person can make all observations. Pieces of apparatus of this kind have also proved very useful for a number of investigations of samples of extremely low specific activity.

To be able to observe an increase of γ-radiation due to storage of radium in the skeleton, or to small amounts of other radioisotopes in human subjects, it is necessary to take into account several factors:

1. The person to be investigated and his clothes must be sufficiently free from decay products originating from the radon and thoron in the air.

2. The normal values representative for the age, weight and physical conditions of the person in question must be known from tests of a sufficient number of similar subjects, if the person's normal γ-radiation has not been tested earlier.

3. It seems correct to assume that if the water ingested during life has a radium content less than $10^{-12}$ curie per liter, the radium stored in the body cannot, even in old people, give a higher contribution to the γ-radiation than 20 per cent of the $K^{40}$ γ-radiation.

What is said here refers to methods which do not permit the separation of γ-rays of different energy. If it seems possible to improve measurements of γ-radiation from living subjects by using scintillation counters[16,17] it will be worth while trying to develop methods based on this principle.

The methods described in this paper are intended for observations of the natural radiation and of the additional radiation due to development in atomic

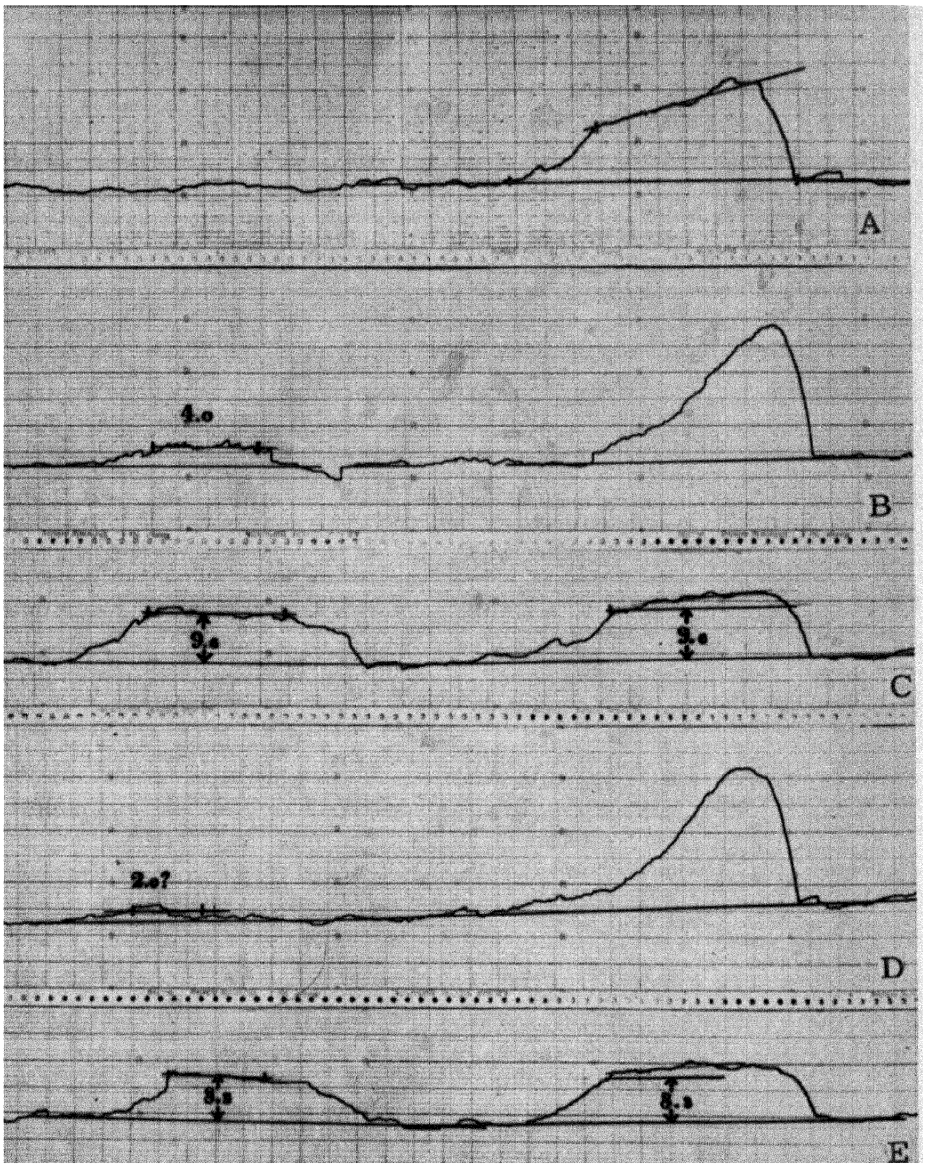



Figure 10. Records of measurements of a person with clothes contaminated by decay products of radon. From the right (½ hour between two vertical lines): *a*, subject having decay products from radon in his clothes; *b* and *d*, the clothes tested under the same conditions as in *a*; *c* and *e*, the same subject as in *a* without clothes, taken simultaneously with *b* and *e* respectively. Eight hours between the right and left record

energy work. A careful study of the doses received which are representative for the present population, and a still more careful follow-up of their possible increase in the future, must be regarded as an unconditional requirement for our generation.

As long as we are doubtful about the genetical consequences of small doses of ionizing radiation in man, it is our duty to prevent every undertaking that is found to be followed by an increase of radiation which may raise the irradiation of any population above an amount considered as safe. By studying the variations of the dose caused by natural radiations, it seems possible to arrive at a reasonable maximum permissible dose which can be provisionally used until we know more about the genetical hazards.

For a great many radiation protection purposes we already possess recommendations made by the International Commission on Radiological Protection and adopted by the International Congress of Radiology in Copenhagen, 1953. Most of these recommendations, however, are intended only for occupational work with radioactive material and they need completion with regard to the general radiation hazards problem of the world.

For that purpose it seems necessary to have an international protection organization of experts in

**Table III. Potassium Content in Living Subjects**

| Author | Method | Number of subjects | Age | Average weight kg | % K |
|---|---|---|---|---|---|
| Corsa *et al.* (1950)[8] | Isotope dilution (exchangeable potassium) | 30 M | 21–32 | | $0.18_1 \pm 0.01_9$ |
| Aikawa *et al.* (1952)[9] | Isotope dilution (exchangeable potassium) | 20 F | | | $0.12_8 \pm 0.01_1$ |
| Edelman *et al.* (1952)[10] | Isotope dilution (exchangeable potassium) | 14 F<br>33 M | | | 0.16<br>0.18 |
| Blainey *et al.* (1954)[11] | Isotope dilution (exchangeable potassium) | 17 M<br>7 F | | | $0.17_9 \pm 0.01_4$<br>$0.13_7 \pm 0.01_9$ |
| Rundo and Sagild (1955)[12] | Isotope dilution (exchangeable potassium) | 6 M<br>4 F | 22–31<br>17–28 | 76<br>61 | $0.19_0 \pm 0.02$<br>$0.16_7 \pm 0.01_7$ |
| Shohl (1939)[13] | Chemical analysis (total potassium) | | | | $0.21_5$ |
| Hawk *et al.* (1947)[14] | Chemical analysis (total potassium) | | | | 0.35 |
| Burch and Spiers (1954)[15] | Total body radioactivity (total potassium) | 10 M+<br>3 F | 19–20 | | $0.21_5 \pm 0.01$ |
| | Total body radioactivity (total potassium) | 11 M<br>4 M | 26–41<br>60–79 | | $0.21_2 \pm 0.01$<br>$0.21_5 \pm 0.02$ |
| Rundo and Sagild (1955)[12] | Total body radioactivity (total potassium) | 6 M | 22–31 | 76 | $0.21_5 \pm 0.01_2$ |
| | Total body radioactivity (total potassium) | 4 F | 17–28 | 61 | $0.19_7 \pm 0.01_5$ |
| Sievert (1955) | Total body radioactivity (total potassium) | 20 M | 10–13 | 40 | $0.19_1 \pm 0.008_3$ |
| | Total body radioactivity (total potassium) | 11 M | 20–29 | 70 | $0.19_6 \pm 0.01_0$ |
| | Total body radioactivity (total potassium) | 10 M | 30–49 | 71 | $0.20_6 \pm 0.01_3$ |
| | Total body radioactivity (total potassium) | 14 M | 62–84 | 72 | $0.15_5 \pm 0.008_2$ |
| | Total body radioactivity (total potassium) | 17 F | 10–13 | 40 | $0.16_4 \pm 0.01_1$ |
| | Total body radioactivity (total potassium) | 4 F | 25–28 | 69 | $0.14_5 \pm 0.005_2$ |
| | Total body radioactivity (total potassium) | 5 F | 36–56 | 72 | $0.13_5 \pm 0.009_3$ |
| | Total body radioactivity (total potassium) | 18 F | 66–86 | 71 | $0.12_9 \pm 0.008_6$ |

**Table IV. Preliminary Comparison of Radium Content in Subjects from a City (A) with Pipe Water Containing about $10^{-15}$ gm Ra/ml and from Stockholm with Water Containing about $0.2 \times 10^{-15}$ gm Ra/ml**

| City | Number of subjects and sex | Average age | Average weight | Radiation in ng γ-eqv. | Difference in γ-radiation* |
|---|---|---|---|---|---|
| *A* | 9 M | 18.3 | 61.6 | 8.13 | |
| Stockholm | 9 M | 19.1 | 61.4 | 6.94 | + 16.7% |
| *A* | 5 M | 20.0 | 71.0 | 7.98 | |
| Stockholm | 11 M | 24.5 | 70.3 | 7.18 | + 10.2% |
| *A* | 5 M | 18.0 | 83.4 | 8.25 | |
| Stockholm | 6 M | 26.5 | 78.7 | 8.37 | − 7.2% |
| *A* | 5 F | 20.2 | 51.8 | 5.51 | |
| Stockholm | 22 F | 18.1 | 50.7 | 5.04 | + 7.0% |
| *A* | 3 F | 20.7 | 60.7 | 7.18 | |
| Stockholm | 16 F | 17.7 | 58.7 | 5.54 | + 25.2% |
| Average | | | | | + 10.4% |

* Corrected for differences in body weight.

radiophysics and radiobiology including radiogenetics, empowered to promote essential research work in its field and to organize continuous general supervision of the radiation level in the world. An organization with this program must have a high degree of independence, sufficient means at its disposal, and should not be under any restrictions as to publication of its findings. These should be reported periodically in some suitable way, with summaries in a form that would be understood even by the man in the street. It is to be hoped that the present International Commission on Radiological Protection may be given possibilities to adopt such a programme.

The world problem of general radiation hazards is interesting and fascinating. There are many young scientists who would enter wholeheartedly into international teamwork in this important field if organizational and financial requisites could be created for the purpose.

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# Permissible Exposure to Ionization Radiation

**By Lauriston S. Taylor,* USA**

This short note, reflecting the general opinions of most members of the National Committee on Radiation Protection, directs attention to a major problem that must be faced when we consider large-scale, peaceful developments of nuclear energy. The problem directly influences our whole philosophy with regard to radiation protection and centers about the fact that the genetic effects of low-level exposures of a large fraction of the population may ultimately decide the permissible dose for all persons. Until relatively recently the prime consideration in deciding upon the maximum permissible dose of ionizing radiation has been the exposure of the single individual. Future considerations of the maximum permissible dose may conceivably involve an entirely new feature; whereas past decisions have been based on scientific principles alone, future decisions may have to include political and economic factors.

The current maximum level of permissible exposure for the single individual rests on the philosophy that exposure at this level throughout his adult lifetime is believed unlikely to cause him bodily injury at any time during his lifetime. Based on this major premise, the present permissible exposure levels are acceptable both from the plant and the individual's viewpoint, and do not appear to involve an unreasonable working risk. Such levels are, however, also based on the additional premise that only a small portion of the world population will be so exposed up to the close of their reproductive lifetime.

On the other hand, where large population groups may be exposed, the preservation of the genetic balance of the population may require that the exposure per individual be limited to only a very small fraction of the individual occupational exposure.[1] Up to the present, most of the pertinent data is from animal rather than human experiments, yet we are forced to accept tentatively the animal data as applying to man.

For purposes of discussion, let us take Muller's[2] statement that an exposure of 80 r to the gonads would double the natural mutation rate and that such exposure repeated generation after generation might seriously upset the genetic equilibrium. Presumably this would be genetically unacceptable in view of the double-heavy genetic load thrust upon the unexposed population, and in view of the present trends in reproductive practices. Muller suggests a maximum average exposure per individual per reproductive lifetime of 20 r, which would result in an increase in the mutation rate of only 25 per cent. Other authorities have arrived at figures as low as 3 r for a permissible average "lifetime" exposure. Either figure is much lower than the presently accepted individual occupational exposure limit which may be as high as about 400 r per lifetime.

If we are to adhere to the 20 r average for the whole population, not more than 5 per cent of the people could be permitted to receive a full occupational exposure of 400 r to the gonads. For the United States this would be some eight million persons—a figure that we are unlikely to attain for many years to come. One should also consider that most radiation workers do not receive exposures over periods as long as 25 years, and that in fact only a very small number receive more than about one-third of the permissible exposure. On the other hand, there must be added to this, medical and diagnostic exposures wherein radiation may reach the gonads.

Before facing the problem of determining how much additional radiation may be received by non-occupational population groups, it is essential to make a careful evaluation of existing exposure patterns. Since the prime consideration will be genetic effects, such exposure evaluation should be limited to the gonads of persons before the close of their reproductive lifetime.

Concurrent with, or a part of, such a study should be certain sociological investigations. Reproduction habits will play an important role, and these will vary markedly depending upon such factors as race, education, inbreeding within certain geographical limits, etc. In averaging the exposure of population groups, erroneous results would be obtained by equal weighting of say the population of New York City and an Indian Reservation in the West.

Since the size, distribution, and nature of a population group may influence the pattern of cross-breeding, it may be worthwhile even within a single country such as the United States, to consider very different average exposures for different parts of the country. Such differentiation might present almost impossible administrative problems because of population movements, yet there are relatively large and different population groups between which cross-breeding is negligible. This same consideration will, however, be likely to necessitate lowering of average exposures in some areas where inbreeding is high

* National Bureau of Standards.

within a population group that remains somewhat static in location. (This may be somewhat less of a problem in cities of the United States as compared with those of Europe and Asia.) There will undoubtedly be special problems of this sort in certain areas and it might be worthwhile to treat them specially, rather than inflict unnecessarily low permissible exposures over the country as a whole. Analysis of individual situations, while costly to perform, may nevertheless be sound economy in the end.

From the point of view of power reactors, the limits set for radioactive wastes will vary considerably, dependent upon location. This, of course, has been one of the prime considerations in locating reactors thus far, and each case has been treated on its own merits.

We have to face the high probability of an enormous growth in the uses of nuclear energy. It is doubted whether anyone has ventured to predict this for the next 20 or 100 years. Looking backward at the enormous changes in our civilization brought about by technological advances in the past four or five generations, indicates the futility of planning ahead in detail a similar range of time. This does not imply, however, that we should stand passively by and let nature take its course. There is much that can be done in preparation for the future.

As the invention of the internal combustion engine revolutionized the world, so also may we expect controlled nuclear energy to do so again. The engine has brought vast improvements into our material ways of life. One wonders whether, if in 1910 it had been anticipated that the engine would be responsible 40 years later for 30,000 deaths a year on United States highways alone, there would have been a hue and cry to curtail its further use. In spite of the fact that we somehow accept this carnage, we might have been able to have held it down, had the problem been attacked while automobiles were in their infancy.

With nuclear energy the situation is importantly different—even if more complicated. We know a great deal about its potential hazards and to some extent how to cope with them. In addition, we have some time within which to solve the special problems. However rapid the growth in the use of atomic energy, there is still some leeway that will allow us to proceed with technological developments before we outrun our practical limits in the methods of protection. The greatest and most serious limitation is that imposed upon us by genetic considerations. Our most substantial advances in knowledge of genetics have been within the past two or three decades—a very short time. The results of this knowledge have been a major consideration in discussion leading to our present concepts of permissible dose, yet we have avoided any attempts to rigorously define the genetic limitations. (It is presumed that the geneticists, to be safe, assume the most pessimistic conditions.)

At some time, probably not within the immediate future, man will be faced with making an inescapable decision. At what point may the advantages of atomic energy be offset by the disadvantages to the future man? And who will have the abundant wisdom to recognize that point and do something about it? Will it be known, in time for such a decision, just what radiation may do to man's future? I believe that at some point a decision involving an educated gamble with man's future will have to be made, and history of the past indicates that such a decision may be made on the less- rather than the more-conservative side. The decision does not need to be made tomorrow or probably for some years. In the meantime, we can continue our present pace with relatively little risk. But in that same meantime, it is felt that we should start to condition our thinking for a change in philosophy with regard to radiation exposure. On the basis of today's knowledge of ourselves, we may be expected to show a willingness to accept more rather than less radiation exposure insofar as its effects ten or twenty generations hence may be felt.

In this same time, we should also devote our every energy to keeping radiation exposure of persons to the minimum compatible with reasonable progress and good sense. Through education and the dissemination of wisely chosen information, we can do much better than at present in matters of radiation protection, without at the same time fettering a source of great benefit to mankind. The better we do the ordinary job of protection today, the longer we postpone the fateful decision on man's future.

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# Maximum Permissible Exposure Standards

**By Masanori Nakaidzumi,* Japan**

The maximum permissible radiation dose which is at present in use originated from a report made by Mutscheller in 1925. As a result of his studies of health conditions of those working in X-ray laboratories he concluded in his report that if dosage does not exceed $1/100$ skin erythema dose per month it will not cause injury even after a long period. He said in conclusion that this amount is the tolerance dose. This tolerance dose of Mutscheller corresponds to 200 mr per day. Judging from the condition of X-ray applications in and around 1925, as well as from the term "skin erythema dose" itself, doctors were concerned with those X-ray injuries mainly restricted to the skin. This tolerance dose was adopted as that of international recommendation at the Third International Congress of Radiology of 1931 in Paris. In 1937 at the Fifth International Congress of Radiology held at Chicago this dose was replaced with 100 mr per day on the condition of measurement in the air. Since then the quality of radiation used and its applications in radiology have changed considerably. Besides, if the dose of 100 mr/day is observed strictly by X-ray workers there will not be any radiation injury, but from the animal experiments and genetics this dose of international recommendation seems to have no such safety factor as it was supposed to guarantee. Thus the international recommendation of 1937 has been revised to the present dose of 300 mr a week which is widely used today. However, this dose of 300 mr per week is not the result of revisions necessitated by the actual radiation injuries experienced by workers but is the value adopted after the result of Mutscheller's field study having been twice revised. Therefore, we do not know what safety factor this dose includes. To know exactly what safety factor is provided by a certain standard is indispensable for planning protective precautions against radiation injury. Meanwhile, in Europe and the United States no radiation injury to workers is reported as the result of adherence to the maximum permissible dose for human beings, and it is impossible to study and decide what safety factor this permissible dose provides; whereas in Japan, radiation injury still occurs in the X-ray laboratories. Therefore, we must study the question again by adopting a new method as Mutscheller did in 1925. When the results of this study are clear we shall know the safety factor of the present maximum dose of 300 mr a week and thereby greatly contribute to the planning of the protective precautions. In order to measure radiation dose to which X-ray technicians are exposed while they are engaged in the above study, we are using film badges of Japanese manufacture† together with those of American manufacture, which are to be sent to New York for development and measurement.

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† Anyone interested in knowing the details of the film badges used in Japan is welcome to make an inquiry.

# On the Maximum Permissible Dose of X- and Gamma-Radiation

**By W. Jasinski* and I. Zlotowsky,† Poland**

The biological effectiveness of X-rays and gamma rays has been investigated by carrying out differential blood counts of persons occupationally exposed to radiation in the premises of the Institute of Oncology at Gliwice. The hematological data of 145 employees for a period of over three years have included the counts of red blood cells, granulocytes, neutrophiles, lymphocytes and monocytes as well as the determination of the hemoglobin content of the peripheral blood.

The daily doses of stray radiation received by individual employees were estimated on the basis of ionisation measurements made by means of ordinary radiation monitors, calibrated G-M counters and pocket condenser-type ionisation chambers of our own construction.[1] Radiation monitoring was carried out periodically in various parts of the Institute,[2] while the individual blood counts were repeated regularly every three or, sometimes, every six months.

All 145 employees submitted to the regular hematological studies were divided into four groups comprising *(1)* 5 persons exposed to 0.07 to 0.15 r/day, *(2)* 13 persons exposed to 0.008 to 0.012 r/day, *(3)* 35 persons exposed to 0.004 to 0.02 r/day or occasionally to 0.2 r/day, and *(4)* 93 persons exposed to

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**Table I**

| *Blood element* | *Mean levels of blood elements* | | |
|---|---|---|---|
| | *Initial value (a)* | *3 years after employment (b)* | *Differential counts (a–b) as % of a* |
| 1. Hemoglobin (gm) | 11.39 | 10.46 | −8.1 |
| 2. R.B.C. | 4,007,000 | 3,757,000 | −6.1 |
| 3. W.B.C. | 6378 | 6047 | −5.2 |
| 4. Granulocytes | 3973 | 3811 | −4.1 |
| 5. Neutrophiles | 3796 | 3637 | −4.2 |
| 6. Lymphocytes | 2167 | 1985 | −8.4 |
| 7. Monocytes | 226 | 267 | +18.1 |

**Table II**

| *Group of empl.* | *Occupat. exposure* | *Type of radiation* | *Whole-body dose in r/day (measured in air)* | *Average differential counts at the end of the 3 year period in % of the initial counts* | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | *Hb (gm)* | *RBC* | *WBC* | *Granulocytes* | *Neutrophiles* | *Lymphocytes* | *Monocytes* |
| 1+2+3+4 | Various | X-rays and/or gamma rays | - - - - | −8.1 | −6.1 | −5.2 | −4.1 | −4.2 | −8.4 | +18.1 |
| 1 | 6.5 hr/day over the entire 3 yr period | Gamma rays | 0.07–0.15 | −32.5 | −16.5 | −22.2 | −15.6 | −17.0 | −17.0 | +40.0 |
| 2 | 6.0 hr/day over the entire 3 yr period | X-rays | 0.008–0.012 | −9.5 | −10.0 | −12.7 | −0.8 | −10.3 | −13.6 | −11.0 |
| 3 | 8.0 hr/day over the entire 3 yr period or 5.0 hr/day occasionally over a period of 3 to 12 m | Gamma rays | 0.004–0.02 / less than 0.2 | −6.3 | −6.4 | −17.4 | −25.6 | −25.5 | −4.0 | +18.7 |
| 4 | 8.0 hr/day over the entire 3 yr period | Gamma rays | 0.004–0.02 | −7.5 | −2.7 | −3.8 | −5.8 | +5.8 | −7.5 | +20.3 |

0.004 to 0.02 r/day. The detailed conditions of exposure are indicated in Table II. Table I presents the average values of the differential blood counts for all 4 groups over the entire three-year period.

From the data summarized in Table II it is possible to conclude that in the fourth group of employees the biological response to radiations has been very slight. Also in the second group relatively small changes in the peripheral blood have been observed. In the third group, however, an appreciable decline in the total white blood cell counts has been noted, due mainly to a reduction in the absolute number of granulocytes. In the first group the occupational overexposure is exhibited by a change in all blood counts with a large increase in the number of monocytes.

The data presented in Tables I and II lead to the conclusion that the daily dose of radiation received by persons who are occupationally exposed to gamma rays and/or X-rays should not exceed 0.01 r per day.

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# Validity of Maximum Permissible Standards for Internal Exposure

By R. C. Thompson, H. M. Parker and H. A. Kornberg,* USA

It is well recognized that the validity of maximum permissible standards for internal exposure to radioisotopes is not comparable with the validity of standards for external radiations. The International Commission on Radiological Protection (ICRP), in 1950, did not "consider that there is sufficient information to make firm recommendations concerning maximum permissible exposures to internal radiation from radioactive isotopes."[1]

In 1953, the Subcommittee on Permissible Internal Dose of the National Committee on Radiation Protection (USA) published maximum permissible exposure limits for 70 internal emitters.[2] Recognizing that, "in some cases there is considerable uncertainty about the maximum permissible values given . . .," the Subcommittee felt that it was "desirable to agree upon what are considered as safe working levels for these radioisotopes now rather than wait until more complete information is available."

To establish permissible limits for internal emitters, one of two approaches is presently used. If the radioisotope in question is a bone seeker, its toxicity is compared with radium. The radium permissible limit is derived directly from human exposure experience, and is currently established at 0.1 $\mu$c total-body burden. Although there is considerable uncertainty in the interpretation of the human exposure data, this value for radium is without doubt the most realistic of the recommended permissible body burdens. The comparison of a bone-seeking element, as $Sr^{89}$ or $Pu^{239}$, with radium may be made from the results of animal toxicity experiments in which the ratio of concentrations of radium and the isotope in question, which result in equivalent damage, are determined. In the absence of such a direct comparison, the concentrations which should result in equivalent damage may be calculated from the ratios of energies of disintegration and of relative biological effectiveness of the two emitters and from data on their comparative biological distribution and retention.

For the majority of radioisotopes recourse must be had to experience with external radiation. The limiting dose rate of external radiation adopted by the ICRP of 0.3 r per week was based on the maximum permissible dose rate for irradiation of the blood-forming organs.[1] Despite differences in tissue sensitivities, 0.3 rem per week is presently considered the maximum permissible dose rate to the critical organ, whatever it may be.

Considerable uncertainty attaches to maximum permissible body burdens determined by either method. Additional uncertainties are introduced in the calculation of maximum permissible concentrations (MPC) for radioisotopes in air or water. These limits for air and water are based, of course, upon the permissible body burden, but require, in addition, information on the fraction of inhaled or ingested radioisotope which reaches the critical organ, and information on the retention of the radioisotope in the critical organ.

The problem of establishing MPC's for internal emitters in air or material that may be ingested can be resolved into three factors. (1) Determination of the permissible critical organ burden; (2) Determination of the radioisotope concentration in air, water, or other directly ingested material which will result in this permissible burden upon chronic exposure; and (3) Determination of limiting factors other than direct human ingestion, e.g., damage to nonhuman organisms, concentration along food chains terminating in the human.

The balance of this paper is given over to a description of work carried out in our laboratories which has resulted in a better understanding of certain of the parameters involved in all three of these factors. Wide variations from currently accepted values were found, particularly in those parameters necessary to the establishment of MPC's for air or ingested material. It seems probable that the uncertainties in our present knowledge of factors 2 and 3 are much greater than the uncertainties in the permissible human body burdens.

## PLUTONIUM-239

The presently accepted MPC for $Pu^{239}$ in water is $1.5 \times 10^{-6}$ $\mu$c/ml.[2] This is based, among other parameters, on an assumed absorption from the gastro-intestinal tract of 0.1 per cent of the ingested dose, a figure derived from a measured absorption of 0.01 per cent, with a safety factor of ten included because measurements were made at relatively high plutonium concentrations. In experiments involving the chronic feeding of plutonium solutions to hundreds of rats,

* General Electric Company.

an average absorption of 0.0028 per cent was measured, with 90 per cent confidence limits of ± 0.0008 per cent.[3] This figure holds for solutions varying in plutonium concentration from 10 times lower than the present MPC to 50,000 times higher. Limited studies with pigs have indicated a slightly lower, but not significantly different, value.

These results were obtained by feeding pH 2 solutions of plutonium(IV) nitrate. At pH 2 the plutonium in such a solution is partially hydroxylated and exists as a high molecular weight polymer. This type of plutonium molecule should be typical of that which would exist in contaminated water supplies. By feeding more acidic solutions, or complexed forms of plutonium, much higher absorption can be obtained.

Without considering possible changes in the other parameters involved, these results on gastrointestinal absorption would seem to justify an increase in the MPC for plutonium-239 in water by at least a factor of ten. Considering the over-all validity of the $Pu^{239}$ limit, it is probable that the ratio of Pu to Ra toxicity is reasonably well known, and that the Ra limit itself is conservative. Therefore, it seems appropriate to take advantage of the lower uptake demonstrated here and write the MPC for $Pu^{239}$ in water as $1.5 \times 10^{-5}$ $\mu$c/ml.

## RUTHENIUM-106

Current maximum permissible standards for internal exposure to $Ru^{106}$ are based on limited observations on a few rats. The absorption of ruthenium from the gastrointestinal tract is reported as $< 0.05$ per cent, and on this basis the MPC for $Ru^{106}$ in water is recommended as 0.1 $\mu$c/ml.[2]

In extensive feeding experiments with rats, approximately 3 per cent of fed ruthenium was absorbed from the gastrointestinal tract. This result was obtained with *p*H 2 solution of either Ru(III) or Ru (IV) chlorides. Reducing the acidity to *p*H 6 resulted in an approximately 5-fold reduction in gastrointestinal absorption.[5]

Kidney is presently considered to be the critical organ for ruthenium internal exposure, and the biological half-life for ruthenium in kidney is listed as 20 days.[2] From our studies, it was found impossible to adequately represent the retention of ruthenium in the rat kidney by a single exponential function. Initial loss of ruthenium is quite rapid, but a considerable fraction of the kidney ruthenium is retained with a half-life of about 40 days. In chronic feeding experiments the equilibrium kidney ruthenium burden was found to be about 2.5 per cent of the daily dose fed.[4] Using this datum there may be derived an MPC for $Ru^{106}$ in water of $4 \times 10^{-3}$ $\mu$c/ml, a value 25 times lower than the presently recommended limit.

Our studies of long-term ruthenium retention suggest strongly that bone rather than kidney may be the critical organ. While kidney accumulates the highest initial concentrations, the half-life for ruthenium retention in bone is of the order of 100–150 days.[5] Data from chronic feeding studies of sufficient duration to prove the point are not yet available, but calculations based on existing data indicate that equilibrium concentrations of ruthenium in bone and kidney should be approximately equal. On this basis, bone, in view of its greater radiosensitivity, should be considered the critical organ.

Assuming bone to be the critical organ, an MPC for $Ru^{106}$ in water may be derived by comparison with the accepted value for the $Sr^{89}$ MPC. Such a calculation indirectly relates the $Ru^{106}$ MPC to the human exposure experience with radium and involves no assumptions concerning a permissible dose of 0.3 rem/week. Since both $Ru^{106}$ and $Sr^{89}$ are energetic beta emitters one can assume that they would have similar biological effect. Correcting the $Sr^{89}$ MPC of $7 \times 10^{-5}$ $\mu$c/ml[2] for differences in effective energy, for differences in effective half-life, and for the different fractions of ingested material deposited in bone, the MPC for $Ru^{106}$ becomes $1 \times 10^{-3}$ $\mu$c/ml. This is a factor of 100 lower than the presently recommended MPC. It is recommended that this limit be tentatively accepted.

## IODINE-131

A continuing experiment on the toxicity of $I^{131}$ in sheep has thus far covered a period of five years and utilized some 600 sheep. The experiment has involved daily oral administration of $I^{131}$ at various levels, with exposures continuing at a constant level through succeeding generations.[6]

When animals were fed 5 $\mu$c/day for about two years, mild symptoms of thyroid damage became evident. In second generation animals which were exposed in utero and via milk during the suckling period, slight evidence of thyroid damage was apparent by the eighth month. Approximate dose rate to the adult thyroid was 150 rads/week, and to the fetal and immature thyroid, 200 rads/week.

At the 0.15 $\mu$c/day feeding level there has been no evidence, over a period of four years, of any damaging effect. Thyroid dose was approximately 3 rads/week.

A radiation dose rate 10 times the assumed maximum permissible level has therefore given no evidence of harmful effect, while a dose 500 times the 0.3 rad/week limit has shown marginal effects in adult sheep only after an exposure period of two years. Sheep that have received daily amounts of $I^{131}$ between 0.15 and 5 $\mu$c/day for 18 months have shown no symptoms of toxicity.

Despite its relative radio-resistance, the thyroid is clearly the critical organ. At the 5 $\mu$c/day feeding level, the maximum radiation dose rate to tissues other than those in the thyroid region does not exceed 0.5 rad/week. For truly chronic exposure, the fetal and early post-natal thyroid should be specified as the critical organ, because of its greater concentration of iodine and because of the apparent greater radiosensitivity of the immature organ.

From these studies it can be calculated that a concentration of $2 \times 10^{-5}$ $\mu c$ $I^{131}$ per gram of vegetation eaten by the sheep should result in no damage to the thyroid. To allow for possible species differences for other range animals, a value of $10^{-5}$ $\mu c$ $I^{131}$/gm vegetation would seem to be a reasonable MPC for grazing land. Such a concentration of $I^{131}$ on vegetation would be expected to result from an air concentration of $3 \times 10^{-13}$ $\mu c/cm^3$. The presently recommended MPC for $I^{131}$ in air is $3 \times 10^{-9}$ $\mu c/cm^3$. While no doubt safe for direct inhalation, this limit should be reduced by four orders of magnitude to assure radiation safety of grazing animals. Approximately the same reduction is required for the safety of humans eating large quantities of fresh garden produce and drinking milk from cows grazing on $I^{131}$-contaminated pasture.

## PHOSPHORUS-32

The presently recommended MPC for $P^{32}$ in water is $2 \times 10^{-4}$ $\mu c/ml$.[2] While this is a reasonable, perhaps even conservative, limit for drinking water, it is inapplicable to biologically populated waters. Here the remarkable concentration in small aquatic organisms and their use in food chains by fish and man necessitates lower limits. At $2 \times 10^{-4}$ $\mu c/ml$, plankton receive dose rates up to 70 rads per hour, and some fish may receive up to 50 rads per hour. Humans eating one pound of these fish per day would receive about 200 rads per week, at some seasons of the year. The MPC for $P^{32}$ in such waters should not be higher than $3 \times 10^{-6}$ $\mu c/ml$, or better, $3 \times 10^{-7}$ $\mu c/ml$ to include the conventional safety factor of 10.[7]

## SODIUM-24

The drinking water MPC for $Na^{24}$ is listed as $8 \times 10^{-3}$ $\mu c/ml$.[2] For the protection of large fish, the MPC should not exceed $2 \times 10^{-6}$ $\mu c/ml$.[7]

## HYDROGEN-3 (TRITIUM)

The present maximum permissible standards for internal exposure to tritium are based on the assumption that the tritium encountered will be freely exchangeable with ordinary hydrogen in the aqueous vapor in the lungs.[1] This assumes, effectively, that the tritium is present as the oxide. Although this assumption is practical because of the difficulty in distinguishing between the elemental form and the oxide in routine monitoring of contaminated atmosphere, we have shown in experiments with rats that tritium in the elemental form ($T_2$ or HT) does not exchange to a very significant extent with the hydrogen of water or other hydrogenous substances in the body.[8] Perhaps more academic than practical, exposure to elemental tritium, therefore, constitutes a much smaller hazard than exposure to tritium oxide.

To derive the MPC for tritium oxide in air and liquid media the following parameters were measured: absorption into the body via lungs and skin; distribution into and biological decay from body water; fixation, concentration, and biological decay of tritium bound to organic molecules in the body; and relative biological effectiveness of tritium beta particles.[9]

Absorption of virtually all tritium oxide inhaled into the lung was readily observed. In extensive studies on rats and man, atmospheric tritium oxide vapor was absorbed through the skin at a rate about equal to the rate of pulmonary absorption.

The uniform distribution of tritium oxide in body water was noted in experiments with small and large animals. From many studies with humans, the best value for the average half-life of body water seems to be about 10 days.[10]

A small fraction of body-water tritium becomes incorporated into nonexchangeable positions in the organic compounds of the body and is lost from the body with half-lives characteristic of the compounds in which it is incorporated. In extensive experiments with rats and mice it was shown that this "organically bound" tritium may have a significant effect in increasing the radiation exposure beyond that calculated for body-water tritium alone. In chronically exposed rats the increased radiation due to the organically bound tritium amounted to about 10 per cent of the total dose, averaged over the entire animal. For specific tissues the increase was as much as 20 per cent, and there were indications that certain compounds containing high levels of bound tritium might receive twice the dose calculated from body water tritium alone.

In studies comparing the effectiveness of different beta emitters in inhibiting the growth of microorganisms we have shown that tritium is approximately twice as damaging per unit of energy absorbed as is the more energetic beta emitter $P^{32}$. Other investigators using other biological criteria have reported values for the tritium RBE of from 1.4 to 2.

The present permissible body burden of tritium is $10^{-4}$ $\mu c$, and the MPC's for air and liquid media are, respectively, $2 \times 10^{-5}$ $\mu c/cm^3$ and 0.2 $\mu c/ml$.[2] Application of our findings to the calculations for the several limits yields the following:

Permissible body burden $1.5 \times 10^{+3}$ $\mu c$
MPC in air $3 \times 10^{-6}$ $\mu c/cm^3$
MPC in liquid media $5 \times 10^{-2}$ $\mu c/ml$

From the preceding examples it is evident that at least several of the present "best" standards for internal exposure are uncertain by rather large factors. While some degree of uncertainty is inherent in any description of biological processes, the uncertainty in present maximum permissible standards for internal exposure is due in most cases to simple lack of information on the biological parameters which determine these standards. Careful study of these parameters in a variety of animal species is urgently needed.

Lacking complete information on these parameters, present planning for installations or procedures must

incorporate a degree of flexibility which will permit ready adaptation to future changes in maximum permissible exposure standards. While most present standards may be accepted at face value for control of the customary hazards of infrequent laboratory exposure, any planning for truly chronic exposure, or for the release of large amounts of radioisotopes, should incorporate conservative safety factors.

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# Is the Concept of "Critical Organ" Valid in Determining the Maximum Permissible Level for Exposure to Radioactive Materials?

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Nearly all current recommendations for the maximum permissible exposure of man to the radioactive materials used in industrial nuclear energy plants or in research require rather broad assumptions. These are detailed by the US National Committee on Radiation Protection in the National Bureau of Standards Handbook 52[1] and given even more attention in the forthcoming recommendations of the International Commission on Radiological Protection.[2] Frequently an assumption must be made as to which organ is to be considered the "critical organ." This is necessary because it is desired to limit exposure so that no body tissue will receive a dose exceeding 0.3 roentgen equivalents man (rem)† per week.[1] In the absence of definite information to the contrary, the organ with the highest concentration of material perforce becomes the critical organ. It is the purpose of this paper to review the consequences of the "critical organ" concept, and to show what alterations might occur in present standards if different assumptions were made.

## CHOICE OF CRITICAL ORGAN

Several criteria might be utilized for choosing a critical organ. Among these are: (1) the organ with the highest concentration of radioactive element; (2) the organ of greatest radiosensitivity; (3) the organ with the most vital functions to perform; and (4) the organ showing the greatest amount of damage on histopathological examination of material from experimental animals.

Each of these has certain virtues and defects as discussed in part by the Subcommittee on Permissible Internal Dose of the U.S. International Committee on Radiation Protection.[1] Hematopoietic organs and testes are much more sensitive than the muscle or nerve or growing bone. If concentrations within two organs differ by less than their relative radiosensitivity, it might seem advisable to designate as critical the organ of greater sensitivity. On the other hand, relative sensitivities are not fixed quantities and vary considerably with dosage regimen, dose level, etc., as shown below. Also, the critical organ may change with route of administration (e.g., lungs after inhalation, gastrointestinal tract after swallowing) or type of compound (soluble or insoluble).

In setting maximum permissible exposures to external radiation arbitrary choices regarding the essentiality of various organs have necessarily been made.[3] Thus alterations in fertility and possible genetic changes have been partially eliminated by choosing to consider only the life of the individual exposed, not that of future generations. The blood-forming organs thus become the critical organs for external radiation. Present data permit some extrapolation to internal emitters. Despite the radiosensitivity of testis, no instances can be found in the literature on internal emitters where a lower maximum permissible body burden would result if testes rather than bone or bone marrow were chosen as critical organs. This results, in part, from the fact that the reproductive organs do not concentrate these elements to any great extent. Thus, if concentration alone is used as a criterion it makes little difference whether the possibility of decreased fertility or of mutation potentialities are included in choice of the critical organ.

However, this may be unsound. Instances can be found where testis[4] or even ovary[5] show more histological damage than kidney, spleen or other organs which would be considered "critical" on a concentration basis. Also, there is practically no information concerning genetic changes produced by radioactive materials in the body, whether or not they concentrate in testis or ovary. Unfortunately, histopathological observations frequently reveal little damage in *any* organ at doses in the tolerance range unless neoplastic changes are induced. Thus, extrapolation from higher doses is required and may be risky.

An estimate of the problems involved in extrapolation from histopathological observations is seen in the data of Casarett[4] and of Casarett *et al.*[6] on polonium-210. Following single doses of this element, the degree of damage to the rat kidney and probably the cause of death shift markedly with dosage. At 10 μc/kg single dose there is a progressive nephrosclerosis and

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† Roentgen equivalents physical × relative biological effectiveness of the type of radiation under consideration.

the animals die in uremia. At higher doses the animals do not develop the characteristic lesion while at lower doses it appears but does not progress sufficiently to be a cause of death as it is at 10 μc/kg. At 1 μc/kg the kidney lesion is not serious, yet life span is undoubtedly shortened presumably by some other cause. Furthermore, if the same total doses are given in monthly installments so that a more or less constant body burden is maintained the kidney lesion appears but fails to develop sufficiently to be a cause of death even at 10 μc/kg total dose. By contrast, spleen shows *greater* degrees of damage on the divided dose regimen. Thus, not only the dosage level but its temporal distribution affect the histopathological picture and in turn the choice of critical organ if it were to be made on this basis.

One can conclude from these considerations only that there is no single "best" criterion for choice of a critical organ. In the absence of data contraindicating the choice, use of the organ with the highest concentration is simplest and most direct. It is the criterion used for most of the figures given in the US National Bureau of Standards Handbook 52[1] and all but the bone-seeking alpha emitters in the International Commission recommendations.[2] In most cases no better information is available. But, as pointed out by Morgan recently,[7] the choice on this basis should be examined with great care and for each element considered. Calculation on a simple basis is obviously attractive but can easily become uncritical.

## IS THERE ALWAYS A "CRITICAL ORGAN"?

With elements, like the bone seekers, where concentration in one tissue is orders of magnitude higher than that in any other tissue, there seems little reason to question the validity of using the concentration criterion except for problems of non-uniform distribution in the tissue. On the other hand, evidence is accumulating to indicate that the *average* body content may be as significant as any other figure in certain cases.

Taking examples from work in this laboratory with polonium-210 and consideration of other data some illustrations are available. An influence of dosage regimen has been found in the relative amounts of polonium retained in organs of the reticulo-endothelial system, in the rat. A larger fraction of the body burden remains in liver, spleen and kidney when a body burden is maintained through monthly injections than when a single dose of comparable magnitude is given.[8] This is shown in Table I in terms of the radiation doses received by two organs.‡ It is clear that both spleen and kidney, potential critical

**Table I. Comparison of Radiation Doses to Spleen and Kidney (Male Rats) from Single and Multiple Doses of Polonium-210 (calculated in rep to each gram of organ)**

| | *Single dose* | | *Multiple dose* | |
|---|---|---|---|---|
| *Dose* μc/kg* | *Method A rep* | *Method B rep* | *Method A rep* | *Method B rep* |
| | | **Spleen** | | |
| 0.37 | 175 | 107 | 280 | 280 |
| 1.42 | 686 | 486 | 1116 | 1116 |
| 3.40 | 1474 | 1113 | 1468 | 1690 |
| 10.55 | 4480 | 3708 | 4959 | 9380 |
| | | **Kidney** | | |
| 0.37 | 86 | 72 | 140 | 140 |
| 1.42 | 339 | 292 | 558 | 558 |
| 3.40 | 728 | 670 | 734 | 841 |
| 10.55 | 2213 | 2237 | 2480 | 4682 |

* Effective total dose in case of multiple dose animals, as a single injection in others.

organs, receive significantly larger radiation doses particularly at the lower dosage levels in a divided-dose experiment. Contrary to expectations, however, these organs were found *not* to show parallel differences in histopathologic damage.[6] In fact, as described above, the kidney shows less damage in the multiple-dose experiment than in the single, the spleen more damage with divided doses.

Of greater significance perhaps is the fact that the life shortening per microcurie appears to be about identical for the two regimens.[9] This is especially noteworthy since the lower dosage levels approximated "tolerance" figures more closely than in most experiments.

Another example indicating significance of the total body burden rather than a critical organ is seen in an acute toxicity experiment. Significant differences in distribution of polonium to the tissues occur as a function of route of administration.[10] Blood cells contain a higher fraction of the body burden and liver, spleen, and kidney a much lower fraction of the body content after oral than after intravenous administration. Radiation doses to the critical organ (excluding blood contained therein) differ by a factor of 1.5 to 2. Yet the acute toxicity (30 day $LD_{50}$) of a given body burden is independent of the route of administration.[11] The radiation dose from the circulating blood after oral administration may be sufficient to compensate for the lower concentration in the critical tissues, or the relatively high radiation dose to the gastrointestinal tract from unabsorbed polonium may compensate. In either event a "normal" calculation of expected results on the basis of concentration in a critical organ would not have predicted the result obtained.

‡ For animals receiving single doses the total number of reps is calculated on two bases: (1) as if the loss by decay and excretion can be expressed correctly by a single exponential curve with respect to time; and (2) by arithmetic integration taking into account the actual rates of loss over several time periods. (This latter procedure may be somewhat more precise since polonium loss from the body is not accurately described by a single exponential.) For calculation of the radiation dose to these organs in the maintained body burden group, two methods of calculation are also represented: (1) the integrated dose in the animal of mean life span; and (2) integrated dose in 400 days or more. These are identical at the lower dosage levels since the animals lived beyond 400 days. This adjustment is reasonable because the calculations for the single-dose group assumes sufficient time for complete elimination by decay and/or excretion.

Table II. Variation of Maximum Permissible Body Burden with Choice of Critical Organ* (expressed as microcuries in a 70 kg man)

| *Radioisotope* | *Critical Organ A* | *Max. Perm. Body Burden* | *Critical Organ B* | *Max. Perm. Body Burden* |
|---|---|---|---|---|
| $Sc^{46}$ | Spleen | 6 | Liver | 5 |
| $Sc^{47}$ | Spleen | 15 | Liver | 11 |
| $Sc^{48}$ | Spleen | 5 | Liver | 3 |
| $Mn^{56}$ | Kidney | 25 | Liver | 8 |
| $Re^{183}$ | Thyroid | 37 | Skin | 650 |
| $Ir^{190}$ | Kidney | 23 | Spleen | 21 |
| $Ir^{192}$ | Kidney | 3 | Spleen | 3 |
| $Au^{196}$ | Liver | 8 | Kidney | 32 |
| $Au^{198}$ | Liver | 3 | Kidney | 10 |
| $Au^{199}$ | Liver | 9 | Kidney | 30 |
| $Po^{210}$ | Spleen (sol. cmpd.) | 0.04 | Lung (insol. cmpd.) | 0.02 |
| Th-natural | Bone (sol. cmpd.) | 0.01 | Lung (insol. cmpd.) | $2 \times 10^{-3}$ |
| U -natural | Kidney (sol. cmpd.) | 0.04 | Lung (insol. cmpd.) | 0.01 |
| $U^{238}$ | Bone (sol. cmpd.) | 0.04 | Lung (insol. cmpd.) | 0.016 |
| $Pu^{239}$ | Bone (sol. cmpd.) | 0.04 | Lung (insol. cmpd.) | 0.02 |

* Modified from Reference 2.

Recognition of the gastrointestinal tract as "critical organ" or as a contributing organ when materials enter the body through feed, water or by being swallowed as a consequence of inhalation exposure has been emphasized very recently by Morgan.[7] In fact, the gastrointestinal tract becomes "critical" in over 70 per cent of ingestion exposures and 40 per cent of inhalation exposures even though the critical organ on the basis of absorbed dose may be bone or some other tissue.

Another factor enters when mixtures of radioisotopes are involved. The interesting data of Salerno and Friedell[12] indicate that when mixtures are given or isotopes whose decay schemes involve elements with very different localization tendencies, the net toxicity, on an acute basis anyway, may be much more than the sum of the two doses.

Whether these findings indicate an interaction among organs, the presence of a circulating toxin, or simply that no organ is actually critical or limiting cannot be ascertained from present data. However, these possibilities should be considered, especially for elements which do not show very large differences in their concentration among various organs.

## VARIATION OF MAXIMUM PERMISSIBLE BODY BURDEN WITH CHOICE OF CRITICAL ORGAN

All operating values for permissible air or water concentrations are based on the body burden. In a majority of cases this is in turn calculated as the amount of isotope deposited in the body which will produce the maximum allowable, 0.3 rem per week, dose to the critical organ. Thus choice of critical organ may alter the operating limits in an expensive and complicated installation. For this reason, it is pertinent to examine the alteration in body burden resulting from various reasonable choices regarding critical organ, or, indeed, the choice of alternative methods of calculation of the body burden.

Some examples can be taken from the forthcoming recommendations of the International Commission,[2] and are contained in Table II. In the upper part of the table, the choice of critical organ is based on relative likelihood of damage on the basis of concentration or radiosensitivity. All values are arrived at by direct calculation. The criterion for choice of critical organ is different in the lower part of the table. Here the main variable is the type of compound involved,[4§] and the calculations are based on toxicity relative to radium except in the case of polonium-210.

Examination of Table II reveals differences in body burden from zero to a factor of about 20 when alternative choices of critical organ are made. If, in addition, exposure of the gastrointestinal tract is considered nearly all isotopes would have more than one critical organ at some time after administration by all but parenteral routes. When this is done factors of difference as high as $10^4$ appear, usually but not always with the lowest figure obtaining when the gastrointestinal tract is critical. These are of less importance, however, since the gastrointestinal tract is seldom critical for long periods.

A more detailed analysis for a single element is contained in Table III, based on work in this laboratory using polonium-210 (which concentrates in soft tissues). The maximum permissible body burden and corresponding limits for air and water concentrations are shown after calculation by three independent methods. These are explained in the legend to the table. The body-burden figures range from 0.1 $\mu$c to 0.004 $\mu$c in a 70-kg man. However, both of these are extreme. The gastrointestinal tract is likely to be critical over only short periods. All recent toxicity work negates the possibility that polonium can bear a 1 : 1 relationship to radium on a microcurie basis.

§ Frequently different compounds may be involved in a single exposure and choice regarding critical organ on the basis of type of compound may be meaningless. It is still pertinent to demonstrate the differences in body burden following a given choice.

Table III. Variation of Maximum Permissible Values for Polonium-210 with Criterion Used

| Method* | Criterion | Basis | Maximum in body† μc | Amount permissible in water μc/cm³ | Amount permissible in air μc/cm³ |
|---|---|---|---|---|---|
| A | Spleen as critical organ | Has highest concentration (Ref. 10) | 0.02 | $10^{-5}$ | $3 \times 10^{-10}$ |
| A | Kidney as critical organ | Most damage at moderate exposure levels (Ref. 4) | 0.04 | $2 \times 10^{-5}$ | $5 \times 10^{-10}$ |
| A | Lung as critical organ | Highest concentration after inhalation of insoluble salt (Ref. 1) | 0.01 | — | $10^{-10}$ |
| A | G. I. tract as critical organ | Has highest concentration at certain times (Ref. 2) | 0.004 | $3 \times 10^{-6}$ | $5 \times 10^{-10}$ |
| B | Acute toxicity relative to radium (20×) | Ref. 13, 14 | 0.005 | $4 \times 10^{-6}$ | $7.5 \times 10^{-10}$ |
| B | Probable chronic toxicity relative to radium (5×) | Ref. 13, 14, 15 | 0.02 | $10^{-5}$ | $3 \times 10^{-10}$ |
| B | Minimal toxicity relative to radium (Equal) | Ref. 15 | 0.10 | $5 \times 10^{-4}$ | $1.5 \times 10^{-9}$ |
| C | Threshold for life span shortening in rat | Ref. 9 | 0.07 | $3 \times 10^{-5}$ | $10^{-9}$ |

* Method A—Calculation of dose to produce maximum of 0.3 rem per week in critical organ. Method B—Assuming maximum permissible body burden of radium in man is 0.1 μc. Method C—The rate figures are divided by 50 to account for difference in life span and excretion between rat and man.

† 70 kg man.

It is probably not as high as 20 nor as low as 1. Therefore, the range becomes essentially seven-fold and, if inhalation of insoluble salt is considered as a separate case, the range becomes 0.02 μc to 0.07 μc. This, or even 0.01 to 0.07 μc is a small range indeed considering the many assumptions necessary and the many indeterminate factors in our present data.

## DISCUSSION AND CONCLUSIONS

This paper has reviewed the rationale of the critical organ concept in calculations of maximum permissible exposures to radioactive materials. It is shown that in many instances there is little evidence to indicate the existence of a critical organ. Furthermore, the choice is frequently quite arbitrary and without real evidence of damage to the organ in a chronic exposure situation, or evidence that the organ is critical in producing damage to the body as a whole. Therefore, caution is urged in any given case, especially where large issues such as cost, plant operating practices, etc. are involved.

On the other hand, a review of the quantitative differences seen in operating levels when various possible critical organs are chosen or even entirely independent approaches used reveals that most values will lie within an order of magnitude range. Some of these differences may be important but a range of 10 is actually small in a field requiring so many assumptions and with as many gaps in knowledge as that concerning the long-term effects of radioisotopes taken into the body. Therefore it is concluded that, in the present state of our knowledge, the use of the critical organ concept will not be likely to lead to gross errors in setting maximum permissible exposure levels. It will continue to be the only possible approach in many cases. However, continuous and careful review of all pertinent parameters will be needed as our knowledge of the ultimate causes of chronic radiation damage expands. This is particularly likely in connection with the induction of tumors, and the production of genetic changes by radioactive materials.

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# Lymphocytes with Bilobed Nuclei as Indicators of Radiation Exposures in the Tolerance Range

**By M. Ingram,* USA**

Biological indicators of exposure to ionizing radiation have certain advantages which are not likely to be entirely superseded by physical monitoring devices. This is particularly true of indicators which utilize changes in the worker himself and which are sensitive to exposures likely to be classed as "tolerable" for radiation work. The advantages of biological monitoring are especially pertinent to the medical, medico-legal and morale problems which arise during the operation of health programs for radiation workers.

The major biological changes known to occur following exposures in the tolerance range are: (1) genetic; (2) alterations in sperm production; (3) latent changes such as shortening of life-span; and (4) hematological changes. Of these, hematological changes observable in the peripheral blood are most readily applicable to biological monitoring.

A good hematological monitoring program is one in which adequate control studies are followed by repeated observations at intervals determined by the general nature of the work and the work history of the individual. Such a program will provide a great deal of valuable information, even when the studies are limited to the more commonly performed determinations. If studies designed to detect some of the more sensitive hematological changes are included, the value of the program is considerably augmented.

A particularly sensitive hematological indicator of radiation exposure is the increased incidence of lymphocytes with bilobed nuclei which follows very small exposures to ionizing radiation. Examples of these cells are shown in Fig. 1. It will be noted that the normal lymphocyte contains a single, rounded nucleus. The lymphocyte with bilobed nuclei, however, tends to be slightly larger than the majority of the lymphocytes, and contains two distinct nuclear masses which are typical lymphocyte nuclei in all respects except that two are present in a single cell. The two lobes may be completely separated or joined by a thin bridge of nuclear material.

From the point of view of personnel protection, it is significant that this phenomenon was first noted and studied in a group of cyclotron workers who were receiving exposures believed to be well below the maximum permissible level. This group of 44 individuals comprised the original crew of physicists, technicians, shop workers, and maintenance personnel working in the 130-inch cyclotron laboratory at the University of Rochester, Rochester, New York. The cyclotron itself was nearing completion when the group came under study.

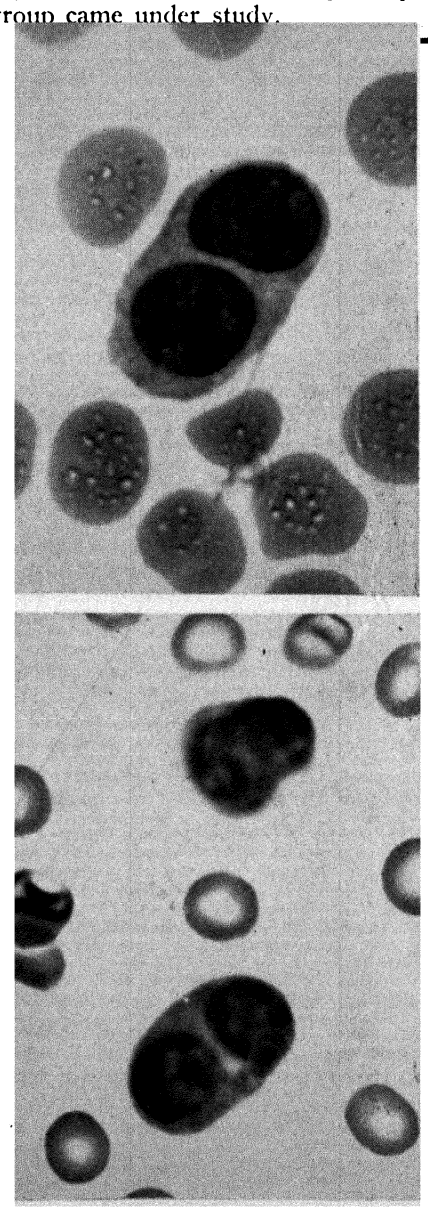


Figure 1. Above: a binucleate lymphocyte (human blood); below: a normal and a binucleate lymphocyte (dog blood)

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Careful preliminary planning on the part of the physicists and engineers responsible for design of the installation had resulted in working conditions in the cyclotron laboratory building which were considered safe, even in terms of conservative radiation safety standards. There were, however, some uncertainties about the precision of the various methods selected for determining radiation levels in personnel areas. Other uncertainties existed regarding evaluation of the biological effectiveness of the various types of radiation which might be encountered, as well as the validity of translating tolerance doses into terms applicable to complex mixtures of radiation.

With these uncertainties in mind, it seemed advisable to augment physical radiation measurements with careful hematological studies. Accordingly, physical examination and control blood studies were carried out before cyclotron operation began, and blood studies were repeated at relatively frequent intervals thereafter until a reasonable period of non-hazardous operation had elapsed. From the outset, emphasis was placed on the leukocyte picture, with particular attention to thorough study of entire blood smears.

Shortly after cyclotron operation began, a few leukocytes which appeared to be lymphocytes with bilobed nuclei were found on blood smears. The observation was sufficiently unusual to warrant a detailed search for these cells, both on control smears, obtained as initial studies on newly hired personnel, and on smears taken after the cyclotron runs began. Peroxidase staining was introduced as a routine procedure to facilitate identification of the abnormal lymphocytes. The leukocytes in all good areas on coverslip blood smears were counted (approximately 1500 WBC per smear) and the position of all abnormal cells were indexed in terms of the settings of a graduated mechanical microscope stage. Bilobed lymphocyte studies are time consuming and tedious, and some provision must be made for review to insure proper classification. Indexing in the manner described provided a means for checking the individual cells.

As reported in previous publications, a definite increase in the incidence of lymphocytes with bilobed nuclei was found in the blood of the cyclotron workers during the first few months of cyclotron operation.[1] Fairly early in this period (and partly as a result of the hematological findings) a decision to install additional shielding was reached, and an earth and concrete dike was constructed between the cyclotron laboratory and the building housing the cyclotron. This dike was completed in mid-April, 1949. The incidence of the abnormal lymphocytes subsequently returned to the control level, where they have remained (Fig. 2).

These observations on personnel were confirmed by experiments in dogs.[2] After control studies extending over a period of about two months, three dogs each received three consecutive exposures to the mixture of radiations outside the cyclotron. Ade-

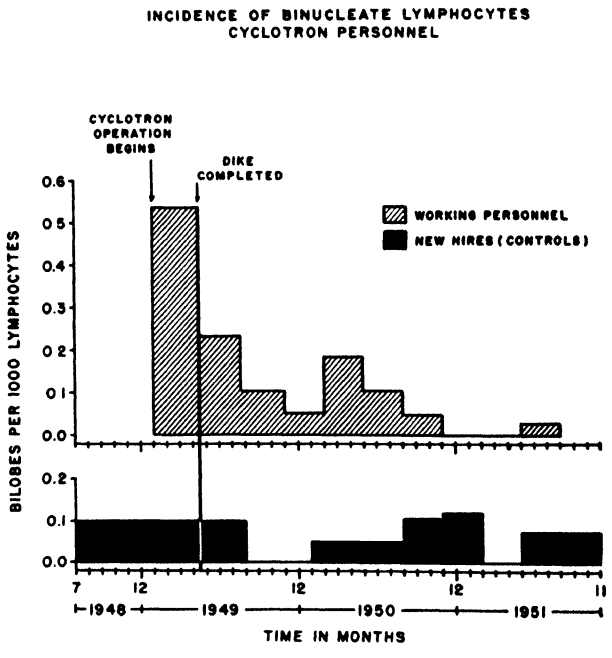



Figure 2

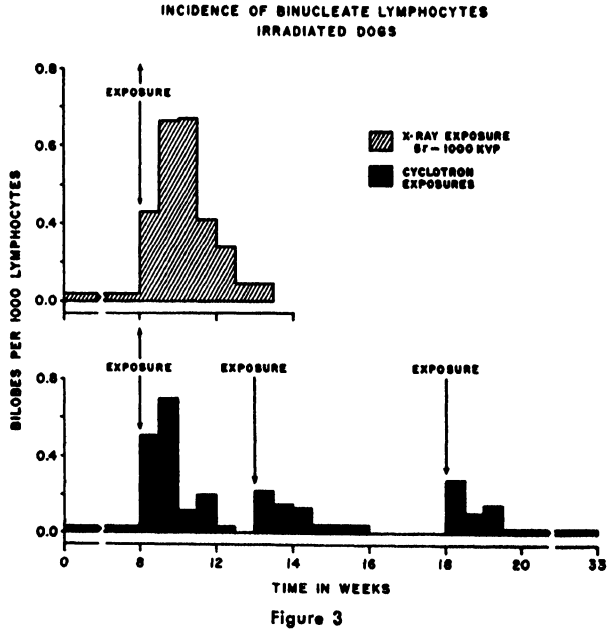



Figure 3

quate periods for recovery were allowed between exposures. Each exposure was followed by an increased incidence of the abnormal lymphocytes, as shown in Fig. 3.

Data accumulated during the years since the observations were first reported have been subjected to statistical study, the results of which are shown in terms of confidence limits in Tables I, II and III.†

The observations regarding bilobed lymphocytes have been confirmed by other investigators, in particular Dr. Ethel Browning in England, Dr. Robert

† The author is indebted to Dr. Arthur Dutton of the Statistics Section, Department of Radiation Biology, University of Rochester School of Medicine and Dentistry, for statistical evaluation of the data.

**Table I. Incidence of Binucleate Lymphocytes in Cyclotron Personnel during First Year of Operation of 130-Inch Cyclotron***

| *Interval* | *No. individuals* | *Bilobes per 1000 WBC* | *95% confidence interval* | *Bilobes per 1000 lymphs* | *95% confidence interval* |
|---|---|---|---|---|---|
| Original control group† (1948–Jan., 1949) | 44 | 0.03 | 0.01–0.07 | 0.10 | 0.03–0.23 |
| Jan., '49 to April 14, '49 | 44 | 0.15 | 0.11–0.19 | 0.54 | 0.39–0.68 |
| April 15–Aug. | 39 | 0.08 | 0.05–0.11 | 0.23 | 0.15–0.34 |
| Aug.–Nov. | 34 | 0.03 | 0.02–0.06 | 0.10 | 0.05–0.20 |
| Nov., '49 to Feb., '50 | 18 | 0.01 | 0.00–0.06 | 0.05 | 0.00–0.29 |
| Feb.–May | 11 | 0.03 | 0.00–0.13 | 0.19 | 0.02–0.67 |
| May–Aug. | 34 | 0.04 | 0.01–0.08 | 0.11 | 0.04–0.23 |
| Aug.–Nov. | 26 | 0.01 | 0.00–0.05 | 0.05 | 0.00–0.18 |
| Nov., '50 to Feb., '51 | 18 | 0 | 0.00–0.08 | 0 | 0.00–0.09 |
| Feb.–May | 3 | 0 | | 0 | |
| May–Aug. | 34 | 0.03 | 0.00–0.10 | 0.08 | 0.01–0.29 |
| | 162 | 0.02 | 0.01–0.04 | 0.08 | 0.05–0.12 |

* Cyclotron operation began in January, 1949. Dike completed mid-April, 1949. Pooled control data through December, 1952.

**Table II. Incidence of Binucleate Lymphocytes in Dogs after Exposure to Radiation from Cyclotron**

| *Interval* | *Bilobes per 1000 lymphocytes* | *95% confidence interval* |
|---|---|---|
| Control period (2 months) | 0.03 | 0.00–0.10 |
| **First exposure at cyclotron*** | | |
| Post-exposure: | | |
| 1 week | 0.5 | 0.37–0.65 |
| 2 weeks | 0.7 | 0.38–1.17 |
| 3 weeks | 0.1 | 0.01–0.39 |
| 4 weeks | 0.2 | 0.08–0.40 |
| 5 weeks | 0.03 | 0.00–0.17 |
| 6 weeks | 0 | 0.00–0.12 |
| **Second exposure** | | |
| 1 week | 0.2 | 0.14–0.30 |
| 2 weeks | 0.2 | 0.04–0.39 |
| 3 weeks | 0.1 | 0.03–0.39 |
| 4 weeks | 0.04 | 0.00–0.21 |
| 5 weeks | 0.04 | 0.00–0.19 |
| 6 weeks | 0.04 | 0.00–0.25 |
| 7 & 8 weeks | 0 | 0.00–0.22 |
| 9 & 10 weeks | 0 | 0.00–0.29 |
| **Third exposure** | | |
| 1 week | 0.3 | 0.20–0.37 |
| 2 weeks | 0.1 | 0.01–0.36 |
| 3 weeks | 0.2 | 0.02–0.56 |
| 5 through 11 weeks | 0 | 0.00–0.15 |
| 12 weeks | 0.04 | 0.00–0.24 |
| 13 through 17 weeks | 0 | 0.00–0.20 |

* Since slight depression of WBC count developed during the first week after this exposure only, it is assumed that dose received in this exposure was higher than that received in second and third exposures.

**Table III. Incidence of Binucleate Lymphocytes in Dogs after Single Total-Body Exposure to 5 r of 1000 kvp X-Rays***

| *Interval* | *Bilobes per 1000 lymphocytes* | *95% confidence interval* |
|---|---|---|
| Control period (2 months) | 0.04 | 0.01–0.14 |
| Post-exposure:† | | |
| 1 week | 0.5 | 0.20–0.92 |
| 2 weeks | 0.9 | 0.48–1.62 |
| 3 weeks | 0.9 | 0.58–1.48 |
| 4 weeks | 0.4 | 0.17–0.86 |
| 5 weeks | 0.3 | 0.09–0.66 |
| 6 and 7 weeks | 0.00 | 0.00–0.52 |

* 1000 kvp, 3.0 ma, 0.75 r/minute.
† Exposed to 5 r of X-rays.

Carter at Los Alamos, and Dr. Lowry Dobson in California.[3,4,5]

Relatively little information was available concerning the kinds of radiations involved in the various cyclotron exposures, or the absolute doses received. It appeared virtually certain, however, that the dose in all instances was very low, probably well below the maximum permissible exposure level. There was a general suspicion that the neutron component of the mixed radiations from the cyclotron might be largely responsible for eliciting the bilobed lymphocyte response. This suspicion, however, was based almost solely on inference.

In order to estimate the magnitude of the radiation dose required to produce an increased incidence of lymphocytes with bilobed nuclei it was decided that animals should be exposed to some well-characterized radiation so that accurate dosage measurements could be made. Accordingly, three dogs, after suitable control studies, received a single whole-body dose of 5 r using 1000 kvp X-rays. It was appreciated that X-rays might prove less efficient than a mixture of radiations with a significant neutron component, however, 5 r was considered to be a relatively large dose in comparison to doses probably received in the cyclotron studies, and would, it was hoped, compensate for any decrease in biological effectiveness.

Results of this exposure to 5 r of 1000 kvp X-rays are included in Fig. 3 to facilitate comparison with

the results following the three consecutive dog exposures outside the cyclotron. It will be noted that the incidence of lymphocytes with bilobed nuclei was somewhat greater after the X-ray exposure than after the cyclotron exposure, and that the maximum incidence occurred somewhat later. On an absolute basis, the incidence of the abnormal lymphocytes in this experiment, as in the earlier studies, proved to be very low, necessitating the examination of exceptionally large numbers of leukocytes.

The need for examining extremely large numbers of leukocytes on blood smears is a serious deterrent to the application of this kind of study to practical hematological monitoring. For this reason, efforts were directed toward the development of a simple technique for preparing concentrations of intact leukocytes without altering the differential leukocyte picture. A highly satisfactory procedure for this, utilizing 0.5 ml of whole blood (with ethylene diamine tetraacetic acid as anticoagulant), consists of centrifuging the blood gently in a closed length of plastic tubing, and then removing almost all of the supernatant serum by aspiration from the top of the column. The tubing is then clamped off about one centimeter below the top of the cell layer, and the fraction in the uppermost section, containing the leukocytes, is made uniform and discharged a drop at a time onto coverslips for blood smears.

The leukocyte concentration procedure is illustrated in Fig. 4. The yield of leukocytes in the concentrate is virtually quantitative, and on the average the leukocytes are concentrated about five-fold. The cells are intact, and the differential leukocyte count is unaffected.

Investigation of the relationship between radiation dose and the incidence of lymphocytes with bilobed nuclei in peripheral blood, utilizing this concentration procedure, has been extended to include observations, now nearing completion, on groups of dogs exposed daily over long periods of time to tolerance range doses of X-radiation. These dogs are being studied at the University of Rochester by Dr. George Casarett, Dr. John Hursh, and co-workers to determine the effects of chronic exposure to tolerance range doses of X-rays on sperm production. Hematological studies on these dogs have been possible through the courtesy of this group.

The ultimate biological significance of an increased incidence of lymphocytes with bilobed nuclei in peripheral blood is not yet clear. It is interesting to note, however, that multinucleated cells have been observed in several other tissues after irradiation. Certainly if this type of change occurs in diverse tissues following relatively small exposures, it is not surprising that it should also occur in the highly radiosensitive and diffusely distributed lymphoid tis-

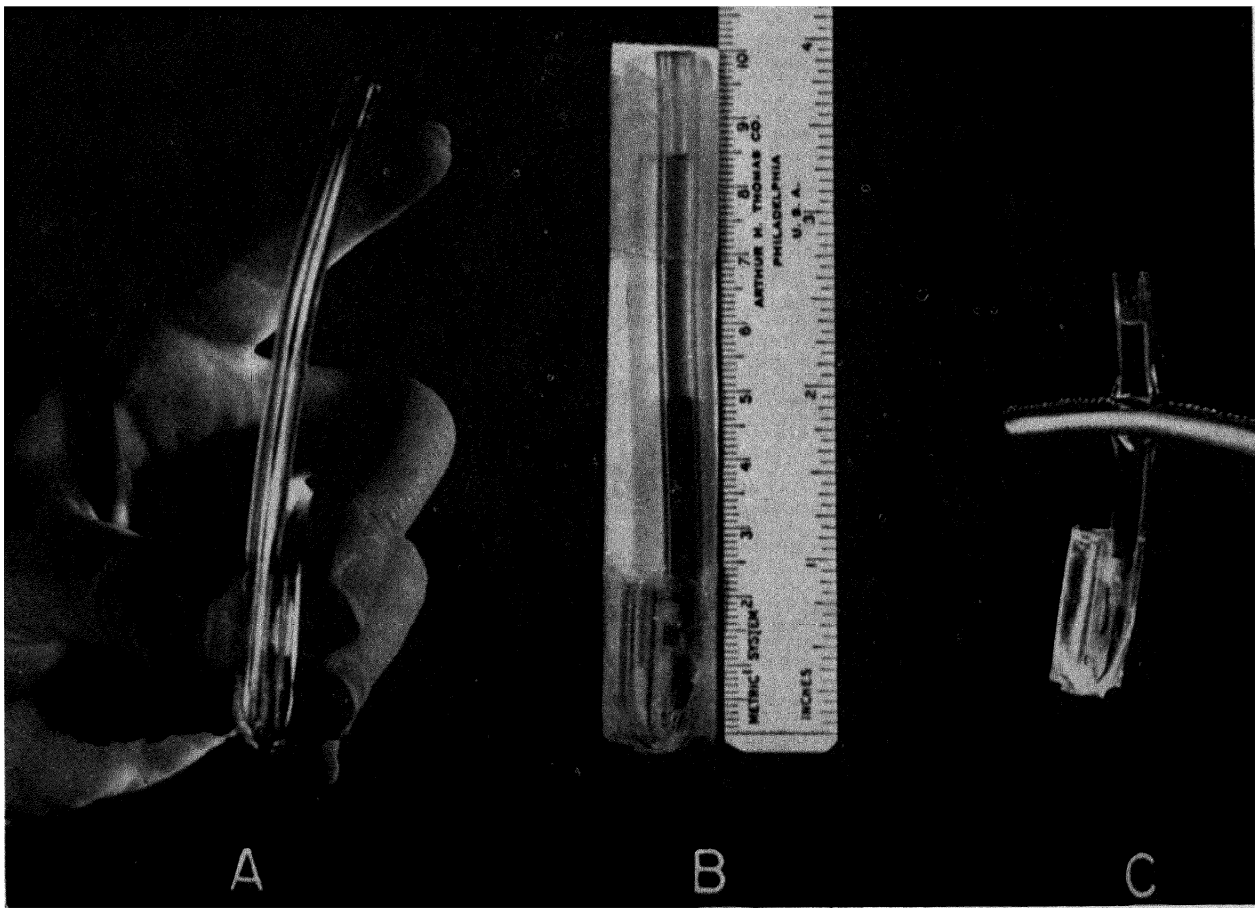



Figure 4

sue, resulting ultimately in the release of occasional lymphocytes with bilobed nuclei into the blood stream.

One might speculate regarding the possibility that the increased incidence of lymphocytes with bilobed nuclei after small radiation exposures may be related to the changes which result in an increased incidence of leukemia among individuals receiving repeated small exposures over long periods. Since an increased incidence of these cells may also be observed in infections, however, the prognostic significance of barely detectable, transient increases of binucleate lymphocytes in peripheral blood probably is not grave. The association of both specific and general alterations of the leukocyte picture with infection, however, emphasizes the need for careful integration of hematological studies with evaluation of general health. This is an essential point in the Rochester hematological-monitoring program.

At present, with most questions related to the biological significance of the phenomenon unanswered, the major significance of the bilobed lymphocyte response lies in its usefulness in biological monitoring for radiation exposures. Studies of the phenomenon have proved helpful on numerous occasions when we have been confronted with situations in which there was a possibility of overexposure but in which conclusive physical evidence of exposure was lacking. In such instances, the personnel in question are likely to develop a sudden interest in the bilobed lymphocyte studies, and have, on occasion, returned to the laboratory repeatedly while waiting for blood studies to be completed to ask, "Do I have any of those two-headed blood cells?" Personnel appear to find it reassuring when told that "no changes characteristic of exposure to ionizing radiation are found," or (when the only finding is an increase in the incidence of lymphocytes with bilobed nuclei, unaccompanied by depression of the total leukocyte or absolute lymphocyte counts) that whatever exposure was received was so slight as to be almost undetectable.

A discussion of sensitive hematological indicators of radiation exposure would be incomplete if it failed to mention studies of abnormal peripheral blood cytology in general, as determined by classification of all types of cytological abnormalities occurring on blood smears representing repeated sampling over a long period of time. An excellent example is Dr. Ethel Browning's investigation of a large group of luminizers in England.[6] In this type of study, there may be considerable difference of opinion among various laboratories as to the classification of cytological abnormalities. Within any one laboratory, however, the general appraisal of cytological abnormalities in leukocytes provides a great deal of information. The bilobed lymphocyte study actually represents a special case of this general type of study. It has, however, definite advantages, in that difficulties arising from varying opinions about leukocyte cytology are eliminated by using this distinctive abnormality and expressing the results quantitatively.

Certain other types of hematological studies may, under proper conditions, also serve as sensitive indicators of radiation exposures. Careful evaluation of the total and differential leukocyte counts at frequent intervals over a long period of time in selected groups of individuals receiving repeated tolerance range doses appears to be a surprisingly sensitive exposure indicator. The most striking example of this type of study in radiation workers was carried out by Dr. N. P. Knowlton, Jr. at Los Alamos.[7] He studied a group of 10 workers who received average weekly doses of 0.2 r of gamma radiation over a period of 77 weeks. At the end of that time, it was found that the group as a whole had developed slight but statistically significant depressions of the peripheral blood leukocyte count, whether expressed as total leukocytes, total granulocytes or total lymphocytes per cubic millimeter of blood. This does not mean that a casual program of routine studies carried out largely as a gesture on large groups of personnel with a variety of work histories will permit adequate evaluation of exposures received. On the contrary, programs of this sort are likely to provide only negative results of questionable significance, and have unfortunately convinced many administrators that routine blood studies are of little or no value.

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# Experimental Data Useful in Establishing Maximum Permissible Single and Multiple Exposures to Polonium

By D. S. Anthony, R. K. Davis, R. N. Cowden and W. P. Jolley,* USA

The element polonium (radium-F) has been of major interest to radiobiologists for many years since it is a virtually weightless source of nearly pure alpha-particle emission. The early studies on the biological effect of the element were carried out chiefly by Lacassagne and his associates in France.[1] Subsequently, improved analytical techniques made possible much more quantitative studies of the metabolism and the acute toxic effects of polonium by Fink and his associates.[2]

Currently, there is much interest in reactors employing circulating molten bismuth. This would yield large quantities of polonium through the $n, \gamma$ reaction on $Bi^{209}$, to produce $Bi^{210}$ which, in turn, would decay to $Po^{210}$. Presumably, safety devices would prevent exposure to very large, acutely toxic amounts of polonium. However, there is a real possibility that personnel working with such reactors might be exposed to small amounts of polonium from time to time. This report is a summary of experimental data obtained with small animals relevant to the chronic toxicity of small amounts of polonium.

An extensive search was conducted for the most sensitive criterion of polonium damage in rats. Among the criteria tested were: clinical tests of liver and kidney function; body weight gain or loss; food and water intake; extensive differential and total blood cell counts; measurements of tumor incidence, type and rate of development; histopathological evidence of damage in vital organs; and reduction of life-span of a population. Only the last two named proved useful at the lower levels, and at the lowest toxic doses of polonium only the reduction in life-span was a sufficiently sensitive criterion to detect an effect of polonium. It follows then, that this study of chronic effects of small doses of polonium will be almost entirely a report of life-span shortening and of histopathological effects. This work should sound a note of caution to those responsible for establishing maximum permissible exposures to any relatively short-lived radiation. For, although the radiation exposure from a single dose of polonium is virtually complete within 100 days, low doses caused irreversible damage that was undetectable for 300 to 500 days, yet resulted in earlier than normal death of the exposed population.

## METHODS

Rats (Sprague-Dawley strain) were employed in all experiments reported here. Polonium–210 was injected into the caudal vein as a phosphate-buffered ($p$H 6.8) solution. The polonium solution was prepared for injection and was sampled for radioactivity measurements in a manner essentially similar to that described by Fink.[2]

Standard histopathological techniques were used. Animals were sacrificed at various times from 1 day to over 300 days after the initial polonium treatment by injection of lethal doses of Nembutal. The usual number of animals sacrificed was 2 males and 2 females from each dose level, including controls. Many additional animals, especially at very late times after polonium treatment were taken immediately after their "natural" death or were sacrificed in a moribund condition. Organs were fixed in formaldehyde and/or special fixatives. Sections of 4 to 5 microns thickness were cut and stained routinely with hematoxylin-eosin and Masson's trichrome. Special stains such as fat stains, reticulum stains, glycogen stains, etc. were used where indicated. The organs or tissues examined included kidney, heart, liver, lungs, lymph nodes, spleen, GI tract, gonads, thyroid, the endocrine glands, salivary gland, skin, brain, bone and bone marrow. Substantial numbers of animals were examined following single doses of 35, 23, 8, 3.6, 0.9, 0.75, 0.5, and 0.25 microcuries of polonium per kilogram of body weight ($\mu$c/kg bw). Large numbers of animals were examined at various intervals following multiple injections (repeated at 14-day intervals) of 2 $\mu$c/kg bw. Additional large numbers of rats were examined following multiple injections (repeated at 28-day intervals) which maintained a body burden averaging 0.01 $\mu$c/kg bw.

Mortality data were obtained at higher single dose levels (81, 73, 70, 61, 51–55, 40, and 30–35 $\mu$c/kg) by the classical acute $LD_{50}$ type of experiment. In the middle dose range (23, 8, 3.6 and 0.9 $\mu$c/kg and their controls) the mortality information was obtained incidentally to the main purpose of the experiments which was to obtain hematological information. Accordingly these data may be slightly biased by the possible concomitant effects of loss of small amounts

* Prepared by D. S. Anthony and K. A. Busch. Including work by the entire staff, Biology Division, Mound Laboratory, Monsanto Chemical Company, Miamisburg, Ohio.

of blood at each of several sampling periods. The groups in the higher and middle dose ranges normally consisted of 10 rats of each sex at each dose level. The lowest single dosages used (0.75, 0.51, 0.25 μc/kg and their controls) were in experiments deliberately designed to yield chronic mortality data. The large groups in this series (40 males and 40 females at each dose level) were all allowed to live out their life-span in identical fashion virtually without disturbance except for feeding, watering, occasional weighing, and periodic inspection.

Information on mortality following repeated doses of polonium was obtained from a small number of rats (10) that had received from 10–14 consecutive injections of 2 μc/kg bw at 14-day intervals. Attempts were also made to obtain mortality data from the large group of rats previously described that were maintained with an average polonium burden of 0.01 μc/kg bw through replacement injections every 28 days.

## RESULTS

The outstanding feature of the results was the late expression of toxicity, following a long latent period, from a polonium radiation insult which occurred and disappeared hundreds of days earlier.

The pattern of polonium radiation insult to the tissues following a single intravenous injection is illustrated by that in the kidney (Fig. 1). In this organ, as with all other vital organs that carried any appreciable fraction of the retained polonium, the great bulk of radiation exposure occurred in approximately the first 100 days after injection. It should be noted that the peak concentration, approximately maintained for many days, was a little over 10 times the average total body concentration originally injected. The spleen showed a very similar pattern and peak concentration. The liver showed a polonium concentration at 1 day of 16 times the average total body concentration originally injected. This concentration rapidly dropped with time so that at 7 days it was 6 times and at 112 days only 0.3 times the original average concentration. No other organ or tissue ever showed a peak concentration as much as 20% of that shown by the kidney and spleen. For the benefit of those who might wish to make calculations of maximum permissible concentrations by currently fashionable formulae, we have given below for rats the critical organs and best numerical values of the various coefficients from our experiments:

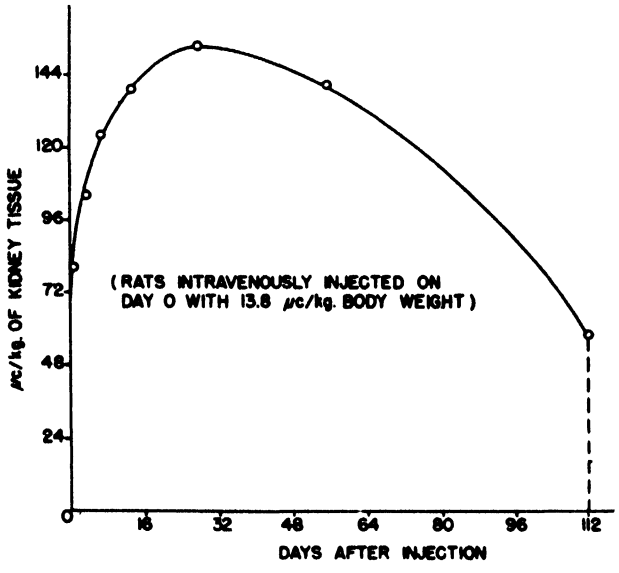



Figure 1. Polonium-210 in rat kidneys

Critical organ: kidney

Fraction of ingested polonium absorbed: 3–6%

Fraction of absorbed polonium in critical organ: (1) following ingestion, 3%; (2) after injection, 7%.

Biological half-life in critical organ: In both injection and ingestion experiments, the period from 0–20 or more days was one of slowly increasing polonium concentration in the kidney. Subsequently, we found a biological half-life of about 70 days.

Biological half-life of polonium in total body: Ingestion; 90% excreted in 15 days. Subsequent biological half-life about 70 days. Biological half-life after injection is about 35 days. Data from limited excretion studies on humans showed a 50% drop in excretion rate in 34 days.

Although the polonium exposure following single injections was of relatively short duration, in doses less than 8 μc/kg bw, there was little histopathological evidence of damage for 100 days or more. The dominant lesion seen at this and still later times was one of tubular damage to the kidneys. It appeared first as a cloudy swelling in the cells of the loops of Henle and progressed through various stages of tubular cell damage to a point where extensive fibrosis occurred and the normal tubular architecture was so greatly disorganized that the kidney damage was certainly a factor contributing strongly to early death. At no time was there serious involvement of the glomeruli. The rate of development and the ultimate severity of the lesions was a function of dose. At the 3.6 μc/kg bw level, 100% of the animals sacrificed 350 days or more after injection showed severe kidney lesions. At lower levels a lower percentage of animals developed less severe kidney lesions at later times. No other lesion occurred with significant frequency at late times following small single doses of polonium except a focal fibrosis of the heart with small round cell infiltration. Neither the kidney nor the heart lesion ever developed in any severity at less than 100 days even at the highest doses. Time, in addition to dose, was required. In any acute toxicity experiments the kidney and heart lesions and the kidney as the critical organ would have been completely overlooked. From the histopathological data on acute toxicity of polonium at high dose levels, it would be impossible to extrapolate correctly to the chronic toxicity situation. One would conclude erroneously that the blood-forming organs (reticulo-endothelial tissue) were the critical ones and that deaths from low but lethal doses would be caused by blood dyscrasias. At the lowest effective

single dose levels (0.75 and 0.51 μc/kg bw) the only histopathological evidence of polonium toxicity seen was an occasional mild form of the kidney lesion. The usual senile changes were observed at very late times in both the injected and control populations. However, these changes appeared at an earlier date and progressed more rapidly in the injected groups than in the controls. This appearance of accelerated aging led our histopathologists to refer to the syndrome as "race-horse senility." The 0.25 μc/kg bw dosage level rats were completely indistinguishable from the controls.

Examination of tissues from animals receiving sizeable multiple injections of polonium (2 μc/kg bw at 14-day intervals) showed two very surprising histopathological changes. The same kidney lesion seen in the single-dose series occurred but it developed very much earlier (quite severe within 60 days of the original injection) in the multiple series. This was true even though the integrated radiation exposure to the kidneys in the multiple series was only a small fraction of that delivered in the same time by high single doses (which did not produce the lesion until much later). The other surprising feature of the histopathological changes following the 2 μc multiple dose was the development of a severe liver lesion. No single doses, no matter how large or how small, ever showed comparable liver damage. In the multiple-dose series, the livers of the rats became cirrhotic, with necroses and with extensive bile duct proliferation. Islets of parenchymous liver cord cells remained but these showed frequent giant cell forms and cloudy swelling of the cytoplasm.

The animals of the low level multiple dose series (maintained 0.01 μc/kg bw average dose) were completely indistinguishable from their controls in all examinations for histopathological changes.

The effect of polonium on life-span, using median lethal time (MLT) as a measure of life-span, is summarized in Fig. 2. The shortening of the MLT over the entire range of doses from 3.6 μc/kg bw to 81 μc/kg bw is obvious. In an attempt to portray more clearly what happened at the lowest single doses (0.9, 0.75, 0.51, 0.25 μc/kg bw and controls), this end of the dose spectrum was replotted on an expanded arithmetic scale in the inset. Statistical analysis was necessary in order to determine the significance of the mean difference in life-span, approximately pictured here as differences in MLT. At the 0.9 μc/kg bw level, sufficient animals died to establish the MLT, but shortly afterward the remaining animals were sacrificed for histopathological examination, so that statistical comparison of mean life-span differences was not possible. In the case of the 0.75 and 0.51 μc level males the reduction in mean life-span when compared with their controls was highly significant. The significance was at less than the 1% probability level as determined by Student's t test. The females at the 0.75μc level did not die significantly earlier than their controls, but did die significantly later than the males. Although the females at the 0.5 μc level did not differ significantly from the males (whose life-span was shortened) at that level, they also did not differ sufficiently from their own female controls for this latter difference to be significant. This apparent anomaly may be a result of the much greater degree of variance

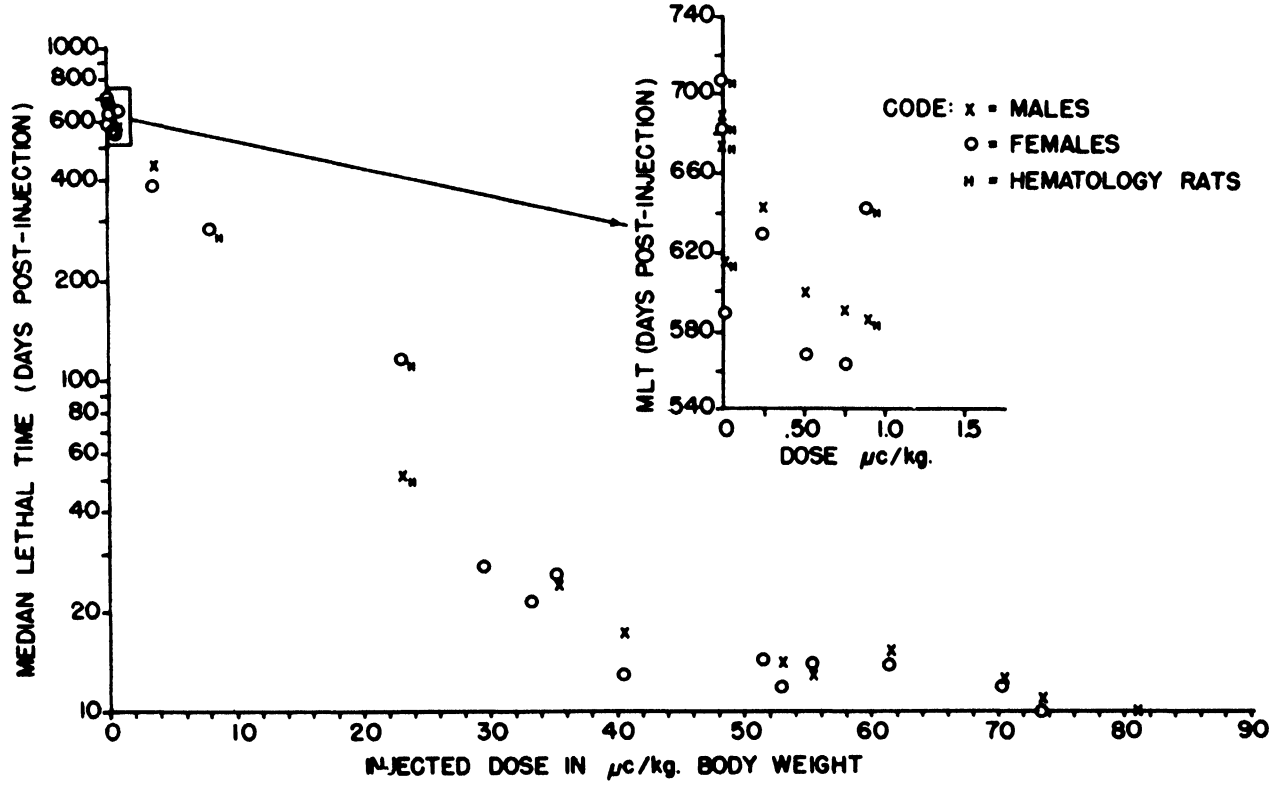



Figure 2. The effect of single injections of polonium on life span

within the female populations than within the male populations. The reduction in mean life-span in the 0.25 μc males is not quite significant, although it is highly suggestive of toxicity, since it is significant at a little greater than the 5% probability level. The females at this level, again, did not differ significantly either from their controls or from the males.

A possible explanation of the apparently greater sensitivity of males to chronic polonium toxicity (clearly established only at the 0.75 μc level) lies in the relative vital organ to carcass ratio in the two sexes. A much higher percentage of the larger male total body weight is in polonium-poor muscle and bone, and conversely, a much lower percentage is in polonium-rich vital organs than in the female. Thus, the males received larger doses per rat because of their heavier total body weight and these larger doses were concentrated in relatively smaller vital organ masses. Accordingly, the radiation dose delivered to the vital organs, including the critical organ (kidney) probably was substantially higher in males than in females.

It is therefore of interest to know the actual radiation dose in roentgens equivalent physical (rep) received by the kidneys at one of the low doses. Given below is a calculation of the radiation dose delivered to the kidneys by the lowest effective polonium injection, viz., the 0.25 μc single injection given to males. Figure 1 shows the mean concentration of polonium in 10 male rat kidneys at each point from animals injected with 13.8 μc/kg bw. (We do not yet have equivalent data on the radiation dose to female kidneys to permit comparison with the males.) The area under the curve to 112 days in Fig. 1 gives a measure of a large fraction of the total radiation exposure to the kidneys in microcurie-days per kilogram. To convert the term microcurie-days per kilogram to the more familiar rep unit, substitution was made in the classical formula

$$\text{rep} = 55\,E\,\frac{C \times days}{M} \qquad (1)$$

In this formula $E$ is the energy of radiation in Mev, and $C/M$ is the concentration of radioactive material in microcuries per gram of tissue. Subsequent extrapolation of the rep value obtained from the 13.8 μc level to the 0.25 μc level suggests that the kidneys of the latter group accumulated about 73 rep. This is an average of 0.65 rep per day over the 112-day measured-exposure period. The dose also can be expressed 0.11 rep per day averaged over the 684-day (± 34 days at the 95% confidence interval) mean life-span of this group. Comparison of this 0.11 rep per day figure with the currently accepted maximum permissible exposure of 0.3 rep of $\beta$ and $\gamma$ per week is difficult. A sure knowledge of the relative biological effectiveness (RBE) of polonium $\alpha$-radiation in this system is not available. Perhaps the above data will be useful to those engaged in determining RBE values.

## CONCLUSIONS

Our data show that, in the rat, the minimum effective single dose of polonium-210 is 0.5 or possibly 0.25 μc/kg bw. Direct extrapolation suggests a maximum permissible single exposure in humans substantially less than 0.5 μc/kg bw (35 μc per standard man).

Our data also show that a polonium body burden of 0.01 μc/kg bw maintained throughout life did not demonstrably affect rats. This suggests that the highest maximum permissible concentration figure (approximately 0.003 μc/kg bw) currently employed for man carries a substantial safety factor.

## SUMMARY

Late expressions of toxicity are observed following relatively short-lived polonium exposures in rats. The manifestations of toxicity, kidney lesions and shortened life-span would have been completely undetectable in acute toxicity experiments.

The minimum effective single dose of polonium in the rat was determined.

Data on toxicity following repeated or maintained polonium exposures are presented.

Recommendations are made for maximum permissible single and maintained polonium exposures in humans.

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2. Fink, R. M. (editor), *Biological Studies with Polonium, Radium, and Plutonium*, McGraw-Hill Co., Inc., New York (1950).

# The Problem of Radon in Uranium Mines

**By H. Jammet* and J. Pradel,† France**

The extraction of uranium minerals is the first stage in the utilization of atomic energy.

The methods currently in use are attended, in all of those mines, by various irradiation and contamination hazards, among which the presence of radon is most significant. The maximum tolerable concentration currently is set at $10^{-10}$ of radon per liter of air, It would appear, even if one considers that the daughter products RaA, RaB and RaC are not in equilibrium, that it would be impossible to set a less stringent limit. In fact, the limit proposed by the International Commission on Radiological Protection gives, for a 40-hour exposure per week (from Karl Morgan[1]), a dose, as computed at the level of the bronchi of: 9.5 rem/week with 100% of RaA and 50% of RaB and RaC; or 19 rem/week with 100% of RaA, RaB and RaC, instead of 0.3 rem.

In addition, it is necessary to allow for the fact that the risk is not the only one present in a miner, exposed to the radiation of the mineral, who also breathes uranium dust.

## RADON DETECTION

In the French mines, systematic measurements have been made, and individual monitoring already is in use in many installations. Starting from the measurements carried out at fixed points and at various times, a computation is made of the average quantity of radon inhaled by each miner. The measurements are carried out in Paris; or, more and more, on the spot, in a small laboratory set up in each mining division.

The radon samples collected (at the bottom of the mine) by means of an empty bottle containing one liter, are measured by measuring the current generated in an ionization chamber.

Currently, another dosage method is used, which consists of passing the air of the mine, by means of a small portable vacuum cleaner, through a cartridge containing activated carbon, which adsorbs the radon. The amount of the latter present then is measured by using three $\beta$-counters embedded in the carbon. This material is inadequate for good monitoring, and a portable instrument which directly gives a reading of the $\alpha$-activity present in the air is needed, along with registering equipment (Babar‡ type) placed at various points.

We are currently testing an individual portable instrument which continuously samples the air by means of a microvacuum device through a cartridge of active carbon. At the end of the day, it is thus possible to compute the average content of the air breathed by the miner.

These monitoring devices have made it possible to obtain a set of results which led to the following findings:

(1) The concentrations found in uranium mines are very high and often exceed $10^{-10}$ c/l, even with substantial ventilation, of the order of 4 to 8 $m^3$/sec for small research centers.

(2) The concentrations depend, not only on the size of the mineralized mass and its uranium content, but also on the nature and compactness of the rock.

(3) The use of explosives to break down the ore releases an important fraction of the bound radon as the rock is broken to pieces. In the blast smoke, concentrations of $5 \times 10^{-8}$ c/l were found.

(4) In the unventilated galleries, radon accumulates in very high concentrations, and by diffusion pollutes the other galleries.

(5) The water which circulates in the areas where there are lodes dissolves the radon formed in the cracks and releases it on contact with the air as it seeps down and drips in the galleries.

(6) Apparently clean galleries contain substantial amounts of radon brought, either by diffusion through the earth, or carried in by the waters.

## RADON CONTROL

These results logically made it possible to control contamination by the radon, by endeavoring first to limit the amount released, and then to evacuate the released radon as fast as possible.

It is of interest to decrease the amount of radon supplied and to accelerate its removal, in view of the quick decay of its radioactivity.

### Elimination of Radon Sources

It is possible to reduce the size of radon sources by the use of judicious operating methods and special arrangements.

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Original language: French.

* Chief, Protection Service against Radiations of the CEA.

† Ingénieur des Mines, in charge of radiological protection on the mining divisions of the CEA.

‡ Babar: a device used to filter air continuously, giving at every moment the activity deposited on the movable filter.

### 1. Operating Methods

The amount of radon released in the mines is reduced: (a) by breaking up the rock as little as possible, and by removing the ore very rapidly; and (b) by reducing to the utmost the research work to be carried out in the mineralized zone, in order to limit the volume released.

These two considerations suggest the use of extraction methods without storage chambers, and small installations, well-insulated from one another, rather than large mines criss-crossed by galleries from all directions.

It would doubtless be very interesting to make a study of an explosive which would break up the rock less, and thus reduce the percentage of released radon.

### 2. Special Installations

Let us study the radon source made up of a part of the gallery (Fig. 1). If $D$ be the output of radon coming out of the rock into the gallery; $d$ the output coming out of the gallery or of the source; $d'$ the output in the gallery due to disintegration; we can then write $d = D - d'$. To reduce the output from this source, we may (1) increase $d'$, (2) reduce $D$.

To this end, we made a study of dams, coverings on the ore, and channeling of the water.

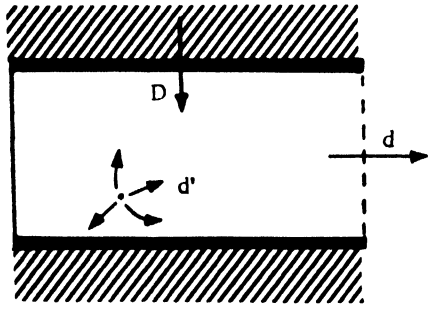



Figure 1

1. Dams. The technique making use of dams consists of closing up any useless gallery by a relatively tight dam. This delays the diffusion and increases the "upstream" radon concentration for an increase of $d'$ and possibly a slight reduction of $D$ (for $D$ is related to a diffusion phenomenon in which the difference of concentration in the rock and in the gallery comes into play).

It is to be noted that the dam need not necessarily be perfectly tight in order to be effective, which is a peculiarity of the radioactive gases.

Before building up a dam, we can write $d_1 = D - d'_1$ and after it has been set up: $d_2 = D - d'_2$. The gain due to the dam is $d_1 - d_2 = d'_2 - d'_1$. Now $d' = \lambda C$, with $C$ the radon concentration; from which, if $V$ be the volume of the portion cut off: $d_1 - d_2 = (C_2 - C_1)\,\lambda V$.

Experiment: The first tests carried out at the BNL mine with dams made up of boards placed edge-to-edge gave good results. More recently, we made use of a plastic dam (cocoon), the results of which confirmed the efficiency of the method.

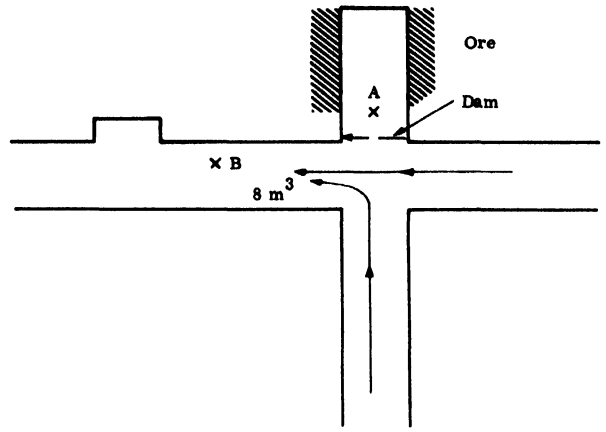



Figure 2

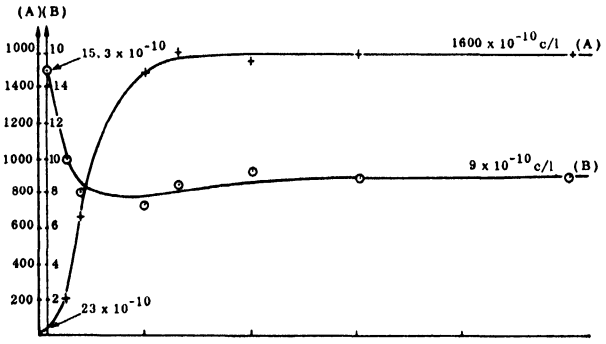



Figure 3. Influence of a boovier on the concentration of radon

There were 8 m³ of air polluted by passing before the heavily mineralized dead end of a cross bank. Referring to Fig. 2, we measured the radon content behind the dam (point $A$), and the content of the 8 m³ of air after they had passed before this cross bank (point $B$).

Here are the results in $10^{-10}$ c/l (see Fig. 3):

| | | | $A$ | $B$ |
|---|---|---|---|---|
| Before the dam was set up, | Wed. | 10 A.M. | 23 | 15.3 |
| After the dam was set up, | Wed. | 4 P.M. | 210 | 10 |
| | Wed. | 7 P.M. | 680 | 8 |
| | Thurs. | 10:30 A.M. | 1480 | 7.4 |
| | Thurs. | 5 P.M. | 1600 | 8.5 |
| First leaks | Fri. | 10 A.M. | 1550 | 9.3 |
| | Sat. | 10 A.M. | 1600 | 9 |
| | Mon. | 3 P.M. | 1600 | 9 |
| Big tears | | 15 days | 410 | 12 |

Interpretation: In this case we had $V = 200$ m³ and $C^2 - C^1 = 1.5 \times 10^{-10}$ c/l. Hence $d_1 - d_2 = 3.8 \times 10^{-6}$ curies/liter, which corresponded to a theoretical decrease, for the 8 m³ or air, of:

$$\frac{3.8 \times 10^{-6}}{8 \times 10^{3}} = 4.7 \times 10^{-10} \text{ c/l.}$$

This theoretical result is in agreement with the experimental results (decrease of 5 to 6 $\times 10^{-10}$ c/l).

2. Covering up of the ore. In order *to reduce D,* we carried out, in a laboratory, a series of tests which consisted of studying the influence, on radon release, of a paint or any other layer applied on a block of mineral.

For the time being, we obtained results only with oil. With those, we have no obstruction in the pores of the rock. With oil, the radon dissolves and diffusion is delayed. The release, for a piece of ore placed under a glass bell decreased, in five days, by approximately 50%. Tests will be carried out in the galleries.

3. Water channeling. The waters which release dissolved radon, constitute at their exit point, an important source of radon and, in order *to reduce D,* it is necessary to channel the water in conduits as soon as it appears, and to dispose of it outside the mine. This phenomenon has been shown by the two following experiments:

Experiment No. 1 — A 25-1 container, half-filled with the water which flows out of the mine, is then closed. At the end of three hours, the air in the jar contained $1.5 \times 10^{-7}$ c/l. One liter of water had released enough radon to contaminate 1.5 $m^3$ of air at the maximum tolerable concentration.

Experiment No. 2 — At the BNL mine, in a cross-bench which was clean, the main air was leaching the water which was running against the air current. We noted an increase in radon contents as the air penetrated further, until several times the tolerance limit was reached after 300 meters, according to the curve of Fig. 4.

The contamination, at the mine face, was mainly due to the water which had just come out of the rock, hence the need to enclose the sources close to the point where they come out. This channeling of the water caused contamination to disappear and following partial drying up of the cross-bank the air reached the working phase with a content of .2 instead of $12 \times 10^{-10}$ c/l.

In another mine, we observed the same pollution at a mine face, nearly clean but very wet, where the air concentrations were 2 to $4 \times 10^{-9}$ c/l.

The waters in these mines may have contained up to $10^{-6}$ c/l of radon in solution and *cannot be used in rams,* where they release their radon.

### Radon Evacuation

Once the radon sources have been eliminated to the greatest possible degree, thought must be given to removing the *radon present in the air* of the galleries.

To this end, we used powerful ventilation, with outputs which may range from 4 to 20 $m^3$/sec according to the size of the working area. Under good conditions, the radon content is approximately inversely proportional to the air output.

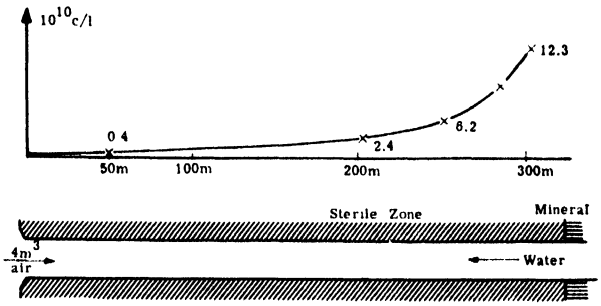



**Figure 4. Contamination of air by run-off water**

Two essential conditions must be met in order to achieve effective ventilation: (1) All recycling must be avoided by installing air intakes outside the mine and at least 10 meters from the air outlets, so placed as to allow for the orientation of the winds; (2) bringing the pure air as close as possible to the mine face.

The heavy concentrations encountered in the blast exhausts require that there be no immediate return to the work face.

Ventilation has another primary merit: it considerably reduces the active deposit on the dust. In fact if the radon released from the rock is quickly removed, the daughter products which have a mean half-life of one half hour do not have time to form, and there is only a fraction of the equilibrium concentrations, mainly in RaB and RaC.

All these steps provide for a considerable reduction in the radon content. In the BNL mine, where all methods have been tried, we obtained very substantial reductions, going up to a factor of 100 in a heavily mineralized crossing of galleries, with abundant water circulation.

However, when all protective measures taken collectively do not sufficiently reduce the radon content, we use individual protection. The best method consists of feeding pure air to the worker: for this, use is made of hoods and masks. The hoods have the drawback of not being entirely self-contained (need for a hose). The dust masks (very large filtering surface and high arresting power) do not stop radon, but rather the active dust and the silica dust.

Resort to these means of protection will be attended by an important transformation in the extraction of uranium: close liaison between the operators and the radiological protection services, systematic organization of the supervision, severe discipline and conscience of the responsibilities involved.

### REFERENCE

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# Industrial Hygiene of Uranium Processing

**By M. Eisenbud and J. A. Quigley,* USA**

The conversion of uranium ores to compounds suitable for reactor operation has become one of the world's important metallurgical industries. In the era of nuclear technology, the refining of uranium will be undertaken in amounts comparable with many of the non-ferrous metals. This report summarizes the knowledge which has been accumulated during more than ten years of large-scale operation of plants which process uranium.

The potential hazards which are incidental to processing this metal will of course depend on the characteristics of the ore and the refining process. Ores from various parts of the world differ in their uranium content and the various isotopes associated with the uranium series. This report will consider the case of a high-grade pitchblende assaying 25% uranium.

## DESCRIPTION OF PROCESS

The ore is ordinarily received in sealed 55-gallon drums and is dried, crushed and blended prior to digestion in nitric acid. Acid insoluble components such as silicates and silica are removed by filtration. Radium, originally present in a concentration of about 100 milligrams per ton, is then co-precipitated with barium sulphate.

The uranium is extracted from solution as uranyl nitrate by an organic solvent, leaving the isotopes of thorium and protactinium in the aqueous phase. The uranyl nitrate is later stripped from the solvent into a water solution which is thermally denitrated to the orange uranium trioxide (orange oxide).

The orange oxide is converted by reduction in a hydrogen atmosphere to uranium dioxide (brown oxide) which is further converted to uranium tetrafluoride by reaction with anhydrous hydrogen fluoride. If metal is desired as the final product, $UF_4$ (a green salt) can be reduced by a thermite type of reaction using metallic magnesium. This step produces a metal ingot which may be purified by recasting under high vacuum. The purified uranium metal can be machined, rolled or extruded into the final form required.

When uranium hexafluoride is desired as the end product, the uranium tetrafluoride is reacted with fluorine gas. Uranium hexafluoride is volatile and passes off as a gas which is subsequently condensed and bottled.

* Including work by M. Eisenbud, A. E. Brandt, J. Alercio, P. Klevin and W. B. Harris, U. S. Atomic Energy Commission; and J. A. Quigley, National Lead Company of Ohio.

## POTENTIAL HEALTH HAZARDS

In Table I are presented the various hazards which may be expected to occur at various places in the process. A detailed discussion of these potential hazards and the methods by which they are controlled is not possible in a single presentation, but some general observations can be made.

### Gamma Exposure

In processing high-grade ores, radiation exposure may be controlled by process shielding, remote operation or rotation of personnel. The latter practice has been employed primarily in the early steps of the process where the radium is still present. By administering the maximum permissible dose as 3.9 r per 13 weeks (¼ year) rather than 0.3 r per week, it has been possible to keep all personnel within acceptable limits at a considerable saving in the cost of process shielding. A simple example is the manual handling of drums of high-grade ore. This is an intermittent operation in which an employee can receive his weekly maximum permissible dose in only a few hours. Rotation of personnel has been accepted as a logical means of controlling individual exposures in lieu of expensive mechanized equipment that would otherwise be necessary.

The kind of ore being processed will of course markedly influence the gamma dosages encountered. High-grade ores are usually correspondingly high in radium content and therefore relatively more active as gamma sources. A worker standing 2 feet from a pile of drums containing pitchblende of 25% uranium content will receive about 25 mr/hour. If he is transferring a drum by an unshielded fork lift truck, he is receiving about 10–15 mr/hour.

### Beta Exposure

The beta dose to the basel epithelium of hands in contact with uranium metal is estimated from extrapolation chamber measurements[1] to be 235 millirads per hour. A horsehide-leather glove will reduce this exposure by about 80%. The maximum permissible dose to the hands is 1500 millirads per week, and therefore contact dose from uranium is not a serious limitation except for those very few individuals who cannot limit their bare-handed contact with the metal to less than 6 hours per week or to 24 hours per week if the protection of leather gloves is available.

$UX_1$ and $UX_2$, being isotopes of protactinium and thorium, are capable of concentration and separation

**Table I. Summary of Potential Hazards from Processing Pitchblende Containing 25% Uranium**

| *Potential hazard* | *Source of exposure* | *Potential exposure under normal operating conditions—but with no controls* | *Remarks* |
|---|---|---|---|
| amma radiation | 1. Handling ore | 50 mr/hr adjacent to stack of drums of ore | Radium content of ore approximately 100 mg per ton |
| | 2. Precipitates containing radium | 100 mr/hr adjacent to a stack of drums | $Ra^{226}$ content approximately 30 mg per ton |
| eta radiation | 1. Conversion of $UF_4$ to $UF_6$ | 2–3 rads per week possible among workers loading and unloading reaction vessels. Dose to hands may be higher | Gaseous $UF_6$ is removed, leaving unreacted residue highly enriched $UX_1$ $UX_2$ |
| | 2. Vacuum casting uranium billets | Furnace operators may receive as much as 2 rads per week. Dose to hands somewhat higher | Source of B are distilled impurities which contain $UX_1$ $UX_2$ and condense on furnace interior |
| | 3. Handling metallic uranium | Dose to basal epithelium of skin approx. 235 millirads per hour when in contact with U metal in equilibrium with $UX_1$ $UX_2$ | 90% of equilibrium beta activity is restored 90 days after vacuum casting |
| adon inhalation | Storage of ores in closed spaces | With no ventilation concentration may range as high as $10^{-8}$ c/l | Conditions much improved by even minimal ventilation in storage and processing areas |
| ihalation of alpha-emitting dusts and fumes | 1. Sampling and crushing ore | Daily average concentration up to 5 milligrams per cubic meter have been experienced at operations in which good process and area ventilation was not provided | Dust control techniques normally employed in processing toxic metals are effective |
| | 2. Mechanical or manual handling of dry U salts and oxides | | |
| | 3. Escape of gaseous $UF_6$ | Massive exposures due to sudden releases of $UF_6$ may occur | High standards of maintenance are required to prevent leaks at valves, flanges, etc |
| | 4. Machining metallic U | Excessive exposures possible due to pyrophoric nature of U | Store chips under oil, use liberal quantities of cooling oils |

y a variety of physical and chemical means. For xample, these daughter products of uranium are ondensed in the cool parts of the vacuum furnace long with other impurities which are low boiling ith respect to the natural uranium. Also when ranium tetrafluoride is converted to $UF_6$, a gas, $JX_1$ and $UX_2$ do not form volatile compounds as oes uranium, but remain behind with other impuries. Residues such as these are relatively active betamitters and as noted in Table I have delivered in ractice whole-body beta doses up to 3 rad per week.

### Radon Inhalation

When high-grade ores of radium-enriched residues re stored in enclosed spaces, the radon concentration ay reach $10^{-8}$ to $10^{-7}$ curies per liter.

These levels are considerably in excess of the aximum permissible dose to radon but they are ctually of little hygienic significance because concen-ations can be reduced remarkably by moderate mounts of general ventilation.

The potential hazard from radon is now regarded[2] o be due to the inhalation of inert dust containing bsorbed radioactive daughters of radon. It has been stimated that when these daughters are in equilibium with radon the dose from radon *per se* is only ne-half of one per cent of the dose from radon plus s daughter products.

When gaseous radon diffuses from a radium containing material, equilibrium with its daughter products will be reached in about two hours. In order for the workroom air to contain both radon and its daughter products in equilibrium, it is necessary that the air, radon and inert dust which serve as nuclei for adsorption of the radon daughters co-exist for the period during which equilibrium is being attained. Under many industrial conditions, this equilibrium is not achieved and it is therefore probable that concentrations of radon far higher than the maximum permissible dose of $10^{-13}$ c/cm$^3$ can be safely inhaled under many conditions.

### Insoluble Alpha Dust Inhalation

The potential risk from inhalation of insoluble alpha-emitting dusts is that retention of the dust within the lung may produce lung cancer over a long period of time. The levels of exposure which are apt to be encountered under this category of hazard are far greater in comparison with the maximum permissible dose than the other exposures encountered in the processing of uranium. In the crushing of ores and in the dry handling of the oxides and fluorides, it is possible to exceed the maximum permissible dose several hundredfold. Techniques of process ventilation developed to control the hazards from metals such as lead and arsenic are satisfactory for uranium.

In the machining of metallic uranium, the pyrophoric properties of this metal must be recognized. Chips of uranium frequently ignite at the cutting point and their burning produces a dense uranium-oxide fume. When uranium is hot rolled into strips or plates, the surfaces tend to oxidize rapidly and the scale thus produced gives rise to dust.

### Inhalation of Soluble Uranium Compounds

When soluble uranium compounds are inhaled, the radiological risk is negligible but one does become concerned with the possible toxic effects on the kidneys. Consequently the maximum permissible concentration for exposure to soluble uranium is based on chemical toxicity rather than radiological hazard.

Uranium occurs in soluble form as $UO_2F_2$, derived from $UF_6$ which hydrolyzes upon contact with the air. $UF_6$ is very corrosive and leaks from valves and flanges are apt to occur if the plant maintenance is not of a high quality.

## STUDIES OF INDUSTRIAL WORKERS EXPOSED TO URANIUM

Many hundreds of employees of the companies processing uranium have been exposed to the dusts and fumes of this metal. When the plants first undertook this process the hazards were not well-understood and in many places the workers were exposed to concentrations of dust and fume which exceeded the levels now recommended by the National Committee on Radiation Protection as the maximum permissible concentration. Studies of these workers in relation to their exposures to uranium has provided useful information concerning the storage, excretion and effects of uranium in humans.

### Observations Concerning Storage and Excretion of Uranium

#### Lung Retention of Insoluble Dust†

In the United States, the maximum permissible concentration for 8-hours exposure to insoluble uranium dust in air is $5.1 \times 10^{-11}$ microcuries per milliliter.[3] This value was obtained from experiments with dogs[4] which indicate that lungs of animals exposed to this concentration for 8 hours per day will, at equilibrium, contain 14 parts per million of uranium. This tissue concentration will deliver 300 mrem per week to the lung, assuming an RBE of 20.

We have had an opportunity to analyze the tissues of 2 men who died of non-occupational causes after being exposed for a long time to high concentrations of dust of $UF_4$ and $UO_2$. The data relative to the autopsy findings and exposure history are contained in Table II where it will be seen that the measured lung concentrations are exceedingly small compared to the concentrations which were predicted from the dog experiments which serve as the basis for equating the above concentration in air to 14 ppm in lung.

It is not possible to reconcile these differences. However, one notes that the animal results are being applied directly to man without any knowledge as to the comparative relationship of deposition and clearance rates in the two species. Also, the assumption that an atmospheric concentration of $5.1 \times 10^{-11}$ microcuries per milliliter is equivalent under equilibrium conditions to a tissue concentration of 14 ppm is based on extrapolation of data actually obtained at very much higher concentrations. It is of course possible that our estimate of the average exposure of the two men is in error, but this is not likely to account for a factor of more than 2 or 3.

There is an obvious need for more data of this type.

†Dust concentrations in this report are sometimes expressed as microcuries per milliliter and sometimes as milligrams per cubic meter. $5.1 \times 10^{-11}$ microcuries per milliliter is roughly equivalent to 50 micrograms per cubic meter.

Table II. Uranium Contents of Lungs of Two Workers

| | Months exposed | Average exposure (micrograms/M³) | No. of months between last exposure and death | Uranium content of lung tissue at time of death (micrograms per gram) | |
|---|---|---|---|---|---|
| | | | | Measured | Predicted |
| J.B. | 24 | 17,000 | 10 | 0.35 | 600 |
| T.W. | 12 | 5000 | 15 | 0.23 | 56 |

#### Storage in Bone

Five determinations of the uranium content of bone have been made in individuals who had been exposed to relatively high concentrations of dust. Four of the specimens were from autopsy of individuals who died of natural causes. The fifth specimen became available during a surgical procedure. Exposures are given as micrograms of uranium per cubic meter of air for a given number of days and the bone data are given as micrograms of uranium per gram of ash. The data are given in Table III below.

Table III. Summary of Exposure of Five Men and the Uranium Content of Their Bone

| Man | Period | Days exposed | Level (micrograms/M³) | Uranium in bone (micrograms per gram of ash) |
|---|---|---|---|---|
| D. N. | I | 252 | 18,750 | |
| | II | 69 | 8200 | |
| | III | 17 | 18,750 | |
| | IV | 548 | 350 | |
| | | | | 16.7 |
| J.H. | I | 90 | 1000 | |
| | II | 1200 | 150 | |
| | III | 60 | 0 | |
| | | | | .5 |
| J. B. | I | 730 | 17,000 | |
| | II | 304 | 0 | |
| | | | | 17.0 |
| R. R. | I | 61 | 4500 | |
| | II | 45 | 0 | |
| | III | 228 | 4500 | |
| | IV | 578 | 850 | |
| | V | 152 | 0 | |
| | VI | 1399 | 850 | |
| | | | | 4.9 |
| T. W. | I | 365 | 5000 | |
| | II | 548 | 0 | |
| | | | | 2.0 |

An estimate of each man's skeletal uranium burden was made assuming exponential build up and diminution functions. It was assumed that the biological half-life was 450 days. It will be noted that this is consistent with data to be presented later in this report. Because the constant of proportionality could not be determined in the assumed exponential relationship between exposure and skeletal burden, the calculated estimates yield units which are only in proportional relation to the actual concentration. For this reason the predicted bone concentration, in arbitrary units, and the observed values, have been normalized in the following way. The subjects were arrayed according to predicted bone concentration in arbitrary units. It was found that they were then also arrayed according to observed bone concentration. The lowest value of each variable was given the value one and the other values as represented as multiples of it. The results are summarized in Table IV and are presented graphically in Fig. 1.

The linearity in the relationship of the predicted to the observed values are striking.

### Relationship of Exposure to Urinary Excretion

It is a relatively simple matter to estimate an individual's exposure to external radiation, using either a film badge or pocket dosimeter. Unfortunately there are no comparable devices with which one can integrate exposure to radioactive dust. This is an old problem in industrial hygiene which has been partially solved by the development of air-sampling techniques with which one can approximate the average exposure of a group of workers who are performing the same repetitive operation.

It is generally assumed that the urinary excretion of a substance is potentially the most satisfactory method of estimating the dust exposure of an individual. Efforts to calibrate this relationship for uranium have encountered the same difficulties which are well known to investigators who have attempted to establish similar relationships for the heavy metals.[5] Because of differences in working habits, it is virtually impossible to estimate the dust exposure of an individual employee with any degree of accuracy. Moreover, for any given exposure there are apt to be individual differences in both absorption and excretion. For example, a mouth breather under some conditions may absorb far more than a worker breathing through his nose.[6]

Table IV. Comparison of Observed and Predicted Bone Concentration

| Man | Uranium in bone (measured) micrograms/gram of ash | Uranium in bone (measured) Relative | Uranium in bone (predicted) Relative |
|---|---|---|---|
| J. B. | 17.0 | 34.0 | 31.7 |
| D. N. | 16.5 | 33.0 | 22.6 |
| R. R. | 4.9 | 9.8 | 7.6 |
| T. W. | 2.0 | 4.0 | 3.6 |
| J. H. | 0.5 | 1.0 | 1.0 |

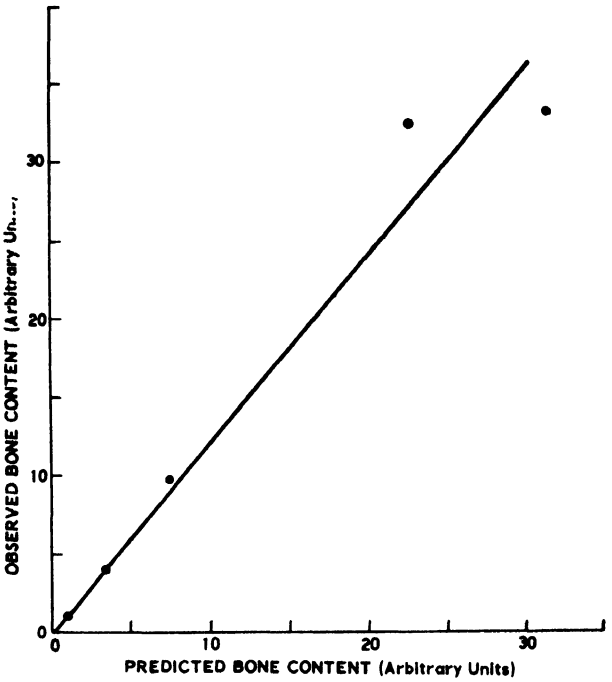


Figure 1. Predicted vs observed uranium in bone

Although it does not appear practicable to use the urinary excretion of an individual as a measure of his exposure, it is quite feasible to estimate the exposure of a group in this way. The average results of urine analysis from many individuals tend to mask the effect of individual differences. In Table V are summarized the average exposure and the urinary uranium excretion for 7 groups of employees exposed to uranium oxide dust. It will be noted that there is a coarse relationship between the severity of exposure and the rate of urinary excretion and on the average those having more severe exposure excrete higher concentrations of uranium. However, it will also be noted that because of the wide range in the reported values for any one group it is not practicable to assign a level of exposure on the basis of any individual's urine sample. A general conclusion which may be drawn from these and other data is that a group of individuals exposed to the maximum permissible concentration of uranium will excrete about 50 micrograms per liter. This figure which is based on human experience is in excellent agreement with the prediction of Neuman[7] based on animal data.

Table V. Urinary Uranium Excretion of Industrial Workers

| No. of men | Average exposure (multiples of $5.1 \times 10^{-11}$ microcuries/ml) | Urinary excretion (micrograms/ml) Low | High | Avg |
|---|---|---|---|---|
| 24 | 0.23 | 0.008 | 0.058 | 0.025 |
| 4 | 0.80 | 0.017 | 0.118 | 0.060 |
| 38 | 0.88 | 0.005 | 0.088 | 0.028 |
| 19 | 1.05 | 0.003 | 0.119 | 0.050 |
| 30 | 1.4 | 0.008 | 0.143 | 0.054 |
| 5 | 1.7 | 0.054 | 0.084 | 0.065 |
| 22 | 4.9 | 0.032 | 0.389 | 0.100 |

### Excretion Following Massive Accidental Exposure

An individual who was working with powdered uranium metal was exposed to massive amounts of uraninum oxide fume when the powder suddenly exploded. Figures 2 and 3 are plots of the daily urinary excretion of uranium for 70 days after the accident. During this period the subject was hospitalized because of extensive thermal burns and it was then possible to collect all the urine voided. The curves give uranium excretion in milligrams per liter and milligrams per day. One notes that the plot of uranium concentration consists of three distinct exponential components, each representing elimination from a distinct storage reservoir.

Of particular significance is the rapid elimination during the first few days. The exposure was to uranium oxide fume, which, on the basis of measurements under simulated conditions, is estimated to be completely less than 0.5 microns in diameter.

### Excretion of Uranium after Cessation of Long-Term Exposure

An individual who resigned after several years employment in one of the uranium plants submitted a urine sample at approximately weekly intervals over a period of about 900 days after leaving his uranium exposure.

The diminution in uranium excretion during this period of time is shown in Fig. 4 where it will be noted that the rate of change of the excretion rate is

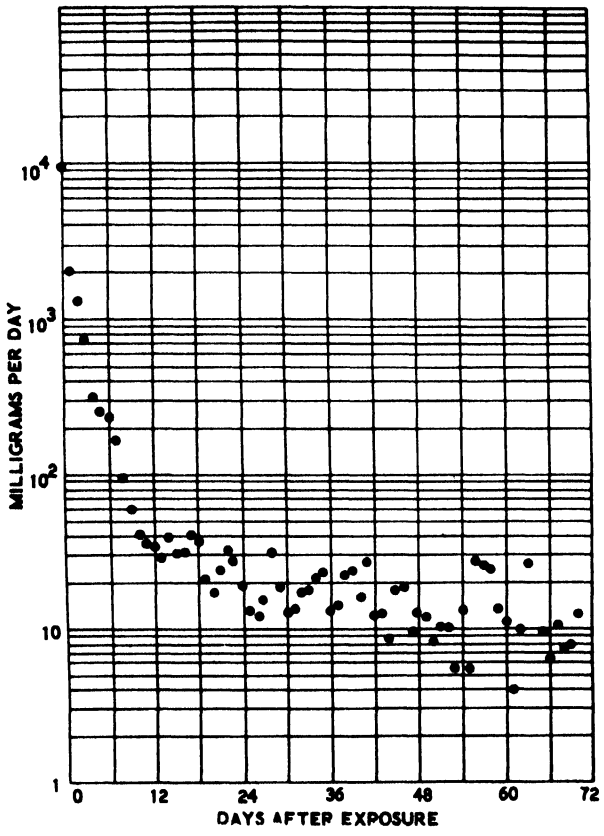



Figure 2. Daily excretion of uranium by an individual subjected to a single massive exposure

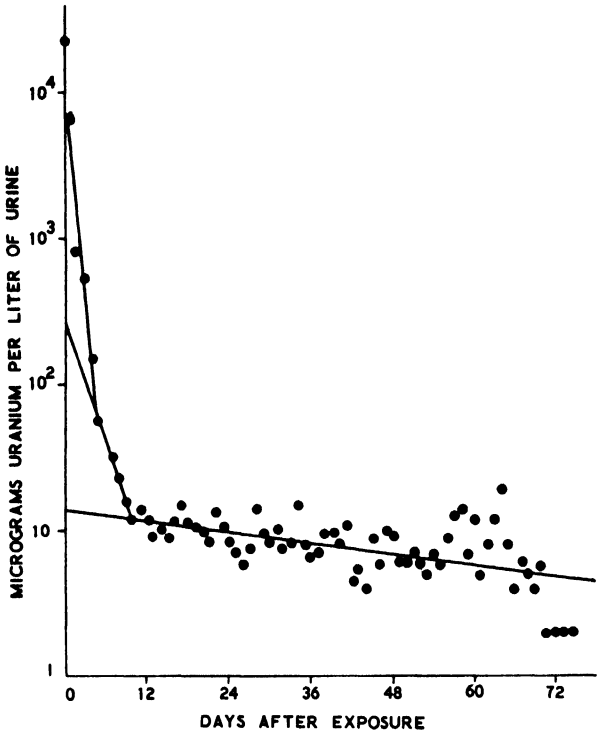



Figure 3. Concentration of uranium in urine of individual subjected to single massive exposure

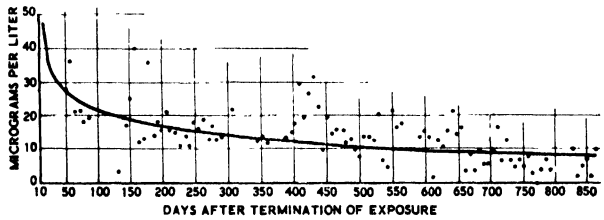



Figure 4. Urinary uranium excretion following prolonged industrial exposure

almost unchanged after about one year. During the latter two years of this study, the urinary concentration of uranium was diminishing at the rate of about 50% in 450 days. The scatter about the least-squares fit of the data may be due less to biological factors than the lack of precision in the methods of uranium assay then in use. The standard error of a determination at 10 micrograms per liter was approximately equal to symbol 10 micrograms during the period when this study was under way. With fluorimeters of more modern design this has been reduced to approximately equal to symbol 1 microgram.

The more rapid elimination of uranium during the first weeks following cessation of exposure appears to be due to clearance of uranium from the soft tissues of the body. This is the period covered under accidental exposure above. This clearance is apparently completed after about 150 days following which the large reservoir of uranium which has been deposited in the skeleton is then eliminated at a relatively slow rate. As noted, the half-life of uranium from bone appears, from these data, to be approximately 450 days.

### Evidence of Kidney Injury

The maximum permissible dose for soluble uranium compounds was determined by their toxic effects on the kidney of experimental animals.[8] In the United States, the only exposures to soluble uranium are in processes involving $UF_6$. This substance is gaseous at room temperature but upon release to the atmosphere it hydrolyzes, forming uranyl fluoride ($UO_2F_2$) a soluble fume.

The early history of $UF_6$ production involved frequent overexposures due to leaks that occurred because of the corrosive properties of the $UF_6$ and the free fluorine often associated with it.

### Single Massive Exposure

An incident occurred at one plant in which 5 employees received a short massive exposure of uranium hexafluoride. These men all had recent complete physical examinations with repeated laboratory examinations and were considered entirely normal in every way.

The exposure resulted from approximately 28 lb of hot $UF_6$ which was released in their immediate vicinity within a matter of two minutes. Table VI summarizes the urinary finding in the days following exposure. Three of the five excreted over one milligram per liter on the first day of the accident. Abnormal findings persisted for a few days but in all cases the urinary findings returned to normal, indicating only minimal effects on the kidney despite the relatively high exposures encountered.

### Chronic Exposure to Soluble Uranium

In 1950 seven cases of nephrosis due to chemical toxicity of soluble uranium were diagnosed in a plant employing approximately 130. This plant had been in operation since 1942, but had a high turnover among the employees due to induction into the Armed Forces and transfer to other employment. Prior to 1950, although a medical program was in existence, no cases of renal injury had been encountered.

The seven cases considered here all had pre-employment physical examinations and repeated laboratory examinations from the time of their employment. With one exception, the individuals in the group had been employed for periods ranging from several months to just over two years. In each case abnormal urinary findings, consisting of red blood cells, fine and coarsely granular casts and albumin were the only findings encountered; all were asymptomatic. All were given complete physical examinations, including careful histories, in an attempt to rule out other possible causes of their abnormal findings. Uranium determinations made on urinary samples showed the presence of as much as 3.46 mg/l. Each of the individuals was then hospitalized in a university hospital for a careful evaluation of their renal status. Examinations included cystoscopics, retrograde pyelograms, intravenous pyelograms, and renal-function tests. These tests indicated no diminution in renal function and, because of the absence of other discernible causes for the abnormal urinary findings and the fact that these men had been exposed to soluble uranium which is a known renal irritant, a diagnosis of nephrosis due to exposure was made. The patients were removed from further contact with uranium when the abnormalities were first encountered and, in all cases in which follow-up was possible, urinary findings became normal within a few months.

The exposure of the men was severe, as indicated by the summary of exposures in Table VII.

Table VI. Urinary Findings in Five Individuals Exposed to $UF_6$

| *Subject* | *Day* | *Urinary uranium concentrations mg/l* | *Albumin* | *RBC* | *Casts* |
|---|---|---|---|---|---|
| 1 | 1 | 2.60 | 0 | 0–2 | Rare |
| | 2 | 0.60 | 0 | Occ. | 0 |
| | 3 | 0.67 | 0 | 0–2 | 0 |
| 2 | 1 | 2.10 | 0 | 0 | 0 |
| | 2 | 0.05 | 0 | 0–2 | 0 |
| | 3 | 0.11 | 0 | 0 | 0 |
| 3 | 1 | 1.30 | 0 | Occ. | 0 |
| | 2 | 0.06 | 0 | 0–2 | 0 |
| | 3 | 0.07 | 0 | Occ. | Rare |
| 4 | 1 | 0.28 | 0 | Occ. | 0 |
| | 2 | 0.01 | 0 | Occ. | 0 |
| 5 | 1 | 0.110 | 0 | 0 | 0 |
| | 2 | 0.028 | 0 | 0 | 0 |

### Exposure to Insoluble Dust

A large number of industrial workers who were exposed for many years to high concentrations of insoluble dust have been under careful medical surveillance, but all findings to date have been negative. Included in this group are approximately 100 men who were exposed for about 5 years to from 0.5 to 2.5 milligrams of uranium per cubic meter. It is estimated that 50 men were exposed during this period to from 2½ to 10 milligrams per cubic meter. There has been no evidence of renal injury among these workers nor have any lung pathology or blood changes been observed.

Although it is possible that insufficient time has elapsed to obtain observable lung pathology, this should be weighed against the fact that the exposures of these men began about 13 years ago. It is of particular interest that the two lung studies referred to earlier in this report were from this group of men. It will be recalled that the observed lung concentration of uranium was very much lower than would be expected in view of the heavy exposures sustained.

Table VII. Exposure to Soluble Uranium Compounds

| *Exposure mg/M³* | *Year* | | | | |
|---|---|---|---|---|---|
| | *1948* | *1949* | *1950* | *1951* | *1952* |
| 0–0.1 | 9 | 13 | 38 | 33 | 55 |
| 0.1–0.5 | 13 | 14 | 62 | 55 | 48 |
| 0.5–2.5 | 44 | 31 | — | 30 | 22 |
| >2.5 | 34 | 61 | 32 | 8 | — |

### Summary of Medical Observations

There has been sufficient experience with severe exposures to both the soluble and insoluble compounds of uranium to permit the conclusion that uranium has a low order of chemical toxicity in man. Many of the non-radioactive heavy metals such as lead, arsenic and mercury would produce very severe, perhaps fatal, injury in the levels of exposures reported here.

The negative findings relative to renal injury among workers exposed to insoluble compounds are particularly significant in view of the high levels of exposure reported. Although some injury has been noted among individuals who had been exposed to soluble compounds the injury has been minimal and disappeared completely when the exposures were reduced.

The relatively low concentrations of uranium in the lungs of the two workers autopsied suggests that the safety factor in the maximum permissible dose for insoluble compounds is unnecessarily conservative. However, more data of the type presented are needed before any upward revision of the maximum permissible concentrations could be permitted on this basis alone.

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# Mechanism of Uranium Poisoning

**By H. C. Hodge,* USA**

Uranium has many distinctions, principally as a source of atomic energy. The biological effects of uranium have been the object of the most intensive research. In Dr. Voegtlin's words: "it is fair to say that the study of the toxicology of the uranium compounds represents the most comprehensive experimental investigation of an industrial poisoning ever carried out."

When uranium gets into the body it is reactive, quite apart from its radioactivity. Uranium is one of the most toxic elements chemically, more toxic than arsenic or mercury. Providentially, uranium is absorbed into the body only with difficulty. If it were not so, certain workmen in the early days of the Manhattan Project must have been injured severely. For example, there was one man whose job in the process of converting the brown oxide, uranium dioxide, into the green salt, uranium tetrafluoride, involved at one step shoveling the green salt into a wheelbarrow, a dusty operation in which he daily became liberally coated with the green powder. So much so that he was known to his comrades as the "Green Hornet." Yet, this man was not injured by uranium. In fact even when uranium is swallowed, only traces are absorbed into the body.

## ANALYTICAL METHOD

To discover this simple fact, it is necessary to have a good analytical method for uranium. The reaction between the uranyl ion and ferrocyanide in solution to give a beautiful port wine color forms the basis for a reliable analytical method if milligram quantities of uranium are to be measured. For a highly toxic element like uranium, tolerated doses are measured in tenths or hundredths of milligrams per kilogram body weight. The first requirement in the study of the biological effects of uranium was to find a method sufficiently delicate to trace quantitatively the very small amounts of uranium capable of producing injury to the body. A satisfactory method was gained when Neuman and Bloor perfected Hoffman's procedure utilizing the fluorescent light emitted when a sodium fluoride glass fused with a small amount of uranium is exposed to ultraviolet light. Under the proper experimental conditions the intensity of emitted light is directly proportional to the amount of uranium present. By this method, it is possible to measure one-two billionth (1/2,000,000,000) of a gram of uranium in a biological sample with an error of ± 5% (The first atom bomb cost two billion dollars.)

## TISSUE DISTRIBUTION

Having a good analytical method, the distribution of uranium in the body can be described when a small dose is given intravenously. Uranium given into the blood stream rapidly leaves the blood. *Forty minutes* after an intravenous dose, a quarter to a third of the uranium has been deposited in the skeleton, nearly a half has been excreted in the urine and, of the balance, approximately half is in the kidney and half in the other soft tissues. Only a trace (1% or so) remains at this moment in the blood. The trends established so promptly continue. The uranium deposited in the kidney tends to move into the urine; the uranium deposited in the other soft tissues tends to move into the kidney. Consequently, when the pattern of distribution is examined *forty days* after the administration of an intravenous dose, a fifth to a quarter of the uranium still is retained in the skeleton and the balance essentially entirely has been excreted from the body via the urine. Thus, the bone and the kidney are the two tissues of major interest in the study of uranium poisoning.

## PROTEIN AND BICARBONATE COMPLEXES

Uranium in the body tends to be converted into the hexavalent uranyl ion; this is the stable valence form. In the discussion that follows when biological effects are being considered, "uranium" usually means "uranyl ion." The uranyl ion in the blood stream is carried in part complexed with protein and in part with bicarbonate; almost none exists as uranyl ion ($UO_2^{++}$). The uranium protein complex is non-diffusible, the uranium bicarbonate complex is diffusible. Approximately 40% of the uranium is protein bound and 60% is bicarbonate bound. The uranium protein complex is an anion, the uranium bicarbonate complex (1 $UO_2^{++}$ to 3 $HCO_3^-$) is an anion; it is an interesting fact that the uranyl cation is actually carried in the blood in anionic forms. Uranium is a strong protein precipitant reacting apparently principally with carboxyl groups. This is a non-denaturing precipitation and the addition of bicarbonate is followed by a prompt re-solution. The diffusibility of the uranium bicarbonate complex is a key fact in understanding the mechanism of uranium poisoning.

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## SKELETAL DEPOSITION

The deposition of uranium in bone involves both organic and inorganic bone constituents. Amprino has shown that trace quantities of uranium in the body may be deposited in part in the osteoid, the organic bone matrix. While this organically bound uranium may be important, because of the permanence of such binding, when exposures to highly radioactive uranium isotopes occur, this locus of deposition is negligible for exposures to the natural mixture of isotopes. When chemically detectable amounts of uranium are administered, Neuman has shown that the bone *mineral* is the principal site of deposition. Uranium is deposited on the surfaces of the extremely minute crystals of hydroxylapatite by exchange with surface calcium ions. Experiments using $Ca^{45}$ in solution in contact with bone crystals have shown that uranium-treated bone has less exchangeable calcium than "normal" bone. Numerically, for every uranium ion deposited on the crystal surface, two calcium ions are no longer available for exchange with calcium in solution. Similar experiments with radioactive phosphate show that there is simultaneously a reduction in exchangeable phosphate of the crystal surfaces; for every uranium ion bound to the mineral, there are two phosphate groups no longer available for exchange with solution phosphate. A reasonable explanation can be made by assuming that each uranyl ion complexes tightly with two adjacent phosphates of the crystal surface thus (1) preventing their escape into solution and subsequent substitution by tagged phosphate ions, and (2) simultaneously releasing from the crystal surface two calcium ions previously associated with these complexed phosphate ions.

## QUESTION OF RADIOLOGICAL HAZARD

When the Manhattan Project was in the stages of rapid development, and when for the first time it could be seen that hundreds of workmen were going to be exposed to quantities of uranium, one of the questions presented to those responsible for health protection was whether uranium would ultimately constitute a hazard because of its radioactivity in the same way as radium had been to the famed dial painters of World War I. Since a total of 1 microgram of radium had been found in the skeleton of a woman who had died from osteogenic sarcoma many years after exposure, it was assumed that a danger level could be set at the amount of uranium equivalent. Uranium is so much less radioactive that 3,800,000 micrograms must be deposited in an adult skeleton to constitute a calculated equal radiation hazard. It has now been established from the massive accumulation of animal toxicity studies that *uranium deposition in bone is no radiological hazard.* The toxic effects of uranium on the kidney would be fatal long before enough uranium was absorbed into the body to give a radiologically hazardous concentration in the bone. Animals inhaling atmospheres containing 2000 micrograms of soluble uranium dusts per cubic meter of air for a period of a year only deposited on the average 6 micrograms of uranium per gram of bone or less than 0.01 of the amount judged capable of inflicting radiological injury. When insoluble uranium dusts are inhaled, there is no chemical hazard to the kidneys; by the same token not enough uranium is absorbed into the blood stream to furnish hazardous amounts for bone deposition. The inhalation of insoluble uranium compounds offers the possibility of building up in the lungs total amounts of radioactive particles sufficient to exceed the calculated tissue tolerance. This point is under active investigation at the present in our laboratory.

## CHARACTERISTIC KIDNEY DAMAGE

Uranium produces a characteristic histological injury in the kidney. This organ is the only site of injury following small or moderate doses of uranium. Uranium poisoning follows a well-defined pattern. There is a lag period of hours to days (depending inversely on the dose) after the administration of the uranium before the damage becomes evident either histologically in the kidney tissues or functionally in the various indices of kidney function (e.g., elevated blood NPN, diuresis or oliguria, etc.).

From the glomerular capillary, diffusible uranium bicarbonate enters the glomerular urine. Passing through the proximal convoluted tubule, bicarbonate is resorbed into the venous blood freeing uranyl ion in the tubular urine. The uranyl ion reacts with the protein membranes of the columnar cells producing injury and—if the dose is sufficient—death of these cells. The cells rupture and the cell contents are discharged into the urine. The severity and extent of uranium poisoning may be followed by examining the excreted urine for proteins and enzymes once in the cell cytoplasm. The most delicate indices of uranium poisoning are: (1) the concentrations of urinary proteins; (2) the concentrations of urinary amino acids (especially the amino acid nitrogen to creatinine ratio), and (3) the catalase activity. From studies of kidney function and renal clearances combined with histological and autoradiographic studies, the site of major uranium injury has been localized in the distal third of the proximal convoluted tubule. If the injury has not been too severe and cells of the tubule lining remain, repair and regeneration are promptly established. Although the regenerated cells are frequently atypical in certain details, recovery is prompt and a few weeks or a month following the administration of uranium an animal may be almost completely normal by every test. There is no chronic form of uranium poisoning. Only repeated bouts of acute injury are found in animals chronically exposed to uranium and, save in cases of overwhelming doses, the histological injury is limited to the kidney. There is some evidence of the development of tolerance to uranium. In the rat, this has been linked to the excretion of citrate in the days immediately following an acute uranium injury.

From studies of large numbers of animals exposed to a variety of soluble uranium compounds for periods of one or two years, it has been conclusively demonstrated that there is no kidney injury when animals breathe an atmosphere containing fifty micrograms of uranium per cubic meter of air. This value has been set as the maximal allowable concentration. If exposures are exclusively to insoluble uranium compounds, somewhat higher dust concentrations can be inhaled without evidence of kidney injury. For insoluble compounds, "it is evident that exposures to concentrations of 50 or 100 micrograms per cubic meter would lie within the negative zone with wide margins of safety."

## BLOCKADE OF CARBOHYDRATE METABOLISM

The mode in which uranium injures or kills cells has been examined in some detail by Rothstein. It was found early that uranium blocks carbohydrate metabolism. Uranium reacts with the cell surface in such a way as to prevent the absorption of glucose. It is ironical that uranium, the source of nuclear energy, should poison living cells by blocking the cells' source of energy. Most of the work on the metabolic effects of uranium has been carried out using yeast cells. Metabolic activity of yeast is blocked with extraordinary promptness by the administration of a very low concentration of uranium ($10^{-6}M$) into the medium. It should be stressed that it has not been proven that the blockade of carbohydrate metabolism is the only toxic effect that uranium can exert on a cell. Or even, that this is the means by which kidney tubule cells are injured and killed. The known uranium effects, however, are limited to the blockade of the absorption of glucose (and to a nearly equal extent of fructose or of galactose). Simpler substances such as alcohol, acetate, pyruvate or lactate are absorbed and metabolized as usual. Furthermore, stored glycogen within the cell can be metabolized without diminution in rate.

Uranium blockades at the surface of the cell. This localization has been established by demonstrating (1) that the blockade is extremely prompt, (2) that the blockade can be instantaneously reversed, (3) that the reversal can be accomplished with concentrations of phosphate in the medium only 1/100th those in the cell cytoplasm, (4) that uranium ($U^{232}$) is removed from the cells when the blockade is reversed, and (5) that the uptake of uranium by cells is a saturation phenomenon involving approximately sixty million surface sites per cell.

The surface sites with which uranium reacts are polyphosphate in nature. When uranium in small amounts is added to a suspension of yeast cells, most of the uranium can be found in the packed cells after centrifugation. If to the original suspension are added various materials known to complex with uranium, e.g., orthophosphate, pyrophosphate, and polyphosphates, these dissolved materials compete with the yeast cell surface sites for the uranium and more of the uranium remains in solution. Certain polyphosphates (hexametaphosphate, for example) appear to be able to compete on nearly equal terms with cell surface sites.

The ability of the cell site to bind uranium increases with increasing *p*H (from *p*H 2.5 to 4.5). Changing the *p*H of the medium in the presence of hexametaphosphate does not disturb the *p*H dependency of the cell binding of uranium. This is in contrast to the alteration by other complexers such as citrate in the *p*H dependency of uranium binding. In other words, whatever changes occurred in the binding power of the yeast cell surface sites by changes in the *p*H have been exactly matched by changes in the uranium binding by hexametaphosphate in solution. These facts indicate that the cell surface-site binding uranium either is a polyphosphate or acts much like one.

## HEXOKINASE INHIBITION

Uranium specifically blocks carbohydrate metabolism apparently by blocking an enzyme system. The evidence can be marshalled as follows:

1. Glucose metabolism is blocked but not by glucose complexing uranium.
2. The polyphosphate nature of the cell surface-binding site fits in the concept since a high energy phosphate source is required in the first step of glucose utilization, the formation of glucose-6-phosphate.
3. Magnesium inhibits and reverses the uranium blockade of carbohydrate metabolism. Magnesium is the preferred co-factor for the first reaction in the utilization of glucose.
4. The great dilution at which uranium is able to act suggests that the reaction is enzymatic.

In addition, data obtained (1) from studies of the kinetics of the inhibited reaction, (2) from the temperature coefficient and from the specificity of the reaction, and (3) from the effects of other ions, for example, hydrogen or potassium, all point not only to the enzymatic nature of this reaction but to the hypothesis that uranium specifically inhibits hexokinase in the surface of the cell. The inhibition can be represented schematically as follows. In the absence of uranium, glucose tends to enter the cell membrane through an appropriate portal assumed to bear a negative charge. Glucose is adsorbed on the surface of the hexokinase, a surface in which there is a characteristic binding of adenosine-triphosphate (ATP) through magnesium to hexokinase. The adsorption permits a phosphate from ATP to be taken by the sixth carbon atom of glucose. Glucose-6-phosphate thus formed bears a negative charge and therefore cannot return to the medium through the negative charged portal. Each glucose-6-$PO_4$ molecule is further acted upon in the cell membrane, ultimately reaching the cytoplasm broken down into fragments no larger than three carbon atoms. In the presence of uranium, the uranyl ion competes with and replaces the magnesium ion so that the surface of the enzyme now becomes ATP bound through uranyl ion to hexokinase. When glucose penetrates and is adsorbed the ATP-uranyl-hexokinase complex will not release a

phosphate to the glucose. Consequently the first step in glucose utilization cannot be taken and the glucose returns unchanged to the medium.

## SUMMARY

More is known about the toxicity of uranium than about the toxicity of any other element. Uranium in the body is highly toxic: providentially, it is hard to absorb into the body. Once absorbed, uranium is carried partly as a protein complex and partly as a bicarbonate complex. In the bone, uranium is deposited principally on the surface of the mineral crystals by exchange with calcium. The kidney is the sole site of the characteristic histological injury. Uranium kills cells by blocking carbohydrate metabolism, apparently inhibiting specifically hexokinase in the cell surface.

Finally a tribute should be paid to the vigilant medical supervision of the Manhattan Project and the Atomic Energy Commission. This supervision is so successful in all their installations that severe uranium poisoning has never occurred and mild uranium poisoning is so rare that it is practically unknown.

# Radiation Dosage to Lungs from Radon and its Daughter Products

**By W. F. Bale and J. V. Shapiro,* USA**

In most programs for the utilization of atomic energy, control of radiation exposure to personnel is based upon a maximum permissible dose equivalent to 0.3 rad per week of combined beta and gamma radiation. It is also usually assumed that alpha radiation is ten times more damaging due to higher specific ionization, and that the maximum permissible dose for this radiation should be 0.3 rad per week. For calculating permissible dosage from internal radiation with beta-emitting isotopes, or alpha-emitting isotopes such as polonium, it is usually assumed that dosage to any organ should not exceed 0.3 rad for beta and gamma emitters, or 0.03 rad per week for alpha emitters. In the operation of chain-reacting piles, in the associated fuel-processing plants, and in the industrial, medical, and research uses of radioactive materials, health-physics techniques have been so well developed that human exposures to radiation are usually negligible or limited to a few per cent of the maximum permissible values.[1]

For two radioactive elements, more direct methods have been used in setting maximum human tolerance levels. Human ingestion of radium in the past, before its danger was fully appreciated, has produced many cases of bone cancer. Many of these cases have been intensively studied. The maximum human-body burden of radium is set below the smallest known cancer-producing dose. A high percentage of deaths among miners in certain areas of the Schneeberg mining region of southern Germany and the adjoining Joachimstahl region of Czechoslovakia has been commonly attributed to radon present in mines. Evans and Goodman,[2] after reviewing radon measurements made in these mines, concluded that the average concentration was about $1 \times 10^{-9}$ curies per liter. On the basis that radon concentration in this range probably was the cause of lung cancer, Evans suggested that the maximum permissible level for radon averaged over working hours ought not to exceed one per cent of this value, $1 \times 10^{-11}$ curies per liter air. This value came to be generally accepted in codes regulating the use of radium in producing luminous indicator and watch dials.

It is however recognized that the level of radon which is cancer-producing is not nearly as reliably established as for radium. Arsenic and other tumor-producing agents also perhaps occur airborne in these mines.[3] Because of the long latent period associated with radiation-induced cancer the relevant exposure period for individual miners probably occurs ten to thirty years before the development of symptoms from cancer. Radon concentrations were not determined at that earlier time, and later values are of uncertain reliability extrapolated to that period since they were made after the possible hazard from radon was realized, perhaps after steps had been taken to reduce mine radon levels. Thus, some uncertainty perhaps exists even regarding the importance of radon as a cause of lung cancer in these miners, and substantial uncertainty as to the level and duration of exposure that must occur to produce harmful effects in human beings.

Investigations conducted by the US Public Health Service have indicated that radon concentration levels in continental United States uranium mines commonly exceeded the $1 \times 10^{-11}$ curies/liter level, sometimes by substantial factors.[4] Mine-ventilation techniques recommended by this agency and put into use have reduced radon levels to the point that no short-term exposure hazard is believed to exist. However, the possibility does exist that cumulative effects for individuals engaged in uranium mining for many years might be harmful. Additional procedures necessary to achieve radon levels substantially less than those obtained by currently recommended practices will add substantially to the cost of uranium fuel for reactors. It has therefore seemed worthwhile to determine experimentally the radiation dose to the human respiratory system by appropriate experiments with animals and with human subjects.

Radon in the atmosphere is accompanied by airborne radioactive daughters, RaA through RaC′ (the active deposit of Rutherford, Curie and Soddy), which often approach radioactive equilibrium. The suggestion by Bale that most of the radiation dosage to the human respiratory system was, in fact, due to the retention in the lungs of these daughters was confirmed for human subjects by experiments informally reported by Harley and by Bale, and for rats by the work of Cohn, Skow, and Gong.[5] No actual respiratory system dosage measurements or estimates based upon experimental data were made. The following experiments reported here are part of a continuing program designed to lead to maximum allowable con-

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centration figures for radon and radon daughters in the air based upon experimental determination of radiation dosage. The experimental data are in part taken from a thesis in which the experimental procedures used are fully discussed.[6]

## EXPERIMENTAL PROCEDURES AND RESULTS

### Dependence of Radon Daughter-Product Level upon Dust Load and Air Circulation in Chamber

All experiments were performed in a rectangular exposure chamber of 1800 liters capacity. The radon concentration in most experiments was about $2.5 \times 10^{-8}$ curies per liter. Radon was measured by introducing it into an ionization chamber, or with a well-type scintillation counter after quantitative absorption on charcoal. Daughter-product concentrations in air were sampled using millipore filters or tubes packed with Fiberglas, and radioactivity determinations made by alpha measurements with a sulfide-screen scintillation counter or by gamma measurements with a well-type counter. Radioactivity measurements on biological samples were made with the well-type scintillation counter or, for larger samples, with a Texaco well-type Geiger-Müller counter.

When the air in the chamber was supplied as outdoor air with its normal dust load, airborne daughter products, through RaC', reached 70 to 90 per cent equilibrium shortly after radon was placed in the chamber. This level depended very little on whether or not the air was stirred with a fan.

When the dust level in the chamber was made very low by careful cleaning, the content of airborne daughters fell to a very low value compared with the equilibrium value: 20 per cent of equilibrium in quiet air, 2 per cent when the air was stirred. Experiments showed that radon daughters not trapped on dust particles had a much greater motility and were more rapidly deposited on chamber walls.

### Determination of Biological Half-Life of Radon Daughters in the Lungs of the Rat

Groups of four rats were exposed in the chamber to radon and radon daughters in air with a normal dust load, and in other experiments to radon and radon daughters in carefully cleaned air. Pairs of rats were sacrified immediately after a few minutes' exposure, and other pairs 80 to 140 minutes later. The lungs were removed and gamma activity measured. In each instance the bilological half-life was two hours or greater, a long time compared with the effective radioactive half-life of about 35 minutes.

### Measurement of Lung and Tracheal Dosage in Rats

Rats were placed in the exposure chamber at least 2.5 hours, removed, immediately sacrificed, and the radioactivity of lungs and tracheae determined by gamma measurements with appropriate calibration experiments and decay corrections. Different groups of animals were exposed to air with a normal dust load, to cleaned air, to cleaned air with daughter products kept at low levels by fanning, and to cleaned air to which were added artificially produced nuclei. These nuclei were produced by commutator sparking of a defective motor inserted in the chamber and operated for a few minutes 18 hours before the animal exposure. In the air with normal dust load, the radon daughters were attached to dust particles with a diffusion coefficient of about $4.8 \times 10^{-6}$ cm$^2$/sec; for the daughters on spark nuclei this diffusion coefficient was about $9 \times 10^{-7}$. Table I shows the results of these experiments. Radioactivity to trachea epithelium was calculated on the assumption that alpha energy was dissipated uniformly in the inner surface of the trachea to a depth of 66 microns. The actual chamber radon concentration, mentioned earlier, was about $2.5 \times 10^{-8}$ c/l.

### Experiments with Dogs

Experiments carried out on dogs had, for an additional purpose, the measurement of the distribution of ventilation and perfusion in the various lobes of the dog lung. These experiments were done in collaboration with Rahn, Sadoul, and Farhi.[7] Four dogs were exposed to radon and radon daughters contained in outdoor air introduced with its normal dust load into the chamber a few hours before the experiment. For nine additional dogs the radon daughters were on nuclei generated by a sparking motor. The animals were anesthetized with sodium pentobarbital in all experiments. The dogs were kept outside the chamber, and breathed through tracheal cannulae. Minute volume per kilogram of the first three dogs averaged 250 cm$^3$. Experimental results are shown in Table II. In this instance, also, dosage to the trachea epithelium was calculated from the measured radioactivity on the assumption that alpha energy was dissipated uniformly on the first 66 microns of the tracheal lining. The tracheae, about 1 cm internal diameter, approximated in size the larger human bronchi. One notes that trachea epithelial dosage is about 8 times the average dose to the lungs as a whole.

### Radon-Daughter Retention in Man

The fraction of the inhaled radon-daughter products retained by two human subjects was measured for chamber air with a normal dust load, and by one subject for air largely cleaned of particulate matter before the radon was introduced. The general principle of the experiment was, first, to sample radon-daughter radioactivity in the air to be breathed by the subject, second, with the subject breathing chamber air to collect and measure the radon-daughter activity of the air exhaled by the subject, and, finally, again sample the air breathed by the subject. Reduced pressure behind the millipore paper used for removing degradation products was so regulated that the subject could breathe under essentially physiological conditions.

The results are given in Table III. The average retention of the daughter products in air containing normal atmospheric dust was 25 per cent. The average retention for cleaned air was 75 per cent. A

Table I. Average Radiation Doses from Alpha Radiation to Lungs and Tracheae of Rats Breathing Radon plus Radon Degradation Products at Radon Concentration of $1 \times 10^{-11}$ curies/cm$^3$

| No. of exper. animals | Average wt (grams) | Measured average dosage to lungs (millirad/hr) | Average deviation | Measured average dose to tracheal epithelium | Average deviation |
|---|---|---|---|---|---|
| | | **Normal outdoor air** | | | |
| 9 | 190 | 0.22 | 0.07 | 0.33 | 0.04 |
| 2 | 184 | 0.36 | 0.07 | | |
| | | **Electric spark nuclei** | | | |
| 2 | 186 | 0.25 | 0.02 | 0.11 | 0.02 |
| | | **Stagnant cleaned air** | | | |
| 5 | 197 | 0.045 | 0.010 | | |
| 5 | 195 | 0.047 | 0.018 | 0.30 | 0.11 |
| | | **Stirred cleaned air** | | | |
| 3 | 184 | 0.0015 | 0.0003 | Too low to measure | |
| | | **Maximum estimated additional dose due to radon gas undergoing radioactive decay in lungs** | | | |
| All above animals | | 0.004 | | 0.003 | |

neoprene balloon, used as an artificial lung in control experiments, gave essentially zero retention for the air containing normal atmospheric dust.

## DISCUSSION

For the experimental data given in the previous sections, and assuming the long biological half-life found for rats also holds for man, one can calculate average radiation dosage to the human respiratory system under those exposure conditions for which the data were obtained.

For radon daughters produced in normal dusty air the following assumptions seem reasonable:

1. $V_m$ = minute volume - 20 liters/minute.
2. Airborne RaA through RaC′ in equilibrium.
3. $C_d$ = concentration of each daughter—$1 \times 10^{-11}$ curies/liter ($1 \times 10^{-8}$ $\mu$c/cm$^3$).
4. $R$ = fractional retention = 0.25.
5. $W_e$ = weight of lungs—1000 gm.
6. All radioactivity retention occurs in lungs.
7. $E_\alpha$ = the energy in Mev due to $\alpha$ radiation from complete radioactive decay of RaA, RaB, RaC, and RaC′, each at an initial activity of 1 $\mu\mu$curie, where disintegration constants are given in units of min$^{-1}$.

$$2.2 \times \frac{6.00}{A} + 7.68\left(\frac{1}{A} + \frac{1}{B} + \frac{1}{C} + \frac{1}{C'}\right) = 1.277 \times 10^{-3}$$

$D_c$, the average $\alpha$-radiation dose to the lungs in millirad units per hour will then be:

$$\frac{C_d \times 10^{12} \times V_m \times R \times 60 \times E_\alpha \times 1.6 \times 10^{-6} \times 10^3}{W_e \times 100} = 0.061$$

where the numerical units in order in the numerator are the conversion factor from curies to $\mu\mu$curies, from hours to minutes, from Mev to ergs, from rad to millirad, and in the denominator from rad units to ergs/gram.

Table II. Average Radiation Doses to the Lungs and Tracheae of Cannulated Dogs Breathing Air at a Level of $1 \times 10^{-11}$ Curies/Liter Radon in Substantial Equilibrium with Its Daughters through RaC′

| No. of dogs | Average millirad/hr to lung | Average deviation | Average millirad/hr to tracheae | Average deviation |
|---|---|---|---|---|
| | **Normal outdoor air** | | | |
| 4 | 0.038 | 0.005 | 0.35 | 0.10 |
| | **Electric spark nuclei** | | | |
| 9 | 0.024 | 0.006 | 0.62 | 0.13 |

The average lung dosage for a 40-hour work week under these conditions would be $40 \times 0.061 = 2.44$ millirad. Assumption of 10 liters per minute ventilation rate gives the 1.2 millirad value quoted in the abstract.

It follows that the level of radon daughters in equilibrium with radon that would give a dose of 0.03 rad per week alpha radiation averaged over the lung would be $12.3 \times 10^{11}$ c/l. The data presented in Table II suggest alpha-radiation dosage to the epithelium lining the bronchi may be substantially higher than to the lung as a whole. Most lung cancer seems to arise from bronchial epithelium. A reduction of the above level by a factor of 5 will largely take into account this non-uniform distribution of radiation dosage. Auxiliary experiments indicate the amount of bronchial deposition is not strongly dependent upon ventilation rate. The present data confirm suggestions that the primary radiation hazard to the lungs is from the radon daughters rather than radon itself. These data suggest a level of airborne RaA through RaC′ in amounts equivalent to equilibrium with $2.5 \times 10^{-11}$ curies per liter radon ($2.5 \times 10^{-8}$ $\mu$c/cm$^3$) as a maximum permissible limit for human exposure averaged over a forty-hour work week. These conclusions are based on the assumption that alpha-radiation dosage to human bronchi should not greatly exceed 0.03 rad per week.

**Table III. Results of Human Retention Experiments**

| *Subject* | *Tidal volume* | *Per cent retention* |
|---|---|---|
| | **Uncleaned outdoor air** | |
| 1 | 1.3 | 27 |
| 1 | 1.1 | 23 |
| 1 | 1.1 | 25 |
| 2 | 1.0 | 25 |
| 2 | 0.72 | 22 |
| 2 | 1.5 | 30 |
| Artificial lung | | 0 |
| | **Cleaned air** | |
| 1 | 1.5 | 72 |
| 1 | 1.5 | 78 |

Radon degradation product concentrations can be measured by determinations of the $\alpha$, $\beta$, or $\gamma$ activity of molecular filters through which known volumes of mine air have been drawn. Methods for making these measurements, including the necessary radioactive decay corrections, have been previously discussed by Holaday and his colleagues,[4] by Shapiro,[6] by Harley,[8] and will be more explicitly described in a US Public Health Service bulletin now in preparation.[9]

Experimental studies have shown that uranium miners store radiolead derived from radon degradation products, and that polonium derived from this radiolead is excreted in measurable quantities in the urine.[10] Research in progress is directed at determining the usefulness of urine polonium determinations as a measure of integrated polonium exposure in man.

Surveys by the US Public Health Service indicate that past exposures of miners to radon and radon daughters in the Colorado Plateau area, even in non-uranium mines, have often exceeded this permissible concentration figure suggested above. Measurements of radiolead in bone obtained at autopsies of lung cancer victims are being made as a part of a cooperative program with the US Public Health Service, the University of Utah Medical School, and Dr. Geno Saccomanno of Grand Junction, Colorado. This program is aimed at establishing whether a correlation exists between these radon exposures and liability to develop lung cancer.

## SUMMARY

Experimental studies suggest that the primary hazard from breathing radon-containing air is from the accompanying airborne daughters, RaA through RaC′, of which about 25 per cent are normally retained in the human respiratory system.

On the basis that dosage from alpha radiation to the bronchial epithelium should not exceed 0.03 rad per week, a level of radon daughters RaA through RaC′, in amounts equivalent to equilibrium with $2.5 \times 10^{-11}$ curies per liter radon ($2.5 \times 10^{-8}$ $\mu c/cm^3$) is suggested as a maximum permissible level for human exposure averaged over a forty-hour work week.

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# Use of Radioisotopes in Studies of Health Hazards in the Uranium Mining Industry

**By E. C. Tsivoglou,* USA**

This paper describes the use of radioisotopes in a a series of experiments designed to evaluate the health hazards resulting from inhalation of atmospheric mixtures of radon and its daughters, and to test theoretical expressions which were developed to serve as the basis for control of these hazards in the uranium mining industry. In the laboratory, radioisotopes in both purified and natural form were used according to the purposes of the individual experiments, the latter in experiments designed to represent natural environmental conditions insofar as possible. In the field studies radon and its daughters were used in their natural state, i.e., in a uranium mine. Within reasonable limitations the experiments provide a sound basis for the practical control of the inhalation hazard in this important industry.

## RADIOACTIVE EQUILIBRIUM AND THE RADIATION DOSE

The radiation dose delivered to the lungs as the result of inhalation of radon alone is negligible compared to the dose resulting from inhalation of the same amount of radon together with all of its daughters.[1] Radon is a gas having a 3.8 day half-life, while its daughters are not gaseous and their half-lives are much shorter (see Table I). A substantial fraction of the inhaled gas is expelled in breathing before it can decay. On the other hand, a relatively large fraction of the inhaled daughter elements is retained in the lungs and decays there. Since the relative biological effectiveness of beta particles is small as compared to alpha particles, those daughters which decay by beta emission contribute only a small part of the total radiation dose. As a result, our main concern is with the alpha emissions from RaA and RaC'.

**Table I. Properties of Radon and Its Daughters**

| *Isotope* | *Emission* | *Energy, Mev* | *Half-life* |
|---|---|---|---|
| Rn ($Rn^{222}$) | Alpha | 5.49 | 3.8 days |
| RaA ($Po^{218}$) | Alpha | 6.00 | 3.1 min |
| RaB ($Pb^{214}$) | Beta | 0.65 | 26.8 min |
| RaC ($Bi^{214}$) | Beta | 3.15 | 19.7 min |
| RaC' ($Po^{214}$) | Alpha | 7.68 | $2.5 \times 10^{-6}$ min |

Atmospheric radon concentrations as high as $10^{-5}$ microcuries† per cubic centimeter are not rare in uranium mines, and concentrations of $10^{-7}$ microcuries per cubic centimeter are found in metal (non-uranium) mines of the United States.[2] A maximum permissible concentration of $10^{-8}$ microcuries per cubic centimeter has been suggested as safe for continuous exposure.[3] This suggested standard is based on the assumption that all of the radon daughters are present in the atmosphere in concentrations reflecting radioactive equilibrium with the parent gas.†

Estimates of the radiation dose resulting from continuous inhalation of a mixture of radon and its daughters can be made by use of a formula developed by Morgan:[1]

$$W = CQ \Sigma (bE) f_e T \qquad (1)$$

where $W$ is the radiation dose in rem (roentgen equivalent man) per week from a specific daughter; $C$ is a constant; $Q$ is the atmospheric concentration of radon in microcuries per cubic centimeter; $\Sigma (bE)$ is the total energy in Mev of the alpha particles which will be emitted in the lungs (13.68 Mev for RaA and 7.68 Mev for RaB and RaC); $T$ is the half-life of the daughter in minutes, and $f_e$ is the fraction of equilibrium with radon represented by the specific daughter concentration. Separate calculations are made for each of the daughters and these individual doses are then added together to obtain the estimate of total dosage. It is noted that the radiation dose for each daughter is directly proportional to its particular equilibrium fraction, $f_e$.

A suitable technique for measuring the individual atmospheric daughter concentrations was not available until recently. Earlier estimates of the inhalation hazard therefore involved the conservative assumption that the daughters occurred in equilibrium concentrations.[4] Based on this assumption, the total hazard was estimated on the basis of a measurement of the atmospheric radon concentration alone.

Theoretical considerations indicated[5] that ventilation of a uranium mine would cause wide departures from the equilibrium situation, with resulting small values for the factor $f_e$. If true, the actual hazard in a ventilated mine would be significantly less than the hazard estimated from observed reductions in the atmospheric radon concentration and calculations as-

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† If in an atmosphere radon and its daughters occur in equilibrium concentrations, then the activity of any one of the daughters (measured as microcuries per cubic centimeter) is equal to the activity of the radon present.

suming equilibrium daughter concentrations. In order to provide for sound control of the inhalation hazard in the uranium mining industry, it was necessary to evaluate the actual hazard in a ventilated mine in terms of measured atmospheric daughter concentrations.

## EVALUATION OF THE INHALATION HAZARD

The first experiments were designed to develop a method for measuring the individual atmospheric concentrations of the daughter elements. In brief, the method involved mathematical analysis of the observed alpha decay curve from a filter paper sample of the atmosphere. Depending on the relative atmospheric daughter concentrations, such decay curves are significantly different in shape during the first 45 minutes after the completion of sampling. They become quite similar after about an hour of decay (see Fig. 1).

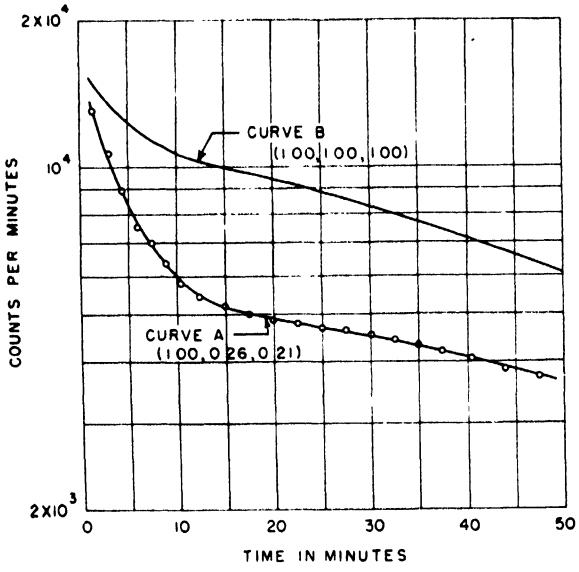



**Figure 1. Observed alpha decay curve for sample number 5**

Any specific set of atmospheric daughter concentrations results in only one possible set of initial numbers for decay on the filter paper after sampling is completed. This in turn implies only one possible observed alpha decay curve. A general equation for the family of all possible alpha decay curves was derived on the basis of the known constituents and decay constants. It was then possible to use alpha activities observed during the first 45 minutes after sampling in order to solve the general equation for the initial numbers of the daughter elements present on the filter paper at the end of the sampling period. The individual atmospheric daughter concentrations were then obtained from these initial numbers. This method is described elsewhere in detail.[6]

The laboratory source of radon and its daughters was a closed box of volume 145 liters containing a quantity of rich uranium ore. The box was equipped with stoppered holes to permit the introduction of sampling equipment and the return of the exhaust air from the sampling device. Atmospheric samples were collected by drawing a specific amount of air from the radon box through a millipore filter paper of one-inch diameter. A hand-cranked pump was used for this purpose and operated for 5 minutes at a rate of about 17 liters per minute. During this sampling period the exhaust air from the pump was returned to the radon box. At the end of the sampling period, the filter paper was immediately removed from its holder and placed in a methane flow proportional counter. An alpha decay curve was then developed over a period of about an hour. The gas radon is of course not retained on filter papers, and the foregoing method yields only the atmospheric concentrations of the daughters. In the laboratory experiments the volume of air sampled was large in relation to the volume of the radon box. Therefore the laboratory data is significant only as an example and test of the methods. This is of course not so in the case of subsequent field samples, which were very small in relation to the volume of air in the uranium mine section sampled. The proportional counter was calibrated and periodically tested by the use of a purified standard source of known activity and half-life.

Figure 1 illustrates the type of data obtained. Curve *A* is an observed alpha decay curve. As shown, calculations based on the observed activities indicated that, if RaA is considered as the parent isotope, then RaB and RaC were present in atmospheric concentrations representing only 26 per cent and 21 per cent, respectively, of their possible equilibrium concentrations.‡ Curve *B* was calculated to illustrate the course of alpha decay which would have been observed had RaB and RaC been present in the atmosphere in equilibrium concentrations.

The foregoing experiment was repeated a number of times in the laboratory in order to perfect the technique for field use. It was desirable, however, to carry out a second set of experiments as an independent test of the reliability of the decay analysis method, as it was intended that this method would be used in basic studies of radiation dosage and control of hazards in the uranium mines. Accordingly, the following laboratory experiment was devised and carried out to serve this purpose.

It was desired to determine the individual alpha activities of RaA and RaC′ from a single atmospheric sample. From this information the relative concentrations of RaA, RaB and RaC could be derived and compared with the same data obtained by the alpha decay method from a second sample taken simultaneously. In view of the markedly different energies possessed by the alpha particles emitted during decay of

‡ As radon is not collected on the filter paper, it is necessary here to consider RaA as the parent member of the chain so as to express the daughter concentrations as equilibrium fractions. In the field studies radon concentrations were determined independently, and all of the daughter concentrations could be expressed in relation to radon as the parent element.

RaA and RaC′ (6.00 and 7.68 Mev, respectively), and because of the relatively short half-life of RaA, alpha pulse-amplitude analysis of atmospheric samples appeared to be the most practical and direct means of obtaining the desired information.

Atmospheric samples of the daughters of radon were collected from the previously described radon box on one-inch diameter copper planchets by use of an electrostatic precipitator of recent design.[7] By this means it was possible to collect a very thin sample (usually not visible to the eye), thereby minimizing the effects of self-absorption. At the completion of the five minute sampling period one planchet was counted as usual by a fellow worker in the proportional counter and its alpha decay curve developed.

A second planchet was at once taken from the sampler and placed in a scintillation counter. This planchet was separated by a thin collimator from a thallium-activated sodium iodide crystal. The scintillations produced in the crystal by the sample were amplified by means of a photomultiplier tube and transmitted to an oscilloscope. The oscilloscope controls were set to permit deflections in the Y-direction only. The resulting pulse amplitudes were photographed from the screen of the oscilloscope by means of a continuous motion camera. The shutter of the camera remained open during the entire film run. This made it possible to record all events which occurred during the run. A cross-sectioned grid was used in front of the screen of the oscilloscope. This permitted photographing a background scale in conjunction with the recording of the pulse amplitudes, and made measurement of the pulse amplitudes a simple matter. The exposed film strips were developed, projected on a large screen, and the amplitude of each pulse was recorded. Finally, the distribution of pulse amplitudes was plotted for the film run.

Alpha particles emitted by radium (4.79 Mev) were used to calibrate the equipment. For this purpose the radium sample was prepared by evaporation of a few drops of a radium salt solution on a standard copper planchet.

Figure 2 is an enlarged photograph of a typical section of exposed film. The four longest pulses are due to the alpha emissions of RaC′, and the group of pulses next in length reflects decay of RaA by alpha emission. The average lengths of these two groups of pulses are in near-direct proportion to the known energies of the alpha particles emitted by RaA and RaC′. The two short pulses are due to high-energy beta emissions from RaC, lower energy events having been eliminated from the film record by discrimination in the oscilloscope.

Figures 3 and 4 are typical pulse-amplitude distributions obtained from a single sample. As can be seen, satisfactory resolution of the RaA and RaC′ peaks was obtained. The RaA activities observed at the two decay times (2.13 and 9.88 minutes after completion of sampling) indicate a half-life for RaA of 3.12 minutes, in good agreement with the known

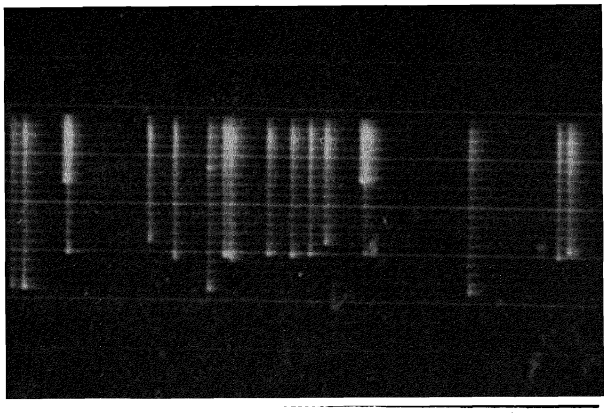


Figure 2. Typical pulse amplitude record

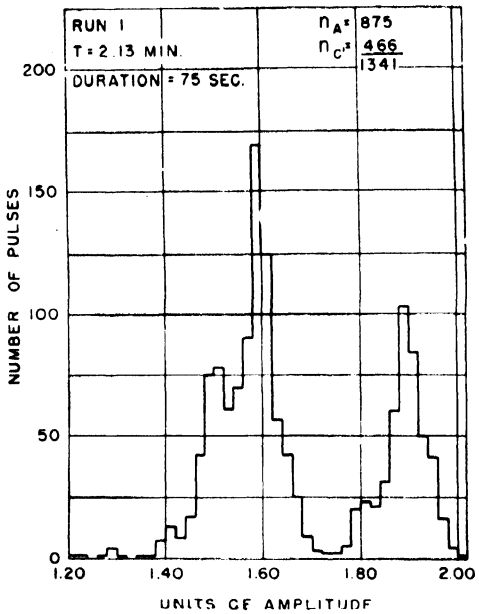



Figure 3. Pulse amplitude distributions, sample number 5

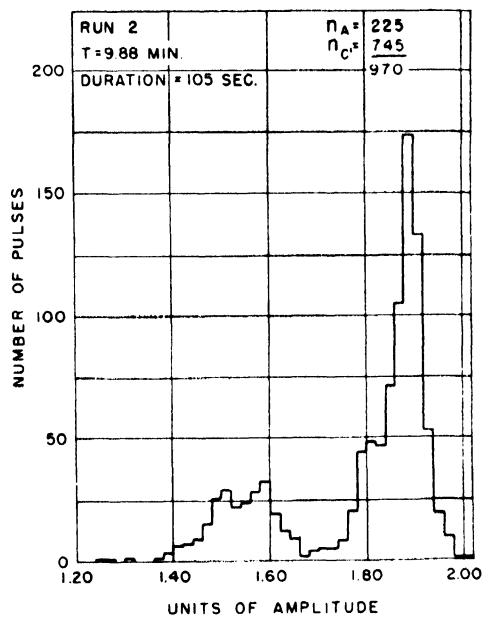



Figure 4. Pulse amplitude distributions, sample number 5

value. The relative concentrations of RaA, RaB and RaC were calculated from the pulse-amplitude distribution data and compared to relative concentrations derived from the simultaneously obtained decay curve sample.

The foregoing experiment was repeated a number of times. In all cases agreement between equilibrium fractions obtained by both methods was quite good. Having confirmed the reliability of the alpha decay analysis method, plans were made for field studies in the uranium mines.

## CONTROL OF THE INHALATION HAZARD

### Ventilation

Before ventilation could be advised as a general measure for practical control of the inhalation hazard in uranium mines it was necessary to develop a satisfactory means of estimating required rates of air turnover and the extent of beneficial effects. Theoretical expressions were developed for this purpose and field experiments were carried out to test the validity of the theory. Details of the theory and field experiments have been reported elsewhere.[5]

The atmospheric radon concentration in a particular section, or drift, of a ventilated uranium mine is dependent upon the following major processes: radon is liberated as a gas from the surrounding ore body at an essentially steady rate, it decays according to its known half-life, and some of it is removed from the mine section by the outgoing air stream. The daughter elements are formed in the air through decay of the atmospheric radon. They decay according to their respective half-lives and are also removed by the outgoing stream of air. A system of differential equations was developed to describe the operation of these factors; its solutions expressed the atmospheric concentrations of radon and each of its daughters as functions of the rate and duration of ventilation. It was then possible to predict the effects of ventilation as a control measure and therefore to estimate the amount of ventilation needed in a specific case to reduce the radiation dosage to any desired level. The theory indicated that ventilation would cause wide departures from the equilibrium condition. It further showed that the working section of a mine should be ventilated for a period of about an hour before personnel enter the mine to begin work.

A set of experiments was carried out in a uranium mine in the western United States to test the reliability of the foregoing theory. A section of the mine which was not at the moment being worked was selected for purposes of this study. Outside air was blown into the mine through several hundred feet of flexible tubing by means of a large blower mounted on a truck located just outside the mine entrance. The diameter of the tubing was one foot. The section was ventilated at a number of different rates by means of a control at the blower. Five-minute filter paper samples of the air in the ventilated section were collected at desired intervals, and were carried out of the mine for analysis by the alpha decay method. Alpha decay curves were developed by use of a methane flow proportional counter mounted in a vehicle near the mine entrance. This counter took its necessary power from a gasoline-operated portable generator, since electric power was not available at the mine.

Separate volumetric samples were taken simultaneously with collection of the filter paper samples, and were transmitted to the main laboratory for determination of the atmospheric radon concentration. The daughter concentrations were determined in the field by analysis of the observed alpha decay curves from the filter paper samples.

There was good agreement between predicted and observed atmospheric concentrations of radon and its daughters at all rates of ventilation studied, thus providing good support of the theoretical developments. At the highest rate of ventilation used (one air change about every 3 minutes) the atmospheric radon concentration was reduced from the unventilated condition by a factor of 200; the concentrations of RaA, RaB and RaC were reduced by factors of 1000, 2500, and 2500, respectively. Ventilation was therefore shown to be a very effective means of controlling the inhalation hazard in a uranium mine.

For the highest rate of ventilation studied, the observed atmospheric concentrations of radon, RaA, RaB and RaC were 58, 12, 4 and 2 micromicrocuries per liter, respectively. Using Equation 1 for calculations of radiation dosage, it is easily shown that the actual radiation dose calculated on the basis of these concentrations is only 7 per cent of the dose calculated on the basis of the assumption that all of the daughters are present at their equilibrium concentrations (58 micromicrocuries per liter). In another sense, the actual radiation dose in this observed case is less than half of the dose calculated on the basis of the suggested maximum permissible concentration of radon (10 micromicrocuries per liter) and the assumption of equilibrium daughter concentrations. It therefore appears clear that control of the inhalation hazard in the uranium mining industry should be based upon observed daughter activities, rather than upon observed radon concentrations.

### Air Cleaning

In extensive deep uranium mines ventilation with outside air may prove to be very expensive. A less costly means of reducing atmospheric daughter concentrations to acceptable levels is most desirable. Accordingly, other workers at the Salt Lake City, Utah, Field Office of the Public Health Service Division of Special Health Services have begun investigating the use of air cleaners as a means of control of the inhalation hazard.[8] Instead of reducing atmospheric daughter concentrations by dilution with outside air, the mine air is continuously recirculated through the air-cleaning device, which removes some

of the daughter elements during each passage of the air through the cleaner.

The gas radon is not removed by air cleaning. Its atmospheric concentration is at a constant level which depends on its decay rate and on the rate at which it is liberated from the ore body. RaA is therefore produced in the mine air at a constant rate by decay or radon; it decays as usual, and in addition is removed by the air cleaner. The subsequent daughters are produced by decay of their respective parent isotopes, decay according to their specific half-lives, and are also removed by the air cleaner. If one postulates an air cleaner whose efficiency is approximately constant, then atmospheric daughter concentrations can be predicted by the same equations as those which describe the effects of ventilation. The only difference between the two cases involves the physical interpretation of the constant which describes the removal process.

Preliminary laboratory experiments by Holaday's group[8] indicate that removals of the daughters by air cleaning are approximately those which are predicted by the theory. As in previous laboratory work, the source of radon and its daughters was a quantity of rich uranium ore.

It is planned to undertake field studies of the reliability and feasibility of the use of air cleaners in the near future. Since air cleaning does not remove radon, the hazard due to inhalation of the gas itself will be of greater importance once the daughter concentrations have been reduced to acceptable levels by air cleaning. It may be that in some situations a combination of air cleaning and ventilation will offer the most practical and effective means of hazard control.

## SUMMARY

Measurement and control of the inhalation hazard in the uranium mining industry were studied in a series of experiments using both natural and purified radioisotopes. A method was developed for measuring the individual atmospheric concentrations of the daughters of radon. This method was verified by an independent experiment in which the atmospheric concentrations of the alpha-emitting daughters were determined by alpha pulse-amplitude techniques.

Experiments in a uranium mine showed that ventilation is a very effective means of reducing atmospheric concentrations of radon and its daughters to acceptable levels. It was further shown that ventilation caused the daughter concentrations to be reduced well below values corresponding to radioactive equilibrium with the observed radon concentration. The experiments also indicated the desirability of preliminary ventilation for about an hour before workers enter the mine. The observed results were in good agreement with predictions on the basis of theoretical considerations. The experiments indicate that estimates of radiation dosage and control of the inhalation hazard in uranium mines should logically be based upon observed daughter activities and not solely upon radon measurements.

## ACKNOWLEDGEMENTS

The work referred to in this paper was performed while the author was assigned to the Salt Lake City Field Station of the Occupational Health Program, Division of Special Health Services. As noted in the References, Messrs. H. E. Ayer and D. A. Holaday of that office participated in certain phases of the work. Their assistance is gratefully acknowledged.

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# Health Protection of Uranium Miners and Millers

By S. E. Miller, D. A. Holaday and H. N. Doyle,* USA

Between 1881 and 1887, ores, later found to be radioactive, were discovered on the Colorado Plateau in the Rocky Mountain area of the United States. These were carnotite ores which contained vanadium and uranium, with a small quantity of radium. Several mines were developed during this period, but production was small since there was little or no demand for these materials.

In 1912, the US Bureau of Mines surveyed the carnotite deposits of the Colorado Plateau and reported them as a practical source of uranium ore. The Bureau of Mines also made a study of methods for recovering radium, vanadium, and uranium from these ores and operated a pilot plant to demonstrate methods of production.

The development of atomic energy focused attention on the carnotite deposits of Colorado and Utah. Beginning about 1946, the discovery of many new ore bodies in the Colorado Plateau caused the industry to mushroom in this area. In August 1949, when the industry had reached a sizeable production rate in the State of Colorado, the Colorado Department of Health appointed an advisory board to advise the State Division of Industrial Hygiene on the potential hazards associated with the mining and milling of uranium ores. Among the conclusions drawn at the first meeting of this group were that little or nothing was known of the health hazards of the uranium-producing industry, and that a medical reconnaissance survey should be made by a physician from the Occupational Health Program of the US Public Health Service.

Such a reconnaissance survey was made shortly after this meeting. On August 25, 1949, a second meeting of the advisory group was called at which was also represented the management of the larger companies mining and producing uranium in the State of Colorado. The group concluded that, in view of the dearth of available information on the health hazards associated with this industry, the Occupational Health Program of the US Public Health Service should be requested to conduct a study of the uranium mines and mills. Accordingly, on August 30, 1949, a formal request for such a study was made to the Surgeon General by the Colorado Department of Health, the Colorado Bureau of Mines, the Colorado Industrial Commission, and several companies engaged in uranium mining and production. Shortly thereafter, the Occupational Health Program, US Public Health Service, replied to these agencies, agreeing to undertake the study.

While these negotiations were underway, the Occupational Health Field Station of the Public Health Service conducted a very limited study of the mining problems in the mines located on the Navajo Indian Reservation. The preliminary information obtained in this brief study indicated that the miners were exposed to external radiation, radon gas, and a dust with a high silica content.

Shortly thereafter, a preliminary survey was made of a mill producing uranium oxide and vanadic acid. The findings revealed that the mill workers were exposed to uranium- and vanadium-containing dust and to fume and dust of the isolated uranium and vanadium oxides. It was determined that radon was of little significance in the mills because of the large area available for dilution of the gas; that external radiation was not an apparent problem; but that it would be necessary to consider the internal radiation hazard residing in the air-borne dust from the processing of the ore.

Information available in the medical literature of this country indicates there was an attack rate of about 1 per cent per year of lung carcinoma among the miners working in Joachimsthal and Schneeberg mines. It is also reported that 30 to 50 per cent of all the deaths of the workers in these mines were due to a primary cancer of the respiratory system. This information was the only material available which indicated the health hazards associated with uranium mining.[1] It must be pointed out, however, that this disease usually has developed only after an average exposure of seventeen years. Moreover, in any attempt to use these findings as a guide, cognizance must be taken of the fact that, in contrast to European practices, American operations are more intermittent. Generally only one shift is employed, and the mines are not worked on a round-the-clock basis. Consequently, workers are not exposed immediately following the blasting, when dust and radon concentrations are generally highest.

The medical and clinical laboratory examinations of workers in the uranium mining and milling industry in the Colorado Plateau were begun in the summer of 1950. At the present time, over 1300 men have undergone physical examinations with emphasis upon occupational history, chest roentgenograms, urinalyses, and blood studies, which include erythro-

* US Public Health Service, Washington, D. C.

cyte counts, total and differential leukocyte counts, hemoglobin estimations, and hematocrit values. In addition, a selected group of approximately 200 miners and millers was examined for atypical blood forms by using a peroxidase-staining technique on blood smears.

At this time, no clear-cut etiologic or pathologic patterns have been uncovered among the workers examined. Since the majority of the workers had been exposed for a period less than three years, this observation is not entirely surprising. It does, however, point to the need for repeating the medical studies at frequent intervals. At the present time, therefore, it is planned to re-examine these workers periodically using the present medical findings as a baseline.

Moreover, it is felt that a great deal of valuable information could be obtained by an epidemiologic study of persons who were employed in the uranium mines and mills before 1950. At this time, we have not had the opportunity to examine a sufficient number of former uranium miners to determine whether or not there has been an excess of lung cancer among them.

As a note on biologic effects reference is made to work which proposes that urinary polonium measurements may serve as a rough measure of cumulative lung exposure to radon and its daughters since the daughters decay to $Pb^{210}$, a lead isotope with a long effective half-life in the body and a decay scheme[2] producing $Po^{210}$. In a comparison of the urine findings of 19 uranium miners of the Colorado Plateau with those of unexposed laboratory personnel it was found that the miners chronically exposed to high radon concentrations had amounts of $Po^{210}$ varying from 2 to 38 micromicrocuries per liter while no polonium was found in the urine of the laboratory group. Thus, it would appear that the miners had inhaled and retained radon and its degradation products.

## OCCURRENCE IN URANIUM MINES OF RADON AND THE DAUGHTER PRODUCTS OF RADON

Uranium ores contain, in addition to uranium, all of the other members of the radioactive series of which it is the parent. The elements of this family which are considered to be potentially most hazardous to uranium miners are: radon, $Po^{218}$(RaA), $Pb^{218}$(RaB), $Bi^{218}$(RaC), and $Po^{214}$(RaC').

Radon enters mine atmospheres by diffusion from the ore bodies, or by being carried into the mines by ground water. Radon is slightly soluble in water and can therefore travel long distances underground to be released when the pressure on the water is lowered. The amount of radon that will be present in any part of a mine is determined by the rate of emanation from the ore body into the mine, the rate of removal by ventilation, and radioactive decay of the gas. The immediate daughters of radon all have short half-lives and thus will rapidly tend to reach equilibrium in an atmosphere containing radon even though freshly emanated radon is free of these decay products. However, it requires about three hours for equilibrium to be reached in this series, and therefore the atmospheric concentrations of these elements are readily affected by ventilation. Under these conditions, it is rare to find equilibrium amounts of radon and its daughters present in mines.

During the study of the uranium mines, an effort was made to obtain atmospheric samples for radon and for RaA and RaC' in all the operating mines in the Colorado Plateau. This developed into a major project since many of the smaller mines are worked only sporadically. In the absence of central records that would show which mines were operating at any particular time, it was necessary to find them by local inquiry and any other available means. However, during the summer of 1952, samples were obtained in 147 mines located in Colorado, Utah, Arizona, and New Mexico. While not all existing uranium mines were reached, it was believed that those that were surveyed employed a large percentage of the workers and that the concentrations of radioactive gas and dust found in them were representative of those occurring throughout the industry.

The results for the atmospheric samples are given in Tables I and II. The data have been consolidated and are presented by mining districts, showing the median, maximum, and minimum concentrations that were found in the mines in each district. Table III shows the number of mines and miners that fall into each of several ranges of atmospheric concentrations of radon daughter products.

Tables I and II show that the atmospheric concentrations of radon and its degradation products in the mines varied over very wide ranges. The median values for each mining district are more representative of the situation existing at the time of survey and are given to assist in interpreting the data. Table III, which gives the distribution of mines by ranges of radon daughter concentrations, shows what conditions can be anticipated in uranium mines in which there are no planned measures to control the amounts of radioactive elements in the atmospheres. If 100 micromicrocuries per liter of RaA plus RaC' (equivalent in alpha activity to 50 micromicrocuries per liter of radon at equilibrium with its daughters) is taken as a tentative working level, 84.2 per cent of the miners were employed in areas showing concentrations above this figure. Thus, it was apparent that extensive control measures were needed in almost all of the mines.

## CALCULATION OF DOSE TO LUNGS AND EFFECT OF REMOVING DEGRADATION PRODUCTS

The radiation hazard involved in the mining of uranium ores comes from the radioactive gas, radon, and two of its most important daughters, RaA and RaC'. All of these elements emit alpha particles which are very energetic and will damage body cells with which they interact. Since radon is a gas, it is breathed in along with the air in the mine and while in the lungs will continue to decay, emitting alpha

**Table I. Summary of Concentrations by Mining Areas, Colorado Plateau Survey, 1952**

| *Area* | *No. of mines* | *Median conc.* RaA plus RaC'* | *Max. conc.** | *Min. conc.** |
|---|---|---|---|---|
| Bull Canyon | 11 | 4500 | 32,000 | 36 |
| Calamity Mesa | 10 | 1800 | 17,000 | 66 |
| Cottonwood Wash | 7 | 520 | 26,000 | 2 |
| Durango | 5 | 18 | 120 | 3 |
| East Reservation | 9 | 160 | 3200 | 11 |
| Eastern Slope | 1 | 8 | 8 | 8 |
| Gateway | 8 | 1000 | 6000 | 6 |
| Grants | 3 | 2800 | 7000 | 38 |
| Gypsum Valley | 4 | 9000 | 18,000 | 500 |
| Long Park | 25 | 4600 | 37,000 | 15 |
| Marysvale | 4† | 2500 | 59,000 | 180 |
| Moab | 10 | 380 | 3400 | 2 |
| Monticello | 2 | 950 | 1900 | 15 |
| Monument Valley | 3 | 1200 | 11,000 | 9 |
| Outlaw Mesa | 3 | 3000 | 3800 | 18 |
| Paradox Valley | 4 | 800 | 6500 | 37 |
| Polar Mesa | 7 | 2400 | 7700 | 38 |
| Slick Rock | 25 | 1100 | 30,000 | 2 |
| Temple Mountain | 9 | 300 | 2800 | 2 |
| Uravan | 7 | 820 | 12,000 | 160 |

* In micromicrocuries per liter. † Working mines.

**Table II. Concentrations of Radon in Mine Atmospheres by Mining Areas, Colorado Plateau Survey, 1952**

| *Area* | *No. of mines* | *Median conc. Rn** | *Max. conc Rn** | *Min. conc. Rn** |
|---|---|---|---|---|
| Bull Canyon | 11 | 4100 | 59,000 | 1085 |
| Calamity Mesa | 6 | 570 | 23,000 | 70 |
| Cottonwood Wash | 4 | 4500 | 9900 | 3100 |
| East Reservation | 6 | 170 | 2200 | 100 |
| Gateway | 4 | 1100 | 3200 | 160 |
| Grants | 3 | 940 | 2000 | 870 |
| Gypsum Valley | 3 | 14,700 | 18,000 | 1200 |
| Long Park | 17 | 8300 | 48,700 | 1300 |
| Marysvale | 2 | 3360 | 25,900 | 840 |
| Monument Valley | 3 | 5900 | 6100 | 170 |
| Paradox Valley | 2 | — | 3800 | 520 |
| Polar Mesa | 3 | 6800 | 7300 | 3000 |
| Slick Rock | 11 | 3900 | 22,000 | 130 |
| Uravan | 4 | 4900 | 7100 | 1085 |

* In micromicrocuries per liter.

**Table III. Distribution of Mines and Miners by Radon Daughter Concentrations, Colorado Plateau Survey, 1952**

| *Concentration in Micromicrocuries per liter (RaA plus RaC')* | *Mines* | | *Miners* | |
|---|---|---|---|---|
| | *No.* | *Per cent* | *No.* | *Per cent* |
| 0–99 | 35 | 22.3 | 116 | 15.8 |
| 100–499 | 27 | 17.2 | 117 | 16.0 |
| 500–999 | 8 | 5.1 | 31 | 4.2 |
| 1000–1999 | 17 | 10.8 | 140 | 19.1 |
| 2000–4999 | 36 | 22.9 | 153 | 20.9 |
| 5000–9999 | 18 | 11.5 | 60 | 9.2 |
| Over 10,000 | 16 | 10.2 | 116 | 15.8 |
| Total | 157 | 100.0 | 733 | 100.0 |

Average, all mines = 4200 micromicrocuries per liter (RaA plus RaC')
Median, all mines = 1200 micromicrocuries per liter (RaA plus RaC')

particles and producing RaA, RaB, RaC, and RaC'. The daughters of radon will also decay in the lungs, likewise emitting alpha particles besides those of the gamma and beta variety. Furthermore, some of the radon enters the blood stream. Potential hazard to the lung tissue arises mainly from the alpha particles from radon, RaA, and RaC'.

Under usual mine conditions, large numbers of dust particles and water droplets are present in the atmosphere to which the solid decay products of radon will become attached. This dust will be inhaled and carried into the lungs where a portion of it will be retained and decay as outlined above, thus delivering additional radiation to the lungs. The amount of this dust-borne radioactivity that is present in mine atmospheres will depend on the ventilation, air turbulance, and probably other factors. Studies have shown that where ventilation is provided the ratio of radon decay products to radon may be as low as 2 per cent of the theoretical value. The facts that the solid daughters of radon will become attached to dust and be inhaled and that the ratio of radon to its decay products is profoundly affected by ventilation are important in assessing the potential hazards from the elements.†

Experimental measurements made in atmospheres containing, besides radon, normal dust loads in the range that human beings are likely to encounter, show that RaA, the first product of radioactive decay, an alpha emitter with a half-life of 3.05 minutes, is essentially in radioactive equilibrium with the radon producing it. The abundance of the next alpha-emitting radioactive daughter substance, RaC', separated from RaA by two generations of intermediate isotopes, is much more variable but is typically present in amounts in the range of 20 to 80 per cent of the equilibrium value. In the following calculations and discussion it is arbitrarily assumed that this value is 50 per cent. Experimental measurements have shown that when these degradation products are breathed into the human respiratory system appreciable percentages are retained there. Values have been given that range from 25 to 75 per cent.[3,4]

The radiation dose from radon-degradation products through RaC' breathed into human respiratory system has been calculated, using the following assumptions—Radon level: $1 \times 10^{-11}$ curie per liter. RaA concentration, same as radon: $1 \times 10^{-11}$ curie per liter. RaB concentration, ½ radon: $0.5 \times 10^{-11}$ curie per liter. RaC concentration, ½ radon: $0.5 \times 10^{-11}$ curie per liter. RaC' concentration, ½ radon: $0.5 \times 10^{-11}$ curie per liter. Retention of these degradation products in respiratory system: 25 per cent. It is further assumed that they remain in the respiratory system until radioactive decay through RaC' has occurred.

Under these conditions and certain assumptions which cannot be given in detail here, the total alpha-ray dose from RaA, RaB, and RaC-RaC' inhaled as particulates into the lungs is:

0.00178 RaA
0.00441 RaB
0.00330 RaC
Total 0.00949 rep/week

This is larger than the dose due to inhaled radon plus the daughters of this inhaled radon decaying in the lungs by a factor equal to:

$$\frac{9.49 \times 10^{-3}}{4.43 \times 10^{-4}} = 21.4$$

This is to say, if the degradation products could be filtered from the air breathed, the average radiation dose to the lungs due to radon and its daughters would be reduced to 4.4 per cent of the value for unfiltered air.

Thus, if the air that is breathed is cleaned of these daughter products, the permissive radon concentration could be increased several times. Because of the rapid build-up of RaA from radon, probably the only method that will give substantially complete cleaning of breathed air will be a respiratory filter that will remove essentially all particulate matter. However, other effective air cleaning or ventilative methods may, by removal or dilution of degradation products, decrease the hazard of breathing a radon-containing atmosphere below that indicated by the measured radon concentration value.

## CONTROL OF RADON BY VENTILATION

In the previous section it was indicated that the greatest potential health hazard is due to the decay products of radon, particularly RaA, RaB, and RaC'. Thus, control measures would necessarily have to be designed to prevent the inhalation of these nuclei and to allay the dusts which attract and hold the nuclei by electrostatic forces. Wet drilling and wet handling of the ore are common practices and required by State law in uranium mines as a control for silicosis.

It was therefore apparent that there were three possible control methods: (1) respiratory protective devices to remove radioactive particles in the inspired air; (2) filtration of the air in working areas to reduce the concentration of radon decay products and to prevent the attainment of equilibrium between radon and its decay products; and (3) dilution of the radon and its decay products by forced ventilation.

Previous experience in metal mine operations indicated that ventilation was the only feasible method and involved principles familiar to mining practices. Preliminary studies had also shown that ventilation was effective in reducing the concentration of radon decay products. Consequently, studies were designed to determine quantitative information on the effects of ventilation. The reports on these studies, in addition to presenting the results of ventilation experiments, developed methods for determining ventilation rates which are required to prevent the concen-

† The rest of this section is taken from unpublished material prepared by Dr. W. F. Bale, University of Rochester.

tration of atmospheric radon from exceeding any selected allowable value in a working section or a drift of a mine, for estimating the actual radon-emanation rate from an ore body, and for determining atmospheric concentrations of the decay products and their equilibrium ratio.[5,6,7] These studies coupled with others performed by the Public Health Service have led to the following conclusions:

1. Proper mechanical ventilation will reduce the concentration of radon and its decay products to an acceptable level.‡

2. The approximate amount of air necessary in a working area to reduce the radon decay products to a given value can be readily determined by methods developed in the study.

3. In the absence of radiation equipment to measure the concentration of decay products, it is suggested that the following minimum air volume be used:

(a) 500 cubic feet per minute of fresh air to dead-end drifts except where the initial radon concentration was in excess of 10,000 micromicrocuries per liter; in this case a minimum of 1000 cubic feet per minute is necessary.

(b) For larger areas, such as rooms or stopes, there should be at least 1000 cubic feet per minute with 2000 cubic feet per minute where initial concentrations of radon decay products in excess of 10,000 micromicrocuries are found.

(c) The fresh air should be delivered within 30 feet of the working area.

(d) A minimum of 2000 cubic feet per minute of ventilation capacity should be available at any mine even when not required by the above standards.

4. Maximum use should be made of natural ventilation, but this cannot be relied upon to bring concentrations in dead-end drifts down to acceptable levels even under the best of conditions.

It should be emphasized that the above recommendations apply only to uranium mines which have shallow and limited underground workings. As uranium mine operations approach deep-level mining and other metal mines in size and activity, it will be necessary to use standard metal-mine ventilation practices to control the hazards from radioactive materials.

## HAZARDS AMONG MILLERS

Since the process for the separation and isolation of uranium and vanadium is different in each mill, it is impossible to generalize the findings except in a few operations which are common to all plants. In general, it may be said that there are no health hazards in the mills which cannot be controlled by accepted industrial hygiene methods.

‡ A conference called in Salt Lake City, Utah, in February 1955, sponsored by the Utah Industrial Commission, adopted the proposed value of the National Committee on Radiation Protection of 100 micromicrocuries of radon in equilibrium with its immediate daughters (RaA, RaB and RaC').

As pointed out earlier, radon and its short-lived decay products are usually not problems in the milling plant because of the large volume of air available for dilution, and the open-type construction which is generally used throughout the industry.

Dust control at the crushing operation has been found to range from fair to poor. The control of dust at this operation can be accomplished by standard engineering methods, but the installation should be designed by competent and skilled persons. Standards set by the State for silicosis control, using the ore of highest free-silica content (70 per cent), should be used as a guide in determining the effectiveness of dust control systems. This value (5–20 million particles per cubic foot) should also control other health hazards associated with the dust. Since the radon has not been confined at this operation, the decay products have not had the opportunity to reach equilibrium. Until adequate dust control has been established at this operation, the workers should be required to wear approved dust respirators. Daily baths and frequent changes of clothing by the workers in this area are also indicated.

Relatively high concentrations of uranium and vanadium fume were found around the fusion furnaces. In practically all plants the workers in this area were found to be suffering from a chronic irritation of the upper respiratory tract, apparently resulting from exposure to vanadium fume. Several cases of a transitory illness were observed among workers welding or cutting vanadium-coated pipes and metals.

Since the fusion operation is different in each plant, it is impossible to make specific recommendations applicable to each establishment. In general, it may be said that all fusion furnaces should be constructed so as to prevent fume leakage. Local exhaust ventilation should be provided at the transfer point (from furnace to casting wheel) and at the bagging operation. The workers should be provided with fume respirators for emergency and temporary exposures to vanadium and uranium fumes.

Portable exhaust blowers should be located in the fusion area and used by maintenance workers whenever it is necessary to cut or weld metal coated with uranium or vanadium.

Good personal hygiene should be practiced by the workers in the fusion area. This should include daily baths and freshly-laundered work clothes each day.

Only certain specific hazards in the mill are dealt with here. Exposures to other health hazards, such as acids, alkalis, certain gases, and other agents, were also noted, however. Each mill should therefore consider the advisability of a general industrial hygiene survey by a competent authority.

## SUMMARY

1. Atmospheric concentrations of radon and its degradation products in uranium mines of the Colorado Plateau varied widely. It was determined that

radon was of little significance in the mills because of the large volume of air available for dilution.

2. Up to the present, over 1300 miners and millers have been examined medically. No clear-cut etiologic or pathologic patterns have therefore been uncovered probably because the majority of the workers had been exposed for less than three years. The need for repeating the medical studies is clear.

3. Reference is made to the calculation of dose to the lungs and the effect of removing degradation products.

4. A number of conclusions are presented referring to the control of radon by ventilation.

5. Recommendations are made with regard to certain hazards to which mill workers may be exposed.

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# Record of Proceedings of Session 17C

## WEDNESDAY MORNING, 17 AUGUST 1955

*Chairman:* Mr. W. V. Mayneord (UK)

*Vice-Chairman:* Mr. J. Coursaget (France)

*Scientific Secretaries:* Messrs. I. D. Rojanski, L. F. Lamerton and C. Polvani

### PROGRAMME

**17C.1. Radiation safety recommendations and control**

| | | |
|---|---|---|
| P/451 | Radiation injury and protection—maximum permissible exposure standards | W. Binks |
| P/89 | Maximum permissible exposure standards | R. S. Stone |
| P/79 | Maximum permissible concentration of radioisotopes in air and water for short period exposure | K. Z. Morgan *et al.* |
| | Discussion | |
| P/689 | Health protection of workers exposed to ionizing radiations | A. A. Letavet |
| P/907 | The protection of workers against ionising radiations | ILO |
| | Discussion | |
| P/792 | Measurements of low-level radioactivity, particularly the γ-radiation from living subjects | R. M. Sievert |
| | Discussion | |

**17C.2. Hazards related to uranium mining**

| | | |
|---|---|---|
| P/370 | The problem of radon in the uranium mines | H. Jammet and J. Pradel |
| P/85 | Industrial hygiene of uranium processing | M. Eisenbud and J. A. Quigley |
| | Discussion | |

The Chairman: During the last few days, we have been discussing the biological and physical sciences in relation to the effects of radiation. This morning we have to consider the very difficult task of trying to embody these scientific concepts into practical recommendations or regulations.

Circumstances are very different in different countries, and legislation must necessarily fit the needs of the individual countries. Nevertheless, I am sure we are all agreed that particularly in these matters the greatest measure of international agreement must be reached, for these are, after all, international problems.

We have several main tasks: first, we have to arouse world interest in these subjects; secondly, we have to collect information; thirdly, we have to develop a kind of philosophy in relation to these problems; and fourthly, we have to create or develop world organizations to deal with them. All these aspects have indeed been studied for some time and we have started. The dangers are now very widely and well recognized.

We realize that our job essentially is to protect workers and populations to the maximum possible extent but compatible with the task of bringing the benefits of atomic energy to the maximum number of people in the shortest possible time. It is important that we recognize these balancing considerations.

It has perhaps been insufficiently recognized that for thirty years there has been international collaboration on these problems. Certainly this collaboration has been on varying scales at varying times, and we wish very much to see it extended. One obvious example of such collaboration was shown in the recently published report of the International Commission on Radiological Protection. Many of you are aware of the provisions of this document, and I think we can agree that it is a reasonable and indeed a good basis on which to start our further developments.

Finally, let me say that I am sure we all feel that this meeting may well be the beginning of wider and more fruitful international co-operation.

Mr. W. Binks (UK) presented paper P/451.

Mr. R. S. Stone (USA) presented paper P/89.

Mr. K. Z. Morgan (USA) presented paper P/79, as follows: The National Committee on Radiation

Protection agreed on the maximum permissible concentrations of radioisotopes in air and water for *continuous* exposure. For most radioisotopes equilibrium is reached between the concentration of the radioactive material in the environment and in the body organs after only a few weeks or months of continuous exposure. Many radioactive materials are relatively insoluble and the fraction entering the bloodstream following ingestion or inhalation is so small that the gastrointestinal tract becomes the critical tissue rather than kidney, bone, liver, or some other body organ. Of the 355 maximum permissible concentration values for continuous exposure listed in the International Commission on Radiological Protection Handbook, 71 per cent for ingestion and 41 per cent for inhalation, refer to the gastrointestinal tract as the critical body organ.

No maximum permissible values for *single* exposure have been agreed upon, and it is the purpose of this report to extend the method of calculation used in the International Handbook to estimate the Maximum Permissible Intake of these radioactive materials for a *single* exposure.

Twelve years of experience at Oak Ridge National Laboratories have indicated that *ingestion* of radioactive material following an accident is seldom the major problem compared with hazards of inhalation. Therefore, in this study particular attention is given to the *inhalation* of radioactive materials where the *lung* or the gastrointestinal tract is the critical organ.

Reasons for obtaining the Maximum Permissible Values for single exposure to the radionuclides are as follows:

1. *To indicate hazards and necessary protective measures.* This may require special building designs, remote control equipment, hood enclosures, etc., and rigidly enforced working rules must be established for work with some of the radioactive materials.

2. *To aid in estimating damage and selecting medical treatment when radionuclides are taken into the body as a result of accidents.* After such accidents, urine and fecal analyses aid in estimating the burden of the radioactive material in the body. In some cases, such as with plutonium, it may be desirable to take therapeutic measures or in case of puncture wounds to excise the contaminated tissue from the wounds.

3. *To aid in classifying radionuclides as Very Dangerous, Moderately Dangerous and Slightly Hazardous Materials.* This information is useful in preparing legislation, in adopting rules and procedures of safe handling, in applying adequate radioactive waste disposal and in establishing safe national and international regulations for the shipment of radioactive material.

The cases considered are:

*Case 1. Inhalation of soluble radioactive material.* The characteristics of the Standard Man are assumed; namely, 25 per cent retained in the lungs where it enters the blood stream, and 50 per cent swallowed, a portion of which passes from the gastrointestinal tract to blood stream and critical body organ.

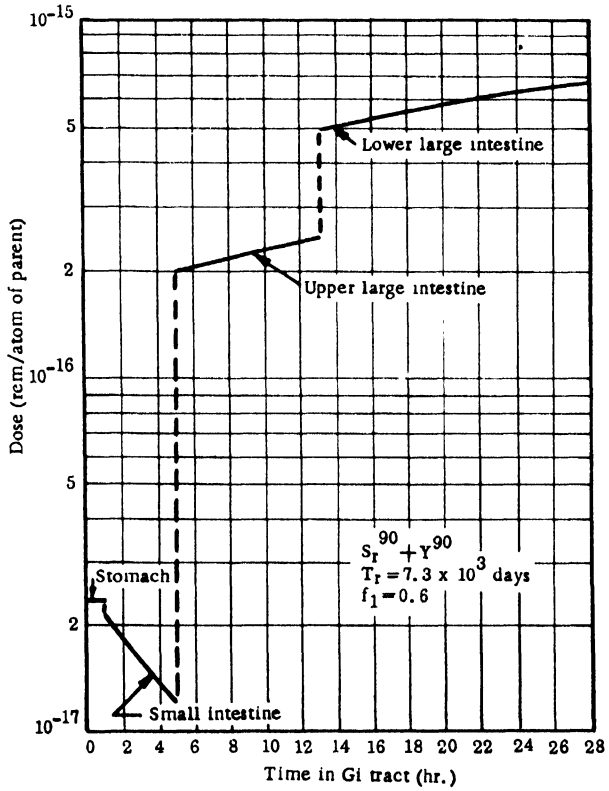
*Case 2. Injection of soluble radioactive material by way of a wound.* It is assumed that all this soluble radioactive material enters the blood stream, a portion of which reaches the critical body organ.

*Case 3. Injection of insoluble radioactive material.* Here it is assumed that all the radioactive material remains at the wound site with a biological half-life of 120 days and that it is localized in 1 milligram of tissue in the wound site.

*Case 4. Inhalation of insoluble radioactive material with the lung as the critical organ.* In this case it is assumed that one-half of the 25 per cent remaining in the lungs is retained with a biological half-life of 120 days and that this radioactive material is distributed uniformly throughout the lung.

*Case 5. Inhalation of insoluble radioactive material with the gastrointestinal tract as the critical organ.* It is assumed that 62 per cent of the inhaled material is swallowed and that it irradiates various portions of the gastrointestinal tract in direct proportion to the time spent in each section. Some of the radioactive material passes through the wall of the small intestine, resulting in a biological half-life. In the case of the daughter products, their contribution to the absorbed dose is taken into account. The Oak Ridge digital computer was used in making these calculations.

Slide 1 indicates the results of these calculations in the case of $Sr^{90}$ and its daughter $Y^{90}$. The radioactive material spends only one hour in the stomach so the dose here is quite small. The dose in the top




Slide 1

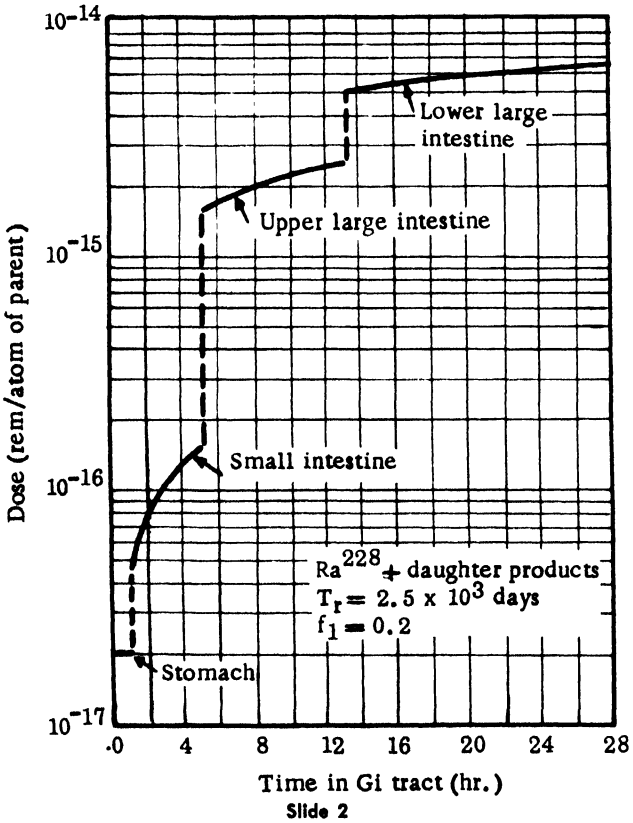
portion of the small intestine is the same as that of the stomach. This dose decreases, however, because of the 60 per cent uptake from the small intestine into the blood stream. The dose to the large intestine is much greater than the dose to the small intestine, because the radioactive material spends a longer time here, i.e., 8 hours in the upper large intestine and 18 hours in the lower large intestine, and the material irradiated here is about ⅛ that of the small intestine. The slow rise in the dose as the radioactive material moves along the large intestine is the result of the growth of the 61-hour daughter product $Y^{90}$.

Slide 2 shows the situation for the 6.7-year $Ra^{228}$. The 6-hour daughter, $Ac^{228}$, makes a large contribution to the dose delivered by the parent. This rise is accentuated by reason of the fact that the effective energy of $Ac^{228}$ is 80 times that of the parent $Ra^{228}$ and there is only a 20 per cent absorption from the small intestine.

The maximum permissible intake, MPI, is given by,

$$\text{MPI} = \frac{CDM}{fT\Sigma(bE)(\text{RBE})Nf_n(T_r, T_b, t_2)} \ \mu\text{c}$$

in which $C$ = constant; $D$ = maximum permissible dose of 0.3 rem/wk, 15.7 rem/yr or 150 rem/70 yr, whichever gives the smallest value of MPI; $M$ = mass of the critical organ; $f$ = fraction arriving in critical body tissue; $T$ = effective half-life; $\Sigma(bE)(\text{RBE})N$ = effective energy weighted by relative biological effectiveness, RBE, and non-uniform distribution factor, $N$; $f_n(T_r, T_b, t_2)$ = insensitive function of radioactive half-life, $T_r$, biological half-life, $T_b$, and period of exposure, $t_2$.



Slide 2

The values given for inhalation in Tables I and IV of paper P/79 were for the MPI per day which if taken for ⅓ of a day would give the indicated dosages. These values (in Tables I and IV) should be multiplied by the period of exposure, i.e., ⅓ of a day, in order to make them directly comparable with the values in the other tables. This change will be made in the published Proceedings of the Conference.

Table I shows some typical values of maximum permissible intake of radionuclides for a single exposure. These values have been corrected for direct comparison. It is to be noted that the MPI for injection of insoluble radioactive material is several orders of magnitude smaller than for the other cases. In most cases the inhalation of insoluble radioactive material gives lower MPI values than inhalation of soluble radioactive material. Usually the lower large intestine is the most critical portion of the gastrointestinal tract.

The last objective of this investigation was to list the radioactive materials in the order of decreasing hazard.

It is considered that the relative hazard, $H$, of a radionuclide is given by, $H = P/\text{MPI}$ in which MPI = maximum permissible intake, and $P$ = probability of inhalation or injection into the body.

$$P = \frac{10}{(\text{mg}/100\,\text{mc})} \leq 1 \text{ for inhalation}$$

$$P = \frac{1}{(\text{mg}/100\,\text{mc})} \leq 1 \text{ for injection}$$

There are over 50,000 domestic shipments of radioactive material from our laboratory each year, and the average activity per shipment (exclusive of $Co^{60}$) is 100 mc. The average industrial air has a dust-loading which corresponds to inhalation of 10 mg of dust in an eight-hour work day. Therefore, this equation assumes there is a probability of unity of inhaling all the radioactive material in a spill if there are less than 10 mg/100 mc involved in the spill. Likewise, it is assumed that not more than 1 mg of material gets into the average contaminated wound and if there is less than 1 mg/100 mc of material involved in the accident, all the material may get into the wound.

Table II is a summary of the more dangerous radionuclides listed in order of decreasing hazard. A wound site contaminated with insoluble radioactive material presents by far the greatest hazard as you can see by the magnitude of these numbers. In general, the injection of soluble radioactive material presents a somewhat greater hazard than the inhalation of this material. When insoluble radioactive material is inhaled, the hazard is to the gastrointestinal tract about ⅓ of the time and to the lung about ⅔ of the time. I have indicated by asterisk the radioisotopes that consistently appear in this list of the most hazardous radioactive materials. It is to be noted that in four out of five cases the bone-seeking $Ac^{227}$ is the most hazardous of all the radioisotopes considered. Astatine-211, polonium-210, americium-241, and cerium-144 also appear in all five groups.

**Table I. Typical Values of Maximum Permissible Intake (MPI) of Radionuclides for a Single Exposure**

| | | *MPI or μc taken into body† to deliver permissible dose by.* | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | *Inhalation of soluble* | | *Injection of soluble* | | *Injection of insoluble* | *Inhalation of insoluble* | | | | |
| | | | | | | | *To lungs* | | *To GI tract* | | |
| *Z* | *Isotope* | | | | | | | | | | |
| 1 | $H^{3}$ | $1.5 \times 10^{4}$ | (*TB*) | $1.1 \times 10^{4}$ | (*TB*) | $1.4 \times 10^{-4}$ (*W*) | $1.1 \times 10^{3}$ | (*L*) | $1.2 \times 10^{4}$ | (*LLI*) | |
| 15 | $P^{32}$ | 53 | (*B*) | 34 | (*B*) | $1.5 \times 10^{-6}$ (*W*) | 12 | (*L*) | 19 | (*LLI*) | |
| 26 | $Fe^{59}$ | 14 | (*Bl*) | 9.1 | (*Bl*) | $7.5 \times 10^{-6}$ (*W*) | 9 | (*L*) | 68 | (*LLI*) | |
| 33 | $As^{76}$ | $9 \times 10^{2}$ | (*K*) | 100 | (*K*) | $3.6 \times 10^{-6}$ (*W*) | 25 | (*L*) | 5.4 | (*LLI*) | |
| 37 | $Rb^{86}$ | 143 | (*M*) | 100 | (*M*) | $1.6 \times 10^{-6}$ (*W*) | 11 | (*L*) | 130 | (*LLI*) | |
| 38 | $Sr^{89}$ | 10 | (*B*) | 5.5 | (*B*) | $1.6 \times 10^{-6}$ (*W*) | 13 | (*L*) | 17 | (*LLI*) | |
| 38 | $Sr^{90} + Y^{90}$ | 5* | (*B*) | 2.7* | (*B*) | $8.5 \times 10^{-7}$ (*W*) | 7 | (*L*) | 21 | (*LLI*) | |
| 42 | $Mo^{99}$ | $9.3 \times 10^{4}$ | (*B*) | $5.9 \times 10^{4}$ | (*B*) | $1.4 \times 10^{-5}$ (*W*) | 28 | (*L*) | 72 | (*LLI*) | |
| 44 | $Ru^{106} + Rh^{106}$ | 20 | (*K*) | 5.1 | (*K*) | $6.3 \times 10^{-7}$ (*W*) | 4.7 | (*L*) | 2.7 | (*LLI*) | |
| 53 | $I^{131}$ | 0.70 | (*T*) | 0.52 | (*T*) | $6.3 \times 10^{-6}$ (*W*) | 22 | (*L*) | 290 | (*SI*) | |
| 74 | $W^{181}$ | $9.3 \times 10^{3}$ | (*B*) | $2.1 \times 10^{3}$ | (*B*) | $2.9 \times 10^{-5}$ (*W*) | 6.3 | (*L*) | 16 | (*LLI*) | |
| 84 | $Po^{210}$ | 0.60 | (*S*) | 0.12 | (*S*) | $1.6 \times 10^{-8}$ (*W*) | 0.12 | (*L*) | $6.4 \times 10^{-2}$ | (*LLI*) | |
| 85 | $At^{211}$ | 0.10 | (*T*) | $5.3 \times 10^{-2}$ | (*T*) | $1.8 \times 10^{-7}$ (*W*) | 2.1 | (*L*) | 2.4 | (*St*) | |
| 88 | $Ra^{226} + dr$ | 0.32* | (*B*) | 0.11* | (*B*) | $5.3 \times 10^{-9}$ (*W*) | $2.8 \times 10^{-2}$ | (*L*) | $5.1 \times 10^{-2}$ | (*LLI*) | |
| 89 | $Ac^{227} + dr$ | $3.7 \times 10^{-3}$ | (*B*) | $1.2 \times 10^{-2}$ | (*B*) | $2.5 \times 10^{-9}$ (*W*) | $2.1 \times 10^{-2}$ | (*L*) | 1.1 | (*LLI*) | |
| 94 | $Pu^{239}$ | $2.0 \times 10^{-2}$* | (*B*) | $5.2 \times 10^{-2}$* | (*B*) | $1.6 \times 10^{-8}$ (*W*) | 0.13 | (*L*) | $7.1 \times 10^{-2}$ | (*LLI*) | |

* Limiting dose is 150 rem in 70 years. In all other cases the limiting dose is 0.3 rem in 1 week.

† *T* = thyroid, *B* = bone, *M* = muscle, *K* = kidneys, *S* = spleen, *Bl* = blood, *W* = wound site, *L* = lung, *LLI* = lower large intestine, *SI* = small intestine, *St* = stomach, *TB* = total body.

**Table II. Summary of the More Dangerous Radionuclides* Listed in Order of Decreasing Hazard**

| *Inhalation* | | | | *Injection* | | *Inhalation* | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| *Soluble radioactive material* | | | | *Insoluble radioactive material* | | | | | | | |
| *The indicated organ as the critical body organ* | | | | *Wound site the critical tissue* | | *Lung the critical body organ* | | | *GI tract the critical body organ* | | |
| 210 | $Ac^{227}$* + dr | *B* | 60 | $Ac^{227}$* + dr | *B* | $4 \times 10^{8}$ $Ac^{227}$* + dr | 48 | $Ac^{227}$* + dr | 56 | $Ra^{228} + dr$ | |
| 9.6 | $At^{211}$* + dr | *T* | 19 | $At^{211}$* + dr | *T* | $7.1 \times 10^{7}$ $Cm^{242}$ | 8.1 | $Pb^{210} + dr$ | 45 | $Ra^{224} + dr$ | |
| 4.8 | $Pb^{210} + dr$ | *B* | 13 | $Cm^{242}$ | *B* | $6.7 \times 10^{7}$ $Pb^{210} + dr$ | 8.1 | $Po^{210}$* | 29 | $Th^{228} + dr$ | |
| 3.3 | $Cm^{242}$ | *B* | 11 | $Pb^{210} + dr$ | *B* | $6.3 \times 10^{7}$ $Po^{210}$* | 3.6 | $Ra^{226} + dr$ | 21 | $Th^{227} + dr$ | |
| 1.6 | $Po^{210}$* | *S* | 8.3 | $Po^{210}$* | *S* | $5.6 \times 10^{6}$ $At^{211} + dr$* | 2.5 | $Am^{241}$* | 19 | $Ra^{226} + dr$ | |
| 1.4 | $I^{131}$ | *T* | 1.9 | $I^{131}$ | *T* | $2.0 \times 10^{6}$ $Am^{241}$* | 0.48 | $At^{211} + dr$* | 17 | $Cm^{242}$ | |
| 0.9 | $Am^{241}$* | *B* | 0.53 | $Th^{234} + Pa^{234}$ | *B* | $1.9 \times 10^{6}$ $Ra^{226} + dr$ | 0.23 | $Ba^{140} + La^{140}$ | 16 | $Po^{210}$* | |
| 0.3 | $Ra^{226} + dr$ | *B* | 0.43 | $Ce^{144} + Pr^{144}$* | *B* | $1.6 \times 10^{6}$ $Ru^{106} + Rh^{106}$ | 0.23 | $Ag^{105}$ | 6.5 | $Pb^{212} + dr$ | |
| 0.3 | $Pu^{239}$ | *B* | 0.42 | $Ba^{140} + La^{140}$ | *B* | $1.4 \times 10^{6}$ $Ce^{144}$* + $Pr^{144}$ | 0.21 | $Ru^{106} + Rh^{106}$ | 4.6 | $Am^{241}$* | |
| 0.2 | $Sr^{90} + Y^{90}$ | *B* | 0.36 | $Am^{241}$* | *B* | $1.2 \times 10^{6}$ $Sr^{90} + Y^{90}$ | 0.21 | $Co^{60}$ | 0.91 | $Ac^{227}$* + dr | |
| 0.13 | $Th^{234} + Pa^{234}$ | *B* | 0.34 | $Sr^{90} + Y^{90}$ | *B* | $9.1 \times 10^{5}$ $Th^{234} + Pa^{234}$ | 0.21 | $V^{48}$ | 0.42 | $At^{211}$* + dr | |
| 0.11 | $Ba^{140} + La^{140}$ | *B* | 0.28 | $Ir^{192}$ | *S* | $7.7 \times 10^{5}$ $Ba^{140} + La^{140}$ | 0.20 | $Ce^{144}$* + $Pr^{144}$ | 0.37 | $Ru^{106} + Rh^{106}$ | |
| 0.11 | $Ce^{144}$* + $Pr^{144}$ | *B* | 0.25 | $Y^{91}$ | *B* | $7.1 \times 10^{5}$ $Te^{129}$ | 0.19 | $Te^{129}$ | 0.34 | $Ce^{144}$* + $Pr^{144}$ | |
| 0.10 | $Sr^{89}$ | *B* | 0.20 | $Ru^{106} + Rh^{106}$ | *K* | $6.7 \times 10^{5}$ $P^{32}$ | 0.18 | $Sc^{46}$ | 0.23 | $Th^{234} + Pa^{234}$ | |

* Parent radionuclides with radioactive half-lives less than one-half hour have been excluded in this comparison. *B*-bone, *T*-thyroid, *S*-spleen, *Bl*-blood, *K*-kidneys.

## DISCUSSION OF P/451, P/89, AND P/79

Mr. W. F. BALE (USA): There is little distinction between a permissible exposure and an exposure slightly over permissible. Would you care to speculate upon what may reasonably be described as an overexposure, i.e., one requiring special attention?

Mr. BINKS (UK): In the United Kingdom we have been considering this question of overexposure, and have introduced for administrative purposes what we call a thirteen-week rule, that is, if a person has an overdose during a period of one week we then look back over his radiation history for the past thirteen weeks, and if that total is less than 13 × 0.3 r no action is taken; he continues with his work. If he exceeds that dose, then we suggest that he be put under medical supervision and that the dose in the following weeks should be reduced so that over the whole period we bring it down below the 0.3 r per week. Therefore the answer is that we are rather regarding four roentgens as the criterion.

Mr. H. M. PARKER (USA): I think perhaps Mr. Binks answered the first question in his talk, but I will read the question with your permission. Lifetime exposure is not the relevant factor for genetic effect. What counts is the cumulative exposure up to the date of conception of each child. This is normally considerably less than the lifetime dose. Did Mr. Binks intend to take account of this in computing the average dose in the United Kingdom up to the age of thirty, and should we not in any case emphasize that we do make a differentiation between lifetime dose and dose to average conception?

Mr. BINKS (UK): We took into consideration the fact that in the United Kingdom it appears that on

the average a person receives one diagnostic examination before he reaches the age of thirty years. This was the estimate that we made. An investigation was made from one of the leading London hospitals which carries out a very large amount of diagnostic work to find out the doses received during specific diagnostic procedure, integrating this up to the period of the first thirty years.

Mr. PARKER (USA): In the case of partial exposures, the dose to the lens of the eye is to be limited to 0.3 rem per week. If neutron radiation is involved, the conversion of the physical dose from rads to rem in the lens of the eye will be different from that in the surrounding tissue. Can we assume that the RBE factors used in our tables have been taken liberally enough to cover this point?

Mr. BINKS (UK): This is a difficult matter; it is one of the weaknesses in our knowledge. Those people who have seen the ICRP report have criticized this section on partial irradiation anyway. It is very difficult indeed to shield the head under certain circumstances to get the 0.3 rem and let the rest of the body tolerate 1.5 rem. Here I think there are both practical and academic difficulties which will have to be overcome in the future.

Mr. MORGAN (USA): I should like to ask Mr. Stone two questions. First, why do you feel so strongly that blood counts should be discontinued as a method of monitoring? Second, do you believe the minor changes in the blood are not significant?

Mr. STONE (USA): The use of blood counts as a method of monitoring was started in the old days when we had no other method of monitoring and it was the best that was available. I believe that we hang on to it now because of tradition rather than value. The experience that we had with the Metallurgical Project and all of its ramifications showed us that using blood counts, as they are ordinarily done by the ordinary technician throughout a large project when he is doing thousands of them per day, does not give you sufficiently good results to justify their use as a monitoring method. In addition, we found that it was almost impossible to keep track of when people had colds that were due to bacteria, which shot the count up, or infections with viruses which shot the count down; it was extremely difficult to separate the effects of infections from the effects of radiation.

With regard to the minor changes that are found in the blood, no one has yet shown that they are of real significance. Those that have found them have sought diligently for some change that might be significant, and the changes that are now being detected as a result of small doses are very difficult to separate out from the changes that come, as I mentioned before, from other conditions that may be affecting the population. It has not been shown that any of them are of significance. We have shown that the great majority of the people who were exposed in the early days to much larger doses—and we know, now that we have correct monitoring methods, that people in hospitals got much more than we thought they were getting as permissible doses—have recovered and have shown no deleterious effects; even though many of them in days gone by did show definite changes in the blood. Therefore, I think it is necessary to prove that these minor changes such as bi-lobed lymphocytes and increased granules and so forth, mean something to the person, that is that there are appreciable bodily injuries, before we accept them as a method of lowering the standards that we use.

Mr. W. D. CLAUS (USA): Mr. Morgan's report indicates that the gastrointestinal track receives the greatest absorbed dose of radiation in many cases and as a consequence is used in this report as the critical body organ. I should like to ask two questions: first, why was the gastrointestinal tract not used as the critical organ in Handbook 52? If Handbook 52 revision is contemplated, how will the present maximum permissible concentrations be affected?

Mr. MORGAN (USA): In answer to the question of Mr. Claus, at the time Handbook 52 was prepared in 1952 we had little knowledge about the effect on the gastrointestinal tract and it was considered that damage to the bone, kidneys, liver and other organs was more significant than that to the gastrointestinal tract; also, at that particular time the calculations were made on the basis of the dose to the entire abdomen. Since that time Mr. E. E. Pochin in England has given us information on the distribution of material within the gastrointestinal tract, and other data has become available indicating that the gastrointestinal tract is just as critical as some other parts of the body; therefore the gastrointestinal tract has been included in the International Commission on Radiological Protection Handbook, and values for this organ will be included in the revised edition of Handbook 52 which should be available next year.

Mr. B. N. TARASOV (USSR): 1. What do you consider the permissible external and internal radiation dose?

2. What allowance do you make for the fact that in a number of cases, depending on its physical and chemical properties, an isotope may, while giving a low external distribution coefficient, concentrate in large quantities in individual cells and produce a large dose in them? For example, when one or another property makes a particular product attractive to phagocytes, they absorb it avidly and thus create large local concentrations measurable in tens of roentgens. Do you allow for this, or have you tried to do so?

Mr. STONE (USA): I would refer this question to Mr. Morgan because I think it is a matter of distribution of the dose—the calculation of the dose on a particular distribution.

Mr. MORGAN (USA): In answer to the first question about the admissible external and internal radiation dose, the handbooks on permissible exposure of the International Commission on Radiation Protec-

tion used two criteria: (1) as explained by Mr. Binks, it is considered that the values given do not give to the skeleton more exposure than would be received from one-tenth of a microgram of radium; and (2) that no organ of the body or no large segment of any organ of the body would receive more than 0.3 rem exposure in any one week. The second part of the question had to do with the possible localization of the radioactive material in organs of the body and in various other units within the body. In the International Handbook we use a non-uniform distribution factor $N$ and set that equal to 5 for the radioisotopes that are localized in the skeleton. We realize that other factors should be used for other organs of the body and segments of them, but we do not at the moment have sufficient information to incorporate this particular information.

The CHAIRMAN: I have several other questions but I am afraid we have just come to the end of our time. Mr. Wu has sent in a little note. Mr. Wu, would you make your point quickly, and then we must pass on?

Mr. WU (China): The problem of radiation safety is very important. I have some experience of radiation injuries of miners for 20 years; therefore I would like to say a few words. I suggest, first, the establishment of an international organization or law concerned with radiation injuries—about the dosage, about the international unit, and so forth; secondly, well-trained technicians to avoid careless work, for radiation protection. It is important to have international co-operation in this problem. Thirdly, there should be a longer period of vacation for radiation workers. Thank you.

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Mr. F. G. KROTKOV (USSR) presented paper P/689.

Mr. K. L. GOODALL (ILO) presented paper P/907 as follows: This Conference hardly needs to be reminded of the insidious and dangerous nature of ionising radiations, which cannot be seen or felt and whose effects on bodily health are normally very considerably delayed in their onset. This, in the view of the International Labour Office, makes codes of practice and national protective legislation, impartially and efficiently administered, essential for the protection of radiation workers, especially in countries with little industrial experience. Additional reasons are that in the course of time familiarity may breed contempt for necessary radiation precautions, and competition to reduce the costs of nuclear power production and of industrial radioactive devices may tend to narrow present margins of safety. The cost of protection is the price we must pay for unleashing the giant forces of the atom.

The ILO, one of whose most important tasks is to promote the protection of workers against every kind of occupational accident and injury to health, has a considerable interest in radiation protection dating from 1923. To take but one example, the Office, as long ago as 1949, convened a meeting of experts on dangerous radiations to draw up a Model Code. Their recommendations, over 200 in number, which we set out in an appendix to our paper, constituted the first set of international safety and health precautions for industrial users of X-rays and radioactive substances. The ILO remains anxious to assist workers everywhere, especially in those countries less advanced in this field, to make safe use of ionising radiations, and by "workers" we mean all persons in public and private employment whether manual or salaried, and whether working in factory, field or mine, in hospitals, research laboratories or transport. To all these the developments of atomic energy have brought or will bring, additional and novel types of hazard.

Our paper offers a wide survey of the whole field of radiation injury and protection, with special emphasis on the industrial field. Part II reviews the history of radiation injury since the time of Röntgen and Madame Curie. Though injuries occurred to the early medical and scientific experimenters, knowledge of the harmful effects of ionising radiations was incomplete and slow in spreading. Methods of monitoring for personal exposures were unknown by present standards, and heavy doses, particularly from uncontrolled direct X-ray beams, must have been received. Further serious cases of injury arose with the advent of radium luminizing. By about the early 1920's it was well recognised that excessive radiation exposures from without and within the body were capable of causing a wide range of acute illnesses and diseases—anaemias, radiation burns and dermatoses, bone and tissue necrosis, sterility and glandular disfunctions, and cancers of the bone and blood. To this list was later added the adverse genetic effects which can arise from radiation doses much smaller than those needed to affect fertility.

From 1920 onwards therefore ionising radiations were treated with much greater respect. Ample evidence existed of the imperative need for control of radiation exposures, and unofficial and official bodies began the fruitful task of framing recommendations and legislation for protective measures. As a result, the vast atomic energy industries which began in the 1940's got off to a splendid start and have maintained an almost exemplary standard of health and safety among their workers. General industry and hospitals have followed closely behind. Part II of our paper concludes with a description of the physical properties of all types of radiation met industrially and of the types of injury to which they can each give rise when absorbed in excessive amount.

We are supremely lucky in the case of ionising radiations to have what hardly any other branch of industrial health has—namely, practical, extensive and clearly defined standards to work to. How this fortunate state has come about is the subject of Part III of our paper. This recalls what has resulted from the early pioneer work of the British and American Röntgen Ray Societies in the 1920's. The First International Congress of Radiology held in 1925 greatly

stimulated national activities in the protection field and led in 1928 to the formation of the International Commission on Radiological Protection whose latest Recommendations, published this year and which we summarise, cover the whole range of permissible doses for external and internal radiation and standards of protection against X- and gamma rays, beta rays and heavy particles. It is of great interest to note how the International Commission is turning its attention more and more to the preparation of Model Codes of practice for the use of ionising radiations in various occupations; we hope to receive its collaboration when the ILO Model Code of Safety Regulations for Industrial Establishments is revised.

Part IV of our paper discusses the tremendous growth in the use of ionising radiations and gives many examples of the multifarious uses to which they are now put in medicine, agriculture and industry. The spectacular rise in the use of radioisotopes occurring after 1946 when the atomic reactors of the USA and the United Kingdom came into production is illustrated by a table in Appendix IV showing United Kingdom deliveries year by year from 1947 to date. The rate of growth is substantially linear and has now reached nearly 20,000 deliveries a year, a third being exported. American figures are very similar and the implication is that over the whole world there must by now be many hundreds of thousands of workers with X-rays and radioisotopes—perhaps a million—whose work regularly exposes them to some amount of ionising radiation and whose radiological protection must be our especial care.

How this is to be achieved is set out in detail and in a practical way in the main section of the paper—Part V. The first half gives a summary of those general precautions which are applicable to all users of ionising radiations and includes a discussion of such matters as planning and equipment, inhalation and ingestion precautions, monitoring, film and dose meter tests of radiation exposure, use of safe handling equipment, radioactive waste disposal, supervision and training and medical supervision. Reference is made to the limitation of working periods for ionising radiation workers. Some countries favour a maximum of 48 hours a week, others as low as 24, with 20 consecutive days' leave every six months. The 1949 ILO Model Code suggested a five-day working week of 35 hours with a minimum of four weeks' annual holiday. But if the proper precautions as set out in our paper are followed, we cannot but feel at present that no radiation worker need feel aggrieved or in any danger with a 40-hour five-day week and three to four weeks' consecutive holiday.

On these matters and the related ones of age and sex, however, we await a firm lead from the International Commission on Radiological Protection. Likewise, from the same source we await authoritative guidance on what we are to do with our industrial and hospital radioactive wastes now accumulating in ever increasing quantities.

The ILO attaches great importance to supervision and training by safety officers and others in connexion with radiation hazards since it is useless if the blind lead the blind. Workers will usually only take full precautions and take them intelligently if the underlying reasons are understood. In this connexion there is urgent need in many countries for more specially trained members of impartial inspectorates, such as factory and mines inspectorates, to see that control measures are being properly implemented in practice. Medical supervision is no substitute for effective technical measures but has its place, and attached to our paper is a specimen medical record form considered suitable by the ILO for standardised use throughout the world for recording clinical and haematological examinations of radiation workers.

We mention also the interesting possibilities of providing chemical and biochemical protection of humans against the effects of radiation by the administration, before or after irradiation, of certain chemicals or biochemicals which can inhibit bodily processes of oxidation. This opens up the prospect of providing ameliorative treatment for workers accidentally exposed to heavy doses of radiation which might otherwise produce severe injury, though it is most unlikely that as a form of protection it will ever supplant the basic methods of protection by shielding and distance.

The second part of Section V deals in considerable detail with the special precautions appropriate to the main categories of radiation uses in factories, hospitals, atomic energy plants and radiochemical laboratories and covers the risks from X-rays and from both sealed and unsealed radioactive sources. In passing let us not forget those old stagers of the atom world, the radium luminisers. They have now been steadily at it for 40 years and we can still learn a lot from their experiences. Radium is still, in our view, the most dangerous of all the unsealed sources used outside atomic energy establishments. Some four to five grammes of it are painted on to various articles in the United Kingdom alone every year and it possesses most of the radioactive vices. It is a dust, it is long-lived physically and biologically, it concentrates specifically in bones if inhaled or ingested. It decays to a dangerous radioactive gas and with its disintegration products emits alpha, beta and gamma rays. No wonder the luminisers, of all categories of radiological workers, work closest to the health limits.

In Part VI of the paper we refer to the radiological and other hazards in uranium mining and review the tenuous published data available on health conditions in such mines. The position appears very disquieting. On the one hand there seems to be firm evidence over a long period of years that uranium miners are especially prone to lung cancer, which appears to be due to the inhalation of radon and radioactive dust, although the arsenic, cobalt, chromium and other dusts present may also play a part. On the other hand recent authoritative measurements, which we quote, indicate that in the mines sampled radon concentrations were

found representing between 20 and 250 times the internationally agreed safe permissible level. The ILO appeals to all countries concerned to initiate research to resolve this serious problem, to remove secrecy restrictions and to make available for study all results of health and environmental surveys in such mines. In the paper itself and in an appendix the ILO sets out the relevant precautions advocated by its own Committees of mining and other experts which, if adopted, would go far to eliminate this scourge.

Finally, in our last section we deal with safety precautions in the packing, labelling and transport of radioactive materials. On the question of labelling, the ILO has been very active in trying to secure international agreement on a uniform system of marking and on a standard sign or symbol to identify radioactive danger. The ILO-suggested symbol and other suggestions in this field have found wide acceptance and the paper sets out the present position.

To conclude: In the second great industrial revolution which is upon us and which will be brought about by the peaceful uses of atomic energy, we must not repeat the mistakes and miseries which occurred in the industrial health field in the first. Nor need we do so, for the nature of potential injuries from the various radiations is well established. Clear and authoritative international standards of protection also exist which we know how to implement in practice, as our paper has tried to show.

With some mental reservations about genetic effects and uranium mining, we can say that the present position in the radiological health protection field is good. But let us improve on it still further, realising that if there is no room for alarm there is equally none for complacency. We should go forward confident that we can meet and overcome all challenges. In all this good work the ILO hopes to remain in the vanguard of progress.

## DISCUSSION OF P/689 AND P/907

Mr. L. S. Taylor (USA): I should like to ask Mr. Krotkov whether they have obtained in the USSR some tangible evidence showing that the permissible exposure rate of 0.3 roentgens per week is in fact more harmful than an exposure rate of 0.05 roentgens per day. As a second part of the question, I would point out that monitoring radiation exposure on a daily basis is usually difficult both technically and administratively, and I should like to know whether Mr. Krotkov could give us some information as to how they monitor or how they measure this daily exposure rate on a mass basis.

Mr. Krotkov (USSR): In reply to Mr. Taylor's question, clinical observations and experimental data indicate that patients can recover from fractional doses of irradiation better, i.e., more quickly, than from concentrated doses. There is therefore every reason to think that small, fractional doses are less hazardous than concentrated ones. This fact has also been the subject of investigations by Soviet health experts working in this field.

In undertakings and institutions which work with radioactive substances to any considerable extent there are special dosimetric services equipped with a wide range of dosimetric devices. Part of the duties of these services is the systematic monitoring of radiation. The harmlessness of the total dose is established from the readings of radiometric monitoring instruments of various types. The most hazardous types of work are carried on under the supervision of a dosimetrist who may order the work to be stopped if the dose exceeds the standard level.

Mr. Morgan (USA): 1. You indicate, Mr. Krotkov, that higher hygiene requirements are in force with regard to drinking water of centralized water supplies as well as to atmospheric air of residential sections of towns and workers' settlements. Does this mean that the values listed in Table III of your published report are reduced by a factor of about 10 in such cases?

2. In your discussion, you refer to a committee of physicians and physicists who are consulted on the observance of safety rules for work with radioactive substances. Do you have a professional group of health physicists in the USSR?

3. You indicate that particular attention is paid to neurological examinations with respect to early damage from ionizing radiation and you state that treatment at night sanitariums is widely applied to persons working with ionizing radiations. Do you expect to find any neurological changes or a need for a stay at a night sanitarium for workers who stay below the maximum permissible exposure rate of 0.05 roentgens per day?

Mr. Krotkov (USSR): I will answer Mr. Morgan's questions. You understood me correctly, Mr. Morgan, in speaking of a dose ten times smaller for the population. In this instance we share Mr. Binks' view that the maximum permissible dose for the general population should be ten times smaller than that for the workers. We have a large group of health experts concerned with questions of industrial hygiene and occupational diseases. There are institutes of industrial hygiene and occupational diseases in all the industrial areas of our country. The public health faculties of our medical schools have chairs in these subjects. There is no committee of physicians and physicists in the Soviet Union. The functions of a controlling body in the field of industrial hygiene and occupational diseases are carried out in the USSR by the Institute of Industrial Hygiene and Occupational Diseases of the Academy of Medical Sciences of the USSR. It should be mentioned that as a rule all work by physicians and biologists in the Soviet Union requires the participation of physicists. I do not think it can be said that there is a separate profession of health physicist, but physicists are widely associated with all research and practical work done by the medical establishments.

I will now turn to Mr. Morgan's third question. This question was also asked in Mr. Mawson's note, and I propose, with your permission, Mr. Chairman, to answer both these questions together.

My colleagues, Mr. Lebedinsky and Mr. Domshlak, have spoken from our Conference rostrum on disturbances of a neurological nature. They are both members of our delegation and are present here. I would ask Mr. Morgan and Mr. Mawson to meet my colleagues during the recess and clear up all the details raised in their questions in private discussion.

Mr. H. JAMMET (France): The question I wanted to ask was similar to Mr. Morgan's. I wanted to know what early clinical indications of a neurological nature were looked for in detecting occupational disease due to radiation. Perhaps I could join my American colleagues in meeting our Russian colleagues after the session.

Mr. KROTKOV (USSR): I should be glad if Mr. Jammet would meet me after the session and have a discussion with my colleagues, who are better qualified to deal with clinical questions. I hope that my colleagues will be able to give exhaustive replies to all the questions on neurological symptoms.

Mrs. P. CAMBEL (Turkey): I should like to know whether, in view of the radiation hazards, the Soviet Union gives higher salaries to all kinds of radiation workers than to other employees—in the form, perhaps, of some indemnity to cover longer vacations, cures, and so forth. Secondly, is the Ministry of Health of the Soviet Union also responsible for the health of radiation workers under the Ministry of Labour or any other Ministry? In other words, does the Ministry of Health have control over all radiation hazards?

Mr. KROTKOV (USSR): Special wage scales are established for workers employed in undertakings and scientific research establishments. These scales are higher than those applicable to workers and employees in other categories. With regard to vacations and other preventive measures, I may say this. All workers and employees of undertakings and establishments connected with sources of radiation or with radioactive substances as a rule are given longer paid vacations. In addition, dietary standards have been laid down in the Soviet Union for persons handling highly radioactive substances. Lastly, we have institutions to which workers and employees may go for periods of two weeks to a month to rest and convalesce at doctor's orders. So far as sanatoria and rest homes are concerned, workers in the establishments mentioned enjoy important privileges. Thus, for instance, these workers or employees pay only 30 per cent of the cost of travel to and maintenance in sanatoria and rest homes, the remaining 70 per cent being paid out of social insurance funds.

The second question was whether the Ministry of Health is responsible for the health of workers handling radioactive products and working at undertakings belonging to other departments, and the answer is that it is responsible.

Mr. R. M. SIEVERT (Sweden) presented paper P/792.

## DISCUSSION OF P/792

Mr. TAYLOR (USA): I should like to point out to this Conference that, because of Mr. Sievert's long efforts in this field, in Sweden they probably have at the present time some of the most complete information on radiation exposure to all kinds and varieties of individuals. I should like to ask Mr. Sievert if he has some plans in mind with regard to analyzing this large amount of exposure data, particularly with reference to finding out something about the genetic problem. If he does not have any particular plan in mind at the moment, does he have any suggestions that he would like to offer in this regard?

Mr. SIEVERT (Sweden): It is very difficult to say exactly. We have discussed whether it is possible to make genetic investigations and test the different material we have, but at the present time we feel it would be very difficult as we would need a tremendous amount of material for that. We certainly wished to make such investigations, but I think it should be done on an international scale as it would be a very difficult thing for a small country to do.

The CHAIRMAN: Are there any further questions? If not, may I thank again all the speakers up to the present time. I now propose to hand over my responsibility to my colleague, Mr. Coursaget, for the rest of the meeting concerning the hazards of uranium mining.

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The VICE-CHAIRMAN: This second part of the session is devoted to the hazards incurred by miners in uranium mines. There can be no doubt that at the present time these miners are more exposed than any other workers to the harmful effects of radiation. The pooling of knowledge acquired in various countries and the publication of the safety methods employed and results achieved will undoubtedly help us to improve working conditions in the mines and to reduce the danger involved in this work appreciably. We must hope that, in this field perhaps more than in any other, the widest possible international co-operation will be achieved; in particular, such co-operation would make it possible to lay down safety regulations for work in underground uranium mines similar to the regulations which already exist for coal mines.

Mr. H. JAMMET (France) presented paper P/370, as follows: I have the honour to present to you, on behalf of Mr. Pradel and myself, a few observations on our experience with radiological protection in French uranium mines.

These mines are worked by permanently settled miners, many of whom may have been working in the pits for several decades. Our policy has therefore been to give these workers conditions in which the safety

standards recommended by the International Commission on Radiological Protection are observed.

I am responsible for the entire radiological protection system to the French Atomic Energy Commission (*Commissariat à l'Energie Atomique*). The uranium mines cause us the greatest concern.

I do not think that my colleagues from abroad will deny the fact that medical requirements are much more difficult to enforce in uranium mines than at nuclear reactors.

In this case, we are not concerned with installations in which artificially created hazards can be limited with ease; on the contrary, a great deal of ingenuity has to be used to combat the natural hazards inherent in this type of work.

These radiological hazards are many, and are created both by direct radiation from the ore-bearing rock and by atmospheric contamination by radioactive dusts and radon. We intend to confine ourselves now to the single problem presented by the contamination of the air by radon and its daughter elements, radium A, B and C, in spite of the fact that the hazards of direct irradiation are such that we are often obliged to restrict the miners' working hours to a very limited period.

In the French mines, systematic measures are enforced and there is individual supervision in operation at the working face.

The radioactivity of samples of radon is determined by measuring the current they produce in an ionization chamber. Another determination method also used is performed by adsorption of radon on activated charcoal. These techniques are not enough to ensure good supervision, and a portable apparatus for the direct measurement of the $\alpha$-activity in the air is needed, as well as permanent recording apparatus at different points.

Apart from these statistical results, we wish to ascertain the quantities absorbed by each worker. The average quantity of radon inhaled by each miner is therefore determined by means of group measurements taken at fixed points and at various times. We are at present trying out an individual portable apparatus for continuously sampling the air by means of a small suction device which draws the air through a cartridge of activated charcoal. This, at the end of the day, will give us the average radioactive content of the air actually inhaled by the miner.

These checking methods have produced many data from which we have drawn the following conclusions:

1. The concentrations encountered in uranium mines, in terms of a typical present-day metallurgical mine, are very high and frequently exceed the maximum permissible concentration established at $10^{-10}$ curies per litre.

2. The concentrations depend on the extent of the ore-bearing rock and on its uranium content, but also on the density of the rock.

3. Fragmentation of the rock caused by blasting the ore releases a large proportion of the radon imprisoned in it. Blasting fumes have been found to contain as much as 500 times the maximum permissible concentrations.

4. In galleries which are not ventilated, the radon concentrations are very large and they contaminate the other galleries by diffusion.

5. Even when the ventilation is of the order of 4 to 8 $m^3$ per second, it is in most cases impossible to get the concentrations below the maximum permissible.

6. The water circulating in the ore-bearing levels dissolves the radon formed in the cracks and releases it as it flows through the galleries on contact with the air at the working face.

7. Galleries driven through dead levels contain radon in large quantities brought there by diffusion through the ground or carried by the water.

These data of manifest importance compel us to organize very energetic countermeasures against radon. If these countermeasures are to be effective they must be carried out on a planned basis. It is useful to slow down the appearance of radon and speed up its removal in order to take advantage of its rapidly decreasing radioactivity. Accordingly, all our measures are designed first to restrict the evolution of radon and then to remove the radon which has been released as quickly as possible.

The slowing-down and reduction of radon appearances in the galleries can be secured by judicious working methods and by special arrangements. The recommended working methods make it possible to reduce the evolution of radon at the working face, firstly, by breaking the rock as little as possible, secondly, by removing the ore very quickly and, thirdly, by reducing exploration work in the ore-bearing levels as much as possible. These two considerations have led us to devise working methods not involving storage chambers, and working faces of small dimensions isolated from each other rather than long galleries branching out in all directions.

The special arrangements are intended to slow down and reduce the evolution of radon in the pit, and they are all based on the fact that the quantity of radon which finally appears at the face is equal to the difference between the quantity which penetrates through the walls and the quantity which disintegrates while the gas is being held back temporarily.

We can therefore use one or another of these factors to reduce the final quantity of radon which appears in the gallery. It is for this purpose that we have experimentally studied the effectiveness of gallery screens, water drainage and wall-linings. Here are some data on the experiments we have made.

The screening method consists in closing every gallery not in use by means of a fairly gas-tight partition. The screen slows down the diffusion of radon and increases the radon concentration behind it. The result is a large increase in the quantity disintegrating behind the screen and a small reduction in the quantity penetrating the walls, resulting in a net reduction in the quantity of radon finally appearing. It should be noted that the screening partition does not need to

be perfectly gas-tight to be effective, owing to the fact that the gas held back is radioactive and disintegrates.

You will find in our written paper (P/370) the very simple theory which yields the formula for the effectiveness of the screen as a function of the volume of gallery isolated, increase in radon concentration behind the screen and the disintegration constant.

I would just like to illustrate my remarks with a diagram (Slide—Figs. 2 and 3 of P/370) giving the results of a typical experiment to show what can be expected from such a method. The experiment consisted of isolating a dead-end gallery by means of a "cocoon" screen (plastic material which can be sprayed from a pistol). The pit was swept by an air current of eight cubic metres per second. The radon concentration was measured in the gallery at point *A* and at the face at point *B* before the screen was installed and in the hours and days following. The two curves show that the screen multiplied the radon concentration behind it by a factor of 80 while the concentration in front of it was reduced by a factor of 2. It should be noted that at the end of a fortnight some large tears in the screen caused by tests of its strength made it only partially ineffective.

The experimentally verified efficiency could be predicted to a close approximation by using the theoretical formula.

Our second method, water drainage, suggested itself as a second method of reducing radon when experience revealed what an important part in the evolution of radon was played by this process. The underground waters dissolve the radon as they pass through the ore-bearing zones and release it on emerging into and flowing through the galleries. Draining the water and disposing of it through conduits outside the mine have the effect of considerably reducing the quantity of radon evolved and consequently of reducing the quantity which finally appears in the gallery.

I am going to illustrate this method by the results of one of the final experiments which we made to prove its efficiency (Slide 4—Fig. 4 of P/370). In a gallery laid out in a completely dead level with the air coming directly from outside, one might imagine that the atmosphere would be free of radon. However, measurements showed a concentration which increased as we went deeper into the gallery, until it reached 12 times the maximum permissible concentration after 300 metres. The air was contaminated on contact with the counter-current of water. By draining the water we were able to reduce the radon concentration considerably—by a factor of 60—bringing it down to ⅕ of the maximum permissible concentration.

Quite plainly, the use of these run-off waters in inserting the driving hammers must be strictly prohibited so as to prevent a large evolution of radon through pulverization.

Lastly, we considered lining the walls as a third way of counteracting radon evolution. This method has not yet been employed in current practice, but we have been able under laboratory conditions to show that radon evolution can easily be halved by painting the ore face with oils. This result is chiefly due to the fact that the radon dissolves in the oil and remains there long enough to deposit its solid daughter elements radium A, B, and C.

However effective the application of these methods or reducing radon evolution may be, it still does not enable us to bring the concentration down below the maximum permissible in cases of heavy contamination. In these cases it is necessary to remove the radon present in the air of the galleries by means of a well-designed system of ventilation.

For that purpose, we use powerful ventilating systems with an output of between 4 and 20 $m^3$/sec, according to the size of the working chambers. Under favourable conditions, the radon content is more or less inversely proportional to the rate of flow of air.

However, two essential conditions must be observed for effective ventilation: (1) any recycling must be eliminated by installing the air inlets outside the mine; and (2) the pure air must be brought as close as possible to the working face.

The strong concentrations found in blasting fumes make it essential that the workers should not return to the face immediately after blasting.

Ventilation has a further advantage of great importance: it appreciably reduces active deposits on dust particles. If the radon coming from the rock is removed rapidly, its daughter elements, which have a mean half-life of thirty minutes, do not have time to form and the final result is that there is only a fraction, mainly consisting of radium B and C, of the content found in the equilibrium state.

By combining the method of restricting the appearance of radon with that of forced evacuation, it is possible to reduce the radon content in the atmosphere considerably, by a factor which may easily exceed 100.

Often, however, the contamination of the atmosphere is such that these combined safety methods do not succeed in bringing the radon content down below the maximum permissible concentrations. In such cases we have to resort to individual protection of the miners by supplying them with air filtered through respirators or with fresh air fed through masks. The respirators filter out the radon daughter elements deposited on the dust particles. The masks afford complete protection but have the disadvantage of not being independent. The inconvenience of these individual protection methods is too obvious to need stressing.

Our experience therefore shows that if safety standards are to be observed, as is our policy, in uranium mines, a farreaching change in working techniques must be considered.

This purpose can only be attained by close liaison between working engineers and radiological protection services as soon as projects are begun, and a planned system of supervision and strict discipline at the face.

Both collective and individual protection measures are bound to affect the cost price of uranium to an

extent that can only be reduced by some logical scheme such as that we have just mentioned. We do not think it is too great a price to pay for the health of the workers, who are among the most exposed.

We are faced with a serious problem, the fact that it is impossible in many cases to reduce radon concentrations below the permissible maximum by collective measures. The officials responsible for production may tend to ignore standards which are so often unattainable, and the officials responsible for health protection must on no account permit this.

In conclusion, therefore, we may express the hope that the information, hitherto very scanty, on the hazards of uranium mining and the precautions taken against them will be more widely disseminated so that we can judge of the comparative effectiveness of the safety methods employed.

Mr. M. EISENBUD (USA) presented paper P/85.

## DISCUSSION OF P/370 AND P/85

Mr. EISENBUD (USA): In the United States, it has been observed that the air in many of the non-uranium mines contains radon in excess of $10^{-10}$ curies per liter. Have you, Mr. Jammet, had an opportunity to make radon measurements in mines other than the uranium workings?

Mr. JAMMET (France): We have not had an opportunity to take measurements of this kind in mines other than uranium mines; we have, however, found that atmospheric air at ground level in the neighbourhood of uranium mines and in certain areas may exceed the maximum of $10^{-10}$ curies per litre, in normal weather or in certain atmospheric conditions.

A study of radon distribution at ground level according to local weather conditions and geology would seem to be very useful and, in particular, would perhaps, make it possible to map the radon surface distribution and try to discover whether there is any correlation between concentrations above the recommended standards and certain faults prevalent in the areas concerned.

Mrs. CAMBEL (Turkey): I should like to ask Mr. Eisenbud about the effects on the kidneys of the experimental animals. I should also like to know the exact conditions in which these effects were produced. Were there any effects on other organs, particularly critical organs such as the spleen and the intestines?

Mr. EISENBUD (USA): I should like to pass that question on either to Mr. Neuman, who I believe is here and who actually did that work, or to Mr. Farr, who is one of our colleagues who is familiar with the kidney effects both in animals and in man.

Mr. L. E. FARR (USA): I do not recall the exact conditions of the animal experiment. I think, however, that we may go back to the work of MacNider, which was done in the early 1920's, on the effects on the kidney. When soluble uranium salts are given to dogs, there is a destruction of the tubular elements. When the dogs recover from the initial insult, the type of renal epithelium changes from a high columnar type to a low and rather stratified type. This new cell is much more resistant to uranium intoxication than the original cell.

It may be that in workers who are exposed to uranium, this type of protective response occurs, just as it does in dogs. We studied some patients at the Brookhaven National Laboratory hospital and found no changes in the clearances. No matter whether we used inulin, urea, para-amino-hippurate, or any other type of clearance, it was essentially normal. However, the maximum specific gravity of the urine, after twenty-four hours on a dry regime, appeared to show an increase to a specific gravity above 1.035, which is a very high figure. This seemed to be rather characteristic of those individuals who had an exposure to uranium which might be thought of as hazardous. In each instance, however, the follow-up of the patients, as Mr. Eisenbud has stated, gave no evidence that any progressive renal damage had occurred.

Mr. BALE (USA): In one of the early slides, the discrepancy between the predicted and observed concentration of uranium in the two lungs was striking. I wish to ask Mr. Eisenbud if he has any thoughts on how the data and predictions can be reconciled.

Mr. EISENBUD (USA): I do not think that it is possible completely to reconcile these differences. However, I can suggest a reason for possibly 10 per cent of the differences; in other words, I can explain a factor of 50 but not of 1000. That reason is the following: The density of the uranium dust is about 9. The density of the dust and fumes used in the experimental work from which we derived our knowledge of lung retention is around 1. It is possible to normalize the curve developed with the light-density dust or the heavy dust by utilizing the so-called effective diameter of the uranium dust, which is its diameter times the square root of its density. When one does that for a two-micron particle—which is what one usually deals with in industry—one ends up on a very steep part of the curve, in which the retention of two-micron dust, rather than being around 50 per cent as it is in the published data, is more nearly between 1 and 5 per cent.

The VICE-CHAIRMAN: These papers have shown what great efforts have been made in most countries to ensure the safety of persons exposed to radiation. There can be no doubt that these efforts must be intensified to keep pace with the development of the peaceful uses of atomic energy. Even now, however, it may be said that international co-operation has made a large contribution towards shielding humanity from most of the poisonous effects of ionising radiations, and this is an achievement of which we may legitimately be proud.

# Session 18C

## SAFETY STANDARDS AND HEALTH ASPECTS OF LARGE-SCALE USE OF ATOMIC ENERGY (*concluded*)

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# Health and Safety Activities in Reactor Operations and Chemical Processing Plants

By A. J. Cipriani,* Canada

Radioactive materials became an industrial health problem when radium and mesothorium were used in the dial painting industry. Unfortunately the toxic nature of these substances was not fully appreciated until they had affected the health of those working with them. Within the last decade the industrial health problem has grown considerably as a result of the production, from nuclear reactors, of large amounts of radioactive nuclides. There has also grown a philosophy that the occupational damage which occurred in the past from ionizing radiation should not be repeated. This poses the following questions:

1. How can workers handle large radiation sources without suffering harmful radiation exposures?
2. How can work be carried out with large amounts of radioactive materials without getting harmful amounts into the body by ingestion and inhalation?
3. How can one work with loose radioactive material and not contaminate the working environment so there is ultimately a health hazard?
4. How can one deal with radioactive effluent so that the safety of the surrounding population is ensured?

The exposure of workers to radiation levels above the natural background is now an accepted situation in nuclear energy plants. The prevention of overexposure to radiation is a function of proper plant design, the existence of an adequate system of hazard control and adherence to a proper code of practice, in this case the recommendations of the ICRP.[1] Experience at Chalk River shows that exposure to radiation can be controlled within acceptable limits even under emergency conditions.[2]

## EXTERNAL RADIATION EXPOSURE

External exposure data, for the Chalk River Project of Atomic Energy of Canada, is presented as a guide to what may be expected in practice in atomic energy work. The frequency of occurrence of annual total exposures is tabulated in terms of class of work and for a three-year period. The figures for 1952 and 1954 represent normal operations while those for 1953 represent an abnormal emergency period wherein a considerable effort was directed towards decontaminating and rebuilding a seriously damaged reactor.

During the period covered by Table I, the total employment averaged 1800 persons. Certain observations are pertinent. Despite work in high radiation fields, (in several cases involving entry of personnel into fields greater than 50 r/hr) only one person received more than the recommended upper limit of 15 r/year. This exposure of 16.1 r occurred accidentally to an individual lacking any experience in active work and under supervision which also had little experience with radiation.

The abnormal situation in 1953 involving the restoration of NRX shows the value of careful planning. The experienced reactor personnel were kept working throughout the year with relatively modest exposures by calling on personnel normally in inactive work to carry out the tasks involving the higher radiation exposures. As a result, the figures for 1953 show that the moderately high exposures were distributed widely among all plant personnel.

**Table I. Frequency of Occurrence of Various Exposure Levels**

| *Class of work (normal)* | *Year* | *Exposure range in roentgens year* | | | | |
|---|---|---|---|---|---|---|
| | | *1–3* | *3–5* | *5–10* | *10–15* | *>15* |
| High activity: | | | | | | |
| Reactors | 1952 | 14 | 6 | 14 | – | – |
| | 1953 | 11 | 13 | 25 | 2 | – |
| | 1954 | 23 | 4 | 1 | – | – |
| Chemical process | 1952 | 38 | 16 | 5 | – | – |
| | 1953 | 35 | 34 | 62 | 8 | – |
| | 1954 | 34 | 34 | 18 | – | – |
| Maintenance | 1952 | 59 | 5 | 2 | – | – |
| | 1953 | 68 | 57 | 64 | 3 | – |
| | 1954 | 78 | 24 | 14 | – | – |
| Moderate activity: | | | | | | |
| Research | 1952 | 25 | – | – | – | – |
| | 1953 | 6 | 2 | 2 | – | – |
| | 1954 | 6 | 3 | – | – | – |
| Others | 1952 | 17 | – | – | – | – |
| | 1953 | 88 | 39 | 20 | – | – |
| | 1954 | 18 | 5 | 2 | – | – |
| Low activity: | 1952 | 17 | – | – | – | – |
| | 1953 | 61 | 22 | 14 | – | 1 |
| | 1954 | 19 | – | – | – | – |
| Totals | 1952 | 170 | 27 | 21 | – | – |
| | 1953 | 269 | 167 | 187 | 13 | 1 |
| | 1954 | 169 | 70 | 35 | – | – |

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It is interesting to note that chemical operations personnel who were not directly involved in pile restoration were responsible for the majority of exposures over 10 r/year, although the pile restoration work involved five times the total exposure received in all other work combined. This in part could be a result of concentration of protective measures and attention on pile restoration with relaxation of supervision in areas considered less critical.

Since this plant is of early design and has suffered from some "growing up" the problem should be more easily dealt with in better designed installations, which would have the benefit of past experience.

At Chalk River it has been found that in order to preserve adequate Radiation Hazard Control standards, it is necessary to keep the organization of the control personnel separate from that of the operation groups. A similar situation exists at all other places in Canada where radiation workers receive this service, at the moment, from the Department of National Health.

In general, the figures presented justify the belief that an efficient radiation protection service well integrated into the over-all project organization can result in a large output of work in high radiation fields with no need for serious overexposure.

## INTERNAL RADIATION EXPOSURE

The entry of radioactive materials into the human body where they might remain and irradiate specific tissues with adverse effects presents a more complex problem. A great deal of provisional information exists on what amounts of the various radioactive nuclides may exist in the body without harmful effects.[3] From these figures are derived maximum permissible amounts of radioactive materials in air and water for continuous intake but this is of a form more useful to the plant designer than to those working with health hazards, because, in practice, the first indication of internal contamination is often the presence of radioactive material in the excreta of the worker, without a knowledge of the exposure conditions. Since the correlation between excretion levels and hazard estimation is at present unsatisfactory and since there are at present no good methods of usefully increasing excretion of the wide range of elements involved, a conservative approach is taken. This involves very strict measures to prevent intake and the removal of internally contaminated individuals from working areas when the activity of their urine reaches arbitrary low levels.

Urinary assays are carried out by the Medical Division at Chalk River. The samples are collected at home because it is not practicable to collect them during working hours and not desirable due to the strong possibility of contamination. Since the daily creatinine excretion is relatively constant and related to body weight, this is used in estimating the 24 hour excretion of radioactive material from that measured in the partial sample of urine. The practice is to treat internal radioactive contamination conservatively and to rotate personnel who have radioactive materials in the urine. Workers are removed from contact with loose radioactive substances when the urine activity reaches the following levels:

| *Material* | *Removed from work* | *Ret'd to work* |
|---|---|---|
| 1. Mixed fission products | 150 dpm in a 24 hr sample | 50 dpm |
| 2. Plutonium | 3–4 dpm | 3–4 dpm |

The arbitrary level chosen for mixed fission products is based on the following:

1. It is a level which can be easily measured. It is usually not measured immediately after exposure since examples are not done daily and the time of exposure is not usually known. At this time the urine level would be much higher.

2. The contaminant is presumed to be strontium, in all cases, based on the results of many spot analytical checks. It can be said that mixed fission products in the urine is the most frequent cause of urinary contamination which has been encountered.

Urine tests are carried out on all personnel in certain critical areas every three months. Spot checks are done on some of these individuals every week. Additional tests are done promptly on individuals suspected of being accidentally exposed.

A table of results from the bio-assay laboratory for the year 1953 to 1954 (Table II) is attached.

With this conservative policy regarding internal contamination, a serious attempt is made to prevent such occurrences. This in brief involves the control of air-borne contamination by installation of proper ventilation systems and the prompt cleaning up of spilt radioactive material. Filtration of potentially contaminated air is an important consideration. At Chalk River the Cambridge absolute filter or its equivalent is used for filtering out the fine particles.[4] To preserve the life of this filter it is usually preceded by roughing filters and if acid or moisture is likely to be present, the air is scrubbed and dried. Together with this there are strict rules controlling eating and smoking in the active areas. A controlled respirator and protective clothing programme is in force when work must be done in contaminated areas. The use of respirators gives a certain confidence to the worker so that this equipment must function properly mechanically. Since respirators are uncomfortable to wear for long periods, a certain amount of discipline has to be exercised to prevent their misuse.

## RADIOACTIVE CONTAMINATION

The insidious spread of radioactive contamination is such that original backgrounds are never restored even after prompt and thorough decontamination efforts. Due to the experimental nature of the work at Chalk River equipment and other failures leading to substantial contamination occur from time to time. It is possible to live with such incidents if they are dealt with promptly and to accept under certain circumstances an increased radiation background from fixed contamination.[5] Such an increased radiation

**Table II. Bio-Assay Results (April 1st, 1953—March 31st, 1954)**

| | | *Results: mixed fission products* | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | *Excretion d/m/24 hours* | | | | *Removed from work* | | | | *Plutonium* | |
| *Branch* | *Total tests* | *Neg.* | *100–150* | *150–500** | *500–over** | *Total** | *1–3 weeks* | *3–6 weeks* | *>6 weeks* | *Neg.* | *Pos.* |
| Pile operations | 1833 | 1512 | 139 | 92 | 30 | 122 | 96 | 20 | 6 | | |
| Chemical extraction | 451 | 214 | 73 | 49 | 26 | 61 | 38 | 12 | 11 | | |
| Chemical control | 243 | 83 | 39 | 24 | 19 | 33 | 19 | 8 | 6 | | |
| Process and development | 108 | 60 | 8 | 5 | 4 | 7 | 6 | 1 | 0 | | |
| Isotope production | 221 | 162 | 15 | 13 | 5 | 18 | 11 | 6 | 1 | | |
| Radiation hazards control | 319 | 218 | 31 | 22 | 9 | 30 | 23 | 5 | 2 | | |
| Totals | 3175 | 2249 | 305 | 205 | 93 | 271 | 193 | 52 | 26 | 292 | 31 |

* The three starred columns which should check on addition do not because an individual worker's lost time is accumulated.

background from fixed contamination, in working areas, is acceptable if it does not approach the level which is considered unsafe from the health point of view. In practice a worker receives his weekly radiation exposure by working in radiation fields which are higher than those which would result in the weekly exposure if received at a uniform rate. If the ambient radiation in working areas approached the acceptable uniform rate of 6.5 mr/hr no work in higher fields could then be done without receiving an overexposure. Although it is not the philosophy that each worker gets his full 0.3 r per week, it is often impractical to avoid this in operations, during some weeks.

Incidents leading to local contamination are not difficult to deal with unless the radiation fields are extremely high; however, widespread more or less uniform contamination is a different matter. In this case, it is necessary to have a rapid monitoring method which would indicate the progress of the cleanup over a relatively small area within the contaminated field. This was one of the main monitoring problems encountered in the decontamination of the NRX reactor building; directional monitors and differential monitors were tried and a proper solution has not yet been found. As far as decontamination procedures are concerned, some foresight exercised in the construction of buildings in which radioactive materials are to be used is an important factor. The main part of decontamination is concerned with the removal of radioactive material from surfaces. At one time the use of strippable coatings was favoured and is still worthwhile if such coatings are impervious. Experience has now shown that the important factor is the preparation of the base surface especially in the case of concrete. With a proper paintable concrete surface, any good paint which will bond to the surface can be readily decontaminated.

The use of special agents for decontamination will not be treated here, but suffice it to say that as yet there is no general substitute for a thorough scrubbing with soap and water and detergent. Any decontamination operation results in contaminated material and solutions. These are of the class which can usually be disposed of to the ground.

## CONCLUSION

There have been no significant radiation accidents at Chalk River since operations started. Constant surveillance and strict control are largely responsible. An independent radiation hazards control organization with well-trained personnel in critical positions is considered essential to good control. At Chalk River it is the practice to consider radiation hazards in the early planning of operations. This step has done much to reduce the hazards which always accompany improper planning and design.

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# Radiation Exposure Experience in a Major Atomic Energy Facility

**By H. M. Parker,* USA**

The Hanford Works is one of the major facilities of the US Atomic Energy Commission. It includes a number of high-power nuclear reactors, chemical separation plants, plutonium purification facilities, associated control laboratories and major research and development laboratories. Collectively, the facilities present a good cross section of the radiation hazards that may be expected in peaceful applications of atomic energy. The magnitude of the radiation hazard can be described as the equivalent of handling some millions or tens of millions times the world's available supply of radium.

These facilities have been operated by major industrial organizations under contract to the AEC; specifically, the operator was the DuPont Company from 1944 to 1946, and the General Electric Company from 1946 to date. The radiation hazards have been managed to similar high standards as are reflected in the conventional industrial safety records of such organizations. The exposure experience should thus be broadly similar to that which may be anticipated under private industrial management.

This paper summarizes the experience in exposure of personnel in in-plant operations.

## EXTERNAL EXPOSURE

Exposure to external radiation is measured by methods that are now quite conventional. The X-ray and gamma-ray components are primarily determined by pocket ionization chambers, which are worn in pairs to reduce the incidence of spurious readings due to chamber leakage. It can be shown that the lower of two readings is a valid index, unless an outright recording error is made; high accuracy is confirmed by introducing many chambers of known exposure into the system. The daily exposures so obtained are supplemented by film-badge readings integrated over two-week intervals.

The same film badges record beta-ray exposures through an unshielded aperture over the film. The interpretation of beta-ray data depends on the geometry and extent of the sources. The calibration system is adjusted to give readings equal to or higher than the actual exposure. The separation of beta and gamma exposures in the presence of soft gamma radiation is difficult. For selected operations, such as those involving routine exposure to the 17 kev X-radiation of plutonium, special badges are used.

Some operations, such as laboratory manipulations, involve significantly higher exposure risk to the hands than to the rest of the body. Measurements in these cases depend on film rings, which are essentially miniature film badges separating beta and gamma exposures. In the local experience, data of this type are influenced by such defects as pressure sensitivity of film, chemical effects, and light leaks. The recorded results undoubtedly contain many spurious high readings.

Neutron exposures are measured by means of nuclear track emulsions for fast neutrons, and by boron-lined chambers for slow neutrons. Neither system has the reliability of the gamma-ray measurements. The potential hazard for neutron exposure is low, and the incidence of significant readings has been negligible.

The average annual exposure to gamma radiation for all persons entering radiation areas is given in Table I. It is probable that the entry for 1944–45 is too high, due to initial difficulties with personnel meters and a tendency to record high if in doubt. Also, the average exposure can be held low by a conservative policy of requiring the wearing of meters in areas of low-exposure potential hazard. The data reported assume that 20% of the work in which it is forced to wear, personnel meters, has no real risk of exposure. Despite these defects, the table does show

**Table I. Average Annual Exposure—Gamma Radiation**

| *Year* | *Annual exposure roentgens* | *Percentage exceeding 1 r* | *Percentage exceeding 3 r* |
|---|---|---|---|
| 1944–45 | 0.9 | 0.2 | 0 |
| 1946 | 0.6 | 0.2 | 0 |
| 1947 | 0.4 | 0.3 | 0.02 |
| 1948 | 0.3 | 0.2 | 0 |
| 1949 | 0.2 | 0.08 | 0 |
| 1950 | 0.1 | 0.06 | 0 |
| 1951 | 0.1 | 0.4 | 0 |
| 1952 | 0.2 | 3 | 0.02 |
| 1953 | 0.2 | 5 | 0.07 |
| 1954 | 0.2 | 5 | 0.2 |

* Hanford Atomic Products Operation, General Electric Company, Richland, Washington. Prepared by H. M. Parker; including work by D. P. Ebright, W. A. McAdams, H. A. Meloeny, and other members of the radiological records and standards section, General Electric Company.

the influence of planned control of exposure. This has consisted of an attempt in the early years to reduce exposure as far as possible, and since 1950 to operate to a limit of 3 r per year. The sharp increase in the percentage of exposures above 1 r is a good index of planned relaxation. That the average annual exposure has not risen much, means that the exposure risk of many personnel groups is low.

Table II shows in more detail the number of cases of doses in excess of one roentgen. This illustrates more clearly the effects of the planned relaxation to a 3 r annual target.

Another useful index is the annual record of the ten highest individual annual exposures given in Table III for beta radiation and gamma radiation separately. The combined highest total does not necessarily represent the same cases as given separately.

Table III refers to so-called whole-body exposure. It is known that exposures of hand and forearm may considerably exceed these levels, but the data are not available in form for tabulation.

Table IV summarizes the experience in selected years with neutron exposures.

**Table II. Annual Whole-Body Gamma Exposures above 1 r**

| | *Number of cases** | | | | |
|---|---|---|---|---|---|
| *Year* | *>1 r* | *>2 r* | *>3 r* | *>4 r* | *>5 r* |
| 1944 | 0 | 0 | 0 | 0 | 0 |
| 1945 | 8 | 1 | 0 | 0 | 0 |
| 1946 | 8 | 2 | 0 | 0 | 0 |
| 1947 | 13 | 2 | 1 | 1 | 1 (6.1 r) |
| 1948 | 10 | 2 | 0 | 0 | 0 |
| 1949 | 4 | 0 | 0 | 0 | 0 |
| 1950 | 3 | 0 | 0 | 0 | 0 |
| 1951 | 23 | 0 | 0 | 0 | 0 |
| 1952 | 179 | 22 | 1 | 0 | 0 |
| 1953 | 323 | 42 | 4 | 0 | 0 |
| 1954 | 372 | 68 | 16 | 3 | 1 (14.4 r) |

* Number of cases in any column for a given year includes all the cases in the columns to the right for that year. For example, the 42 cases >2 r in 1953 include the 4 cases which were >3 r. Figures in parenthesis give the highest individual total in the years in which this exceeded 5 r.

**Table III. Yearly Averages of Ten Highest Accumulated Exposures**

| *Year* | *Beta radiation rads* | *Gamma radiation r* | *Beta plus gamma rads* |
|---|---|---|---|
| 1944* | 0.9 | 0.06 | 0.9 |
| 1945 | 4.2 | 1.4 | 5.1 |
| 1946 | 7.5 | 1.4 | 7.9 |
| 1947 | 8.2 | 1.9 | 8.7 |
| 1948 | 9.2 | 1.4 | 9.7 |
| 1949 | 7.5 | 1.0 | 8.1 |
| 1950 | 5.5 | 1.0 | 5.8 |
| 1951 | 6.5 | 1.4 | 7.2 |
| 1952 | 6.9 | 2.6 | 7.5 |
| 1953 | 4.3 | 3.1 | 5.5 |
| 1954 | 4.7 | 4.8 | 6.6 |

* 1944 was an incomplete year.

**Table IV. Neutron Exposures**

| | *1952* | *1953* | *1954* |
|---|---|---|---|
| Fast neutrons: | | | |
| Number of readings | 5034 | 6418 | 6770 |
| Number above 50 mrem | 0 | 0 | 8 |
| Slow neutrons: | | | |
| Number of readings | 5971 | 7101 | 7763 |
| Number above 50 mrem | 0 | 0 | 3 |

**Table V. Relative Hazard of Typical Operations**

| *Type of work* | *Per cent above 1 r* | *Per cent above 3 r* |
|---|---|---|
| Engineering, research | 1.7 | 0.2 |
| Reactor operations | 38.5 | 0.8 |
| Reactor monitoring | 36.9 | 11.9 |
| Reactor maintenance | 10.9 | 0 |
| Separations operations | 4.8 | 0 |
| Separations monitoring | 8.7 | 0 |
| Separations maintenance | 2.0 | 0 |
| Radiation instrument calibration | 58.2 | 0 |

By comparison, approximately 3-million gamma-exposure readings are made per year. The highest recorded neutron exposures in one week have been 220 mrem for fast neutrons, and 176 mrem for slow neutrons.

In reviewing the general picture of external exposure, it is useful to know the relative contributions of different radiation types, and the relative exposure potential hazard of different tasks. For the former, beta and gamma radiations predominate. Near an operating reactor, the transmission through the shield gives approximately equal hazard for gamma radiation and neutron radiation. Both these exposures should be low. The actual exposures come in operation when the reactor is shut down. Exceptions are experimental work on reactor test holes and the operation of particle accelerators. In separations work there is a theoretical neutron hazard from $(\gamma, n)$ reactions, but these are essentially eliminated by shielding for gamma radiation.

Table V gives some indication of the relative hazard for different operations, with gamma radiation as the index. (Per cent annual exposure above 1 or 3 r).

## RADIOACTIVE CONTAMINATION

Experience has shown that the control of contamination is emphatically more troublesome than the control of external radiation. The performance is much less amenable to systematic presentation; only a general discussion is given here. It is compulsory to record the contamination of hands and feet on leaving a radiation area. The frequency of such contamination is shown in Table VI.

In this period, the annual total of cases requiring skin decontamination has ranged between 900 and 1700. The striking reduction in incidence of high counts in Table VI means then that the more recent desirable practice of making frequent contamination

**Table VI. Hand and Shoe Contamination in Typical Years**

| Year | Alpha radiation | | Beta-gamma radiation | |
|---|---|---|---|---|
| | Number of readings | Per cent of high readings | Number of readings | Per cent of high readings |
| 1947 | 332,303 | 0.16 | 410,529 | 0.24 |
| 1948 | 429,163 | 0.29 | 562,166 | 0.23 |
| 1950 | 487,897 | 0.08 | 507,813 | 0.09 |
| 1954 | 563,772 | 0.006 | 720,358 | 0.006 |

checks at the work site has detected most cases before they come to the recording contamination counters. A quick pass over the body with a suitable meter additionally locates small contamination spots other than on hands or feet.

Of major importance to the future atomic energy program is the accumulation of radioelements of long half-life in the body. Such deposits must be detected by bio-assay procedures and there is considerable uncertainty in interpretation especially when the intake may have been progressive over several years. Table VII shows the experience with plutonium.

Significant features of the table are:

1. The high percentage of cases from inhalation, and the insignificance of ingestion or injection (puncture wounds).

2. Forty-five per cent of the cases not associated with known incidents, which may represent slow accumulation under apparently normal working conditions. That the number of such cases has increased sharply in the last two years supports this viewpoint, and emphasizes the need for the most sensitive bio-assay procedures.

Similar data for fission products appear in Table VIII.

In both Table VII and VIII, MPL means the accepted maximum permissible limit (NBS Handbook 52) and "Partial interpretation" means that the urinary elimination has not yet been followed long enough to characterize the body content. In Table VIII, F.P. stands for a general fission product mixture.

In addition, there is a significant incidence of uranium intake in operations involving uranium dust or fume, despite generally high standards in industrial hygiene. This element is rapidly eliminated. Some specialized work with radioisotopes other than fission products has also led to demonstrable depositions. Collectively, these have led to about 3 technical overexposures per year (i.e., deposition *temporarily* exceeded the permanently permissible body burden). It is noteworthy that for no radioelement of long persistence in the body has the deposit exceeded the permissible limit.

## RADIATION INCIDENTS

Untoward incidents with radiation or contamination are recorded in 4 degrees of severity.

1. Serious overexposures—those in which the tolerance status of an individual is affected (as used in NBS Handbook 59). None has occurred; one hand exposure closely approached the limit of 125 r.

2. Technical overexposures—those in which exposures exceeding any recommendation of Handbook 59 has occurred. These have averaged about 4 per year, with an additional 3 per year if the same concept is applied to contamination and internal deposition.

3. Potential overexposures—cases in which technical overexposure could have occurred under similar circumstances. These, averaging 11 or 12 per year for radiation and about 24 per year for contamination, are used for radiation safety training.

**Table VIII. Confirmed Fission Product Deposition Cases in 10 Years**

| Per cent of MPL | Detected by routine samples | Detected after known incident | Radio-element | Route of intake |
|---|---|---|---|---|
| <0.5 | 1 | 40 | 24 Ru | Inhalation |
| | | | 3 Cs | Inhalation |
| | | | 14 F.P. | 13 Inhalation |
| | | | | 1 Absorption |
| 0.5 to 1.0 | 0 | 3 | 3 Ru | Inhalation |
| 1 to 10 | 0 | 1 | Sr | Inhalation |
| 10 to 20 | 0 | 0 | | |
| 20 to 50 | 0 | 1 | Sr | Inhalation |
| 50 to 100 | 0 | 0 | | |
| >100 | 0 | 0 | | |
| Partial interpretation | 8 | 3 | F.P. | Uncertain |

**Table VII. Confirmed Plutonium Deposition Cases in 10 Years**

| Per cent of MPL | First detection by routine samples | First detection after known incident | Route of intake on known incident cases | | | | |
|---|---|---|---|---|---|---|---|
| | | | Inhalation | Ingestion | Absorption | Injection | Unknown |
| <0.5 | 4 | 29 | 25 | 1 | 3 | | |
| 0.5 to 1.0 | 7 | 4 | 2 | | 2 | | |
| 1.0 to 10 | 13 | 13 | 8 | | 4 | 1 | |
| 10 to 20 | 4 | 6 | 5 | | | 1 | |
| 20 to 50 | 0 | 1 | 1 | | | | |
| 50 to 100 | 2 | 0 | | | | | |
| >100 | 0 | 0 | | | | | |
| Partial interpretation | 15 | 2 | | | | | |
| Total | 45 | 55 | 41 | 1 | 9 | 2 | 2 |

**Table IX. Radiation Incidents**

| *Year* | *Radiation* | *Contamination* | *Miscellaneous* | *Total* |
|---|---|---|---|---|
| | **Potential Overexposures** | | | |
| 1944 | 0 | 0 | 0 | 0 |
| 1945 | 3 | 32 | 0 | 35 |
| 1946 | 15 | 24 | 0 | 39 |
| 1947 | 10 | 17 | 0 | 27 |
| 1948 | 16 | 22 | 0 | 38 |
| 1949 | 9 | 27 | 0 | 36 |
| 1950 | 5 | 15 | 0 | 20 |
| 1951 | 8 | 17 | 0 | 25 |
| 1952 | 18 | 46 | 7 | 71 |
| 1953 | 23 | 38 | 8 | 69 |
| 1954 | 30 | 40 | 6 | 76 |
| Total | 137 | 278 | 21 | 436 |
| | **Technical Overexposures** | | | |
| 1944 | 3 | 0 | 0 | 3 |
| 1945 | 5 | 1 | 0 | 6 |
| 1946 | 1 | 3 | 0 | 4 |
| 1947 | 2 | 0 | 0 | 2 |
| 1948 | 2 | 0 | 0 | 2 |
| 1949 | 0 | 0 | 0 | 0 |
| 1950 | 2 | 3 | 0 | 5 |
| 1951 | 9 | 4 | 0 | 13 |
| 1952 | 7 | 4 | 1 | 12 |
| 1953 | 12 | 13 | 1 | 26 |
| 1954 | 9 | 11 | 0 | 20 |
| Total | 52 | 39 | 2 | 93 |

4. Informal incidents — minor deviations from sound control, averaging 140 cases per year, used for training and system improvement.

The incident experience is detailed in Table IX.

A significant feature of the tabulation is the preponderance of contamination incidents in the potential exposure class. If the data are translated to exposed individuals instead of incident numbers, this is even more striking. Each radiation case averages two people and each contamination case averages three.

In the technical overexposure class, radiation cases outweigh contamination cases, and this holds too for numbers of individuals; both types average two individuals per incident.

**Table X. Distribution of Radiation Incidents**

| *Type of work* | *Per cent incidence* |
|---|---|
| Fuel element preparation | 2 |
| Engineering, research | 20 |
| Separations operations | 45 |
| Reactor operations | 20 |
| All other | 13 |
| Reactor operations detail: | |
| Functioning reactor | 4 |
| Reactor shutdown | 11 |
| Auxiliary tasks | 5 |

In the informal incident class, minor contamination cases predominate, with the balance made up of minor infractions of control procedures.

The relative frequency of radiation incidents in different classes of work is shown in Table X.

Of special interest to peaceful applications is the low incidence of exposure with an operating reactor. This suggests that power-producing reactors should be relatively free from radiation hazard. By contrast, separations plants will probably continue to be plagued by contamination hazards that can be held in bounds only by continuous and detailed attention to decontamination, ventilation and other controls.

In summary, the reported experience establishes that radiation hazards can be adequately controlled by methods that are economically reasonable. Evaluation of the total cost of radiation protection is a controversial field, because in addition to the obvious direct costs, one may wish to include factors for slower performance with special handling tools, time spent in planning radiation tasks, etc. Local studies on costs strongly indicate that the additional cost of operating to high standards is a relatively small increment on the necessary cost for marginal protection. This fortunately removes some of the risk of jeopardizing the future of the atomic energy industry by over-zealous competitive reduction of hazard controls. A sensible approach to the future is to accept radiation as a necessary evil of the business, and to seek engineering advances that will achieve the present or improved standards of exposure at lower cost.

# Control of Radiation Hazards in the Operation of Medium-Powered Experimental Reactors

**By W. G. Marley and B. S. Smith,* UK**

The potential health hazards in the operation of medium-powered research reactors are in many ways different from those in the operation of power reactors. The general radiation levels and the levels of contamination round a research reactor have to be kept sufficiently low so that they do not interfere with the experimental work: neutron beams emerging from holes in the shield for experimental purposes present a particular hazard and need special care. Research reactors may frequently have hazardous experimental features, for instance, coolant loops containing liquid sodium. Moreover, technical personnel need to work in close proximity to such reactors for considerable periods of time in carrying out experimental work, whereas, with reactors designed purely for the production of power, the access of persons can be limited and Health Physics supervision may be necessary only during certain maintenance operations. In all these ways experimental reactors present a greater problem in the control of radiation hazards than power reactors. On the other hand, the potential radiological hazards resulting from mis-operation leading to a burnup of part of the core, are likely to be far greater with a power reactor: the possible extent of this hazard is considered in a separate paper.

The control of hazards in the operation of an experimental reactor requires planning at all stages from the inception of the design to the development of the full code-of-practice for routine operation, and necessitates compliance with the following principles:

1. The routine work round the reactor should not result in the exposure of personnel to radiation doses greater than the maximum permissible level recommended by the International Commission on Radiological Protection[1] for occupational workers, namely, 0.3 rem/week (total of slow neutron, fast neutron and gamma radiation) moreover, the work should not result in personnel, within the reactor building or downwind from the reactor, ingesting or inhaling radioactive material at a rate in excess of the maximum permissible level.

2. The rick of over-exposure of personnel to radiation or of exposure to high levels of contamination and airborne activity due to accident, negligence, or ignorance, must be reduced to the practicable minimum.

3. The pile should continue to operate safely with all possible sources of radioactivity properly canned so that the core and shield remain as free as possible from contamination.

The precautions which have to be taken to ensure compliance with these principles, are illustrated below by reference to experience and current practice with the 6 Mw research reactor known as "BEPO" at the Atomic Energy Research Establishment at Harwell. This reactor is a natural uranium, graphite-moderated, air-cooled reactor, the uranium charge comprising rods 1-ft long, approximately 1 inch in diameter, canned in aluminium and carried in horizontal channels spaced about 7 inches apart in the graphite. Surrounding the core is a further 3-ft thickness of graphite, a 6-inch iron thermal shield and 6½ ft of barytes concrete, designed to reduce the radiation level to $\frac{1}{100}$th of the maximum permissible level at the surface of the shield: experience has shown that this external radiation level could have been increased by a factor of 10 without interfering with the experimental work. The graphite and shield are pierced by a number of holes for experimental purposes and loading and unloading of uranium metal and isotopes. Holes in the shield, other than those in use for experimental purposes, are normally filled with stepped plugs which are only removed when the pile is shut down. The cooling air is sucked from the pile and discharged through a bank of filters up a 200-ft stack. BEPO operates at about 6 Mw when the slow neutron flux at the centre of the pile is about $2 \times 10^{12}$ n/cm²/sec.

## MONITORING OF COOLING AIR

With an air-cooled reactor the activity in the cooling air arises from the following causes:

1. Argon-41 produced by activation of the natural argon in the air (this amounts to 50 curies/hour in BEPO).

2. Activation of graphite and concrete dust dislodged from the surfaces of the moderator and the ducts.

3. Small quantities of fission products from uranium contamination on the outside of the fuel elements: this is always present even when there are no holes in the cans.

4. Escape of fission products through the development of a fault in the canning of the uranium.

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5. Occasional escape of material being activated for isotope production.

Experience has shown that fairly elaborate provision for monitoring the air is of very great operational value in allowing the early detection and correction of any unwanted event in the pile. In this way any escape of radioactivity into the core of the pile is checked before it builds up to such a level that the residual activity would prevent the early detection of similar events later. In BEPO the monitoring provision is as follows:

1. The particulate activity of the air stream is recorded continuously by an automatic sampler with a travelling filter paper and geiger counter.

2. Fission-product activity is detected uniquely by means of a "wire machine" which collects the solid decay products of the gaseous fission products xenon and krypton electrostatically on a moving wire, all other particulate activity having been previously filtered from the air.

3. The whole of the cooling air is filtered before discharge and a gamma-ionisation-chamber is installed alongside the filters to detect the build-up of particulate activity on the filters. These filters prevent any hazard arising from the discharge of particulate activity.

4. A further ionisation chamber is provided to measure the $A^{41}$ activity in the gas being discharged.

5. The air from each channel containing uranium cartridges in the pile is monitored daily, thus enabling any channel giving rise to fission product activity to be readily detected and identified. The canned uranium cartridges must be as free as possible from contamination by uranium (preferably less than 5 $\mu$g uranium/cartridge) so that any device for measuring fission products will not have a high background due purely to surface contamination.

## CONTROL OF CONTAMINATION DURING THE UNLOADING OF URANIUM, ISOTOPES AND EXPERIMENTAL EQUIPMENT

The continuous prevention of contamination in working areas must always be one of the principles governing safe reactor operation and this is particularly true with regard to the methods of removal of irradiated material from the pile.

### Uranium Cartridges

Great care must be taken to prevent contamination by fission products not only outside the pile, but also within the individual channels: it is thus of the utmost importance that any faulty cartridge is detected and removed at a very early stage before the channel is seriously contaminated.

In the BEPO pile the only serious burst cartridge, which proceeded to an advanced stage, occurred about two years after the original start-up of the pile: in this incident the oxidation of the uranium, starting at a small pinhole in the end weld, proceeded to such an extent that it caused a longitudinal split 4-inches long in the can. The extent of the burst arose from failure to locate expeditiously the channel giving rise to the detected activity. The removal of any cartridge damaged to this extent needs considerable care and advance planning in order to prevent the activity spreading over the operating area. In this incident the dust hazard was kept to a minimum by constant vacuum cleaning of the channel and surrounding area, and gamma radiation was minimised by the use of a special shielding container into which the damaged cartridge was sealed. The improved devices already described for burst cartridge detection have resulted in all subsequent bursts being readily detected in their early stages and no precautions beyond those normally taken in the unloading of uranium metal from the pile have been necessary during the removal of damaged cartridges.

### Isotope Irradiations

Aluminium is often used as a container during irradiations on account of its low cross section and short half-life so that it does not acquire a high long-lived activity. The short half-life has the disadvantage, however, of producing a high initial activity which, in the case of the standard 3-inch cans used in BEPO, may be over 2 curies and is often greater than the activity of the irradiated material. It is often possible, when unloading a sample whilst the pile is in operation, to withdraw it into the shield to allow the short-lived activities to decay before finally unloading it.

The design of mechanisms for the irradiation of material requires great care to afford the full facilities for loading and unloading of isotopes, whilst the pile is operating, without significant radiation exposure to the operator and without irradiated parts being unnecessarily withdrawn from the pile shield. An improved irradiation facility, which allows any one of the many samples being irradiated in a single channel to be withdrawn, without subjecting the operator to the radiation from the other samples, has recently been used in BEPO. It consists of a series of graphite blocks in 1 in. $\times$ ½ in. $\times$ 3 in. high hinged together to form a continuous belt. The belt is mounted in a channel reaching from the outside of the shield to the far side of the core. The isotope cans are placed in holes in the graphite blocks and the belt is rotated to bring the cans into the region of high flux in the core. As well as the radiation hazard involved in the unloading of samples and experimental equipment, there is a contamination hazard due partly to the adherence of active graphite (this being greater if the can is greasy) and partly due to the activation of dust on the surface of the can. In either case this contamination hazard can be considerably reduced by thorough cleaning of all the equipment before insertion in the pile.

### Handling of Contaminated Graphite

The handling of irradiated graphite presents potential hazards from gamma radiation and also from contamination. The former arises from the activation of trace impurities and may range up to 1 r/hour at a distance of 1 ft after lengthy irradiation in flux

of $10^{12}$ n/cm²/sec. Care must be taken to prevent spread of contamination in the handling of irradiated graphite. Clothes which have only brushed lightly against a graphite block sometimes give several hundred counts per second on the monitor, without showing any visible mark, and gloves used during actual handling may become contaminated to the order of 0.1 to 1 μc/cm². To prevent the spread of contamination in the handling of graphite, protective clothing and the use of paper covering on the floor, scrupulous cleanliness and an extensive use of the vacuum cleaner are necessary.

### Permissible Contamination Levels in Pile Experimental Area

Health considerations require the fixing of maximum permissible levels of contamination[2] in the working area. The practice in the BEPO pile area is to preserve a level always not exceeding $10^{-4}$ μc/cm² or 5 counts/second on a geiger-counter monitor (AERE Type 1021), except when unloading is taking place during a pile shutdown. At these times a temporary area of contamination is allowed not exceeding $10^{-3}$ μc/cm² (50 counts/second) and operators are required to wear overshoes and protective clothing, and widespread use is made of temporary paper floor-covering.

Monitoring for contamination in a reactor area is complicated by gamma background-radiation which arises from scattered radiation from open experimental holes and from storage of irradiated samples, equipment, etc., in the area. External radiation levels which are biologically permissible may prevent the monitoring for contamination down to the required levels. Smear testing (i.e., rubbing an area of about 100 cm² with a small filter paper and counting it in a shielded counter) is used for the monitoring of surfaces in areas where there is a high gamma-background and is also useful in distinguishing between loose and fixed contamination. A counter shielded by 3 in. of lead and mounted on wheels has greatly facilitated the direct monitoring of floors down to the required levels.

In a reactor running at reduced air pressure such as BEPO, where air is sucked through the reactor, there is little likelihood of gaseous or particulate activity leaking out of the pile into the reactor building. There are therefore no fixed air-sampling devices installed round this reactor. Operations suspected of giving rise to airborne activity are individually monitored using portable air samplers. During the unloading of any materials from the BEPO pile, one auxiliary exhauster is always kept running, thus maintaining the pile at reduced pressure and reducing the escape of any active dust into the working area.

## THE VALUE OF FIXED HEALTH INSTRUMENTS AND THE USE OF PORTABLE MONITORS

It is sometimes argued that fixed health-instruments serve little purpose, as they will not directly pick up stray beams of radiation from the reactor and, unless the general background is kept very low, the scattered radiation from these beams is not intense enough to be detected. The fixed health-instruments comprising gamma and slow-neutron monitors recording in the control room of the BEPO pile have been found to be well worthwhile. Fixed gamma-monitors are particularly useful in areas where a large amount of unloading of isotopes and equipment takes place: they ensure that radioactive material has not been left about unshielded with subsequent exposure of people unaware of its presence. The BEPO health ionisation chamber records are read every hour by the pile operator and any changes in the radiation level in the working area are investigated. Several instances have occurred in which the fixed gamma health-monitors have been of very great value in calling attention to faults which have developed or mistakes which have been made relating to active materials or to shielding; for instance, on one occasion these instruments drew attention to water having drained out of an experimental shielding tank whilst the pile was running steadily. Instances have occurred where holes, although adequately shielded for gamma and slow-neutron radiation, did not have the necessary slowing-down material for fast neutrons and, as a result, high fast-neutron fluxes occurred in the working area. An installed fast-neutron monitor would perhaps have enabled these also to have been immediately detected from the control room during start-up.

The portable health instruments[3] at present in use in BEPO include battery-operated monitors using air-filled ionisation chambers for gamma measurement, similar battery-operated instruments but using boron trifluoride ionisation chambers for slow-neutron measurement, and mains-operated, semi-portable instruments using proportional counters for fast-neutron measurement.[4]

## HAZARD FROM RADIATION BEAMS FROM OPEN HOLES IN THE PILE SHIELD

Many experiments require beams of radiation from the pile. The intensity of gamma, slow-neutron and fast-neutron radiation in such beams depends on the amount of graphite in the hole and on the size of the hole: typical figures for a 2 in. beam from the centre of the BEPO pile where the slow-neutron flux is $2 \times 10^{12}$ n/cm²/sec are: fast neutrons—$10^3$ rem/hr; slow neutrons—200 rem/hr; and gamma radiation—10 rem/hr. The beam path usually terminates in a "beam trap," a typical type being a 30-inch concrete cube with a cavity in one side partially filled with lead and borax. A beam trap can be designed to scatter little radiation but there is no advantage in improving its efficiency beyond the point where it scatters little compared with the experimental apparatus in the beam. Air, alone, scatters about 5 per cent of the neutrons from a beam per metre path, and an ionisation chamber with a 0.5 cm steel wall will scatter about 40 per cent of the incident beam. On the main experimental face of the BEPO pile, where a number of

beams are being used, it is often found that the scattered radiation exceeds the acceptable health levels without causing serious mutual interference between experiments: by the erection of the appropriate shielding round particular experiments the general background about 6 ft back from the pile is kept below the maximum permissible level.

It is difficult to define the extent of the hazard from accidental direct irradiation of body from the radiation beams. It is well established, however, that the eyes are sensitive to fast neutron irradiation so that it is essential to prevent persons sighting along the line of the beam even for a matter of seconds when the pile is in operation. A few seconds exposure of the body to the beam, however, may be tolerated, in conformity with the recommendations of the International Commission of Radiological Protection.

## HAZARDS OF SPECIAL EXPERIMENTAL EQUIPMENT

Facilities are frequently required in a research reactor for tests with experimental heat-transfer circuits in which liquid metal is circulated. In view of the potential hazards, arising from inflammability and consequential inhalation risk and the intense gamma radiation, these experimental circuits require special precautions. The practice with experimental sodium-circulation circuits in the BEPO area is to enclose the whole of the pipework lying outside the pile shield in a special shielding enclosure, no section of which is removed without specific Health Physics supervision.

To guard against the release of fumes into the building in the event of a sodium fire in this shielded enclosure, the enclosure is made air-tight and vented through a filter to the pile-exhaust system. A recording gamma-monitor, installed adjacent to the box containing the filter, operates an alarm bell and warning light on the main control panel if the radiation level exceeds a certain preset limit. An alarm circuit also operates off the header-tank gauge to indicate loss of sodium from the system. Protective clothing and fire-fighting equipment are kept available adjacent to the experimental equipment in conformity with the laboratory "Sodium Safety Code" and a warning system and emergency procedure for dealing with a fire or an escape of sodium was instituted.

## GENERAL RECOMMENDATIONS FOR THE CONTROL OF RADIATION AND CONTAMINATION HAZARDS

For an experimental reactor with its large number and diversity of operations the best method of control of radiation and contamination hazards appears to be by a small health physics staff working in close co-operation with the engineering, operating and experimental staff. All staff working in the area should be trained both in elementary health physics principles and in the use of health-monitoring instruments, so that they can appreciate and avoid the type of hazards which arise in normal operations: they should routinely seek further advice from health physics personnel for any new or unusual type of operation. A very large health physics staff would have to be available if all operations were to be under their direct supervision.

It is necessary to keep careful and accurate records of all modifications which are made to experimental facilities during shut-down periods. These records should be checked before start-up of the pile to ensure that all tasks (particularly those affecting radiation safety) have been properly completed and the engineer in charge should confirm the records by a visual check at the pile face. With the variety of shielding plugs used on BEPO, which are not all equally effective for all types of radiation, experience shows that it is also necessary to check with monitors the actual radiation intensities at the pile faces after reaching operating power.

Fast-neutron (track count) and slow-neutron monitoring films should be worn, in addition to the regular gamma-monitoring films, by all personnel working regularly on the reactor face during its operation.

Direct-reading pocket gamma ionisation chambers should also be worn by all personnel engaged in the unloading of experimental equipment, isotopes and irradiated uranium metal. These are invaluable in helping to keep the gamma-radiation exposure to a minimum, as the dose received can be checked as the job proceeds and additional precautions taken if necessary.

Regular surveys for contamination should be carried out, particularly after unloading has taken place from the reactor. Air samples should be taken during all operations involving the risk of airborne activity.

The control of radiation and contamination hazards around the BEPO pile has been effectively and economically maintained over many years by adherence to the principles outlined in the paper.

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# Approaches to Treatment of Poisoning by Both Radioactive and Nonradioactive Elements Encountered in Atomic Energy Operations

By J. Schubert,* USA

The lethal effects of radioactive isotopes were shown early in 1904 by Pierre Curie and two medical colleagues.[1] They exposed experimental animals to atmospheres of radon and found that death followed in a matter of hours. In the same experiment Curie demonstrated by means of radioautographs that the radioactive daughters of radon were present to varying extents in all the tissues: it is now known that these contribute to the radiotoxicological effects.

Probably the first human death from acute radioactive poisoning was reported by Gudzent in 1912.[2] It was hoped that radium injections would cure a woman's arthritis. Instead, 26 days after her last injection she died after exhibiting the typical symptoms of radiation sickness, including bloody stools, hemorrhages, and diarrhea.

Radioactive isotopes when swallowed, inhaled, or injected may cause acute or chronic injury or death solely because of the ionizing radiation emitted by these isotopes within the body. Radioactive poisoning of a chronic type is a slow insidious disease in which a person is usually in good health for many years before detectable symptoms occur. The resulting injuries include various types of anemias, leukemia, crippling lesions of the bone, and bone tumors.

Radioactive substances can produce injury when either outside or inside the body. However, external irradiation is easily avoided by removal of the source or by surrounding the radioisotope with sufficient shielding. When the radioisotope is contained in the body it constitutes a far greater hazard because the internal tissues are irradiated continuously until the isotope is eliminated in the urine and feces, or else loses its radioactivity by natural decay. The most dangerous radioisotopes are those retained in the body for periods of years and which possess half-lives ranging from a few months to thousands of years.

Until 1939 radioactive poisoning in humans was caused almost entirely by radon ($Rn^{222}$) and three naturally occurring isotopes of radium–radium ($Ra^{222}$), mesothorium ($Ra^{228}$), and thorium X ($Ra^{224}$). The discovery of nuclear fission and chain-reacting piles resulted in a tremendous increase in both the kinds and amounts of potentially hazardous radioisotopes. Consider the following facts. The production of radium from the time of its discovery in 1898 to the present totals about one and a half kilograms or 1500 curies. Death or injury from pure radium retained in the body has been observed with as little as 0.5 microcurie. Nuclear reactors now produce hundreds of different radioactive isotopes in megacurie amounts.[3] In 1954 the United States Atomic Energy Commission shipped 15,000 curies of radioisotopes from Oak Ridge to be used in research, industry and medicine.

The construction of nuclear reactors involves the use of many nonradioactive materials possessing

**Table I. Potentially Harmful Elements Encountered in Atomic Energy Operations**

| *Element* | *Chemical symbol* | *Fission yield %* | *Half-life* | *Principal radiation emitted as internal hazard* |
|---|---|---|---|---|
| **Radioactive Elements*** | | | | |
| Plutonium | $Pu^{239}$ | – | 24,360 yr | α |
| Radium | $Ra^{226}$ | – | 1622 yr | α |
| Radon | $Rn^{222}$ | – | 3.825 d | α |
| Polonium | $Po^{210}$ | – | 138 d | α |
| Iodine | $I^{131}$ | – | 8.1 d | $\beta^-$ |
| Cobalt | $Co^{60}$ | – | 5.3 yr | $\beta^-$ |
| Phosphorous | $P^{32}$ | – | 14.3 d | $\beta^-$ |
| Carbon | $C^{14}$ | – | 5600 yr | $\beta^-$ |
| Promethium | $Pm^{147}$ | 2.6 | 2.6 yr | $\beta^-$ |
| Cerium | $Ce^{144}$ | 6.0 | 282 d | $\beta^-$ |
| Cesium | $Cs^{137}$ | 5.9 | 33 yr | $\beta^-$ |
| Ruthenium | $Ru^{106}$ | 0.4 | 1.0 yr | $\beta^-$ |
| Strontium | $Sr^{90}$ | 5.8 | 28 yr | $\beta^-$ |
| **Nonradioactive Elements†** | | | | |
| Beryllium | Be | | | |
| Zirconium | Zr | | | |
| Gadolinium | Gd | | | |
| Cadmium | Cd | | | |
| Lead | Pb | | | |
| Barium | Ba | | | |
| Uranium | U | | | |
| Thorium | Th | | | |
| Molybdenum | Mb | | | |
| Bismuth | Bi | | | |

* Data on fission yields and half-lives of fission products provided by Glendenin and Steinberg, Personal Communication. The half-lives of the other elements are taken from the tables by Hollander, *et al.*, Rev. Mod. Phys. 25: 469-651 (1953).

† Based on information in References 4 and 5.

* Argonne National Laboratory. Including work done by Marcia White Rosenthal, Argonne National Laboratory.

varying degrees of toxicity. Chemically poisonous materials used in the construction of nuclear reactors can be classified with respect to their function in a nuclear reactor: (1) nuclear fuels; (2) moderators and reflectors; (3) coolants; (4) structural materials, and (5) shielding materials.

In Table I are listed those elements which are the most serious health hazards in atomic energy operations, both from the standpoint of quantities available and amount of use. The associated health problems exist potentially for miners and millers of uranium and other radioactive ores, personnel of atomic energy plants, residents of areas in which radioactive sewage and wastes are released, people administrating or receiving radioactive isotopes in medicine, users of radioactive isotopes in research and industry. Industrial uses where health hazards may exist include[6] the preparation and application of luminous paints, static eliminators, and gas mantles made from thorium. Other uses of radioactive isotopes are in radiographic units, nasopharyngeal applicators, and as radiation sources for food and pharmaceutical sterilization and other purposes.

Despite the widespread and increasing handling of radioisotopes it is possible to handle even megacurie amounts safely if suitable precautions are taken. This is proven by the remarkable safety records of atomic energy plants in the United States[7] and other countries. The numerous cases of radium poisoning in the self-luminous dial painting industry reported in the medical literature between 1925 and 1931[8] nearly ceased when proper ventilation and other safety measures were instituted. However, even under the best conditions, accidents can and do happen. As a result, radioactive isotopes gain entrance into the body by ingestion, inhalation, or by injection into the bloodstream usually through the broken skin.

The necessity for the development of methods of treating radioactive poisoning—particularly the chronic type—is demonstrated by the fact that even in the past decade significant, though not necessarily harmful, levels of radioisotopes have gained entrance into numerous individuals, including the following cases:

1. A group of uranium miners were reported in 1954 to have accumulated in the body polonium ($Po^{210}$) derived from daughter products of radon deposited in the lung.[9]

2. In 1953 it was reported that in 27 individuals the body levels of plutonium (Pu) ranged from 0.1–1.2 micrograms.[10]

3. Strontium ($Sr^{90}$) was inhaled during a laboratory operation.[11]

4. Radium sulfate dust was inhaled by a total of seven persons in three different accidents.[11,12,13]

5. A solution of radium salts was injected intravenously during a suicidal attempt.[13]

## SCOPE OF PAPER

In most cases, the poisonous elements with which we are concerned are metals. Consequently, the discussion is limited mainly to poisoning by radioactive and nonradioactive metals. Effective treatment for chemical poisoning by metals is available in many cases. Successful treatment, however, for persons who have accumulated radioactive metals in the body is still severely limited except in the earliest stages after exposure. It is our intention to discuss: (1) similarities and differences in approach inherent in the treatment of nonradioactive metal and radioactive metal poisons; (2) the potential advantages and disadvantages of different treatments; and (3) new or untried possibilities for treatment.

## GENERAL APPROACHES TO TREATMENT

By treatment we mean the prevention, elimination, or alleviation of the harmful effects caused by a poisonous substance. In the treatment of radioactive and nonradioactive poisoning we must deal with many interrelated factors.

Once the route of entry into the body and the chemical nature of the poisonous substance is known, then choice of treatment is partially governed by a time element. Immediately after exposure the object of treatment is to minimize absorption of the poison into the bloodstream. After absorption is complete the object of treatment may differ depending on whether the poison is radioactive or nonradioactive. In the case of a nonradioactive poison one can either hasten its rate of elimination from the body or inactivate the poison *in situ* so that it can no longer react with surrounding tissue. However, with radioactive poisons the main object is to accelerate excretion or possibly to shift the radioelement from radiosensitive organs to less radiosensitive organs in acute cases, however, from the chronic standpoint all tissues may be roughly equivalent in susceptibility to radiation effects. It is not possible to change the radioactive properties so that inactivation *in situ* is impossible. A treatment designed to remove a radioisotope from the body may either remove it from different organs or may only cause the excretion of the circulating fractions. It is always helpful to determine the total amount of a radioisotope in the blood just before the onset of treatment.

In the sections to follow the problems of treatment are described in more detail. It is well to emphasize that no one treatment can always be generally applied but the proper course must be evaluated in each case; good treatment for one case may be harmful in another, even though the same poison is involved. One must take into consideration factors such as the chemical nature of the metal poison, route of entry into the body, age and health of the patient, time elapsed after exposure, and other factors which will become evident.

The treatment of chronic radioactive poisoning is an extremely difficult problem because the excretion rate of the radioisotope is very low—0.002% a day or less in many cases. Even if one induces a tenfold increase in the rate of elimination, the actual amounts retained in the body remain essentially unchanged.

In order to reduce appreciably the deposits in the body, treatment for chronic cases would have to be sustained for several years. This presents many practical problems, such as cooperation of the patient and the effects of prolonged treatment on health.

The chances of reducing the body content of a radioisotope are far greater in the early stages after exposure. During the first few weeks after exposure it is generally found that significant fractions of the deposited radioisotopes are present in the soft tissues and bloodstream from which they are more readily mobilized than from the bone. Also, those fractions in the bone, at least in the case of radium and the other alkaline earths, are more readily mobilized at this time. This was explained by Aub and his associates as follows:[14]

"At first, a heavy metal is stored largely in the bone trabeculae, there being from 10 to 16 times as much per gram in the trabeculae as in the cortex of the bone. During the following months there is a redistribution and the concentration becomes equal in both trabeculae and cortex. Because the cortex is far heavier than the trabeculae, this means that most of the radium is stored in cortical bone. . . . The wide distribution in soft tissues and then the accumulation in the bones implies considerable circulation in the blood and, therefore, a chance for rapid excretion. The first large storage in the trabeculae (where inorganic salts are readily deposited and also readily liberated) would allow a continued though less rapid excretion. When the radium finally accumulates in the cortex, it is to be expected that excretion would be slow and relatively poorly influenced by therapy."

A striking illustration of the transport of calcium and lead from trabeculae and cortex in animals has been reported recently by Aub *et al.*[15] In these experiments alizarin staining was used to follow calcium metabolism while radiolead enabled lead distribution to be followed.

It is not certain, however, that a significant degree of translocation of radioisotopes from the trabeculae to the cortex occurs in adult humans. Radioautographs of bones in humans taken 20 or more years after exposure show highly concentrated deposits of radium in trabecular bone.[28] Also, deposition of an alpha-emitting isotope tends to destroy the trabeculae.[8]

## TREATMENTS FOR RADIOACTIVE POISONING

### Minimization of Absorption

#### Oral Ingestion

Treatment, if necessary, must start as soon as possible. If results from experiments on rats can be extrapolated to man, the only way to minimize absorption of soluble radioisotopes, such as radiostrontium, is to start treatment immediately. Treatment is ineffective if begun only 10 minutes after ingestion.[17] After oral ingestion, there are three obvious approaches to treatment: (1) use of an emetic; (2) minimization of absorption from the gastrointestinal tract by the use of precipitating agents, and (3) hastening elimination of the radioactive element by the use of selected cathartics. A knowledge of the chemical properties of radioactive elements is extremely important when choosing a suitable precipitating agent.

In the case of elements which form insoluble hydroxides at physiological *p*H, treatment, except for a cathartic, may not be needed because absorption from the gastrointestinal tract is very small. In the case of plutonium and the rare earths, for example, absorption is usually less than 0.01%. Therefore, for Pu, Po, Ce and Pm the use of precipitating agents can probably be omitted. When very large amounts are swallowed it might be advisable to decrease the solubility of the radioisotope by raising the *p*H of the gastric juice by oral ingestion of non-systemic antacids such as magnesium oxide, "Sippy powders," or aluminum hydroxide. An antacid in combination with a cathartic would appear to be the best approach. In the case of the rare-earth elements, which are more soluble than plutonium, it is possible that oxalate-containing foods such as rhubarb and spinach or a finely divided carboxylic-type cation exchange resin such as Amberlite IRC-50† may bind the radioisotopes. In cases of oral ingestion it may be advisable to avoid the use of solubilizing chelating agents. Nonspecific adsorbents such as zirconium oxide might be of value.

The alkaline-earth group of elements are absorbed to a significant extent from the gastrointestinal tract. Since Ra and Sr form insoluble sulfates, the treatment of choice would appear to be magnesium sulfate in combination with a laxative and possibly an antacid as well. In rats, MacDonald and associates[18] have shown that oral ingestion of magnesium sulfate by rats *immediately* following the administration by stomach tube of strontium as $Sr^{90}$ decreased the skeletal accumulation of $Sr^{90}$ from 16% to 5%. Also effective were the ammonium salt of an amido-polyphosphate and carboxylic-type cation exchange resins. Certain substances, including powdered milk, actually enhanced the absorption of $Sr^{90}$ from the stomach. The chelating agent, ethylenediaminetetraacetic acid (EDTA) was ineffective.

Copp and Greenberg[17] found that a 5% suspension of tricalcium phosphate in 12½% disodium phosphate solution reduced the intestinal absorption of Sr only when given immediately after the Sr. None of the following orally administered agents were effective when given 10 minutes after the ingestion of radiostrontium by rats: carrier strontium, magnesium sulfate, sodium silicate, sodium bicarbonate, or disodium phosphate.

Experiments with rats have indicated that the maintenance of a high level of calcium intake decreases absorption of Sr and presumably Ra from the stomach.[19]

† Produced by Rohm and Haas Co., Washington Square, Philadelphia, Pennsylvania (USA). The resins are available in low cross-linked forms which undergo considerable swelling in solution. The bulk furnished by the swelled resins may increase the efficiency of removal of the radioisotope from the gastrointestinal tract.

### Inhalation

The inhalation of soluble forms of radium and strontium is nearly equivalent to an intravenous injection. Absorption from the lungs is so rapid that treatment is largely similar to that required following injection into the bloodstream. However, the upper nasal passages do retain some of the radioisotope. In one case[11] where a human inhaled $Sr^{90}Cl_2$ from an aqueous solution, the upper nasal passages were sprayed with 0.25% neosynephrine solution with the thought that vasoconstriction might lessen absorption and that shrinkage of the mucous membranes would facilitate a thorough washing. The nasal passages were then washed out with large quantities of isotonic saline solution. About 95% of the $Sr^{90}$ retained in the nose region appeared to have been removed. The use of nasal swabs to remove material clinging to the nasal hairs before irrigation should be valuable. However, as shown by high urine counts, most of the inhaled $Sr^{90}$ was absorbed.

Following inhalation of insoluble types of radioactive elements, retention in the lungs depends on the particle size of the inhaled substance. The first step in treatment of fractions retained in the lung is to swab thoroughly around the vestibule of the external nares with a short applicatory stick around which is wrapped moistened cotton or filter paper.[10] In addition it appears advisable to administer a laxative, as was done by Rajewsky and Muth,[13] because a large amount of the insoluble material in the lungs is propelled to the mouth, swallowed, and excreted in the feces. The use of expectorants may also help to hasten elimination.

Drugs which enhance macrophage movements might accelerate the elimination of insoluble particles from the lungs. Easton[20] claims to have accelerated the elimination of intravenously injected thorium dioxide deposited in the lungs of mice. He used carbon tetrachloride—obviously impractical for humans—as a general accelerator of macrophage movements.

The biological half-life of insoluble substances in the lungs may be months. During this time a slow absorption of the radioelement from the lungs to the bloodstream occurs.[16,17] In such an event it would be helpful if the excretion of the circulating radioisotope could be increased. For Pu and the rare-earth elements, at least, this could probably be done by periodic intravenous administration of a combination of zirconium citrate and ethylenediaminetetraacetic acid as is described later. However, the usefulness of these agents in such cases is difficult to assess without experimental evidence.

### Puncture of Skin

Following puncture of the skin with an object contaminated with radioisotopes one can only minimize absorption of the fraction retained at the puncture site. Excision and irrigation of the wound site appear most practical. Details concerning the treatment of contaminated wounds are described by Hathaway and Finkel.[21]

## Absorbed Radioisotopes

### Decalcification Therapy—Interference with Calcium Metabolism

In 1926 Aub and Minot[22] postulated that lead metabolism in the body parallels that of calcium and that the circulation of lead could be controlled by influencing calcium in the bone. The metabolism of the alkaline earths including radium and strontium is also very similiar to that of calcium. Consequently it occurred to Flinn in 1926[23] that the decalcification techniques of Aub could increase the excretion of radium. He treated three patients with parathormone extract which, he claimed, increased radium excretion in patients who had accumulated radium in their bodies several years previously.

In 1931 Flinn[24] attempted to cause repair of radium-produced necrotic bone by the administration of viosterol. Since viosterol presumably promotes the laying down of calcium on the bone, Flinn was surprised to find, according to his measurements, that as much as 75% of radium in the body was removed.

The observations of Flinn were not confirmed in studies on four radium dial painters made by Craver and Schlundt.[25] They found that viosterol plus a high calcium diet produced no increase in radium elimination, but that the administration of parathyroid extract with a low calcium diet produced about a 50% increase in radium elimination during the period of treatment.

The definitive, classical study of the effects of alterations in calcium metabolism on radium excretion in human subjects was made by Aub and co-workers.[14] It was pointed out—and rightly so—that the assay techniques of Flinn were not reliable and that the doses of parathyroid employed by Craver and Schlundt were not large enough to affect calcium metabolism sufficiently.

In a carefully controlled series of experiments on three patients Aub *et al.*[14] found that decalcification therapy increased the urinary excretion of radium four- to eight-fold. Treatment consisted of a low calcium diet plus ammonium chloride, thyroid extract, and parathyroid extract. Magnesium gluconate administration also raised the excretion of radium in proportion to its effect on calcium excretion.

One case did not respond to parathyroid extract. The explanation given[14] was that "her osteoclasts were sufficiently damaged so that they were unable to respond to the normal stimulus of parathyroid extract, and were, therefore, unable to liberate bone salts."

When the patients were placed on a high calcium diet with no medication the calcium excretion returned to a very low level but the radium excretion continued high. It was thought that this might have been caused by previous administration of Vitamin D. It is important to note that radium excretion does not always parallel the change in calcium excretion. The excretion of radium does not change as rapidly as that of calcium. One reason given[14] is that when

radium is liberated from bone some is stored in the soft tissues. The rate of elimination from soft tissues, as observed in acute cases of radium exposure, is continuously high.

Decalcification therapy has been tried in experimental animals in attempts to promote the excretion of strontium. Small increases in excretion rate were observed but these were at too slow a rate to have any appreciable effect on the residue left in the body.[19] A low calcium diet unaccompanied by other medication, such as ammonium chloride, appears to have little value for increasing removal of radioactive alkaline-earth metals. In one experiment[27] a group of rats were injected with $Ca^{45}$ and placed on a diet adequate in phosphate but very low in calcium. In these animals the excretion of $Ca^{45}$ was actually reduced. In human subjects, Aub *et al.*[14] also found that the sole use of a low calcium diet had no effect on radium excretion. These findings are reasonable when it is considered that on a low calcium but adequate phosphate diet the body attempts to conserve its store of calcium.

No effect at all of decalcification therapies has been observed on the excretion rates of plutonium and the rare-earth elements.[16,17] Many experiments have shown that the uptake and excretion of Pu and the rare earths are unaffected by age of the animal or by alterations in the degree of mineralization of the bone. Even in bones of rats which are demineralized to the point where the bone contained only one-fourth of the bone salt normally present, no effect on the retention of injected Pu, Y, or Ce is found.[19] In the same experiment, when the rats were placed on a phosphate-deficient diet immediately after injection of the radioisotope, the body retention of Sr is reduced from 76% to only 26%. Similar findings were reported in comprehensive reports by Van Middleworth[26] and Copp and co-workers.[27]

In summary, decalcification therapy is capable of increasing the elimination rate of radium and presumably of strontium in acute and chronic poisoning. In acute exposures to Ra and Sr the increases in elimination caused by decalcification therapy could lower the body content appreciably. However, such therapy appears impractical in the treatment of chronic poisoning because treatment can be given only for limited periods, so that the net depletion of body content of a radioisotope is insignificant. Decalcification therapy has no effect on the retention or excretion of plutonium and rare-earth elements. There is no conclusive evidence to show that decalcification therapy might be useful clinically even if it did not accelerate the elimination of radioisotopes.

### Complexing or Chelating Agents

When a metal is complexed by different chemical compounds, usually organic in nature, a poorly dissociated molecule is formed. When the complexed metal becomes bound in such a manner as to become part of a ring system it is called a chelate. We will use the terms complex and chelate interchangeably, though the term complex is broader, i.e., all chelates are complexes but not all complexes are chelates.

A chelated metal ion does not undergo its normal chemical reactions except with substances which may possess a stronger affinity for the metal ion than that possessed by the original chelating agent. If the chelate is insoluble and non-diffusible it prevents the injurious effects of the metal ion in the animal body. If the chelate is water soluble and readily diffusible it is readily excreted. Consequently, there are two rationals for the use of complexing agents in the treatment of metal poisoning: (1) The complexing agent selectively combines *in situ* with the toxic metal without removal; and (2) it removes the metal ion from combination with cellular constituents with subsequent elimination of the complexed metal from the body. Only the latter is useful for the treatment of radioactive metal poisons.

### Historical Notes

In general, only the earliest papers are mentioned in this section. The more recent literature is covered in the following sections in which specific results are described.

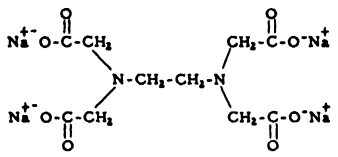
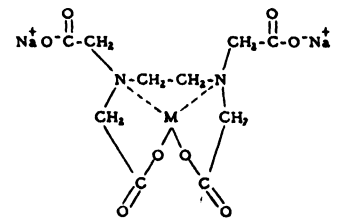
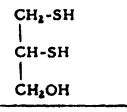
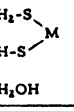
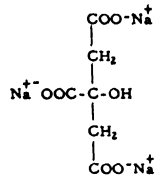
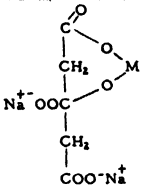
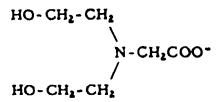
In the treatment of chemical metal toxicity, many investigators in the past had used complexing agents, but for reasons other than their ability to react directly with the metal. The deliberate use of complexing agents because of their metal-binding properties for the treatment of heavy-metal poisoning was begun in 1940.

During the war years the British developed 2,3-dimercaptopropanol, called BAL (British anti-lewisite), to control the harmful effects of arsenical war gases.[29] BAL was subsequently used successfully for the treatment of poisoning by those heavy metals, including arsenic, mercury, and bismuth, which apparently exert their toxic actions through combination with the thiol groups in enzyme proteins. BAL competes successfully for the –SH groups through the formation of a chelate (see Table II). The BAL-metal combination is readily excreted.

About the same time BAL was employed, Kety[30-32] attempted to treat lead poisoning by the administration of sodium citrate. He observed that lead forms a fairly strong complex ion with citric acid and measured its stability.[31] It occurred to him that the phenomenon of complex formation "might be applicable to the problem of dissolving the insoluble lead compounds of the body tissues, making more lead available for excretion with no increase in lead ion concentration."[31]

In recent years there have become commonly available an extremely powerful group of chelating agents —the polyamino acids. One of the most noted of these agents is ethylenediaminetetraacetic acid (EDTA), shown in Table II. This compound forms especially strong chelates with polyvalent cations. The resulting chelates are water soluble and un-ionized. As early as 1942 biological experiments were carried out with EDTA; its ability to inhibit blood coagulation *in vitro*

**Table II. Four Important Chelating Agents Used in Biology and Medicine**

| Chelating agent | Molecular weight | Structure | Type of metal chelate (M = metal ion) |
|---|---|---|---|
| Tetrasodium salt of ethylenediaminetetraacetic (EDTA) | 380.2 |  |  |
| 2,3-Dimercapto-1-propanol or 1,2-Dithioglycerol (BAL) | 124.2 |  |  |
| Sodium citrate | 258.1 |  |  |
| Fe-3 (Commercial chelating agent manufactured by Bersworth Chemical Co., Framingham, Mass.) | |  | |

by complexing calcium was studied.[38] In 1949 Bersworth suggested that EDTA be used for removing metals from the body (p. 509 of ref. 34). It has been found that EDTA causes marked increases in excretion of several metals. It has been especially successful for the treatment of lead poisoning (pp. 506–510 of ref. 34; 35).

Probably the first deliberate attempts to remove radioisotopes from the body by means of complexing agents were made in Chicago beginning about 1945.[36,37] Painter *et al.* (pp. 50–56 of ref. 36), used oral and intravenous administration of sodium citrate in an attempt to accelerate plutonium excretion from dogs injected 53 days previously with plutonium. Their reason for the use of sodium citrate was that "plutonium might be dislodged from the tissues by formation of plutonium citrate complex in the blood and subsequent excretion." A 35% increase in the urinary excretion of plutonium was observed during intravenous administration of sodium citrate. Experiments by Schubert[37] showed that administration of sodium citrate within two hours after plutonium injection raised the urinary excretion of Pu several fold. It was pointed out[37] that "the action of sodium citrate in promoting the excretion of plutonium can be attributed to its ability to increase the diffusibility of plutonium and thus facilitate its clearance through the kidney." With regard to the therapy of radioisotope poisoning in general it was stated[38] that "the use of complexing agents . . . may be of . . . value for removing these radioelements with which they form very stable complex ions." In other studies early administration of sodium citrate was found to enhance excretion of thorium and strontium.[39]

Sodium citrate has certain disadvantages for use as a complexing agent. It is rapidly metabolized and, in addition, it does not generally bind metal ions under physiological conditions as strongly as would appear desirable.

It was only natural that the chelating agent BAL be tried for the treatment of radioactive poisoning. In 1947, Weikel and Lorenz[40] employed BAL in the hope that it would enhance the excretion of radium from mice previously injected with radium salts. In work published in 1953[41] but performed several years earlier[42] Kawin and Copp investigated the effects of BAL on plutonium excretion in rats. From chemical considerations (based, for example, on reactivity with sulfide ions, $S^=$) one would anticipate that BAL would not affect Ra and Pu excretion because these elements do not react appreciably with sulfhydryl groups, and, indeed, no effect was found. However, in the case of polonium (Po), which does react strongly with BAL, marked increases in Po excretion were observed in investigations begun early in 1949 by Hursh.[43-45]

In 1949 Scott, Crowley and Foreman in Hamilton's laboratory at the University of California made a survey of the effect of different chelating agents on the metabolism of radioisotopes of the rare earths.[46] The chelating agents tested included citrate, ascorbic acid, nicotinic acid, picolinic acid, isothiopropionic acid, carboxyethyl sulfide, glutathione, cholic acid, dithiocarbanyl, EDTA, BAL, creatine, and in a few

cases combinations of two chelating agents. EDTA appeared the most promising in promoting excretion. These studies were continued by Foreman[47] and extended to cerium and plutonium by Foreman and Hamilton.[48]

### Considerations Involved in the Use of Chelating Agents to Increase Excretion of Metals

The potential effectiveness of a chelating agent in accelerating excretion of a radioisotope *in vivo* can be evaluated in part from chemical considerations: (1) affinity of chelating agent for the radioisotope; (2) affinity of chelating agent for $Ca^{++}$; (3) hydrolytic reactions of the radioisotope; and (4) affinity of radioisotope for cellular constituents.

It is desirable that the chelating agent: (1) form chelates which are water soluble and readily diffusible; (2) does not react with normal body constituents, especially calcium (if possible); (3) be of low toxicity; (4) possess high absorption when taken orally; and (5) that it be not metabolized.

In practice, no one chelating agent meets all the requirements. It is not possible to evaluate quantitatively all the various factors listed above. However, some deductions of value can be made from the known formation quotients, i.e., equilibrium constants (Table III) between the chelating agent and metal ions. The ultimate criterion for success of a chelating agent is the extent and rapidity with which it safely removes absorbed radioisotopes from the body.

**Table III. Formation Quotients Involving EDTA and Citric Acid with Metal Ions***

Formation quotient (log $K_f$) where $K_f = (M\,Ke)/(M)(Ke)$.

| *Metal* | *EDTA* | *Ref.* | *Citrate* | *Ref.* |
|---|---|---|---|---|
| $Na^+$ | 1.66 | 32 | – | – |
| $Li^+$ | 2.79 | 32 | – | – |
| $Mg^{++}$ | 8.69 | 32 | 3.2 | 32 |
| $Ca^{++}$ | 10.7 | 32 | 3.16 | 49 |
| $Sr^{++}$ | 8.63 | 32 | 2.85 | 49 |
| $Ba^{++}$ | 7.76 | 32 | 2.54 | 50 |
| $Ra^{++}$ | 7.0† | – | 2.36 | 50 |
| Pb(II) | 18.2 | 32 | 5.75 | 30 |
| Cu(II) | 18.3 | 32 | 14.2 | 32 |
| Fe(II) | 14.15 | 32 | | |
| Fe(III) | 25 | 32 | | |
| $Fe(OH)^{++}$ | 2.8 | 32 | | |
| Co(II) | 16.1 | 32 | | |
| Cd(II) | 16.4 | 34 | | |
| La(III) | 14.7 | 49 | | |
| Ce(III) | 15.4 | 49 | | |
| Pr(III) | 15.75 | 49 | | |
| Sm(III) | 16.55 | 49 | | |
| Eu(III) | 16.7 | 49 | | |
| Gd(III) | 16.7 | 49 | | |
| Tb(III) | 17.4 | 49 | | |
| Dy(II) | 17.75 | 49 | | |
| Y(III) | 17.5 | 49 | | |
| Pu(IV) | 24† | 50 | | |
| Th(IV) | 23† | 50 | | |

* The quoted values of the formation quotient, $K_f$, are under conditions of ionic strength 0.1–0.16 and temperature of 25°C.

† This value is an estimate.

There are, at present, principally three chelating agents which have been used for treatment of either clinical or experimental metal poisoning: EDTA, BAL, and sodium citrate. EDTA is the most successful in removing rare earths and transuranic elements, while BAL is effective for the removal of polonium, at least under acute experimental conditions. At present, citrates do not appear as promising, mainly because they are rapidly metabolized.

It is particularly important that in the treatment of radioactive poisoning with chelating agents, the amounts administered at a given time be as high as possible. Too small doses can actually result in higher levels deposited in bone than would be the case in the absence of the chelating agent. When $Y^{90}$,[82] or La are chelated with small amounts of EDTA (1–5 mg), enhanced bone deposition is observed. In the case of $La^{140}$, for example, the injection into mice of $La^{140}$ with about 1 mg of NaEDTA resulted in a bone retention of 7.1% of the injected dose compared to 3.0% when $La^{140}Cl_3$ was injected alone.[61] This result is explainable by the increased diffusibility of the chelated La, shown by the marked decrease in La content in the liver of the LaEDTA injected rats. In addition, the binding of bone for La is strong so that higher concentrations of EDTA are necessary for effective treatment.

### Ethylenediaminetetraacetic Acid (EDTA) Considerations Based on Chelation Constants

As is seen in Table III, EDTA forms chelates of exceptional stability. However, since it reacts very strongly with serum calcium it is usually administered as the calcium chelate so as to avoid hypocalcemia. In order to combine with a metal, $M$, it is necessary for $M$ to displace $Ca^{++}$ from this chelate. Therefore, in order for EDTA to be effective in removing a metal, the metal must possess a binding affinity for EDTA greater than calcium. Taking the simplest possible example, the reaction can be written:

$$CaV + M = MV + Ca \qquad (1)$$

where $V$ represents molar concentration of EDTA and Ca the ionized calcium. The calcium chelate of EDTA, $CaV$, is actually $Na_2CaV$.

The equilibrium quotient, $K_q$, for reaction 1 is:

$$K_q = \frac{(MV)(Ca)}{(M)(CaV)} = \frac{K_{MV}}{K_{CaV}} \qquad (2)$$

where

$$K_{MV} = \frac{(MV)}{(M)(V)} \qquad (3)$$

and

$$K_{CaV} = \frac{(CaV)}{(Ca)(V)} \qquad (4)$$

Writing Equation 2 in logarithmic form,

$$\log K_q = \log K_{MV} - \log K_{CaV} \qquad (5)$$

Since $\log K_{CaV} = 10.7$, the value of $K_{MV}$ must be

large, indeed, in order for $M$ to displace $Ca^{++}$. Rearranging Equation 2,

$$\frac{(MV)}{(M)} = \frac{K_{MV}}{K_{CaV}} \cdot \frac{(CaV)}{(Ca)} \qquad (6)$$

For practical purposes it is convenient to modify Equation 6 by eliminating $(CaV)$. Disregarding the small amount of $V$ bound by $M$ we have, where $V_t$ = total $V$,

$$V = V_t - (CaV) \qquad (7)$$

From Equation 4 we have

$$V = \frac{(Ca\ V)}{K_{CaV}(Ca)} \qquad (7a)$$

Substituting into Equation 7 and rearranging, we find

$$\frac{(Ca\ V)}{(Ca)} = \frac{V_t K_{CaV}}{1 + (Ca)K_{CaV}} \qquad (8)$$

A further simplification can be made because $(Ca)\ K_{CaV} \gg 1$, as can be seen from the fact that in blood serum the value of ionized Ca is about $1.3 \times 10^{-3}\ M$ while $K_{CaV} = 10^{10.7}$. Hence

$$\frac{(CaV)}{(Ca)} = \frac{V_t}{(Ca)} \qquad (8a)$$

Equation 6 thus becomes

$$R_{MV} = \frac{(MV)}{(M)} = \frac{K_{MV}}{K_{CaV}} \cdot \frac{V_t}{(Ca)} \qquad (9)$$

The important point for therapy is to obtain as high as possible a ratio of chelated $M$ to unchelated $M$, i.e., $R_{MV} \gg 1$. This is done by maintaining as high and constant a blood level of EDTA as possible, i.e., high $V_t$. In the case of the rare earths the values of $K_{MV}$ vary from $10^{14.7}$ for La to $10^{17.5}$ for Y (Table III). The value of $V_t$ one hour after intravenous administration of 2.5 grams of $Na_2Ca$ EDTA (M. W. = 374) is about 50 mg per liter of plasma or $0.050/374 = 0.00013M$. The latter part of Equation 9 becomes

$$\frac{(V_t)}{(Ca)} = \frac{1.3 \times 10^{-4}}{1.3 \times 10^{-3}} = 0.1$$

and hence for La we have

$$\frac{LaV}{La} = \frac{10^{14.7}}{10^{10.7}} \times 0.1 = 1000$$

With such a large ratio for the rare earth which binds the most weakly with EDTA, one would not expect much difference in the effect of EDTA on excretion of the different rare earths. This is not so.

Equation 9 overlooks at least three other factors which become of extreme importance in the net value of $R_{MV}$. These are the hydrolysis of $M$, the reactions between $M$ and body constituents such as proteins, and the reactions between the hydroxylated forms of $M$ and the chelating agent. Hydrolysis is of particular importance when dealing with trivalent and tetravalent elements. Consider the following possible reactions:

$$CaV + MOH = MV + Ca + OH^- \qquad (10)$$

$$CaV + MOH = (M\ (OH)\ V) + Ca \qquad (11)$$

and

$$CaV + LaP = LaV + CaP \qquad (12)$$

$P$ represents the binding by those substances present in the body.

Consequently, the net value of the over-all equilibrium quotient resulting from the reaction between a metal, $M$, and the chelating agent, $V$, obtained by combining Equations 1, 10, 11, and 12 is:

$$\log K_q = 3 \log K_{MV} + \log K_{(M(OH)V)} + \log K_{CaP} - (4 \log K_{CaV} + \log K_{MP} + \log K_{MOH}) \qquad (13)$$

Keeping in mind that

$$(H^+)\ (OH^-) = K_W = 10^{-14}$$

$$(CaP) = K_{CaP}\ (Ca)\ (P)$$

and the ratio of chelated $M$ to non-chelated $M$

$$R_{MV} = \frac{(MV)^3\ [M(OH)V]}{(M)\ (MOH)^2\ (MP)}$$

the expression for $R_{MV}$ becomes:

$$R_{MV} = \frac{(K_{MV})^3\ K_{(M(OH)V)}}{(K_{CaV})^4\ K_{MOH}\ K_{MP}\ K_W}\left(\frac{V_t}{Ca}\right)^4 \frac{(H^+)}{(P)} \qquad (13a)$$

A rough estimate of $R_{MV}$ can be attempted. Consider the rare earth, La, for example. From Table III the value of $\log K_{LaV} = 14.7$, $\log K_{CaV} = 10.7$, $\log K_{La(OH)^{++}} = 5.6$ as given in reference 49, $K_{MP} \cong 10^6$, $V_t/Ca = 0.1$, $H^+ = 10^{-7}$. Log $(La(OH)V)$ is taken to be of the order of 3 based on the value for $(Fe(OH)V)$ given in Table III. These give a value of $R_{LaV} \cong 10^{-4}/P$, while for yttrium, $R_{YV} \cong 10^{-3}/P$ —a factor of about $10^7$.

The efficiency of EDTA for one rare earth relative to another, i.e., $R_{M1V}/R_{M2V}$ is nearly proportional to $K_{M1V}/K_{M2V}$ because the values of $K_{M1(OH)} \cong K_{M2(OH)}$, $K_{M1P} \cong K_{M2P}$ and $K_{(M1(OH)V)} \cong K_{(M2(OH)V)}$. For example, the difference in stability between the hydroxide of La, the lightest rare earth, $La(OH)^{++}$ and that of the heaviest, $Lu(OH)^{++}$ is only a factor of 10, while the corresponding difference between $LaV^-$ and $LuV^-$ is nearly a factor of 100,000 (actually $10^{4.9}$)—a reflection of chelation involving polydentate ligands[3].‡

‡This is explained by Wheelwright, Spedding and Schwartzenbach[49] as follows: "Larger differences between individual rare earths must be expected in cases of chelating agents in comparison to simple ligands such as $OH^-$ groups. A polydentate complexing agent possesses a number of ligand atoms which are donated to the metal cation during complex formation. Each of these will be bound more firmly by the cation of a heavier member, in comparison to the cation of a lighter member. All of these differences are cumulative in arriving at the stability constants of the two chelate complexes, the logarithms of which are proportional to the free energies to be gained during the addition of the whole polydentate group to the two cations to be compared. In the case of a simple complex such as $M(OH)^{+2}$ one water molecule is replaced by $OH^-$ during complex formation. In the case of $MY^-$ probably all the water molecules of the hydrated cation are expelled and replaced by the atoms of the hexadentate agent $Y^{-4}$. This explains the comparatively large differences between the individual rare-earth cations."

It is found that under identical conditions EDTA is, indeed, more effective against the heavier rare earths than the lighter. With an element like Pu(IV) the situation is more complicated. The very high value of log $K_{PMV} = 24$ is counterbalanced by two important points—the hydrolysis of Pu(IV) to form a nearly completely hydroxylated compound at physiological pH, with concomitant polymerization and disproportionation into Pu(III) and Pu(VI).

The equations presented in this section indicate that under physiological conditions the effectiveness of EDTA can only be improved by increasing the amount of EDTA administered. A better chelating agent, all other factors being equal, would be one which would chelate nearly as strongly as EDTA with trivalent and tetravalent radioisotopes but which would not react with calcium. This would provide a factor of $10^{10.7}$ improvement in the over-all chelating efficiency, as can be seen from Equation 13. We are now exploring such a possibility.

### Metabolism and Toxicity of EDTA

The metabolism of $C^{14}$-labeled EDTA has been studied in rats and humans.[53,54] In both species most of the EDTA passes through the body unchanged after parenteral administration. It mixes rapidly with nearly all the body water, but does not enter the red blood cells and only very slowly passes the blood-spinal-fluid barrier. One hour after intravenous injection in humans, 50% is excreted, and within 8 hours nearly all is eliminated in the urine. The half-time of disappearance from the blood is one hour. After oral administration about 90% is eliminated in the feces within 2 to 3 days. Absorption after oral administration continues for at least 18 hours. Total absorption reaches a maximum of about 5%, as judged by the amounts appearing in the urine. Maximum absorption from the skin is about 0.001%.

The acute and subacute toxicity of EDTA has been extensively reviewed.[55] As the sodium salt, relatively small doses of 20–50 mg/kg produce death in dogs because of hypocalcemia. However, when administered combined with an equivalent amount of calcium ion, EDTA is relatively non-toxic. In treatment of radioactive poisoning, EDTA is administered as the calcium salt.§ Dogs can tolerate as much as 4.0 gm/kg in a single intravenous injection. In humans, following exposure to plutonium, at least 2.5 gm of CaEDTA have been given in 250 $cm^3$ of saline by slow intravenous drip twice daily.[56] When given orally at levels of 30 mg/kg some patients have nausea and abdominal cramps.

When high levels are given regularly by intravenous administration, kidney damage may occur. In 4 of 5 cases treated with sodium EDTA kidney damage has been reported upon autopsy. In at least two of these it is believed that death was the result of severe kidney damage induced by repeated injections of sodium EDTA.[57] Evidence of kidney damage following the repeated administration of large amounts of CaEDTA has been reported in rats and humans.[58] Foreman[58] states, however, that this damage following repeated injections of CaEDTA is reversible and can be prevented by allowing a few days to elapse after a series of injections. His tentative recommendations when long term treatment may be required are to inject once or twice daily by slow intravenous (I.V.) drip for 5 days, followed by 2 days of rest, and so on.

### Effect on Plutonium and Lanthanum Excretion in Humans

A technician at Los Alamos cut herself deeply with glass contaminated with a solution of $Pu(NO_3)_4$ in $1M$ $HNO_3$.[56] This was probably equivalent to an intravenous injection. From the urinary excretion curve it was estimated that she received 25,000 to 30,000 disintegrations per minute (dpm) of $Pu^{239}$. When her urinary excretion of Pu had dropped to 12 dpm per day she was given 2.5 gm of CaEDTA in 250 $cm^3$ of saline, slowly by I.V. drip twice daily. A striking rise in Pu excretion from 12 to 1300 dpm resulted. The effect of EDTA decreased on subsequent days (Fig. 1), but it was estimated that in 16 days of treatment about 20–25% of Pu had been removed, in large part from tissues. This is a reasonable assumption because the amounts of Pu excreted exceed the total amount in the circulating blood at the time treatment was given, as can be deduced from data on metabolism of Pu in humans.[59] The patient developed symptoms of kidney damage presumably caused by the EDTA.

A second test of CaEDTA was made in a chemist who, 7 years previously, had been exposed to Pu.[56] His body burden was estimated to be 1.2 micrograms. Treatment during a 5-day period with as much as 5 gm CaEDTA daily produced an average increase of Pu excretion of tenfold over pre-treatment levels (Fig. 2). This case illustrates the difficulties of treat-

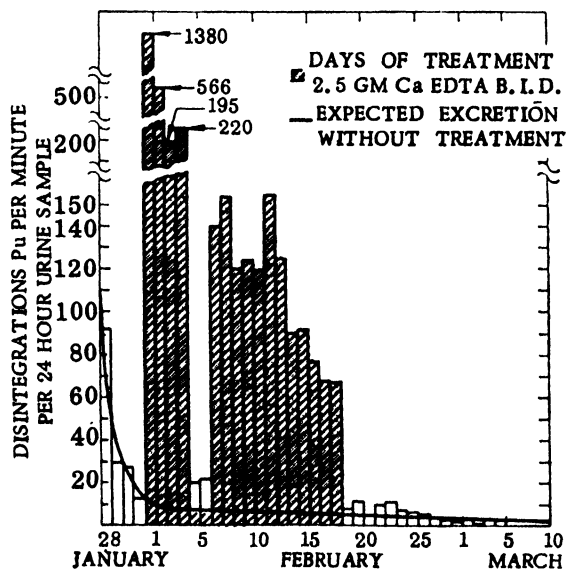



Figure 1. Results of human urine Pu assays following administration of CaEDTA. (Fig. 1 from ref. 56)

§ CaEDTA as Calcium Disodium Versenate® for injection is available commercially as a very stable, sterile solution from Riker Laboratories, Los Angeles, California.

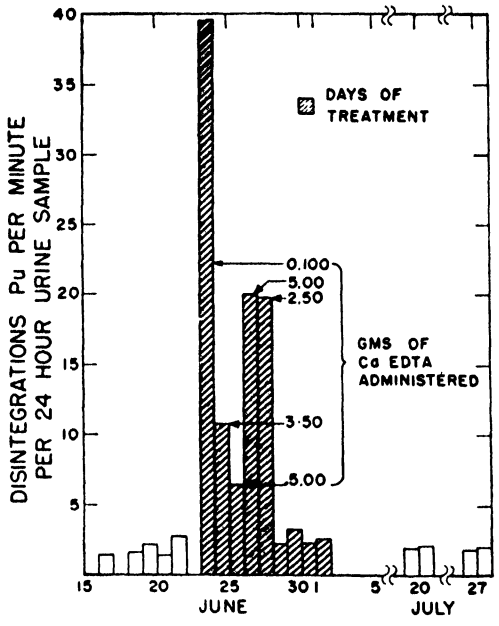



**Figure 2. Results of human urine Pu assays following administration of CaEDTA. (Fig. 2 from ref. 56)**

ing chronic radioisotope poisoning. In the first place treatment has to be given for months or years in order to reduce appreciably the body burden of Pu. Secondly, during this period the tissues are receiving continued radiation so that even if all the Pu could be removed at the end of, say, 5 or 10 years—a most unlikely event—the patient might not be spared lesions or malignancies.

The effect of EDTA on lanthanum metabolism in humans illustrates some interesting points. A tracer dose of $La^{140}$ as the EDTA complex was injected intravenously into a human.[60] The concentration of EDTA was not specified but may be surmised[61] to be a few milligrams. After 48 hours only 5% of the injected La was excreted. At that time 4 gm of CaEDTA in 500 ml of a 5% solution of glucose was infused in 3 hours. A sharp rise in $La^{140}$ excretion was observed. By the 5th day post-injection urinary excretion dropped to negligible levels. A 7-hour infusion of 8 gm of CaEDTA in 1000 ml of glucose solution again raised La excretion, but only one-fifth as high as the previous maximum.

In a second study, in an effort to ascertain the maximum possible increase in excretion of $La^{140}$, CaEDTA was administered prior to, and simultaneously with, the $La^{140}$EDTA injection. A prior dose of 3 gm of CaEDTA in 300 ml of glucose solution was infused in 2.5 hours and was followed by a 1.5-hour infusion containing LaEDTA in 2 gm of CaEDTA. Even in this case the total excretion of La in the first day was only 37%, most of which occurred in the first 4 hours.

These results with lanthanum are reasonably interpreted by the equations previously given. Since La is relatively weakly bound by EDTA it is necessary that the concentration of EDTA in the blood be high. Even when LaEDTA is injected simultaneously with several grams of EDTA the ratio of $(LaV)/(La) = R_{La}$ is insufficient to allow more than about one-third of the La to be excreted. If the reactions are amenable to the laws of mass action, a 1000-fold reduction in EDTA concentration, as is the case when LaEDTA alone is injected, had the effect of reducing $R_{La}$ to the point where, for practical purposes, it is as if uncomplexed La was injected. This, indeed, was observed in an experiment in which La was injected as $LaCl_3$.[60] Approximately 5% of the injected dose was excreted by the kidneys, as was the case following the injection of LaEDTA. This illustrates the importance of the binding strength of metal with a chelating agent in the effectiveness of the latter; a low value of $K_{MV}$ indicates that a large, possibly toxic amount of the chelating agent is necessary to remove a significant quantity of the radioelement.

In the case of yttrium, the affinity for EDTA is approximately 400 times that of lanthanum. This is equivalent insofar as $R_{MV}$ is concerned to a $10^7$-fold excess of EDTA. Consequently it would be expected that EDTA would be far more effective in increasing excretion of yttrium than lanthanum. An illustration of this is shown in experimental animals. Following the injection of uncomplexed Y as $YCl_3$ into rats, about 5% was excreted in the urine in 24 hours. The same amount of La under similar conditions is excreted, as would be expected from the nearly identical chemical behavior of rare-earth ions at physiological pH. However, when therapeutic doses of EDTA are injected simultaneously with the Y, approximately 70% of the Y is excreted in 24 hours,[46] compared to only 37% of La. Thus, a 400-fold increase in the value of $K_{MV}$—at least for rare earths—results in a 2-fold decrease in body retention when EDTA is given simultaneously with the rare earth. It would be of particular interest to learn if quantitative predictions for other radioisotopes can be made from this ratio.

### Effects of EDTA on Excretion and Tissue Distribution of Radioisotopes in Experimental Animals

In general, EDTA can be expected to show varying degrees of effect in removing elements whose $K_{MV}$ exceeds that of calcium. This includes all the rare earths, plutonium, and the transuranic elements. The relative effectiveness of EDTA with trivalent transuranic elements such as americium (Am) and curium (Cm) can be expected to parallel the rare earths. From electronic and chemical observations it is known, for example, that the rare-earth counterpart of Am is Eu, and that of Cm is Gd.[80] A summary of published studies on the effects of EDTA on the metabolism of several different radioisotopes is given in Table IV.

Even under the most favorable conditions, EDTA, alone or in combination with BAL, has no effect on the excretion of $Sr^{89}$ from rats.[69] Since Ra and Ba react more weakly with EDTA (Table III) than does calcium, it is not expected that the excretion of these alkaline earths would be modified by EDTA.

The most striking effects of EDTA are obtained when it is administered soon after exposure. In the case of Am(III), for instance, EDTA forms a chelate whose stability constant, log $K_{AmV}$, is at least 16.7. A single injection of 100 mg of CaEDTA to rats (~400 mg/kg) is given intraperitoneally 30 minutes after intravenous injection of $Am^{241}$ results in a 25% excretion in the urine within 24 hours, compared to 1% in the untreated rats during the same period. Several days later the animals were fed a diet containing 3% CaEDTA. This causes the urinary excretion to increase 5 to 10 times over the control levels.[65] It must be kept in mind that these doses of CaEDTA are much higher than would be administered to humans.

In the case of yttrium (log $K_{YV} = 17.5$), a single intravenous injection of 20 mg (~80 mg/kg) of CaEDTA in rats given two hours after intravenous injection of $YCl_3$ in isotonic saline at *p*H 6[65,66] caused an excretion of 67% of the Y in the urine at the end of 24 hours, compared with 25 to 30% in untreated controls. When EDTA was administered on the 20th through 23rd days after Y administration, the urinary excretion of Y rose from a pre-treatment level of 0.07% per day to as much as 0.7%—a tenfold increase. Of interest is the fact that CaEDTA when given by stomach tube increases Y excretion—a reflection of the large $K_{YV}$ compensating for the low degree of absorption of CaEDTA.

The most striking effect on Y excretion by EDTA occurs when EDTA is given *before* the Y.[68] Rats were first given an intraperitoneal injection of 10 mg of NaEDTA, followed 15 minutes later by 50 mg of CaEDTA. One hour after the injection of CaEDTA the animals were injected intraperitoneally with carrier-free $Y^{91}Cl_3$. Three days later they were sacrificed. The treated rats excreted a total of 93.3% of the injected $Y^{91}$ compared to 25.3% by the controls. Skeletal retention was only 3.9% compared to 58.0% in the controls. The time relationship between EDTA and Y injections are important. Results of intraperitoneal injection of CaEDTA a half hour before Y should not differ significantly from those of one hour because the concentrations of EDTA in the blood and body water are nearly equal at ½ and one hour.[53] However, if EDTA is administered 3 hours before Y, then a sharp reduction in the urinary excretion of Y and an increase in skeletal deposition would be expected because the blood-level of EDTA is 5-10 times less than at the 1 hour point. The effect of time of EDTA administration on the skeletal concentration of Y is shown in Fig. 3.

Note (Fig. 3) that EDTA is less effective in decreasing the skeletal concentration of Y when given 3 hours after Y injection than after ½ or 1 hour. This is easily explained by the fact that the concentration of Y in the skeleton is about 15–20% at ½ hour and increases to about 40% of the injected dose at 3 hours.[81] The increased urinary excretion obtained by EDTA administration at 3 hours or earlier is evidently only partly derived from the skeleton.

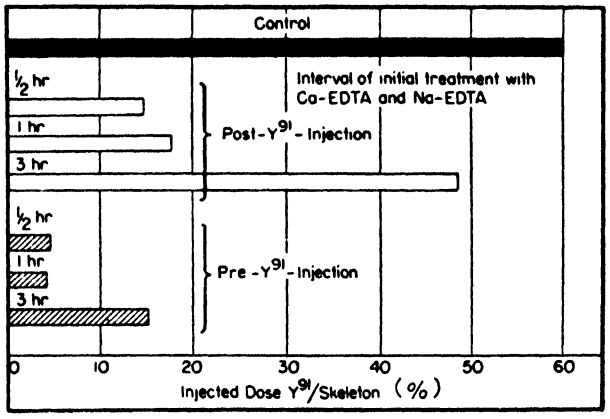



Figure 3. Effect of time of EDTA administration on skeletal concentration of injected $Y^{91}$ in rats. (From ref. 68)

As is to be expected, there is a minimum effective dose of EDTA, even for radioisotopes such as Pu which have a high $K_{MV}$ with EDTA. A dose of 8 mg/kg of CaEDTA is too small to affect significantly the excretion of Pu, even when administered simultaneously.[62] It is conceivable that doses this small could cause a higher retention of Pu in the skeleton as was discussed earlier in the cases of Y and La. On the other hand, the effectiveness of sufficient CaEDTA is independent of Pu concentration, at least in experiments[64] in which the molar ratio of CaEDTA to Pu varied from about $3 \times 10^9$ to $3 \times 10^4$—a factor of $10^5$ in Pu concentration.

The importance of $K_{MV}$ is shown in an experiment involving rats with $Ce^{144}$(III) for which log $K_{MV} = 15.4$. When CaEDTA (~100 mg/kg) was injected intraperitoneally 18 hours after intravenous administration of $CeCl_3$ in a neutral isotonic solution containing citrate, the urinary excretion was increased only about 3 times the control level (from ~3% to ~9% in 24 hours). When CaEDTA was given on the 13th through the 17th day in doses of 200 mg/kg twice daily there was scarcely any increase in Ce excretion over the control levels.[67] Under similar conditions the excretion of Pu and Y was enhanced about tenfold.

When EDTA is administered as a therapy for poisoning by elements with which it reacts only moderately, such as with Ce, then early administration, or even administration prior to the injection of the radioisotope, produces relatively small effects. For example, rats were injected with 255-day old mixed fission products (MFP). The principal fission products were $Zr^{95}$, $Ce^{144}$, $Sr^{89,90}$, $Y^{91}$, and $Pr^{143}$. Under optimal conditions of treatment, EDTA-treated rats excreted 40–50% as compared to 35% in the controls. The skeleton of EDTA-treated rats contained 25% of the injected dose of MFP compared to 30% in the controls. More information on these experiments[70] is given in Table VII in which the effects of EDTA and zirconium salts are compared. Little effect is expected with praseodymium, Pr, because of a relatively low binding capacity for EDTA—log $K_{PrV} = 15.75$.

**Table IV. Published Studies on the Removal of Radioisotopes from Mammals by the Use of Zirconium Salts and the Chelating Agents—Ethylenediaminetetraacetate, Citrate and BAL**

| *Experimental animal* | *Therapeutic agents tested* | *Routes of administration** | *Radioisotopes involved* | *References* |
|---|---|---|---|---|
| Rat | CaEDTA | I.V., I.P., oral | $Pu^{238}$ | 60 |
| Rat | CaEDTA<br>Zr Citrate<br>Zr Malate<br>Zr + CaEDTA | I.P. | $Pu^{239}$ | 63 |
| Rat | CaEDTA | I.V., I.P., I.M., oral | $Pu^{239}$, $Y^{91}$, $Am^{241}$ | 65 |
| Rat | CaEDTA<br>Zr Citrate<br>Zr + CaEDTA | I.P. | $Pu^{239}$ | 64 |
| Rat | CaEDTA<br>Ca Citrate | I.P., I.M., oral | $Pu^{239}$, $Y^{91}$ | 66 |
| Rat | CaEDTA | I.P., I.M., oral | $Pu^{239}$, $Y^{91}$, $Ce^{144}$ | 67 |
| Rat | CaEDTA, NaEDTA | I.P. | $Y^{91}$ | 68 |
| Rat | CaEDTA + BAL | I.P., I.M. | $Sr^{89,90}$ | 69 |
| Rat | NaEDTA<br>Zr Citrate<br>NaEDTA + Zr Citrate<br>Na Citrate | I.P., oral | Mixed fission products (255 days old) | 70 |
| Man | CaEDTA | I.V. | $La^{140}$ | 60 |
| Man | NaEDTA | I.V. | $Ca^{45}$ | 71 |
| Man | CaEDTA | I.V. | $Pu^{239}$ | 56 |
| Man | Zr Malate | I.V. | $Pu^{239}$ | 72 |
| Rat | Zr Citrate, Na Citrate | I.P. | $Pu^{239}$, $Y^{91}$ | 37, 38, 75, 76 |
| Rat | Zr Citrate, Na Citrate | I.P. | $Th^{230}$, $Sr^{89,90}$ | 39 |
| Rat | Zr Citrate | I.P. | $Pu^{239}$ | 77 |
| Rat | Zr Citrate, Na Citrate | I.P. | $Pu^{239}$, $Y^{90}$, $Nb^{95}$, $Sr^{89,90}$, $Ce^{144}$ | 42 |
| Rat | Zr Citrate | I.P. | $Am^{241}$ | 79 |
| Rat | Zr Citrate<br>+ Parathormone | I.P. | $Pu^{239}$, $Y^{90}$ | 42 |
| Dog | Zr Citrate | I.V. | $Pu^{239}$ | 38, 78 |
| Dog | Na Citrate | I.V., oral | $Pu^{239}$ | 36 |
| Rat | BAL | I.M. | $Po^{210}$ | 43, 44 |
| Rat | BAL | I.M. | $Pu^{239}$ | 41, 42 |
| Mouse | BAL | Subcutaneously | $Ra^{226}$ | 40 |
| Mouse | BAL | I.M. | $Sr^{90}$, $P^{32}$, $Ra^{224}$(ThX) | 73, 74 |
| Rabbits | BAL | I.M. | $Ra^{224}$(ThX) | 74 |

* I.V. = intravenous, I.P. = intraperitoneally, I.M. = intramuscular.

An important question is whether prolonged treatment with EDTA can remove radioisotopes once they are fixed in the body, particularly in the skeleton. Partial answers are provided in experiments in which administration of EDTA is begun at least two weeks after the intravenous injection of the radioisotope. In Table V are tabulated the results of such experiments on the effects of CaEDTA on Pu excretion and retention in the skeleton and liver. It will be noted that a *single* massive injection of CaEDTA given 30 days after Pu has no effect. However, if CaEDTA is given daily for one month or for a total of 30 injections starting a month after Pu injection a substantial increase in total excretion is observed and skeletal deposition is reduced from 56% of the injected dose of Pu to 41%. Even under optimum conditions the skeletal deposition of Pu can only be reduced a maximum of somewhat less than twofold (Table V). These results do not appear to offer much hope for those cases of chronic poisoning in which treatment is delayed for a year or even more than that.

In similar delayed-treatment experiments with Y, Cohn *et al.*,[68] began treatment of rats 14 days after Y injection. Rats were injected daily after Y injection with 10 mg of NaEDTA, followed at varying times during the day by 50 mg of CaEDTA, until sacrificed at 29 days. The treated rats excreted 17% of the Y during the treatment as compared to 4.5% in the controls. The increased amount of Y excreted appeared to be nearly all derived from the skeleton since the treated rats had 48% in the skeleton compared to 62% in the controls. The administration of EDTA four times daily gave no improvement in results.

**Table V. Effect of Different Dosage Schedules of CaEDTA on the Excretion and Distribution of Intravenously Injected Pu in Rats**

| *Total number of injections of CaEDTA* | *Time of first injection of CaEDTA after Pu* | *Dosage and schedule of treatments with EDTA* | *Time of sacrifice after radioisotope (days)* | *Per cent of administered dose of Pu* | | | | | | | | *Reference* |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | *Urine* | | *Urine + Feces* | | *Skeleton* | | *Liver* | | |
| | | | | *Controls* | *Treated* | *Controls* | *Treated* | *Controls* | *Treated* | *Controls* | *Treated* | |
| 1 | Immediately | Pre-treatment by addition of 3.5% CaEDTA to diet *24 hrs prior* to Pu. Single injection I.P. of 186 mg of CaEDTA *at time of Pu injection* | 15 | 6 | 51 | 27 | 62 | 55 | 31 | 8.8 | 3.6 | 62 |
| 1 | 30 minutes | *Post-treatment* – 600 mg CaEDTA (2400 mg/kg) given 30 min after Pu. Single injection | 30 | – | – | – | – | 58 | 33 | 15 (soft tissue) | 5 (soft tissue) | 64 |
| 1 | 30 days | *Post-treatment* – 600 mg CaEDTA (2400 mg/kg) *given 30 days after Pu.* Single injection | 30 | – | – | – | – | 46 | 50 | 22 (soft tissue) | 18 (soft tissue) | 63 |
| 64 | 2 hours | *Post-treatment* – 100 mg CaEDTA (400 mg/kg) given I.P. 4 times daily for 16 days starting *2 hrs after Pu.* | 16 | 4 | 32 | 30 | 40 | 55 | 37 | 10 | 10 | 65 |
| 11 | 18 hours | *Post-treatment* – 25 mg CaEDTA given I.P. at *18 hrs after Pu.* 50 mg CaEDTA twice daily on 13th–17th day | 62 | 3.3 | 13 | 39 | 58 | 50 | 34 | – | – | 66 |
| 25 | 25 days | *Post-treatment* – 162 mg of CaEDTA given I.O. daily from 25th to 50th day post Pu | 50 | 6.3 | 15 | 26 | 39 | 61 | 52 | 5.3 | 4.9 | 62 |
| 30 | 30 days | *Post-treatment*–CaEDTA, dosage not stated – but presumably 20 mg CaEDTA – given I.V. daily for 1 month starting *30 days after Pu* | 30 | 3 | 20 | 22 | 50 | 56 | 41 | 6 | 2 | 56 |

It is claimed[68] that the use of NaEDTA followed by CaEDTA helps to improve the final results. This is explained on the basis that the decalcifying action of the tetrasodium salt of EDTA causes some dissolution of bone causing a release of calcium from the bone and a simultaneous release into the blood of bone-fixed yttrium. This explanation does not appear tenable. First, the results may not be unique because no controls were included in which CaEDTA alone was administered. Secondly, it has been thoroughly demonstrated that the uptake and release of yttrium by the bone is independent of the degree of calcification or decalcification of bone.[19] Finally, a survey of the urinary excretion of Y in experiments in which CaEDTA alone was administered with that in which NaEDTA and CaEDTA were given indicates no significant differences (Table IV).

The use of CaEDTA in combination with zirconium salts in treatment of radioelement poisoning is discussed later.

### 2,3-Dimercaptopropanol (BAL)

Administration of BAL to experimental animals previously injected with radium,[40,74] strontium,[78] phosphorus,[73] plutonium[41] or yttrium[46] does not affect the distribution or excretion of these elements. No effect would be expected, since, as mentioned earlier, these elements do not react with sulfhydryl compounds to any appreciable extent. Polonium, on the other hand, belongs to a family of elements which react strongly with sulfhydryl groupings.

Hursh[44] administered BAL to rats previously injected intravenously with a solution of polonium chloride at *p*H 6.5. Beginning 1 hour after $Po^{210}$ administration the BAL was injected intramuscularly in a 10% solution in peanut oil. Three doses, each of 0.04 ml, were given per day for three days. The rats were sacrificed 10 days after receiving Po. The urinary excretion for the ten-day period was increased from 0.64% to 1.39%—a factor of two. The fecal excretion was increased from 25.1% to 52.1%. The Po levels in the bone marrow, blood plasma, spleen and testes were decreased while the concentration in the muscle was increased. Increasing the dosage of BAL or initiating treatments at the time of Po injection did not improve the results.

It is of interest to note that in this case the feces were the principal route of enhanced excretion. This is the reverse of the results obtained with EDTA and other chelating agents acting on other elements. While the blood level of Po was lowered, the dialyzable fraction of Po in the blood of treated rats was seven times that of those untreated. This is reasonably explained by supposing that "BAL exerts its

principal effect by forming a BAL-polonium complex, freeing polonium bound to protein . . . by virtue of the strong binding power of its dithiol group. The BAL-polonium complex is readily diffusible and therefore the complex in the blood stream may diffuse into the muscle tissue, may readily filter through the kidney glomerulus, and may be secreted into the gut."[44]

Surprisingly, few studies have been made in which increased excretion of a radioisotope has been correlated with increased survival or other criteria of effectiveness. Since BAL decreased the total amount of Po in the body and reduced the level in the more radiation-sensitive organs (from the acute standpoint) it would be expected that the survival time of $Po^{210}$-poisoned rats would be increased. Hursh[45] found that control rats injected with a lethal dose of $Po^{210}$ (36 $\mu c/kg$) had a median survival time of 22 days, compared with 89 days in the BAL-treated rats. Eighty-nine days is considerably longer than the median survival time of 57 days which would have been expected from the amount of Po retained in the body after BAL treatment. It was concluded that the "detoxifying effect of BAL cannot depend only upon increased excretion but must also consist in a mobilization of polonium from radiation-sensitive tissue such as bone marrow and spleen into radiation-resistant muscle tissue."

### Citrates

As can be seen from Table IV, citrates have been employed frequently in attempts to enhance the excretion of radioisotopes. This is to be expected in view of the fact that they are normal constituents of the tissues and blood of mammals.

Even under optimum conditions the effect of citrates on excretion of Pu and rare earths is relatively insignificant. Studies on the effect of citrate-metal complex formation on the metabolism of several rare earths have been reported.[41] In the case of intramuscular injection of the radioisotopes, complexing the rare earth with citrate increased the amount absorbed from the injection site several-fold. While citrate treatment reduced the amount of rare earth deposited in the liver, a large increase in skeletal deposition and kidney deposition was noted.

With thorium and strontium, early administration of sodium citrate gave surprisingly large effects.[39] Rats were injected intraperitoneally with 200 mg of sodium citrate (4.0 ml of a 5% solution) 30 minutes after the intraperitoneal injection of $Th^{230}$. They were sacrificed 8 days later. The control rats excreted a total of 28% of the injected $Th^{230}$, compared to 47% by the treated rats. Nearly all of the increased amount of $Th^{230}$ excreted was derived from the liver, which contained 30% of the injected $Th^{230}$ in the controls, compared to 14% in the treated rats. Significant reduction in the $Th^{230}$ content of the spleen, pancreas, and kidney was also observed. When the injection of sodium citrate was made 3 days after $Th^{230}$ administration, no effect on tissue distribution or excretion of the radioisotope was noted. In these experiments only a single injection of sodium citrate was given.

In the same experiment, when sodium citrate was administered intraperitoneally 30 minutes after intraperitoneal injection of $Sr^{89,90}$, the total excretion of $Sr^{89,90}$ was increased from 30% to 46% of the injected dose. In this case most of the increased amount of radioisotope excreted was derived from the skeleton.

The effect of citrate on Th was not unexpected inasmuch as citrates are known to complex Th salts very strongly. This complexing action was further demonstrated by the fact that the diffusibility of Th in blood serum is markedly increased by the addition of sodium citrate.[39]

Citrate forms a weaker complex with Sr than with Ca. By making use of an expression, originally given by Kety[31] for lead citrate, to solve for available citrate, one can obtain the ratio of complexed Sr to free Sr in the blood serum:

$$\frac{(SrCit^-)}{(Sr^{++})} = \frac{K_{SrCit^-}}{K_{CaCit^-}} \frac{(CaCit^-)}{(Ca^{++})} \qquad (14)$$

where $Cit^{---}$ represents the trivalent citrate anion and $K$ the formation quotient. From the known values of $Ca^{++}$ and citrate in blood serum the ratio $(CaCit^-)/(Ca^{++})$ was found to be 0.063. From the values of $K$ given in Table III:

$$\frac{(SrCit^-)}{(Sr^{++})} = \frac{K_{SrCit^-}}{K_{CaCit^-}} \times 0.063 = \frac{710}{1450} \times 0.063 = 0.031$$

Hence, most of the Sr in serum is uncomplexed.

When 200 mg of sodium citrate was injected into a rat whose blood volume was 15 ml, then, assuming no destruction of the injected citrate, the concentration of citrate per ml = 200/15 = 13.3 mg = $4.5 \times 10^{-2}M$. This is a 450-fold increase. The normal blood citrate level is about 18–30 micrograms of citrate per $cm^3$ or approximately $10^{-4}M$.[83] This would raise the value of $(CaCit^-)/(Ca^{++})$ to about 24 and the value of $(SrCit^-)/(Sr^{++})$ to about 11. Despite the fact that injected citrates are destroyed rapidly by the body—probably within 2 to 3 hours—enough Sr is complexed during that time to permit enhanced excretion. It must be realized that most of the increased excretion of a radioisotope caused by a chelating agent occurs in the first few hours after treatment. Thirty minutes after Sr injection, the amount of Sr in the circulation is approximately 10–20% of the amount injected.[39] Consequently, the increased urinary excretion of Sr which followed the citrate treatment given at 30 minutes was derived from circulating Sr which otherwise would have been deposited in the skeleton. In other words, sodium citrate administration did not remove deposited Sr from the bone. It should be kept in mind that calcium excretion is also increased. Actually, the citrate levels must be maintained in large molar excess over calcium so that sufficient amounts remain to react with the radioisotopes.

The prolongation of high citrate levels may modify the complexing action, as shown by an example taken from the same experiment on Th. In this experiment a separate group of rats was treated with zirconium citrate solutions. Just as with early (30 minute) administration of sodium citrate, the early intraperitoneal injection of zirconium citrate caused a marked increase in urinary excretion of Th. The results with Th are explained[39] by the fact that in zirconium citrate the citrate part of the molecule is not immediately available because it is bound to zirconium. Metabolic processes free the bound citrate, whereby it is available to exert a solubilizing effect through complexing action on the Th. In other words, until the citrate is free, the reaction

$$\text{Zr Cit} + \text{Th} \rightleftharpoons \text{Th Cit} + \text{Zr} \qquad (15)$$

is too far to the left because of the strong binding of Zr by citrate.

The prolonged action of zirconium citrate was shown in the case of Sr where the urinary excretion of Sr remained significantly above the control levels for several days after a single Zr injection.

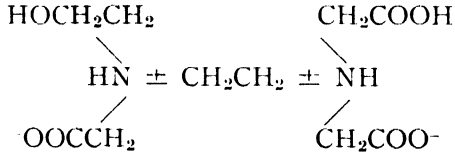
These results suggest that if the metabolic destruction of injected citrates could be reduced or if an elevated citrate level could otherwise be maintained in the tissues and blood, it would be possible to improve the ability of citrates to accelerate excretion of radioisotopes. Under these conditions the citrates could be of value in the treatment of many radioactive poisons. In principle this is possible. Since citrates are involved in metabolic cycles, it is possible to administer drugs which block the destruction of citrates, directly or indirectly. These drugs include non-lethal doses of sodium fluoroacetate which cause large and sustained elevations of citrate levels in the kidney, spleen, and other soft tissues but not in blood.[83] Guanidine and guanidine derivatives increase the blood citrate levels two to threefold.[84] Finally, Vitamin D appears to accelerate citric acid production in bone.[85] We are investigating the use of these agents for the treatment of poisoning by radioactive and nonradioactive metals.

### Miscellaneous Chelating Agents

A compound related to EDTA, called Fe-3 and shown in Table II, causes substantial increases in the excretion of Pu and Ce. In an experiment with rats 100 mg of Fe-3 was injected intraperitoneally each day for 5 days beginning on the 48th day after injection of a mixture of Pu and Ce.[66] The urinary excretion of Pu was increased 6- to 7-fold over control levels during the treatment. Some increase was observed during the three days following the cessation of Fe-3 administration. With the same dosage schedule Fe-3 increased the urinary excretion of Ce as much as 6 times over the level of the control group as compared to a maximum of a 3-fold increase observed when CaEDTA was administered.[48] No effect on fecal excretion of Ce was observed.

It is possible that the effectiveness of Fe-3 is related to its ability to form complexes which are more stable in alkaline solution than those of EDTA. Probably more important, it may form weaker chelates with $Ca^{++}$ while reacting to an undiminished degree with transition elements.

Another compound which shows better promise even than EDTA is N-hydroxyethylethylenediamine triacetic acid (Versenol),¶ which has the structure:



Versenol administration to humans has been found to cause a marked increase of urinary excretion of iron.[86] Therefore it should react strongly with plutonium, cerium, and similar radioisotopes.

In addition to EDTA, numerous other synthetic derivatives of amino acids may well be useful for the treatment of radioactive poisons. A list of possible compounds can be gleaned from tables in Ref. 34, pages 532–541.

Other sulfhydryl compounds besides BAL which have been tried for the treatment of experimental plutonium poisoning are methionine and cysteine. No significant effects have been obtained, although it has been observed that cysteine decreased the amount of intramuscularly injected Pu(VI) absorbed from the injection site.[26] In later studies, no general alteration in tissue distribution of Pu was found following cysteine administration,[42] but the treatment did decrease the absorption of intramuscularly injected Pu(VI). According to Van Middleworth,[26] the latter effect might be explained by a reduction of Pu(VI) to the poorly absorbed Pu(IV).

Among numerous other chelating agents which have been used in conjunction with rare earths but which do not appear promising for therapy are creatine, cholic acid, gluthathione, and trimethylamine tricarboxylic acid.[46]

### Colloidal Carriers and Zirconium Salts

Early in 1945 it was thought that the excretion of plutonium from injected animals would be increased by treatment with large amounts of the soluble salts of various metals.[37] This suggested treatment was based on the displacing ability of one metal for another, i.e., cation exchange. The metals considered were those possessing a high valence and a metabolism similar to plutonium. Salts of lanthanum, cerium, and zirconium were among those tested. Because of their low toxicity and ability to promote excretion of plutonium, zirconium salts were the most promising.

Subsequent studies, summarized in Ref. 87, proved that the injection of zirconium salts, especially zirconium citrate, caused marked increases in the excretion of many different radioisotopes. The mech-

¶ Versenol is available from the Bersworth Chemical Co. Framingham, Mass.

anism of action of zirconium was postulated to be a carrier action rather than a metal displacement action.[42,75] This carrier concept was demonstrated by experiments involving other metals. The mechanism of action zirconium salts is explained as follows:[87] "After the entrance of Zr salts into the bloodstream, collodial aggregates are necessarily formed because of the hydrolyzability of Zr, i.e., above a *p*H of about 2 an insoluble precipitate of Zr hydroxide is formed. The circulating Pu is adsorbed by these aggregates and the subsequent disposition of the Pu is therefore directly related to the metabolism of the Zr.

"If this concept is correct, the salts of other hydrolyzable metals that also form colloids in the blood should be similar in action to the Zr salts. This is borne out by the results with Mn, Fe, Al, and Ti. The salt of a basic, non-hydrolyzable element, magnesium, had no observed influence on Pu metabolism.

"The eventual tissue distribution of the circulating colloidal particles, however, is a function of their particle size distribution . . . Zr colloids of relatively large size are deposited primarily in the liver and spleen . . . Therefore the tissue or organ distribution of that fraction of Pu removed from the blood can conceivably be varied within considerable limits by means of different carriers."

In the sections to follow, we will summarize the results of experimental studies using Zr salts to promote excretion of injected radioisotopes. The limitations and potentialities of Zr treatment either alone or in conjunction with EDTA are explored.

### Metabolism and Toxicity of Zirconium Salts

A thorough review of the toxicity and physiological effects of zirconium compounds has recently become available.[88] The conclusion is that zirconium salts are remarkably low in toxicity. The acute $LD_{50}$ of zirconium citrate is about 2 gm/kg. Rats receiving a total of 15 injections given over a period of five weeks, of zirconium citrate in single doses of 50 mg as Zr (~175 mg/kg) showed no ill effects except a transient weight loss.[77] Histological examinations of the liver showed no evidence of damage.

Doses of zirconium citrate as high as 400 mg/kg as Zr were well-tolerated by dogs.[78] However, the high citrate level in the zirconium solutions does depress the ionized calcium content of the blood. This effect must be counteracted to prevent hypocalcemia. It was found that the administration of 10 ml of a calcium gluconate—calcium glucoheptonate mixture (Abbott)—equal to 10 per cent calcium gluconate w/v for every 0.8 gm of citrate prevented hypocalcemia.[78] Experiments in our laboratories also have shown that calcium gluconate administration controls hypocalcemia. When given with calcium gluconate, the administration of zirconium citrate by intravenous drip under conditions approximating those used clinically did not alter the blood pressure or respiratory rate of a dog injected with 1050 mg Zr (137 mg Zr/kg) (unpublished data).

The lack of overt toxic effects of injected zirconium salts reported above seems at variance with effects observed after injection of zirconium malate[72] and zirconium citrate in man. After several doses of 50 mg of Zr as zirconium malate patients became dizzy and developed symptoms of incoordination and other symptoms of vestibular damage similar to those sometimes observed following 8th nerve damage by streptomycin. A significant observation is that those preparations of zirconium salts which cause these adverse effects are unstable on standing, possessing a very short shelf life.[90] The explanation of these unexpected toxic effects is probably that these solutions were sterilized by autoclaving at elevated temperatures. Heat sterilization causes the zirconium chelate to decompose. Decomposition of hydrolyzable metal salts by heat is well known. For example, heat sterilization of ferric cacodylate solutions (National Formulary VIII) results in the formation of colloidal iron oxide,[89] increasing the toxicity several-fold. Non-sterilized zirconium salt solutions made in our laboratory and by others are stable without any signs of deterioration for indefinite periods—at least a year. It is apparent, therefore, that solutions of zirconium citrate must be prepared in sterile form without autoclaving. This can be done by mixing sterile water and sterile sodium citrate solutions with zirconyl chloride. The latter solution, when prepared in concentrated form, is presumably sterile.

Following the injection of zirconium citrate solutions prepared according to published directions,** more than 90% of the Zr is excreted in the urine, a small amount in the feces, and a small amount is retained in the skeleton and soft tissues.[38,42] Approximately 5% is retained in the muscle and skin—at least for a few days. Kinetic studies on the rates of disappearance of Zr from the blood and rates of incorporation in tissues by Kawin, *et al.*[42] will be referred to later.

Very little zirconium is absorbed from the gastrointestinal tract when it is given orally as zirconium citrate. Hence, this route of administration is not feasible for the treatment of radioactive poisoning.

### Effects of Zirconium Salts on the Excretion and Retention of Radioisotopes

Zirconium citrate treatment has some degree of effect on removal of nearly all of the fission products and transuranic elements as shown by the data in Tables VI and VII.

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** A 100 ml batch of zirconium citrate is prepared as follows:[88] To 18 ml of a filtered solution of pure (iron-free) zirconyl chloride ($ZrOCl_2$), containing 130–140 mg as Zr per ml, add, while stirring, 80 ml of a 12.5% solution by weight of sodium citrate, $Na_3C_6H_5O_7 \cdot 2H_2O$. A precipitate first forms which subsequently dissolves. Two ml of a 20% NaOH solution are added and the zirconium citrate solution is filtered through quantitative filter paper. The final solution should be clear and colorless. It has a pH of ~ 6.3 which can be adjusted to 7 if desired, and contains about 25 mg as Zr per ml and the equivalent of a 10% sodium citrate solution. Because of its hypertonicity ($Na^+ = \sim 1.1M$) it is advisable to dilute it at least 3-fold for intravenous use.

**Table VI. Effect of Intraperitoneal Administration of Zirconium Citrate on the Excretion and Distribution of Different Radioisotopes in Rats**

| Radioisotope | Route of administration* | Dose of Zr citrate (mg Zr) | Time of injection of Zr after radioisotope administered | Time of sacrifice after radioisotope administered (days) | Per cent of administered dose | | | | | | Reference |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | Urine | | Skeleton | | Liver | | |
| | | | | | Controls | Treated | Controls | Treated | Controls | Treated | |
| $Am^{241}$ | I.V. | 40 | 15 min | 4 | 5.7 | 35 | 31 | 22 | 38 | 18 | 79 |
| $Am^{241}$ | I.V. | 40 | 15 min | 16 | 8.1 | 36 | 35 | 21 | 13 | 12 | 79 |
| $Sr^{89}$ | I.P. | 25 | 1 hour | 5 | 34 (+feces) | 40 (+feces) | 61 | 45 | – | – | 69 |
| $Sr^{89}$ | I.P. | 25 | 1 hour (prior to $Sr^{89}$) | 5 | 34 (+feces) | 60 (+feces) | 61 | 36 | – | – | 69 |
| $Ce^{144}$ | I.V. | 40 | Immediately | 3 | 3.6 | 15 | 6 | 6 | 60 | 49 | 42 |
| $Nb^{95}$ | I.V. | 40 | Immediately | 3 | 37 | 44 | 14 | 18 | 9 | 12 | 42 |
| $Pu^{239}$ | I.V. | 40 | Immediately | 3 | 4.7 | 48 | 57 | 16 | 12 | 12 | 42 |
| $Y^{90}$ | I.V. | 40 | Immediately | 3 | 13 | 71 | 21 | 8 | 28 | 1.7 | 42 |

* I.V. indicates intravenously; I.P. indicates intraperitoneally.

**Table VII.* Effect of Treatment with Zirconium Citrate and Ethylenediaminetetraacetate (EDTA) on the Excretion and Retention of Mixed Fission Products by the Rat†**

| Tissue | Per cent of administered dose of mixed fission products | | | | | | |
|---|---|---|---|---|---|---|---|
| | Control | Zr citrate (25 mg Zr per rat) | | NaEDTA (10 mg per rat) | | Zr citrate + NaEDTA | |
| | | 1 hr Pre– | 1 hr Post– | 1 hr Pre– | 1 hr Post– | Zr 1 hr Pre– + EDTA 1 hr Post– | EDTA 1 hr Pre– + Zr 1 hr Post– |
| Skeleton | 30.0 ± 3 | 9.4 ± 0.8 | 18 ± 1 | 25 ± 3 | 25 ± 3 | 12 ± 1 | 17 ± 2 |
| Excreta | 35 ± 4 | 50 ± 3 | 43 ± 3 | 50 ± 9 | 42 ± 7 | 51 ± 4 | 63 ± 2 |
| Liver | 25 ± 5 | 14 ± 3 | 16 ± 2 | 12 ± 2 | 21 ± 2 | 10 ± 1 | 7 ± 0.4 |
| Muscle and skin | 5 ± 4 | 16 ± 2 | 12 ± 2 | 4 ± 0.2 | 9 ± 5 | 19 ± 5 | 10 ± 3 |

* Based on data in Reference 70.

† Each rat received 25 microcuries of mixed fission products, 255 days old, consisting principally of $Zr^{95}$, $Ce^{144}$, $Y^{91}$, $Sr^{89,90}$, and $Pr^{143}$. The rats were sacrificed 5 days after receiving the mixed fission products. Both the mixed fissions and the treatments were injected intraperitoneally.

**Table VIII. Distribution and Excretion in Rats of $Zr^{95}$ and $Y^{90}$ when Mixed with Sodium Citrate or Zirconium Citrate and Injected Intraperitoneally***

| Tissue | Per cent of injected dose† | | | |
|---|---|---|---|---|
| | Zirconium citrate | | Sodium citrate | |
| | Yttrium | Zirconium | Yttrium | Zirconium |
| Femur | 0.2 ± 0.05 | 0.2 ± 0.05 | 2.4 ± 0.3 | 2.4 ± 0.2 |
| Liver | 0.4 ± 0.1 | 0.3 ± 0.05 | 5.1 ± 1.0 | 6.2 ± 0.6 |
| Kidneys | 0.2 ± 0.03 | 0.3 ± 0.08 | 3.4 ± 0.3 | 4.8 ± 0.4 |
| Carcass | 2.0 ± 0.5 | 6.1 ± 0.8 | 65.4 ± 4.3 | 75.8 ± 3.0 |
| Urine | 91.2 ± 4.3 | 86.6 ± 14.8 | 16.4 ± 2.9 | 4.3 ± 0.7 |
| Feces | 6.0 ± 3.5 | 6.5 ± 2.8 | 7.3 ± 2.3 | 6.5 ± 0.7 |
| Total Excreta | 97.2 | 93.1 | 23.7 | 10.8 |

* After table in reference 42. Isotopes were injected in 1.6 ml of 10% sodium citrate solution, or 1.6 ml of a zirconium citrate solution containing a total of 40 mg of carrier zirconium. Rats were sacrificed 3 days after injection.

† Values in the table were adjusted to 100% recovery. Actual recoveries were 78.7 ± 7.0. Each value is the mean from 4 or 5 rats. The deviation is expressed as the standard error of the mean.

The effect of zirconium is greater the sooner it is administered after the injection of the radioisotope, just as was the case when chelating agents were used. An important difference, however, is that a single injection of zirconium given before or after administration of the radioisotope will continue to influence the excretion or retention of the radioisotope days or weeks depending on the initial dose of Zr.

When zirconium citrate is administered in combination with a radioisotope (i.e., simultaneous treatment), the effect on excretion is maximum. In an experiment by Kawin and associates[42] radioyttrium, $Y^{90}$, and radiozirconium, $Zr^{95}$, were mixed either with sodium citrate solution or with zirconium citrate. In three days the zirconium-treated rats excreted 97% of the $Y^{90}$, compared to only 24% by the sodium

citrate-treated rats (Table VIII). Following this simultaneous injection of radioisotope and Zr, the tissue distribution and excretion of the two are nearly parallel, indicating that the zirconium is indeed serving as a carrier for the yttrium.

The effect of varying the time of zirconium citrate administration on the metabolism of Pu and Y is shown in Table IX. The sustained action of a single dose of Zr is shown by the fact that it is possible to decrease skeletal deposition of injected Pu or Y even when Zr is administered 8 days before the radioisotopes. The urinary excretion of Pu can be increased even when Zr is injected 23 days earlier.[76] Aside from possible effects of zirconium citrate on the reabsorbability of radioisotopes during passage through the kidneys, these sustained effects of Zr are probably related to the fact that a small fraction of the Zr circulates in the bloodstream for several days after injection. Kawin, *et al.*[42] have demonstrated that a substantial fraction of injected zirconium citrate leaves the serum with a half-life of nearly 5 hours. However, there is little doubt that if the period of observation had been extended, longer-lived fractions would have been found.

Although it has been shown that the skeletal deposition of Pu or Y is markedly reduced when Zr citrate is administered soon after the radioelement (Table IX), this is not proof that Pu or Y are actually removed from the skeleton. Control animals given radioisotopes, but sacrificed at the time that zirconium was injected (1 hour later), contained the same skeletal levels of Pu or Y as the zirconium-treated animals sacrificed three days later.[76] Consequently, it appears that the Zr just prevented any *further* deposition of Pu or Y in the skeleton after the time of treatment. Additional evidence for this is that Zr causes a very rapid loss of circulating plutonium from the blood. Within 5 minutes after Zr injection the blood level of Pu in treated rats is less than half that of the controls, while one hour later the control rats had 53% of the Pu dose in the blood compared to only 6% in the Zr-treated rats.[76,92] Furthermore, within 1 hour after Zr injection the uptake of Pu by bone reached a plateau while Pu deposition in the bone of untreated rats continued.[92] Similar conclusions as to the mechanism of the effect of Zr on bone deposition of radioisotopes have been reached in the more extensive investigations of Kawin, *et al.*[42] on the kinetic aspects of radioisotope metabolism following treatment with zirconium citrate.

The effect of the amount of zirconium citrate administered on the excretion and distribution of injected Pu in rats is shown in Fig. 4. The amount of Pu excreted into the urine is proportional to the dose of zirconium, while the reduction in skeletal content was nearly independent of the amount of Zr injected or of the number of doses given. As little as 5 mg/kg of Zr is sufficient to cause a several-fold reduction in the Pu level in the skeleton. This means

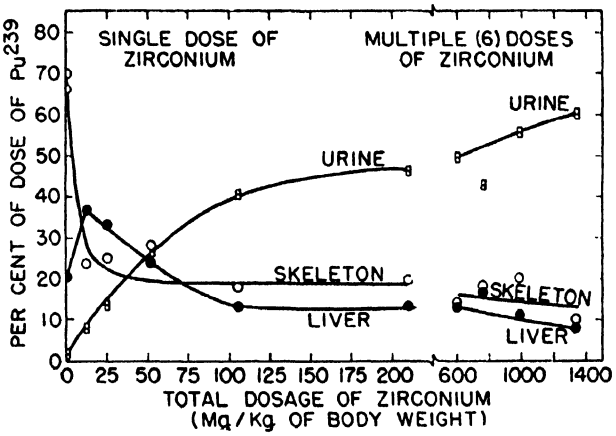



**Figure 4. The effect of different dose levels of intraperitoneally administered zirconium citrate on the distribution of plutonium in the urine and tissues of rats. Zirconium injections were begun 1 hour after intraperitoneal injection of $Pu^{239}$ and all rats were sacrificed at 72 hours. (From ref. 75)**

**Table IX. Effect of Time of a Single Administration of Zirconium Citrate Relative to the Injection of $Pu^{239}$ and $Y^{91}$ on Radioisotope Distribution in Rats***

| *Time of injection of Zr relative to Pu and Y* | *Dose of Zr citrate as mg Zr* | *Time of sacrifice after Pu or Y injection (days)* | *Per cent of administered dose of radioisotopes* | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | *Plutonium-239* | | | | | | *Yttrium-91* | | | | | | |
| | | | *Urine* | | *Skeleton* | | *Liver* | | *Urine* | | *Skeleton* | | *Liver* | | |
| | | | *Controls* | *Treated* | *Controls* | *Treated* | *Controls* | *Treated* | *Controls* | *Treated* | *Controls* | *Treated* | *Controls* | *Treated* | *Reference* |
| Pre-Treated: | | | | | | | | | | | | | | | |
| 1 day | 48 | 3 | – | – | 67 | 12 | 18 | 22 | – | – | 61 | 19 | 4.4 | 4.9 | 76 |
| 2 days | 48 | 3 | – | – | 67 | 23 | 18 | 17 | – | – | 61 | 25 | 4.4 | 2.9 | 76 |
| 3 days | 48 | 3 | – | – | 67 | 46 | 18 | 21 | – | – | 61 | 33 | 4.4 | 3.5 | 76 |
| Post-Treated: | | | | | | | | | | | | | | | |
| 0.5 hour | 51 | 6 | 1.4 | 64 | 73 | 12 | 12 | 11 | 27 | 51 | 42 | 16 | 1.5 | 1.8 | 38 |
| 1.0 hour | 40 | 3 | 1.3 | 46 | 68 | 20 | 21 | 14 | – | – | 47 | 38 | 4.9 | 3.5 | 75 |
| 1.0 hour | 30 | 3 | – | – | 66 | 16 | 21 | 23 | – | – | 49 | 33 | 8.3 | 7.1 | 76 |
| 24 hours | 40 | 10 | – | – | 61 | 40 | 15 | 11 | – | – | 54 | 41 | 2.7 | 2.6 | 76 |
| 72 hours | 51 | 6 | 1.4 | 7.0 | 73 | 61 | 12 | 18 | 27 | 33 | 42 | 43 | 1.5 | 2.0 | 38 |

* All injections were either intraperitoneal or intravenous.

that it is possible to minimize skeletal deposition without a concomitant rise in urinary excretion—indeed an important point in evaluating the effects of Zr treatment in cases of human radioactive poisoning.

The effect of Zr on Pu excretion is largely independent of the concentration of Pu injected[64] and the valence state of Pu.[42,78]

The effects of Zr may be altered by changing the particle size distribution in the Zr injection solution. It is possible to prepare the zirconium citrate solution so that the ratio of citrate to zirconium is decreased. This has the effect of increasing the particle size of the carrier colloids so that a greater percentage of the Zr is found in the liver and much less is excreted in the urine. When such preparations, or similar ones with other metals, are injected into Pu-injected animals it was found that the amounts of Pu in the skeleton are reduced, but that no proportional increase in urinary excretion occurs. Further, the liver possessed higher than normal levels of Pu.[76] An excellent experimental demonstration is given by Gofman *et al.*[91] of the relationship between the distribution and excretion of injected colloids relative to their chemical composition (ratio of complexing agent to metal) and particle size.

Preliminary evidence[78] indicates that zirconium citrate treatment may be far more effective in dogs than in rats. Administration of zirconium citrate to dogs injected intravenously 30 minutes previously with Pu(IV) actually removed most of the deposited Pu from both the skeleton and soft tissues. It reduced the over-all body content tenfold and the skeletal level approximately 20-fold (Table X). In this case Pu was evidently actually removed from the skeleton and other tissues. At the time Zr was injected about 50% of the injected dose of Pu was in the blood (calculation based on extensive metabolic studies on several dogs.)[36] Since 90% of injected Pu was removed from dog, about 40% must have come directly from skeleton, liver, and other organs.

Investigations have been made to ascertain whether the use of parathormone might increase the effectiveness of zirconium.[42] It was thought that if resorption of bone by parathormone could mobilize Pu and Y, they might be more susceptible to the action of zirconium. However, it was found that parathormone action did not enhance the effect of zirconium on the excretion or internal decontamination of Pu or Y. Neither did the parathormone alone have any effect.

The effectiveness of Zr treatment in chronic radioactive poisoning has not been studied adequately. It is known that the administration of Zr citrate weeks, months, or even years after injection of Pu will cause a significant rise in the urinary output. For example, in a dog treated 5 months after Pu injection, the urinary excretion of Pu rose following the use of only 4.15 mg/kg of Zr. A larger injection of Zr (15.8 mg/kg) about a month later produced a 3-fold increase in urinary excretion and kept the urinary Pu at a slightly elevated level for the following two to three weeks.[38] It is reasonable to conclude that optimal doses of Zr could have produced a larger increase in the urinary excretion of Pu. An injection of zirconium citrate into this dog 2½ years after Pu exposure also produced a prompt rise in excretion. However, whether repeated injections of Zr can remove long-standing deposits of Pu or other fission products has not been determined by adequate experimentation. A start has been made by Hackett.[77] Thirty-two days after rats were injected intravenously with Pu(IV), one group received intraperitoneal injections of Zr citrate a week for 5 weeks. The rats were sacrificed 71 days after Pu injection. Some reduction in skeletal concentration of Pu appears to have been obtained but the data were too scattered to establish significance.

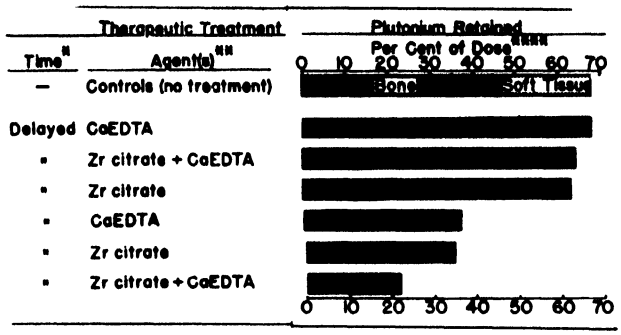
There are several considerations in the treatment of radioactive poisoning in humans by zirconium citrate. The most important is that a sufficient dose of zirconium be given as soon as possible after the exposure in order to obtain a maximum excretion and minimum retention of the radioelement. Treatment should be intravenous. If it is desired only to minimize bone deposition, doses as small as 5–10 mg/kg of Zr may be sufficient. With such small doses the urinary excretion may not be appreciably altered. Since the biological half-life of Pu in the soft tissues is far greater than in the bone and since Pu may be more easily removed from the soft tissues, it should be possible to reduce the long-term retention of Pu by the soft tissues, particularly if Zr treatment is continued with injections administered approximately once a week. If, in addition to the small doses of Zr, CaEDTA is utilized, then there is little doubt that the soft tissue levels of the radioisotope would be decreased. The results in animals of combined CaEDTA-Zr citrate treatment are covered more fully in the next section.

If the results on dogs given in Table X are confirmed and are applicable to humans, then it is conceivable that treatment with Zr citrate given at spaced intervals in doses of at least 50 mg/kg may shorten the biological half-life of Pu by a factor of approximately 50 or more.

The rate at which the blood level of Pu following Zr treatment returns to or approaches the pre-treatment level should be determined. This information would facilitate the design of the most effective treatment schedule and, in addition, allow estimations to be made of the rates at which depletion of the tissue deposits occurred.

### Treatment of Radioactive Poisoning by Combined CaEDTA and Zirconium Citrate

In acute studies with rats injected with plutonium, zirconium citrate causes greater reductions in bone Pu deposition while EDTA is more effective in reducing soft tissue concentrations. A solution containing both agents would therefore be expected to be more effective than either alone. The experimental results[63,64] shown in Table XI and Fig. 5 give striking proof of the greater effectiveness of the Zr citrate-




*Delayed therapy initiated 30 days, prompt therapy 30 minutes, after plutonium administration.

** Zr citrate dose = 50 mg as Zr administered intraperitoneally, CaEDTA dose = 600 mg administered intraperitoneally. Where the two agents were administered, half these amounts were employed.

**Figure 5. Effect of combined CaEDTA and zirconium citrate administered to rats at varying time intervals after plutonium injection on the retention of plutonium by bone and soft tissue. The control group contained 16 animals and the treated groups contained from 4–12 animals. (Adapted from Table 1 in ref. 63)**

CaEDTA combination in reducing retention of Pu as well as in furnishing a comparison of the individual therapeutic agents under the same experimental conditions. The reduction in skeletal deposition following the combined treatment was no different from that obtained with Zr alone.

In the case of 255-day old mixed fission products, the combined use of EDTA and zirconium citrate did not improve skeletal decontamination over that obtained with zirconium citrate alone (Table VII). In this case, Zr alone reduced appreciably the skeletal level of radioelements, while EDTA alone caused a small decrease in the amount of fission products deposited in the skeleton.[70]

Further investigations on the use of combined therapeutic agents for internal decontamination are necessary.

### Miscellaneous Approaches, Chemical and Physiological, for the Treatment of Radioactive Poisoning

Attempts to treat radioactive poisoning by several other methods have been made. None of these has had a significant effect, at least for the specific radioisotope tested. They include:

1. Induced diuresis by injection to dogs of hypertonic saline in the case of Pu.[38]

2. Disturbance of the organic matrix of bone by induction of scurvy using a scorbutogenic diet in the case of Pu, Sr, and Y.[26]

**Table X.* Effect of Zirconium Citrate Treatment on the Excretion and Retention of Injected Plutonium(IV) in Dogs†**

| | *Per cent of administered dose of Pu(IV)* | | | | |
|---|---|---|---|---|---|
| | | *Zr treated (mg Zr/kg per injection)* | | | |
| *Tissue analyzed* | *Controls* | *50* | *100* | *200* | *400* |
| Skeleton | 81.0 | 3.8 | 4.2 | 7.9 | 2.6 |
| Liver | 19.0 | 0.5 | 3.0 | 1.0 | 0.5 |
| Spleen | 0.009 | 0.12 | 0.37 | 0.11 | 0.05 |
| Blood | 0.1 | 0.18 | 0.02 | 0.005 | 0.009 |
| Carcass (less organs and skeleton) | 0.8 | 6.5 | 0.6 | 1.4 | 0.3 |

* Based on data in Reference 78.

† The Pu solution injected was plutonium citrate buffered to *p*H6. The controls consisted of two dogs, while only 1 dog is represented by each Zr level in the treated groups. Treatment was begun 30 minutes after the injection of Pu and was repeated twice each week for three weeks (6 injections total). Both Pu and Zr were injected intravenously.

**Table XI. Comparison of Effects of Zr Citrate and CaEDTA on Pu Retention in Rats* (Per Cent of Administered Pu Retained)**

| | | *Treated rats* | | |
|---|---|---|---|---|
| *Tissue* | *Controls* | *Zr citrate (50 mg Zr per rat)* | *CaEDTA (600 mg per rat)* | *Zr citrate + CaEDTA† (25 mg Zr + 300 mg EDTA)* |
| | | Pu Dose = 0.0014 μg per rat | | |
| Skeleton | 57.7 ± 4.0 | 12.7 ± 1.2 | 33.2 ± 2.6 | – |
| Soft tissue | 14.8 ± 3.6 | 14.8 ± 3.6 | 5.1 ± 0.9 | – |
| Total animal | 72.6 ± 3.3 | 27.5 ± 4.2 | 38.2 ± 2.6 | – |
| | | Pu Dose = 140 μg per rat | | |
| Skeleton | 46.3 ± 5.2 | 13.9 ± 5.6 | 27.5 ± 2.7 | 10 |
| Soft tissue | 24.7 ± 4.8 | 21.2 ± 5.9 | 10.8 ± 5.0 | 12 |
| Total animal | 71.0 ± 4.6 | 35.1 ± 10.9 | 38.3 ± 3.4 | 22 |

* Based on data in References 63 and 64. Rats were given intraperitoneal treatment 30 minutes after the intraperitoneal injection of Pu(IV) citrate. All animals were sacrificed 30 days after Pu injection.

†Values taken from a graph in Reference 63.

3. Use of a protein-free diet in the case of Pu and $Sr^{26}$ to disturb bone protein.

4. Causation of bone resorption with subsequent use of a diet calculated to restore the bones with new, nonradioactive bone, in the hope that the new bone would act as a mass absorber to prevent radiations from deposited Pu from reaching living cells.[19]

5. Use of endocrine changes such as cretinism which was produced by thyroidectomy of rats at birth. The resulting hypothyroidism inhibited the deposition of radiostrontium in the skeleton. Growth hormone and thyroxin were administered either alone or together to the hypothyroid rats. These treatments increased the skeletal uptake of $Sr^{89}$ and $Sr^{90}$ to levels which in some cases exceeded those of the normal controls.[93]

It is conceivable that stimulation of the cells of the reticuloendothelial system may enable them to cope more adequately with phagocytosis of radioactive particles. Substances such as oestradiol benzoate and choline cause stimulation of these cells.[94,95] Choline has been found to enhance the rate of disappearance from the blood of colloidal $CrP^{32}O_4$.[95]

An interesting possibility in the treatment of radioelement poisoning is the modification of tubular resorption processes in the kidney. In the case of radium, for example, more than 90% of the radium passing through the kidney is resorbed back into the circulation. Even a relatively small decrease in the degree of tubular resorption of radium, caused by use of selective blocking agents for the renal tubules, could cause a significant increase in the urinary excretion.

Certain pathological conditions such as diabetes or normally occurring physiological changes of many types, such as those that follow exercise, result in increased blood levels of organic compounds possessing chelating properties. Investigations of the effects of such induced changes in animals, e.g., alloxan-induced diabetes, would be of fundamental interest in the mechanisms of transport and removal of radioactive elements from the body.

## TREATMENTS FOR POISONING BY NONRADIOACTIVE METALS

Treatment of nonradioactive metal poisonings can be approached in two ways: (1) the concentration of the metal in the body can be lowered by promoting excretion, or (2) the metal can be inactivated *in situ* so that it no longer can react with and damage the surrounding tissue. In the case of the first, the same principles apply for nonradioactive metals as have been discussed in the preceding sections.

For nonradioactive metal poisons it is feasible to inactivate a metal *in vivo* by the formation of a chelate without a concomitant change in its distribution or localization. The use of the lake-forming dye aurintricarboxylic acid, which combines specifically with beryllium under physiological conditions has proven successful in the treatment of acute experimental beryllium poisoning.[96] The dye does not change the tissue distribution pattern or the excretion of the metal. It prevents the pathological damage caused by the beryllium in the liver, spleen, and bone marrow which otherwise would kill the animal.[97] Factors which enhance the effectiveness of the dye, in addition to a binding affinity for the metal, are low water solubility, high affinity for proteins, and the fact that it does not react with calcium ion in the plasma. The toxic effects of $Co^{++}$ can be prevented by the administration of cysteine,[31] with which $Co^{++}$ forms an unusually stable complex. The strong complexing action of BAL for metals such as gold, mercury, arsenic and lead is well known. Some protection against poisoning by uranium, vanadium, and cobalt is afforded by sodium catechol disulfonate, which seems to form a chelate with the hydroxy groups.[98,99]

Some of the important features which can modify the action of an agent forming a chelate *in vivo* with metal ions are:

1. The biological behavior of the agent. This includes its rate of metabolism and excretion, its distribution in the tissues relative to the metal, its permeability, and its affinity for tissue protein. These are affected by such things as the acidity of the coordinating groups, i.e., the degree of ionization of the chelating agent, the size and shape of its molecule, and the presence in the molecule of water-solubilizing groups.

2. The stability of the chelate formed with the metal *under physiological conditions*. The influence of these factors is discussed in Ref. 96 with regard to the chelate formed between aurintricarboxylic acid and related compounds with beryllium.

Some of the potential ill effects which may result from the use of chelating agents in the treatment of metal poisoning have been reviewed and documented elsewhere.[100] They may be summarized as follows:

1. The solubilization of insoluble metal deposits by the chelating agent may make more free metal available for reaction with tissues. This point, which we have demonstrated experimentally, illustrates the fact that the toxicity of nonradioactive metals is not always a function of the total concentration of the metal present in the body, but rather of that fraction which is able to undergo chemical reactions *in vivo*.

2. When the metal is chelated it may be transported to tissues to which it normally would not penetrate to an appreciable extent and, if the chelate then becomes broken down, cause tissue damage. It has been demonstrated that the amounts of some metals deposited in bone may be increased by the use of a chelating agent.

3. While the chelate may be stable at pH 7.3, it may decompose during passage through the kidney, where the pH is significantly lower, thus liberating the metal and causing renal damage. It is highly probable, for example, that the calcium EDTA chelate is partially dissociated when passing through the kidney tubules. EDTA does not complex calcium at pH 4 so that a considerable part of the calcium may be free at the pH of the tubules or at the sur-

face of the tubule cells where exchange of cations for hydrogen ions seems to take place. Several examples involving kidney damage caused by certain BAL-metal complexes are known.

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# Environmental Aspects of the Atomic Energy Industry

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Every new industry has a deep-seated responsibility to appraise, as accurately as possible, the impact of its operations on community living and the natural resources of the nations which it serves. Such major components of an industry as its sources of raw materials, its production plants, its research and development laboratories and its channels of transport and distribution, have a wide variety of direct and indirect contacts with people and the communities in which they live. A new industry moving into an established community has, as a new neighbor, a special responsibility to respect the vested rights of people and other industries already there. Even though this industry may enter an isolated unpopulated area it still has an obligation to assess its impact on the natural resources of the region and the long term effect of its operations on future possible communities. It is axiomatic that people follow industry. Conceivably through use of atomic energy in the future, an almost virgin area today may be the site of a city or a vast industrial complex.

The purpose of this paper is to discuss certain environmental aspects of the fast growing atomic energy industry in the United States. Experiences of this industry over the past decade will be discussed in the light of the history of other major industries of this nation over a long time.

In general, the environmental problems which a new industry encounters are inversely proportional to the advance planning for the industry and more or less proportional to the volume and objectionable or hazardous properties of its product and its wastes. Such problems may develop as a result of day-to-day operations or from an unforeseen incident or accident. Both situations must be reckoned with and each is capable of reasonably intelligent evaluation.

In the new atomic energy industry the obligation to assess its environmental impact is all the more profound because of: (1) the unique characteristics of its products and wastes—especially those of long lived radioisotopes; (2) the relatively limited knowledge as to the effect of cumulative radiation on living organisms; (3) the continuing changes in nuclear technology; (4) the relatively large volumes of water used for heat exchange and other purposes; (5) the trend of the industry to expand into more populous areas; and (6) the current lack of knowledge (due largely to security controls) concerning the products and operational hazards of the industry by regulatory public agencies normally responsible for public health, safety and the protection of national resources.

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## EXPERIENCE OF OTHER INDUSTRIES

As this new industry goes forward in all parts of the world—and perhaps in areas where no other industry plant has ever been located—its environmental aspects should be discussed with candor and be evaluated with thoroughness. This conclusion is supported by the experiences of other industries such as textiles and tanning, brewing and distilling, lumber and coal, paper and pulp, by-product coke and chemicals, packing and food products, metals and metal fabrication, smelting and refining, synthetic fibres, pharmaceuticals, plastics and explosives. In its operations and growth each had its special impact on the environment, on public services and on natural resources. In order to comply with reasonable environmental standards, management of these industries has experienced a long and costly series of difficulties in correcting objectionable features of plant location and operation and in treatment and disposal of wastes.

These industries, in their earlier operations, met with objections by the public on various scores including: (1) lowering of ground water; (2) drain on limited surface water resources; (3) contamination of the atmosphere; (4) destruction of crops and vegetation; (5) pollution of ground and surface water resources; (6) contamination of public bathing and recreational areas; (7) damage to public sewers and sewage treatment plants; (8) destruction of shell fish areas; and (9) emission of objectionable odors and noises. Not infrequently these objectionable environmental deficiencies resulted in costly litigation and in some instances in the passage of drastic legislation for regulation and control which unduly handicapped the industry. In recent decades industry and government with commendable courage and foresight have worked together to resolve and to prevent these environmental problems. Through cooperative research participated in by industry, governmental agencies and universities various environmental problems such as those listed above have been and are being resolved at an encouraging rate.

## EVALUATING ENVIRONMENTAL PROBLEMS IN THE ATOMIC ENERGY INDUSTRY

The United States Atomic Energy Commission has sought to benefit by the experience of other in-

dustries in evaluating the environmental problems created by operations of plants under its jurisdiction. It has followed the pattern of these industries in its environmental and public safety research programs. The tempo of research, however, has been much more rapid. It has enlisted the cooperation[1] of specialists in other industries, other governmental agencies, universities and professional experts in private practice.

A start was made by studying the effect on the environment of operations at the plants constructed hurriedly and at isolated places during the war years. Special emphasis was given to (1) the effect of using large volumes of water from ground water sources and (2) release of radioactive and toxic wastes to the ground, to surface waterways and to the atmosphere. In addition, tests were made to determine the effectiveness of treatment facilities and processes installed to remove radioactive and toxic substances from waste streams of all kinds—gaseous, liquid and solid.

### Uses of Specialists

Experience has revealed that a proper appraisal of environmental problems of the atomic energy industry requires competence and experience of a high order and by a wide variety of specialists. They include such professionals as nuclear, health and biophysicists; physical and nuclear chemists; structural and ground water geologists; nuclear, chemical, sanitary and safety engineers; industrial hygienists; biologists; meteorologists; hydrologists; mineralogists; public planners and others.

### Problems Evaluated

Teams of specialists have and still are reviewing and advising in such matters as: (1) site selection for plants; (2) the availability of water for process and domestic uses; (3) the relationship of various units in a plant complex to one another; (4) the type and capacity of safety and waste treatment facilities; (5) the degree of waste treatment required; (6) points of discharge of waste effluents; (7) the natural dilution factors in nature which may be taken advantage of in disposal of wastes; (8) the selection of significant monitoring points for checking the results of operations; (9) the ultimate fate of wastes from day-to-day operations or in case of an accident, loss of product and wastes to the environment; and (10) the effect of releases of this kind on the public health or natural resources.

### Need of Training

A difficulty encountered in enlisting the services of experienced professionals is that many are not too familiar with the technology, the terms and the standards of the nuclear energy industry. This is, however, but another of the challenges the industry must face. It is one which can be met with confidence of success under an aggressive educational and training program. The environmental problems which lie ahead in the development of the atomic energy industry can best be resolved when it becomes more fully integrated into the community and industrial structure of the nation. Policies and programs are bringing this about in a rational and effective manner.

## IMPORTANCE OF SITE SELECTION

Much study of environmental factors should be conducted in advance of selection of a satisfactory site for an atomic energy plant. Advance study of the geological, meteorological and hydrological aspects of a plant site could result in substantial savings in the layout, design and operations of such important units as nuclear reactors, chemical processing, gaseous diffusion and fuel fabrication plants and research laboratories. In the construction and installation of facilities in these plants much care must be exercised in preventing dusts from contaminating piping for process gases, ducts for ventilation and sensitive equipment and instruments. These precautions transcend those of most other industries.

Where water in large volumes or of special quality is required, sources of supply should be carefully investigated as to: (1) adequacy and continuity of supply; (2) the possibility of interference with the supply of other consumers—especially those having prior vested rights; and (3) the physical and chemical characteristics of the water.

Usually, but not always, it is the large plants which present the more serious environmental hazards. Of special importance in the case of nuclear reactors or chemical processing plants is advance knowledge as to the kind, volume, and levels of radioactivity to be handled and the availability of suitable places and conditions for release of waste effluents. This is not to say that selection of sites for such other units as research laboratories, feed material processing and fabrication plants and special test and storage facilities are not also important.

Generally, the problems presented are a function of: (1) the curies of radioactivity involved; (2) the length of time the plant has been in operation; (3) the adequacy of safety and waste treatment facilities provided; and (4) the proximity of the plant to populated areas and sensitive industries. Rarely are the environmental considerations of one site the same as those of another. Each must be studied with respect to its own local conditions.

Appendix A of this report contains a check list of environmental considerations which may be helpful to any who have the responsibility for selection of sites for nuclear reactors and chemical processing plants or others.

### Plant Enlargements

In site selection serious consideration should be given to the possibility or probability that a plant as originally built may be enlarged, or its functional processes changed with resultant greater hazard. When a plant or site planned for one purpose is put to a new use it is important that such basic services as utilities, waste disposal systems and points of release of waste effluents be re-studied to ascertain their adequacy for the newer use. Such modification

should be discussed with those public officials who are responsible for public health and safety in the area. If the original plant was served by public utilities such as water, power, sewers and other drainage facilities this obligation is all the more pressing.

## DISPOSAL OF WASTES

### High-Level Radioactive Wastes

High-level waste may contain as much as $10^2$ curies per liter. Their principal source under normal operations is in the processing of irradiated fuel elements. The cost of treatment and disposal of these wastes is high. Costs have been omitted purposely in this paper because they are dealt with in other papers of this series.

If nuclear power is to compete favorably with other fuels, costs of waste disposal must be reduced. Cutting of costs must be done intelligently for otherwise it might involve risks which could present environmental hazards and reflect unfavorably on the industry.

Disposal policies are especially important in the case of high level radioactive wastes which contain long-lived and biologically significant fission products[2] such as $Sr^{90}$ and $Cs^{137}$ and others of shorter half-life such as $Ce^{144}$, $Ru^{103}$ and certain isotopes of rare earths which may be difficult to control when released to soils.

As the industry grows should disposal of high-level long-lived radioactive wastes by environmental dilution continue to be considered, the amounts of dilution required to meet permissible concentrations in air and water for continuous exposure of humans could be fabulous. W. Kenneth Davis,[3] Director of the Atomic Energy Commission Reactor Development Division, in a paper before the American Power Conference in Chicago in April 1955 estimated that the capability of nuclear power in service in the United States would be:

| *By the end of:* | *Million kw* |
|---|---|
| 1960 | 2 0 |
| 1965 | 5.0 |
| 1970 | 27.0 |
| 1975 | 83.0 |
| 1980 | 175.0 |

This power would of course be the output of our many reactors, probably widely distributed. The estimated rapid rate of increase, however, is striking and serves to give some indication of the importance of resolving the problem of disposal of radioactive wastes which are long-lived and hazardous. In the fissioning of 1 gram of uranium one megawatt day of nuclear heat energy is released and about one gram of fission products is formed. One year after removal from the reactor, assuming continuous uniform operation, one gram of fission products would have a heat power level of about one watt or the equivalent of 500 curies of radioactivity.

My associate, Joseph A. Lieberman, in a recent paper[4] before the American Society of Civil Engineers has calculated that for the above 1980 rate capability the fission product production for one year would require in the order of $1.85 \times 10^{18}$ gallons of water to dilute $Sr^{90}$ (yield 5.3 per cent) to safe lifetime drinking water levels. This is approximately equivalent to the average annual flow of the Mississippi River prior to flood diversion 100 miles above its mouth for a period of 12,600 years.

### Fixation on Soils

Fortunately certain soils[5] and the suspended and bed loadings of most waterways have properties of absorption or adsorption of radioactivity. Nature provides some potentialities for resolving environmental problems. These are being studied in order that they may be taken advantage of in reducing the cost of disposal of wastes from chemical processing of spent fuels and from other sources. The exchange capacities of soils for radioisotopes can, however, be seriously affected by other non-radioactive ions in wastes. This complex should be fully evaluated in deciding on the degree of pre-treatment which is required before wastes are disposed to the ground. The heat in high level waste resulting from gamma radiation introduces an important problem in the disposal of these wastes.

Research in ground disposal is under active investigation at the Oak Ridge National Laboratory as an important environmental problem associated with the development of future reactors for power production. Under contracts with Atomic Energy Commission, staff of the Sanitary Engineering Department of the Johns Hopkins University and the Geological Survey are giving consideration to the feasibility of disposal of these wastes to deep wells or to deep cavities existing naturally or made by dissolving salts from deep dry deposits. The Earth Sciences Division of the National Research Council is also cooperating in this program.

The requirements of environmental protection could be met by fixing radioactivity in clays or other suitable material and then raising the temperature sufficiently to form a solid mass from which the wastes could not be elutriated or leached. Such a mass could then be buried in tight soils designated by a geologist as suitable for waste storage. Research to determine feasibility and cost of such a method is underway at the Brookhaven,[6] Oak Ridge and Los Alamos National Laboratories.

### Separation of Significant Isotopes

Another possibility of lessening the environmental hazards associated with the disposal of high-level radioactive wastes is to remove the long-lived and biologically significant isotopes from the wastes prior to disposal. If this were done disposal to the ground in selected areas could be carried out with greatly reduced environmental hazards. The significance of this consideration may be seen from the fact that in NBS Handbook[7] No. 52 the maximum permissible limits for exposure of people to $Sr^{90}$ is about a factor of $10^3$ more conservative than for most of the radioisotopes. Admittedly this procedure would be costly, but the possibility of selling such isotopes as $Sr^{90}$,

$Cs^{137}$ and $Ru^{103}$ and others as radioactive sources may offset some of this cost. After these sources have served their purpose they still must be disposed of under safe conditions in order to prevent environmental problems.

### Land Burial

Burial is an economically attractive method of disposal of solid wastes but it may present serious environmental problems.

The selection of burial grounds should be made in cooperation with an experienced geologist. Designated burial grounds should be fenced and be well identified. Access to them by unauthorized persons should be prohibited. Provision should be made also to prevent access by animals. A continuing inventory of buried wastes should be maintained and test holes for monitoring the movement of wastes away from the site should be provided. Burial grounds should be a minimum in number as they are dedicated areas so contaminated as to prevent, for all practical purposes, the use of the land for other services for very long periods of time.

### Underground-Tank Storage

Underground storage without fixation of the hazardous long-lived radioisotopes could have long term implications affecting the welfare of future generations. Waste-burial grounds and underground-storage tanks should be located so that in case of leakage pollution of ground water may be minimized. Preferably, they should be set well above the water table and in tight soils from which movement of any leakage would be slow. Provision should always be made for periodic monitoring in the vicinity of these storage areas to detect any leaks.

Storage of high-level radioactive wastes in underground tanks as currently practiced has the advantage of confinement, thus providing time for decay of radioactivity. Provisions to remove heat from tanks when required is costly. Tank storage is not an ultimate solution of the waste disposal problem. The wastes may be radioactive for hundreds of years whereas the tanks in which they are stored may be expected to corrode and leak in 50 to 75 years or sooner. Obviously, therefore, this method of handling still involves a serious potential environmental problem.

### Low-Level Wastes

The release of low-level radioactive wastes from the atomic energy industry also presents environmental problems. These wastes are those whose activity is $10^3$ or $10^4$ in excess of permissible long term limits of exposure for humans. Because the quantities involved are very large, where conditions are favorable for dilution in the atmosphere, in surface waterways or to the ground, such disposal is economically attractive and has possibilities. It is being practiced in the United States—especially in certain remote areas. There are many places even near populated areas where conditions are favorable for disposal by dilution in nature and they should be taken into consideration in selecting a plant site. In appraising these possibilities the advice of experienced meteorologists, geologists and hydrologists should be sought.

Extensive research in determining the significant parameters in making such appraisals is being carried out under Atomic Energy Commission contracts with the Weather Bureau, the Geological Survey and several large universities. In addition, staff of the Atomic Energy Commission and its operating contractors at the Hanford Works in Washington, the Knolls Atomic Power Laboratory near Schenectady, the National Reactor Testing Station in Idaho and at the Brookhaven, Argonne and Oak Ridge National Laboratories are conducting similar research.

### Ocean Disposal of Wastes

There are some who consider the oceans as a satisfactory place for disposal of radioactive wastes of all kinds. To a limited degree this practice has been followed at certain plants and laboratories on or near the Atlantic and Pacific Coasts in the United States. Usually these wastes are enmeshed in a mixture of concrete within steel drums or wooden forms. Considering the pressures they are subjected to and the probable impact on the ocean floor, it is doubtful if these containers hold the waste very long after dumping at sea. The wastes are of a variety of types, some of which are high in activity. The volumes and curies in these wastes are small compared with the total volume and activity in high-level wastes in storage. Disposal is in waters between 500 and 1000 fathoms in depth. Sea disposal of wastes is costly for plants which are not near the sea coast. A study of the cost of sea disposal of low and intermediate level wastes from atomic energy installations conducted by staff of Johns Hopkins University in 1953 and 1954 indicate a range of from $0.30 per pound in the New York area and from $0.80 to $1.00 per pound in the San Francisco area. The difference is reflected by the volumes of waste disposed of in the respective areas.

Some consideration has been given to the feasibility of disposal into the oceans of large quantities of high-level wastes from chemical processing plants. In the summer of 1954 a seminar was held at which experienced oceanographers discussed the problems presented. Sufficient concern was expressed over this method of disposal because of important environmental risks as to call for much field study before it should be approved as standard practice for the industry.

### Disposal in Remote Areas

It is reasonable to assume that within the next decade atomic energy plants may be built in remote places throughout the world where the need of power for special purposes is so important that the factor of cost may not be too significant. Here again the industry has a real obligation to maintain high standards of safety and environmental sanitation. Even

though initially exposure of people and property in these remote areas may be slight, a reckless or capricious attitude in disposal of long-lived wastes should not be permitted. With the advancement in travel and transport to these areas and perhaps unpredictable uses of their natural resources, a careless practice in this generation in the interest of low costs could pre-empt or penalize the use of these resources by future generations. History is replete with examples of the penalties which subsequent generations have paid for the reckless, uncontrolled actions of their forefathers.

## OTHER ENVIRONMENTAL PROBLEMS

### Spills of Wastes

Should a serious spill occur from an atomic energy installation, there is an obligation to notify promptly public officials who may be concerned. In anticipation of such an incident it would be prudent to develop a system of notification of public officials so that they in turn may give adequate warnings to others who may be affected. Through cooperation of this kind the effect of a serious incident may be lessened to such a degree as to prevent damage to others downstream. Serious spills of toxic wastes and products to sewers or streams have occurred in a number of industries. Through prompt notification under a planned arrangement a situation which might otherwise have caused serious damage to property or a threat to the public health was avoided.

### Transport of Radioactive Material

From mining the raw ores used to obtain radioactive materials to the ultimate disposal of wastes care must be taken to prevent exposure of people or property to damaging radiations. Accidents in transport may be of a wide variety. They are unpredictable as to when and where they will take place and the extent of the damage which will result. Therefore, much care must be given in planning: (1) the transport of these materials; (2) the carrier to be used; (3) the routes to be followed; and (4) the packaging of the radioactive or toxic material. In the case of highly radioactive materials shielding to protect handlers and others and packaging under conditions which will avoid criticality are extremely important. These precautionary measures add substantially to costs.

Accidental spills of radioactive materials in transport especially in populated areas, on water sheds of public water systems and on bridges over streams or rivers from which water for industry or public supplies is obtained present real environmental hazards. In the wreck of a train or truck carrying radioactive materials should a fire occur a serious situation could develop spreading radioactive gaseous effluents over a wide area. Crews attending the transport of these materials should be well informed as to hazards en route and to safety measures to follow in case of an accident. They should carry monitoring instruments by which to evaluate hazards which may arise.

The cost factor in transport is one of the reasons why it is desirable to have the chemical processing plant reasonably near the reactor and why in the disposal of radioactive wastes suitable places for release and ultimate disposal should be sought near the processing plant.

### Radioisotopes

Although the levels of activity of manufactured radioisotopes generally used in research are small in comparison with those of materials used in reactors and chemical processing plants, nevertheless, their use is widespread[8] and could create environmental hazards if careful controls are not applied. At institutions using these isotopes care must be given to the release of radioactivity to building plumbing and to public sewers in order that overexposure of maintenance personnel may be avoided. The effect of radioactivity on organisms used in public sewage treatment processes and the ability of these processes to remove radioisotopes from wastes have been studied at the Johns Hopkins, New York, California and Illinois Universities and at the Massachusetts Institute of Technology. No serious effect of low level wastes on these organisms were found. Removal of radioactivity from wastes by biological treatments is not too encouraging.

Destruction of combustible radioisotopes in ordinary institutional incinerators could create environmental hazards. The Bureau of Mines[9] under contract with the Atomic Energy Commission has developed an incinerator especially designed to handle combustible radioactive wastes. From a combustion standpoint the main feature is use of high-velocity tangential over-fire air. Provision will be made for high-efficiency filtration of the gaseous effluent.

### Nonradioactive Wastes

The atomic energy industry has problems in disposal of wastes other than those which are radioactive. In the preparation of reactor fuels large volumes of fluorides are used which appear in waste streams. Since fluorides in drinking water in excess of 15 ppm[10] may have a deleterious effect on the teeth of children the disposal of these wastes present an environmental problem. It is not, however, one too difficult to cope with since in combination with calcium, fluorides can be immobilized in an insoluble form. Substantial quantities of nitrates are also produced in certain chemical processes. If released under conditions where they could contaminate sources of drinking water supplies a serious problem would be presented. Nitrates in drinking water in excess of 50 ppm[11] could cause nitrate cyanosis in children. The physiological effect of nitrates in water drunk by animals needs to be fully investigated. Beryllium fluoride as a gaseous effluent produced in the manufacture of beryllium for the industry has been reported[12] to have serious—even fatal—effects when inhaled by adults. Ammonia used in certain processes for production of zirconium—an important new product used

in the assembly of reactor fuels—is known to appear in wastes in sufficient amounts to be toxic to fish.

## CONCLUSIONS

It is evident from the experience of other industries that as the atomic energy industry expands to serve mankind throughout the world it will present important problems in environmental sanitation and public health. Of special importance will be the disposal of long-lived highly radioactive wastes from nuclear reactors and chemical processing operations for recovery of nuclear fuel. Currently, disposal of these wastes is the subject of much research and development work in the United States. The cost of safe disposal of radioactive wastes is much higher than for wastes in most other industries. Less expensive methods, therefore, must be developed or the industry will be at an economic disadvantage.

There is much evidence that for the large volumes of low and intermediate level radioactive wastes produced factors, of dilution in nature such as disposal to the ground, to the air, and to other surface waterways can with safety be taken advantage of to reduce these costs. Should atomic energy plants be built in remote unpopulated areas of the world the environmental aspects of waste disposal practices should be given serious attention in order that the errors of other industries in damaging natural resources may be avoided. The industry also has environmental problems in the disposal of toxic as well as radioactive wastes especially in wastes containing fluorides, nitrates and beryllium.

Research and development in appraising for the industry its environmental problems calls for a high order of technical competency in many fields. As the industry advances toward more populated areas environmental problems will need to be resolved in full co-operation with federal, state and local regulatory agencies having jurisdiction over public health, safety and natural resources.

In order that staff of these agencies may be in a position to give full assistance to this new industry, training programs must be provided for a better understanding of the technology, the terms and standards of the industry.

## APPENDIX A. CRITERIA

### Appraisal of Environmental Hazards in Treatment, Storage and Disposal of Liquid, Solid and Gaseous Radioactive Toxic Wastes. A. Under Normal Operations. B. Under Abnormal Conditions

1. Wastes
   a. Characteristics—
      1. Physical
      2. Chemical
      3. Radioactivity
   b. Quantities—
      1. Total volume
      2. Production rate
      3. Radioactivity
   c. Treatment facilities—
      1. Quantities handled
      2. Decontamination obtained
      3. Volume reduction
   d. Storage facilities—
      1. Location
      2. Size
      3. Materials of construction
      4. Size
      5. Significant features (cooling requirements, leak prevention and detection, pumps, piping, etc.)
   e. Disposal facilities—
      1. Cribs
      2. Pits
      3. Wells
      4. Outfalls
      5. Sewer connections
      6. Stacks
      7. Others
2. Environmental Conditions
   a. Geology—
      1. General area information
      2. Depth of overburden
      3. Stratigraphy
      4. Soils characteristics pertinent to plant wastes (exchange capacity, size, etc.)
   b. Hydrology—Ground Water and Surface Water—
      1. General area information
      2. Elevation of water table
      3. Direction, rate and nature of underground movement of wastes and water
      4. Effect of other withdrawals or recharge
      5. Watershed characteristics
      6. Streamflow—max., av., min., flow duration, turbulence
      7. Impoundments
      8. Suspended and bed load characteristics—quantity, movements, deposition, exchange capacity, etc.
      9. Stream biology
      10. Seasonal characteristics and effects
   c. Meteorology—
      1. Regional conditions
      2. Surface winds—frequency of direction, velocity by day, night and with precipitation
      3. Winds aloft—above information at various levels
      4. Dry bulb temperatures—max., av., min., monthly and annual averages, frequency and duration of extremes
      5. Precipitation—rain and snow, monthly distribution, average precipitation hour, days in excess of .01″
      6. Inversions—frequency, duration, other characteristics
      7. Diffusion: under normal day and night conditions, with inversions and low wind velocities

8. Atmosphere dust loads—characteristics of dusts of various levels, frequency and duration of dust periods
9. Seasonal characteristics and effects

3. Community Conditions
   a. Downstream users of ground and surface waters—
      1. Distance
      2. Quantity
      3. Purpose
   b. Proximity of industrial and residential installations—
      1. Distance
      2. Kind
      3. Size
   c. Agricultural activities—
      1. Grains
      2. Livestock
      3. Fish
   d. Recreation areas—
      1. Streams
      2. Impoundments
      3. Other
4. Evaluation of Dilution and/or Concentration Factors
   a. Retention in soils and geologic formations
   b. Dilution (or concentration) in streams and surface waters
   c. Atmospheric dilution
   d. Fall-out studies
   e. Increase of dilution by mechanical means
      1. Outlet manifolds
      2. Increased stack height and/or stack heaters
   f. Maximum allowable concentration of contaminant
   g. Emergency aspects
5. Public Relations
   a. Pertinent laws regulating pollution of underground resources, surface waterways, the atmosphere and public facilities
   b. Legal limits as to permissible contamination (feasibility of compliance)
   c. Agencies having jurisdiction
   d. Integration of plant emergency program with disaster planning of local, state and federal agencies

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# Radiation Exposure from Environmental Hazards

**By H. M. Parker,* USA**

Both nuclear reactors and radiochemical processing plants are capable of generating significant radiation hazards beyond the confines of a plant of normal size.

The expected hazards can be classified as follows:

### 1. HETEROGENEOUS REACTORS—AIR COOLED

The governing hazards arise from dispersion of the reactor coolant gas into the atmosphere. The predominant contaminant is $A^{41}$. Due to the short half-life (1.82 hours) and chemically inert behaviour of the gas, there are no appreciable complications in evaluating the hazard in terms of direct radiation from the effluent plume. Other contaminants, which may introduce more insidious hazards, are radioactive particles generated in one of three ways:

1. Direct activation of dust particles drawn through the reactor.
2. Blowing out of reactor structural materials or corrosion debris; because of the long residence of such debris in the reactor, the content of long-lived activation products may be high.
3. Escape of material from ruptured fuel elements; this introduces particles of mixed fission products and plutonium.

### 2. HETEROGENEOUS REACTORS—SINGLE PASS WATER COOLED

The limiting hazard develops from the release of coolant water to a river or lake. The principal contaminants are activation products, which may occur from direct throughput, or be augmented in the more dangerous long-lived components by temporary hold-up on corrosion films in the unit.

Fission products may also arise from four sources:

1. Irradiation of natural uranium in the water.
2. Uranium dust impregnated in fuel element surfaces.
3. Transmission of fission products through a fuel element skin.
4. Rupture of fuel elements.

In typical practice, some 80 radionuclides can be identified in reactor effluent, of which about 16 have separate significance in hazard calculation.

The limiting hazards for the Hanford reactors are $P^{32}$ concentrated through biological chains, and gut-irradiation, and bone deposition from drinking water. In all cases, an extensive radio-ecological program is necessary to validate effluent release practices.

In addition, such reactors may cause atmospheric pollution by leakage of whatever atmosphere is maintained in the reactor, and by irradiation of gas in test hole facilities. Although the escaping gases may be found to contain some unexpected components, this hazard is subordinate.

### 3. HETEROGENEOUS REACTORS—LIQUID RECIRCULATING SYSTEMS

In this case, the environmental hazard is normally low, being restricted to low volumes bled off for freshening the coolant, or arising from an occasional system leak.

### 4. HOMOGENEOUS REACTORS

From the environmental hazard viewpoint, such reactors can be treated as radiochemical processing plants.

Gross atmospheric contamination may conceivably arise from a catastrophic incident in any reactor; this phase is not considered here.

### 5. CHEMICAL PROCESSING PLANTS

The operation may be schematically broken into four steps, not all of which will be used in a specific plant.

#### Step 1. Dissolution of Reactor Fuel Elements

This step leads to periodic release of the rare gas components of the fission mixture, with $Xe^{133}$ as the governing member, and of some volatile fission products, predominantly the radioiodines.

The controlling hazard is a function of the fuel element cooling time. Under normal operating conditions, $I^{131}$ is the significant contaminant.

#### Step 2. Removal of a Specific Isotope

Either for their commercial value or because they create an in-process nuisance, one or more specific isotopes may be removed separately. Particularly if such isotopes have volatile compound forms it may be difficult to retain all the material in the system, and the release of highly active spray, evaporating to particles, is probable.

#### Step 3. General Chemical Separation

Regardless of the details of the chemical separation processes utilized, it has so far proved impossible to maintain all the process materials in the system.

* Hanford Atomic Products Operation, General Electric Company, Richland, Washington. Including work by the staff of the Biophysics Section, Radiological Sciences Department, General Electric Company.

Characteristically, fine mists or sprays of mixed fission products are released. Upon evaporation of the liquid content they form small radioactive particles of high intrinsic activity. Some of these escape through filters to the ventilation stack.

#### Step 4. Purification Stages

After removal of the bulk of the fission products, the product material, for example, plutonium, has to be further purified. The vented air will contain particles rich in this product material.

In Steps 2, 3 and 4 there are potential environmental hazards from the disposal of liquid radioactive wastes. This phase will not be discussed here.

It may be noted that, schematically, Step 2 of this general system is similar to the operations in an isotope factory designed for commercial recovery of a specific isotope.

## PROBABLE TRENDS IN PEACEFUL APPLICATIONS

It is plausible to assume that there will be a substantial field for heterogeneous power reactors together with large central processing plants, receiving fuel elements from many sources.

Such systems place the burden of environmental hazards on the processing plants.

Past experience in this field is therefore of major interest in peaceful applications.

## ATMOSPHERIC POLLUTION AROUND SEPARATIONS PLANTS

Of the available battery of contaminants, those of major concern have proved to be $I^{131}$ in Step 1 and particles emitted in Steps 2 and 3.

The iodine problem can be wholly solved by increased cooling time. This is economically unattractive. In practice, $I^{131}$ is removed by absorption processes. Residual hazard arises from the small fraction normally escaping, and occasionally from malfunctioning of the absorption equipment.

The primary particles escaping from the process have mass median diameter on the order of 0.3 to 0.5 micron. The maximum size is about 3 microns. The emission of $10^8$ to $10^9$ particles per day with activity on the order of $10^{-3}$ $\mu$c per particle is to be expected.

Secondary particles develop in venting systems beyond the filters by attachment to inert substances, which later flake off and escape. Such particles have diameters up to several hundred microns (or conceivably up to several centimeters) with activities up to hundreds of microcuries. These present distinctive hazards.

### Potential Exposure Mechanisms

The principal exposure forms are:

#### 1. Direct Irradiation of Persons or Animals from Highly Radioactive Ground Sources

This situation may develop from depositions of $I^{131}$, primary, or secondary particles. It is associated with isolated single releases of unusual amount. The intensity is greatest for emission of secondary particles, where the phenomenon is necessarily restricted to within a few kilometers of the facility.

#### 2. Adherence of Isolated Particles to the Skin or Clothing

The significant hazard comes from the secondary particles, which in the intermediate sizes may travel up to about 15 km. Such particles on the ground can become airborne again, as in dust storms, so that isolated particles theoretically capable of producing a skin reaction may be found as far as 30 to 50 km from the source. The probability of undisturbed skin contact for the required time (of the order of days) is so vanishingly small, that this is not a pertinent hazard in practice.

Within the 15 km radius, control by radiation monitoring is adequate; there has been no experience which shows injury from contact during a normal work day.

#### 3. Ingestion of Large Isolated Particles

Close to the process stacks, ingestion of a large particle and its retention in a convolution of the gastro-intestinal tract for days is feasible. However, normal personal hygiene makes this hazard improbable; it has not occurred in the local experience.

#### 4. Inhalation of Small Particles

The permissible inhalation of small particles of soluble materials can be deduced from the limits given in such references as NBS Handbook 52. In these terms, the practical inhalation hazard is negligible. With the quoted emission, the average concentration at 15 km would be about $10^{-12}$ $\mu c/cm^3$.

For insoluble particles, there is the residual doubt as to whether a single focus, localized in the lung, may ultimately lead to malignancy. The balance of the evidence makes it improbable that this hazard has significance, at the feasible activity levels.

#### 5. Consumption of Contaminated Vegetation

Although this is a hazard potentially originating from either $I^{131}$ or particles, it is, in fact, limited to $I^{131}$. In ten years of operation the build-up of small particles on vegetation has not produced levels of significance. Areas in which large particles may fall must necessarily be withdrawn from public or grazing use for other reasons.

The hazard from deposited $I^{131}$, on the other hand, has required intensive control and research study. From such studies on sheep, it is determined that the permissible vegetation contamination is about $10^{-5}$ $\mu$c $I^{131}$ per gram. Under equilibrium conditions, this would represent an atmospheric contamination of about $3 \times 10^{-13}$ $\mu c/cm^3$. This is to be compared with the human inhalation limit of $3 \times 10^{-10} \mu c/cm^3$ appropriate for large populations. These values clearly define $I^{131}$ as a ground contaminant rather than a direct air pollution hazard.

As applied to man, the secondary intake is a function of his eating pattern. Two significant routes of

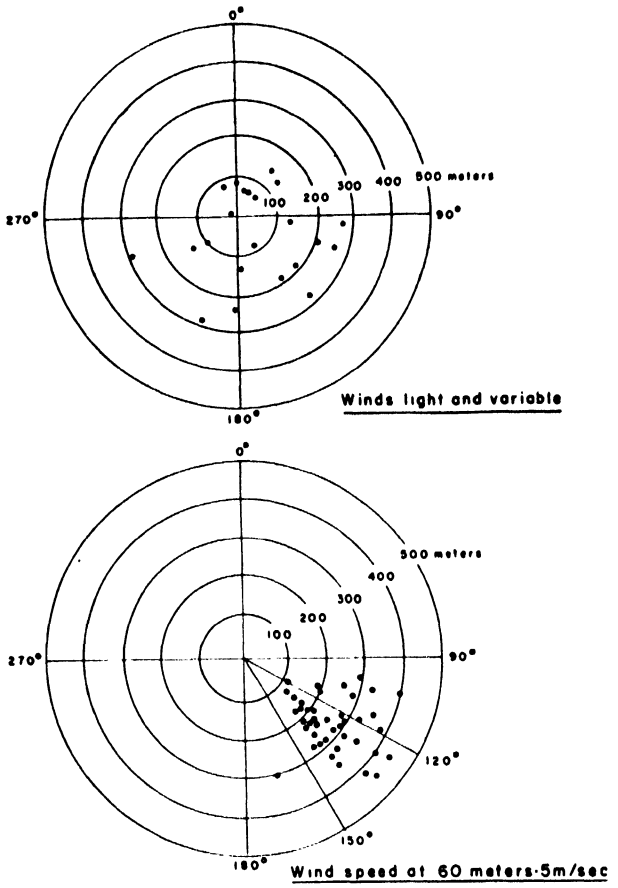



**Figure 1. Distribution of puffs hitting the ground during unstable conditions**

entry are consumption of fresh garden produce, and drinking of milk from cows on contaminated pasture. The limit of $10^{-5}$ μc per gram can be broadly applied to all cases.

### 6. Other Mechanisms

Once a radioactive contaminant has been released to the atmosphere it is necessary to trace its course through a wide variety of natural processes. Typical ones are direct fall into or leaching into a public water supply, uptake by soil and later by plants.

With the exception of the $I^{131}$ case in plants, none of these processes has been found to be critical.

#### Sample Distribution Patterns

The actual distribution pattern of pollutants around a processing plant depends on the time-distribution of effluents, the elevation of the release point, meteorological conditions, and topography of the environs. Although these will require individual study at each site, considerable guidance can be obtained from the local patterns. The Hanford site is a good basic model because the terrain is relatively flat and barren. In site selection, if study does not assure suitability, this can be tested by controlled emissions of smoke, or better, of fluorescent particles from a high tower. An adequate picture cannot be obtained in less than one year, because there are marked seasonal variations.

Of the controlling factors listed, the stack elevation is significant only close to the plant where the area will in any case be controlled. Locally, the maximum ground concentration varies approximately as the inverse square of the stack height. For conventional stacks of 60 to 100 meters, the effect of height is inconsequential beyond 5 km.

Time distribution of effluents is significant chiefly in a discontinuous process that can be interrupted at times of unfavorable atmospheric dilution. The meteorological situations can be crudely segregated into three kinds, which are functions of the vertical lapse-rate and of the wind velocity. These are:

1. *Looping,* characterized by marked vertical instability, and usually fluctuating wind direction. The stack emission loops to ground at random points as shown in Fig. 1.† Instantaneous concentration is high, but the integrated exposure comparable with that under other conditions. As a guide, it appears that a steady emission of 1 curie per day from a 60 m stack is unlikely to give (a) short-period concentration above $10^{-8}$ μc/cm³, (b) short-period dose above $10^{-7}$ μc-sec/cm³, (c) hourly average concentration above $10^{-10}$ μc/cm³. The maximum dose may occur in a zone 2 to 5 stack heights from the source.

2. *Coning,* the elementary picture in which the effluent forms a cone of semi-angle 5° to 7°. The point of maximum ground concentration is 8 to 12 stack heights down-wind. It implies nearly adiabatic lapse rate and strong steady wind. More realistically, the ground concentration can be estimated from the well known Sutton equations.[2]

3. *Fanning,* which occurs during temperature inversion, and usually with low wind speed. This process leads to negligible hazard close to the stack, with maximum ground concentration as much as 40 km away. It is this process that leads to generalized contamination under Hanford conditions. In local experience the standard diffusion equations, when extended to distances in excess of 20 km, yield concentrations that are not reliable to better than a factor of 5. In general, the equations underestimate the concentrations under strong inversion conditions.

Experience over ten years at the Hanford Works suggests that the environmental hazards setting appropriate protective radii are the deposition of $I^{131}$ on vegetation, and the deposition of secondary particles on the ground. In submitting actual patterns, an existing picture will represent a partial history of past emissions. For $I^{131}$ ($t_{\frac{1}{2}} = 8$ days), the historical period is of the order of weeks; for secondary particles, with varied half-lives from months to years, the period is long but indefinite. Existing particles may be washed into the ground, buried in dust storms,

† Figure 1 shows, in the upper figure, the number of ground contacts within 300 meters in 45 minutes in a light variable wind (~2.2 m/s). Contacts occurred in all four quadrants. The lower figure gives the contacts within 500 meters in 51 minutes with a wind speed of 5 m/s. All the puffs fell within one quadrant; 95% of them were within a 40 degree sector, 70% within a 20 degree sector. These data, and those from which Figure 6 was developed appear in Ref. 1.

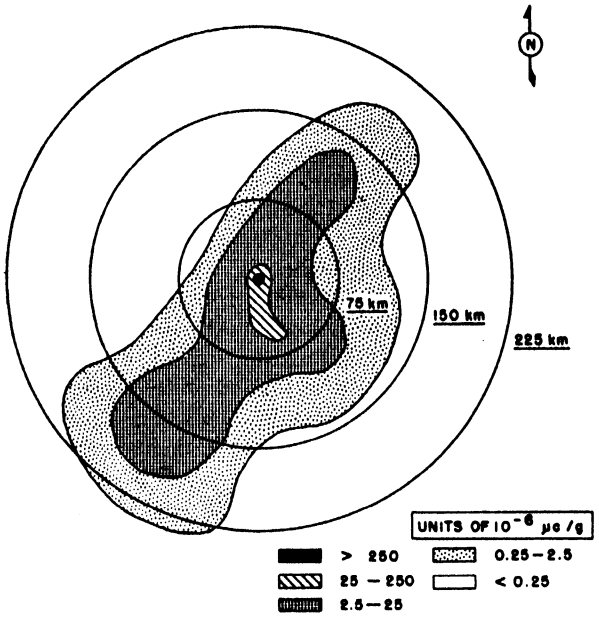



Figure 2. Deposition of $I^{131}$ from single emission of 100 curies

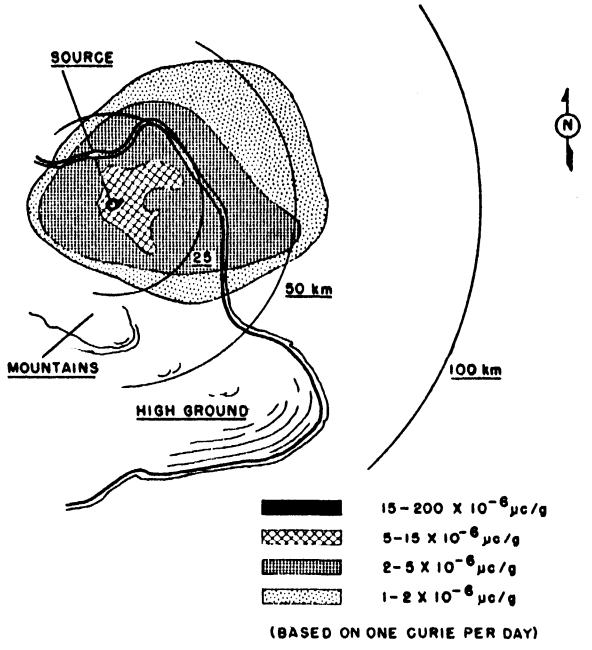



Figure 3. $I^{131}$ deposition—normal spring pattern

or transported by wind. This prevents subtraction of successive pictures to obtain short-term increment.

In the following diagrams, data for single major releases were adjusted to a 100 curie emission. Routine emission of $I^{131}$ was adjusted to 1 curie per day. For other particle maps, the status given is approximately the worst found in 10 years.

Figure 2 relates to a single emission of 100 curies of $I^{131}$ in a few hours, during which time inversion conditions existed near the ground with good dilution above. Effluent traveled north-east in the first half, and south-west in the second half. Wind speed was 2–3 meters per sec with low values near the time of reversal.

With a deeper inversion layer, ground concentrations 15 km and more from the stack could easily have reached 10 times the quoted values.

Figure 3 shows a typical spring condition of vegetation contamination for daily emissions of 1 curie $I^{131}$.

Figure 4 gives the equivalent data for the same season four years later. Figures 3 and 4 serve to measure the variability under broadly similar meteorological conditions.

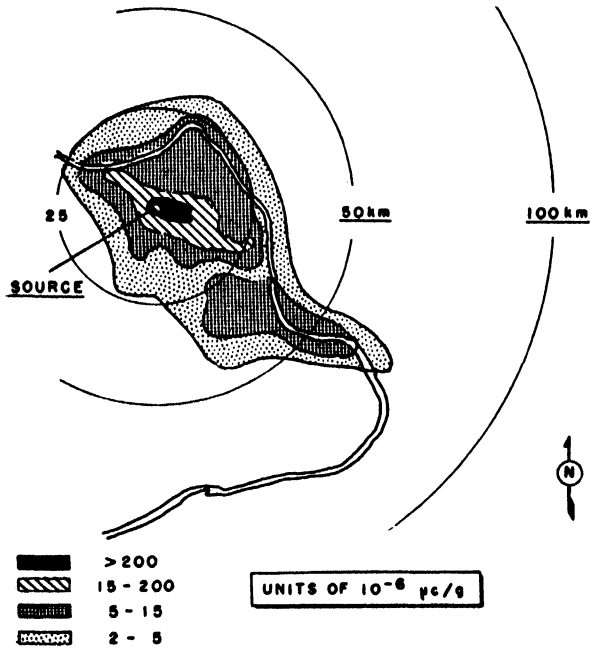



Figure 4. $I^{131}$ deposition—spring pattern another year

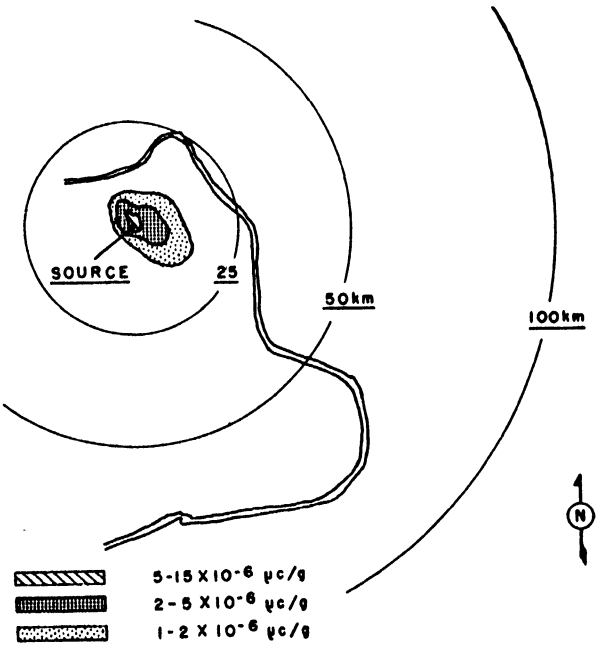



Figure 5. $I^{131}$ deposition—normal summer pattern

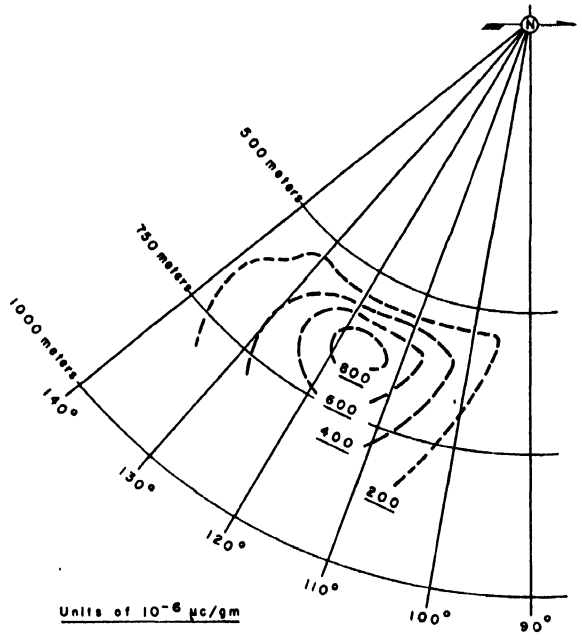



Figure 6. Local isopleths $I^{131}$ deposition in summer

Figure 5 is a typical summer condition. High local concentration due to looping does not show on this scale. Otherwise, the remote environmental hazard is much reduced.

For comparison, isopleths of the local summer condition appear in Figure 6. These data are computed from smoke tests on a 56 m stack, assuming wind speed of 10 m/sec, constant direction, constant lapse rate of −0.01°C/m, and $I^{131}$ deposition constant 2.8 cm/sec.‡

Figure 7 gives a condition resulting from the persistent inversions of the winter season. The large area of significant deposition southeast of the stack and 80 km away is typical of quiet fluid motion down a river valley and contact with vegetation on higher ground in and around a river gorge. With $I^{131}$, temporary depositions above the permanent limit of $10^{-5}$ μc/gm are tolerable, and the location of affected spots tends to change monthly. However, at 10 times the reference emission, such areas would be large and could overlap at distances up to 100 km. Since a plant boundary of this extent would be absurd, there has to be an upper limit for $I^{131}$ emission not greatly different from the reference value.

Figure 8 gives the pattern for a single emission of primary particles, scaled to 100 curies. The unusual narrow band is real up to some 50 km. Beyond this, the conventional width to 0.1 central concentration could not be determined in the actual case, because of lack of instrument sensitivity. The actually detected width is reported. Detailed analysis of this case would provide the best practical test of Sutton-type cloud width calculations up to 50 km travel. Figure 9 shows the worst condition in ten years

‡ A deposition constant of 2.8 cm/sec corresponds with the value $10^8$ μc per meter²/hr/μc/cm³ in the atmosphere used at Hanford since 1948. A. C. Chamberlain and R. C. Chadwick report a deposition constant of 2.5 cm/sec.[8]

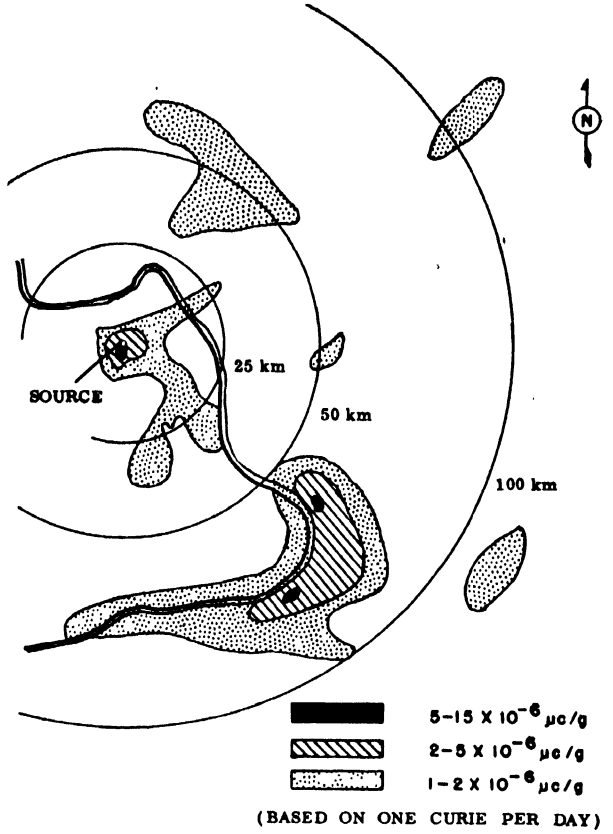



Figure 7. $I^{131}$ deposition—remote contamination by valley drainage —winter condition

for secondary particles in the intermediate size range of about 3–100 μ.

Figure 10 is the similar condition for large secondary particles (>100 μ), which could produce damage on skin contact. Neither Fig. 9 nor 10 can be reliably scaled to a reference emission rate, because

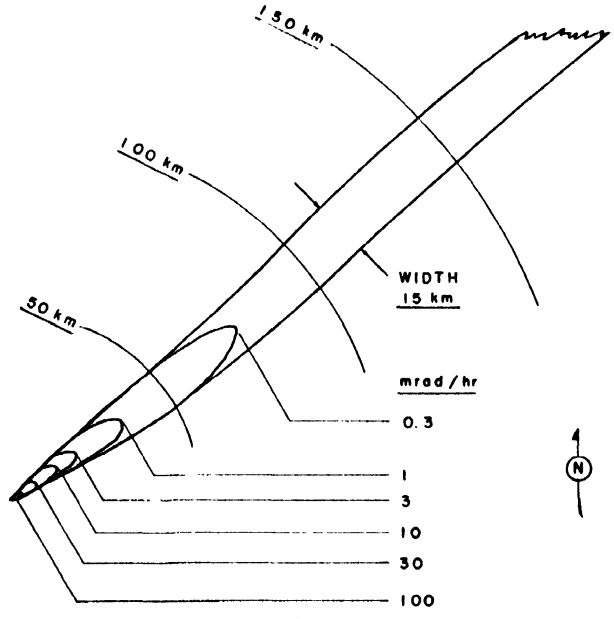



Figure 8. Ground contamination from narrow emission band of radioactive particles (scaled to 100 curies)

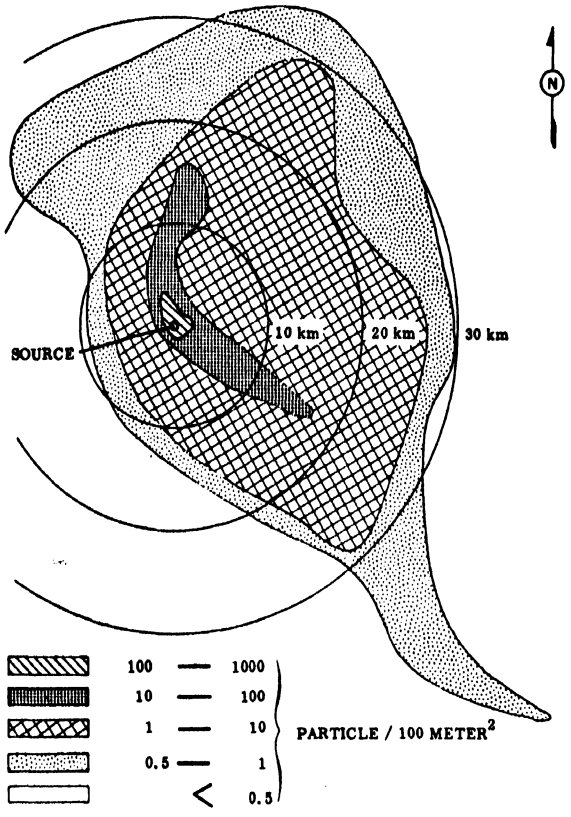



Figure 9. Typical distribution of particles in size range 3–100μ

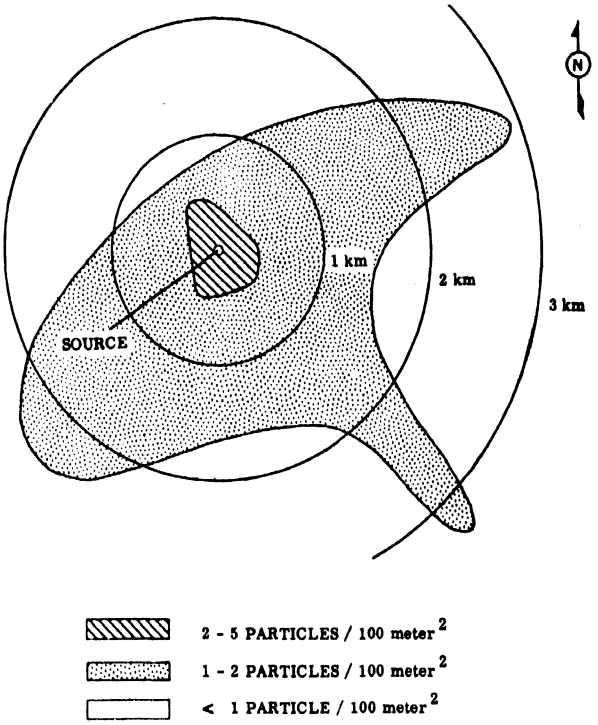



Figure 10. Typical distribution of large particles (Dia. > 100μ)

the pictures are an integration of effects over too long a time, and suitable isokinetic sampling near the stack mouth was not always available.

In reviewing the Hanford experience, one may conclude that a comfortable boundary around a high level processing plant would have a radius on the order of 12–15 km. In areas with a distinctive prevailing wind one could make better utilization of the same area by reserving a shape modeled after the wind rose. At Hanford, for example, the required control area to the west is substantially less than it is to the east.

Reduction of the controlled area by a factor of 2 in linear dimensions would not introduce significant hazard to man; uptake by animals could be borderline and some restrictions would have to be applied from time to time.

With further reduction, the limitations on grazing animals, the potential risk of secondary particle contamination, the loss of morale in an area requiring intensive monitoring, and the potential for damage claims would make the operation unattractive.

As far as real hazard to man, with food growing and animal grazing excluded, a smaller reserved area would introduce no major hazard, provided that intensive monitoring could be practised.

In our opinion, these plants should be operated in remote areas, where land is cheap. With an ample reserved area, the public could be assured of radiation safety, and the healthy growth of peaceful applications of atomic energy promoted.

## BRIEF NOTE ON LIQUID DISPOSAL HAZARDS

A comprehensive survey of this phase is reported elsewhere. Briefly, it is impracticable to create surface lakes or swamps of radioactive liquids. These sources contaminate waterfowl and are prone to create particle hazards, as the water level changes.

Highly active wastes can be retained in underground tanks for long periods; the estimated integrity of tanks is 50 years or more. The intermediate activity, large volume process wastes are most troublesome. These can be safely injected below ground at some sites. With a reservation of the size conditioned by atmospheric pollution, feasible travel time of underground sources to a public area will frequently be 50 years or more. This simplifies the environmental hazard problems. Since this disposal, where permissible, is most economical, it provides an additional incentive to reserving a substantial area around the plant.

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2. Sutton, O. G., *Theory of Eddy Diffusion in the Atmosphere*, Proc. Roy. Soc., *A135*: 143 (1932), and many later contributions.
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# The Criticality Safety of Chemical Plant Producing Fissile Materials

**By H. A. C. McKay and C. M. Nicholls, UK**

## THE NATURE OF A CRITICALITY ACCIDENT

Policy in regard to criticality safety depends on our estimate of the consequences of an accident. At first sight it appears that they might be disastrous. When, however, we reflect on the difficulty of bringing fissile material together fast enough when it is actually desired to cause an explosion, we realise that this may not be so. Even the most rapid redistribution of material occurring in a chemical plant is likely to take place on a time-scale measured in seconds, and this means that any supercritical system which builds up will disperse again through such processes as boiling. For an explosion, the build-up would have to take place in a period of less than a millisecond, and it is difficult to imagine any way in which this could happen.

Design and operating policy in the UK has nevertheless always envisaged the possibility of a catastrophic criticality accident, and has therefore aimed at being able to give a near-absolute guarantee of safety. An accident, even if not catastrophic, might still give rise to a serious radiation and toxic hazard, and cause considerable damage to active plant. Up to the present it has not been felt that a change is possible to a policy of calculated risks.

## CRITICALITY EXPERIMENTS AND CALCULATIONS

A purely empirical approach to the problem is to simulate different parts of a chemical plant, and study the approach to criticality as fissile material is added, or some other parameter is varied. This involves large numbers of difficult and possibly dangerous experiments, but it does yield true criticality limits, free of safety factors. In some cases, e.g., where the geometry is complicated, it is the only possible method. In other cases it may be necessary, as a matter of economics, to weigh the advantage of knowing the true limits against the effort involved in determining them. As an atomic energy industry grows, the increasing demand for maximum throughput in plants of minimum capital cost may call for extensive criticality experiments to provide adequate design data.

The alternative approach is to make a more fundamental investigation relying on calculation from a few key experiments, and making assumptions which tend to underestimate the criticality limits; this has been our approach in the UK wherever possible.

The systems encountered in chemical plant may be classified as follows: (1) slow neutron systems, e.g., solutions of fissile materials in water or organic solvents; (2) fast neutron systems, e.g., pure fissile material as metal or oxide; and (3) intermediate systems, e.g., aqueous slurries of fissile material, or solid salts containing water of hydration.

The most important systems at present are the first-named, both because wet processing plays a big part and because the criticality limits tend to be most restrictive in such systems. Fortunately these are also the systems which are most amenable to fundamental treatment.

## SOLUTIONS OF FISSILE MATERIALS

Our data on solutions are derived from what are popularly called "water-boiler" experiments, in which the neutron flux is measured as the amount of fissile material in solution in a suitable vessel is gradually increased towards criticality. The criticality limits for the vessel in question follow, of course, directly from the experiments. Furthermore we can calculate certain nuclear parameters from the data and then use these parameters to calculate safe limits in other types of vessel; in such calculations we must err on the side of safety at every point of doubt.

Some of the results[1] for plutonium solutions at room temperature are plotted in Figs. 1–3. They apply under certain idealised conditions: it is assumed that the plutonium is pure $Pu^{239}$, that the hydrogen density is equal to that of pure water, and that the solution is homogeneous.* It is also assumed, of course, that no neutron absorbers are present (other than $H^1$ and $Pu^{239}$), and that no other fissile material interacts with the system. Furthermore in the reflected cases the reflector postulated is an infinite thickness of water; heavy water, most hydrocarbons, beryllia and graphite would all be more dangerous, but any other ordinary material, e.g., concrete or steel, would be safer. Comparisons between $Pu^{239}$ and the two other principal fissile materials, $U^{233}$ and $U^{235}$, for spherical systems, are given in Fig. 4.

All the curves plotted pass through a minimum in mass (or mass per unit length or per unit area), all are asymptotic to a minimum radius or thickness.

* It is theoretically possible, within certain limits, to change a safe homogeneous system to a supercritical one by concentrating the fissile material towards the centre.

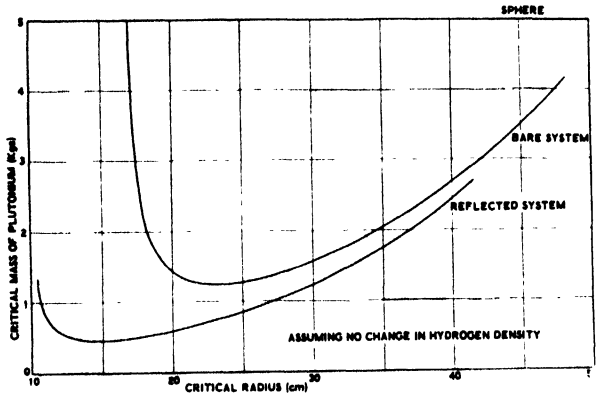



Figure 1. Critical mass of plutonium (kg) vs critical radius (cm)

We can therefore set safe upper limits either on the amount of plutonium in the system, or on the vessel dimensions. Furthermore a sphere usually represents the most dangerous shape; so any vessel of volume less than that of a safe sphere will also be safe. Similar considerations apply to tubes, where there is a safe upper limit to the cross-section, given by the data for infinite cylinders. Finally there is a safe upper limit to the concentration of fissile material in an indefinitely large volume of solution: this may be deduced from the high radius end of the curves for spherical systems. The various safe limits for plutonium are listed in the table. An important point to note is that a system is safe provided it falls within any one of the limits; for example < 464 gm of plutonium is safe in *any* vessel and at *any* concentration. There is also, of course, an infinite number of combinations of mass, vessel dimensions and concentration which are safe; they can be read off from Figs. 1 to 3 as required.

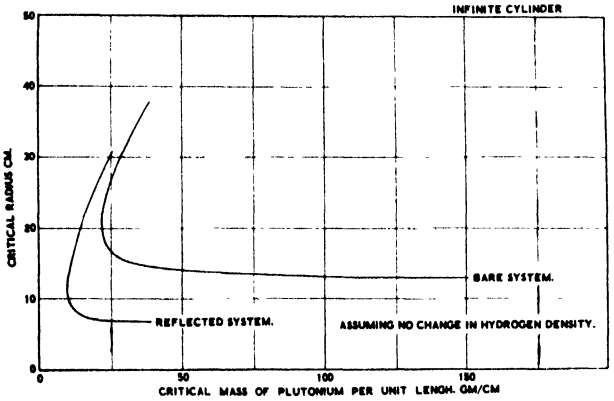



Figure 2. Critical radius (cm) vs critical mass of plutonium per unit length (gm/cm)

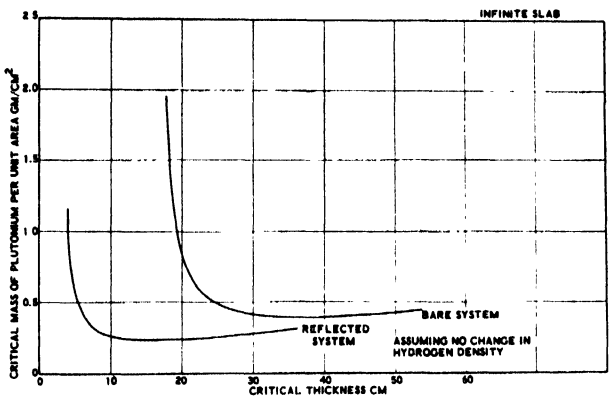



Figure 3. Critical mass of plutonium per unit area (gm/cm²) vs critical thickness (cm)

Table I contains most of the basic data required for the design and operation of wet processing plants for plutonium. The first problem in applying the data is to make sure that the limits laid down are not exceeded whatever abnormalities or accidents

**Table I. Safe Upper Limits for Plutonium Solutions**

| | | *Calculated limits* | | *Limits with ⅓rd safety factor* | |
|---|---|---|---|---|---|
| *System* | *Quantity restricted* | *Water-reflected* | *Bare* | *Water-reflected* | *Bare* |
| Any vessel, any dimensions | Concentration | 6.6 gm/l | | 4.4 gm/l | |
| Any vessel, any dimensions | Mass | 464 gm | 1244 gm | 309 gm | 828 gm |
| Any tube, any cross-section | Mass/unit length | 10.0 gm/cm | 22.4 gm/cm | 6.6 gm/cm | 149 gm/cm |
| Any slab-shaped vessel, any thickness | Mass/unit area | 0.39 gm/cm² | 0.24 gm/cm² | 0.26 gm/cm² | 0.16 gm/cm² |
| Any vessel, any mass (of plutonium) | Volume | 4.5 l | 20.3 l | 3.0 l | 13.4 l |
| Any tube, any mass | Cross-section | 98 cm² | 201 cm² | 80 cm² | 164 cm² |
| Spherical vessel, any mass | Diameter | 20.5 cm | 33.8 cm | 17.9 cm | 29.5 cm |
| Cylindrical vessel, any mass | Diameter | 12.5 cm | 25.6 cm | 10.2 cm | 20.9 cm |
| Slab-shaped vessel, any mass | Thickness | 4.0 cm | 17.6 cm | 2.7 cm | 11.7 cm |

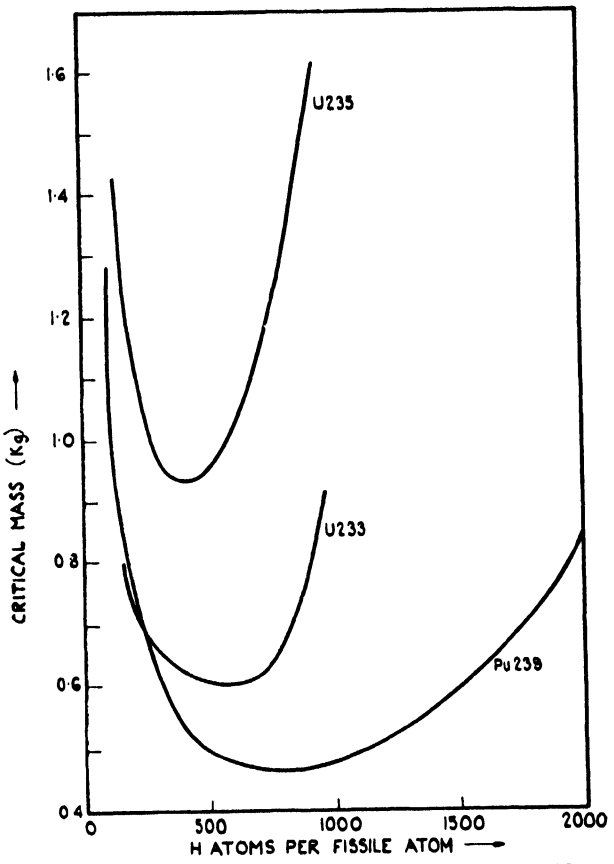



Figure 4. Spherical systems—comparison of results for $U^{233}$, $U^{235}$ and $Pu^{239}$ in water

occur. If, for instance, batch doubling is a possibility, then batch sizes must not exceed one-half of those listed. All such contingencies as blockages, leaks and the gradual accumulation of fissile material as sludge, must be considered. It is comparatively easy, with the aid of the table, to lay down safe conditions for normal operation; much harder to ensure complete safety under all circumstances.

A particular aspect of this problem is the question of reflectors. As the table shows, bare systems are less subject to restriction than reflected, but to take full advantage of this we must be able to guarantee the absence of reflectors. An absolute guarantee is hard to give: we can, for example, visualise the flooding of the whole plant, thereby surrounding it with water reflector, and we have to consider too the approach of human bodies and other hydrogeneous material to the vessels in question. A convenient compromise is to introduce a category of substantially unreflected systems, to which we apply intermediate limits.

However careful the designers may be, it is obviously desirable to introduce some further factor of safety. The UK practice is to reduce the safe upper limits of mass, volume and concentration of two-thirds, which gives the figures in the last columns of the table. (N.B. The radii of spheres and cylinders are then reduced to $\sqrt[3]{2/3}$ and $\sqrt{2/3}$ respectively, of their absolute safe limits). It is also the practice to apply the results in column two (i.e., those for reflected systems without the two-thirds safety factor) to substantially unreflected systems.

Where possible, preference may be given to vessel dimensions as the basis of safety, so that safety is a property of the plant rather than of how it is operated. It is chiefly in the case of long cylindrical vessels that this is helpful; for tanks etc., the 3 l or 4.5 l upper limit is usually far too cramping. In other cases the commonest choice is to place a limit on the total quantity of fissile material permitted in a vessel. The third possibility, the use of an upper concentration limit, is valuable mainly as an assurance of safety in the early stages of the plant, while the fissile material is still very dilute; but it is dangerous to rely too much on a concentration limit owing to such possibilities as the formation of a precipitate in which the limit is exceeded.

## SOLID FISSILE MATERIALS

It is impossible to make such useful generalisations about dry systems containing fissile materials, as about solutions; so much depends on the details of the geometry, etc., that almost every separate problem necessitates fresh calculations. It is helpful to have safe sizes for systems of the metal, and of such compounds as the oxide, both bare and surrounded by reflectors of various kinds. The figures with water as reflector may be used to set the limits for the safe transport and storage of, for example, single metallic billets. (We take water, owing to the possibility of an accident in which the billet is immersed in water). But as soon as we have to consider numbers of billets then we become involved in lattice calculations.

Such calculations can be made with reasonable confidence for fast neutron systems. When, however, we come to intermediate systems, then calculation is of little help in the present state of our knowledge, and direct experiments are desirable. Intermediate systems are indeed of not inconsiderable practical importance; metal turnings mixed with graphite, solid hydrated salts, aqueous slurries and even very concentrated solutions, may all come in this category, having insufficient moderator to thermalise the neutrons completely.

There is fortunately one generalisation which is practically beyond doubt, viz. that the mass limits are always substantially higher for dry than for wet systems. This is certainly true for unmoderated systems, and is strongly believed to be true for intermediate systems. In the absence of other information we might therefore take 464 gm of plutonium, the figure for reflected solutions, as a safe upper limit for all ordinary systems. Furthermore we may conclude that it will be the wet rather than the dry parts of a plant which will need the most careful consideration as regards criticality.

## DESIGN CONSIDERATIONS

Undoubtedly, criticality imposes serious limitations on industrial chemical plant. It is for example un-

fortunately true that the size of vessel in which a supercritical system can most easily be achieved, is very much the size that the chemical engineer would often choose. Attention should therefore be paid to all possible ways of overcoming criticality limitations. Choice of process, choice of vessel size and shape, the addition of neutron absorbers, the use of neutron monitors, and plant layout can all contribute. There may, for example, be instances in which a dry process is to be preferred to a wet one, because it imposes less severe restrictions. Again, continuous processes usually require smaller vessels for a given plant throughput than do batch processes, and may be preferable for this reason.

The introduction of strong neutron absorbers is an important means of obtaining relaxation of criticality limitations. It enables us especially to increase vessel sizes while retaining a safe geometry. This applies particularly to slow neutron systems, when for example cadmium in a stainless steel sandwich can be used. To be most effective, such absorbers are required well to the centre of the system, where their presence may be an embarrassment in design. If applied on the outside they are, however, still a considerable advantage, because the system then becomes very nearly bare. Neutron absorbers may also accompany the process materials themselves, e.g., the nitrogen in nitric acid, which although only a weak absorber may nevertheless be present in very large amounts; but the deliberate admixture of small amounts of strong absorbers with the fissile material is not usually desirable, because the final product must be very free from such substances.

The advantages of introducing neutron absorbers are much less striking in intermediate and fast neutron systems. It is possible, however, to thermalise the neutrons in such systems, e.g., with paraffin wax, and then make use of cadmium or other slow neutron absorbers.

Another valuable technique is the use of external neutron monitors. The most successful are enriched boron trifluoride counters. They can be utilised to detect a gradual build-up of fissile material, e.g., as a sludge, in particular parts of the plant. By reducing our margin of uncertainty they enable us to relax our restrictions. It should be noted that the monitors do not give a direct measure of the amount of fissile material present, but that in any position of constant geometry a rise in the neutron flux indicates an increase in the amount of fissile material.

The final step in design is to examine the plant as a whole. It is essential, of course, to ensure that there is no unsafe interaction between vessels. Wide spacing assists, and another possibility is to place cadmium sandwiched in paraffin wax between vessels; the wax thermalises the neutrons, and the cadmium absorbs them. It is essential, too, to examine the effects of both normal and abnormal operation on vessels in series, especially at points where the basis of criticality safety changes, e.g., where a mass limitation replaces one based on geometry.

## ADMINISTRATIVE CONSIDERATIONS

There is an obvious need for an independent and sufficiently powerful body, charged with the duty of ensuring the criticality safety of chemical plant. In the UK the blueprints for all new designs of chemical plant are submitted to such a body for crutiny, along with such criticality calculations as may be needed by way of support. (The calculations, it should be noted, are the responsibility in the first place of the design side, rather than the safety organisation.) Similar action is taken when the plant operators wish to make changes in the mode of operation. Generally speaking, such "clearances" as are given apply to a particular item of plant operated in a particular way, and a fresh clearance is necessary when a change is made. The safety organisation also reviews from time to time the actual operation of chemical plant, inspecting records and the plant itself as necessary.

The safety organisation is an advisory body, but it can require that an enquiry be held if any of the conditions it lays down, e.g., as to permissible amounts of fissile material, is transgressed for any reason.

Some delegation of responsibility is possible. Relatively small amounts of fissile material, up to one-third of the safe limit, may be handled on the responsibility of any sufficiently senior scientist. Furthermore where a factory has a safety officer sufficiently experienced in criticality matters, clearances up to the limits in the fifth column of the table are left in his hands in straightforward cases, though he is still required to report full details of every clearance he issues.

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# Safety Criteria in Atomic Energy

**By F. R. Farmer,* UK**

At present the industrial development of atomic energy is considered in a special category; not merely the development of one of many industries, such as engineering, mining, chemicals, etc. Furthermore, it is thought that the hazards in this new industry, both to the worker and to the community at large, are exceptional. This feeling was born of a general fear and uncertainty on the one part, and from an unfamiliarity of the scientist and the engineer with the exceedingly small tolerance levels on the other. Already this situation is changing; operating experience shows that radiation tolerance levels can be met and that extremely small quantities of active materials can be detected and therefore controlled. This experience must be shared by the operational and design staffs, and the knowledge disseminated to dispel the fears of the public whose confidence and support are indispensable for a rapid development of the new industry. This industry then will take its part in national and international prosperity and must be considered by the general public as one more major industry.

## HEALTH AND SAFETY CONTROL IN NORMAL INDUSTRY

The development of most of our important enterprises in coal, iron and chemicals took place in an age during which human life was often held in small esteem and when accidents and death were regarded as part of man's misfortune. The hazards of industry were an alternative to death from starvation or disease and it was only when the latter were reduced by a steady increase in the standards of life, by improved sanitation and by medical skill, that a serious attempt to reduce the hazards of industrial life was undertaken.

For this reason, a considerable experience of hazardous conditions has become available which covers the use of machinery, the handling of obnoxious materials, and the effects of dusts, fumes and gases. On this experience, industrial codes of practice have been built and legislation passed defining conditions of employment, and tolerance levels for various materials. The Factories Act (1937, 1948) requires provision for adequate cleanliness, heating and ventilation and for medical supervision in certain cases. In general it imposes an obligation on the employer to take all practical measures to ensure that the buildings, machinery, standard of staff selection and training are adequate for the purposes for which they are to be used.

Under the impulse of growing national consciousness that accidents are not inevitable, and with a control imposed through the Inspectorate of Factories, our industries have reached the stage wherein the design for safe operation is inherent in any new process. Safety, however, is only one aspect of the wider economic field, and few industries can afford to operate in any way other than the most economical available. The big strides toward accident reduction have been made in the course of industrial re-organisation in which each change has led to greater economy, and in many cases the change of process or plant has been forced by competition.

Various conventions are used in quoting the accident rate in industry but these are not an absolute measure as between one industry and another or as between one country and another, because they depend upon the definition of an accident and on its severity. However, the number of industrial accidental deaths has the merit of precise definition although even this figure is not specific to the occupation in that it includes all deaths, including those arising from any mishap or human error entirely dissociated from the industry but occurring whilst at work.

The records of Great Britain in 1950 show the following range in the accidental deaths arising per 100,000 employed:

**Table I. Accidental Deaths Rate (An employed population of 100,000 in 1950)**

| | *Deaths per annum* |
|---|---|
| Fishing industry | 113 |
| Mining and quarrying | 80 |
| Transport industry | 25 |
| Building industry | 16 |
| Chemical industry | 15 |
| Distributive trades | 2.8 |

The fall in the accident rate during the period of 1930/1950 was accompanied by an improvement in national health. In 1930 for every 1000 live male births, 220 died before the age of 45; in 1950 this number fell to 110. The natural death rate in 1950 was reasonably linear for the first 45 years of life at 150 per annum per 100,000 persons. This total includes 30 to 40 deaths from violence, i.e., from falls, road deaths, burns, drowning and suicide.

If industrial activities are viewed against this natural background, it is seen that the probability of acci-

* UK Atomic Energy Authority, Risley.

dental death at work is only a half to one-third that of violent death in leisure hours, and only one-tenth that of death from natural causes—influenza, pneumonia, tuberculosis, etc.

## HEALTH AND SAFETY CONTROL IN THE ATOMIC ENERGY INDUSTRY

The hazards of external radiation were reasonably well known ten years ago and maximum permissible levels were agreed internationally which have not changed appreciably during the initial growth of the new industry. The results of ingestion or the inhalation of radioisotopes are known only in kind, and the estimates for maximum permissible concentrations include uncertainties of a high order. Nevertheless the levels which were agreed before any large scale processes were in operation revealed a hazard incomparably greater than any previously experienced in industry.

For the first time large-scale processes had to be designed and operated in completely unexplored fields of chemistry and physics, confined within a boundary of maximum permissible tolerance levels beyond any previous experience.

In order to ensure safe conditions, as far as practicable, it was essential for the designer to insert additional safety factors to cover the uncertain transition of the hazard from a fissile nucleus to the human body. In applying external radiation recommendations there is a large measure of uncertainty not only in the effect of massive shielding, with its day to day contribution, but also in maintenance requirements on structures and components. The application of internal radiation levels to plant design requires a standard of design, construction, and ventilation that is as perfect as possible—the varying significance of any isotopic tolerance level in the range of $10^{-3}$ to $10^{-7}$ microcuries per ml hardly modifies the design approach. In most cases the radioisotope does not pass directly from the process to the mouth or lung, but passes through many intermediate stages. The activity may be carried by water or air, by surface contamination, or may be absorbed by plants, animals or fish, or may follow a devious route through clothing and hands to the mouth. It is only by experience that such indirect routes can be assessed and adequate control established on a practical basis, thereby obviating the need for the very large safety factors which are now inserted.

The recommendations of the International Commission on Radiological Protection are accepted by the United Kingdom Atomic Energy Authority and the results of the efforts toward safety presented in the following section show our success in this direction. The United Kingdom Atomic Energy Authority further accepts the principle that radiation is harmful and should be kept to a minimum; this principle should be consistently applied, however, having due regard to the radiation already accepted by the human race as part of its background and arising from its other activities.

The present tolerance level structure only defines a framework within which the industry develops. As with conventional industry, ultimate safety lies in good design and enlightened management, and when these conditions are met, the main hazard arises not through encroachment on tolerance levels but from accidents arising from unforeseen circumstances or from human error on the part of either the designer or operator.

We know that in the handling of radioisotopes, it is possible to absorb through a punctured glove from 1000 to 100,000 times as much radioactivity as is normally inhaled in one day's work. As far as individual safety is concerned, it is more important to ensure freedom from such incidents than impose a sense of false security by reduction of maximum air concentrations or exposure levels.

Similarly our responsibility to the population at large to keep the sum total of all radiation to a minimum is not ensured solely by undue emphasis on tolerance levels. These define only top limits, below which there could be considerable variation in total exposure depending on the mode of factory operation, discharge of effluents and choice of sites. It is shown in the following section that the activities of the United Kingdom Atomic Energy Authority make only a fractional addition to the radiation already received by the population of the United Kingdom.

## RESULTS OF SAFETY MEASURES IN UKAEA

The results are naturally divided into the three categories of external radiation and internal radiation received by atomic energy workers and the effects on the population not so employed.

### External Radiation

Control is established both by the use of films and other personnel monitoring devices, and by area surveys. The results are analysed from the film records which now amount to half a million films of weekly, fortnightly and monthly issue. The Authority has followed the recommendations of the International Commission on Radiological Protection and the majority of films fall well below the maximum permissible level. This is shown in Table II by an analysis for 1952, 1953 and 1954, for the works having the highest potential hazard from external radiation.

Furthermore, in the history of the British project, in which all employees have been under medical

**Table II**

57% received an average dose less than 1/20 of the maximum permissible weekly dose.

82% received an average dose less than 1/10 of the maximum permissible weekly dose.

96% received an average dose less than 1/4 of the maximum permissible weekly dose.

99.6% received an average dose less than 1/2 of the maximum permissible weekly dose.

100% received an average dose less than 1 of the maximum permissible weekly dose.

surveillance, there has not been a single case of permanent or even temporary injury from external radiation.

### Internal Radiation

Control in this field is known to be very much more difficult than for external radiation. Regular air samples are taken, surfaces tested for contamination, personnel are monitored for $\alpha$, $\beta$ and $\gamma$ contamination and subjected to periodic urine analysis. Whilst regular air samples are taken throughout the factories and laboratories, the sampling rate is increased in potentially dangerous areas or during periods of maintenance. It follows that the records of air contamination will not describe average conditions but will be biassed toward the high values. Even so, of the 33,000 air samples taken in one year at one works, only 2% gave evidence of a dust concentration requiring protective action. This does not represent 2% of the working time as more samples are taken when high figures have occurred or are expected. Of the 10,000 selective urine analyses carried out at one factory, significant excretion rates were revealed in only four cases. Of these three may be compared with the industrial minor accident, that is an accident in which a potential risk arose but which resulted finally in no ill effect whatsoever. In the fourth case, which occurred very early in the operation life of the works, the dose was not accurately known and no ill effects have yet appeared.

### Population Not Employed in the Atomic Energy Industry

The United Kingdom Atomic Energy Authority undertook detailed district surveys around the factory sites establishing radiation levels on land, sea and shore before and during the operation of any site. The emission of radioisotopes in air, liquids and solid wastes is subject to authorisation by Ministers appointed by the Government under the Atomic Energy Act (1954).

The results of all such effluents have been carefully studied and the present control levels established in light of our early experience to conform with recommendations made by the International Commission on Radiological Protection for large populations.

## GENETIC EFFECT

The genetic effect of radiation has been much publicised and it is commonly suspected that the future development of atomic energy for peaceful purposes could have a significant effect on the mutation rate. The United Kingdom Atomic Energy Authority welcomes the advice of expert geneticists and will collaborate wholeheartedly with the special investigating committee recently set up under Government direction. It was important that the public, as well as our scientists, should be aware of the relative importance of the atomic energy industry in this field as compared with naturally occurring radiations and with the contribution from other activities such as routine diagnostic X-ray examinations and flights at high altitude.

There are several ways in which the radiation of genetic importance could be defined; of these the simplest measure is the total radiation to the gonads received before reaching the age of 30. The figures in Table III are very approximate, but indicate the relative importance of these contributions to the whole population:

**Table III***

| | *Genetically significant radiation per annum to the population of England, Scotland and Wales* |
|---|---|
| Radiation from natural sources | 2,100,000 r |
| Radiation to employees of UKAEA | 2,000 r |
| Radiation received in high altitude flight | |
| (*a*) At present heights of 20,000 feet | 30 r |
| (*b*) Corresponding total at 40,000 feet would be | 300 r |
| Radiation resulting from routine diagnostic X-ray examination | about 50,000 r |

* The above figures are derived from whole body radiation doses i.e., each gram of the whole body receives *r* roentgens of radiation per year.

The figure for routine diagnostic X-ray examination is devised by considerable extrapolation of the work of Stanford and Vance.[1] No great accuracy is claimed for the figure but it is unquestionably the largest source of radiation, for which an estimate has been made, as additional to the radiation already experienced from natural sources.

From these results it can be predicted with confidence that the continued development of atomic energy to ensure our future prosperity and economic health will have a very much smaller genetic effect than routine diagnostic X-ray examinations which are currently practised to ensure our physical well-being.

## NONRADIOACTIVE HAZARDS IN ATOMIC ENERGY

This paper has considered the particular hazards arising from radiations and radioisotopes; it is important to note that accidents of a more conventional type have a far higher probability. Our experience to date has shown a remarkably low death and accident rate in this field in comparison with comparable industries, such as engineering and chemical manufacture. The accidental death rate in United Kingdom Atomic Energy Authority is about a fifth of that in these industries and the non-fatal accident rate is as low as the safest occupation listed in the Industrial Classified Industries. This is undoubtedly the result of the good design, staff selection and training, and supervision by medical, radiation and general safety departments. These are a necessary part of an atomic energy organisation in order to meet the stringent requirements of radioactivity control but our experience has shown that the design and operational precautions against the special hazards lead to enhanced immunity from the more conventional risks in the chemical industry.

## SAFETY COSTS

The safety record of the United Kingdom Atomic Energy Authority has been achieved at a high cost. Most of the charges are intangible and are hidden in plant design and construction and operational charges which constitute by far the largest burden on the industry. The charges which are demonstrably attributable to safety include clothing, monitoring, and laundry; the maintenance of the medical radiation and general safety service and the district radiation survey. The annual cost of this service at a factory concerned with reactors and their products amount to some £75 per head of the factory employees. This compares with the charge borne by normal industry of £3 to £6 per head.

In recent years the nation has increased its expenditure on National Health Services. At the current level our investment runs at £10 per head towards the reduction of a death rate, which in the young and middle-aged is some ten times higher than the industrial risk.

It could be deduced that the initial high safety costs borne by atomic energy are the result of our ignorance in this new and hitherto unexplored field coupled with the public fear which itself arises from lack of information.

Our objective as engineers, physicists and doctors should be to accrue knowledge and experience in the handling and effects of radioactivity and to ensure widespread distribution of our knowledge so that we may proceed with confidence and cooperation of the population at large.

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# The Dangers Presented by Uranium Irradiated in Nuclear Reactors

**By H. Jammet* and H. Joffre,† France**

The danger due to uranium slugs radioactivated by their use as fuel in nuclear reactors is threefold: (1) long distance irradiation, while handling the slugs; (2) contamination of the air when they are taken out of the cladding; and (3) contamination of the air by burning uranium from an operating reactor running away.

The first two risks are always present, and can be met by the safety rules in common use in the nuclear industry.

The third is of an accidental type, and requires special and exceptional rules going beyond the industrial field, and extending to neighboring populations.

## DANGER OF γ-IRRADIATION

Upon examination of the disintegration spectra of the fission products,[1] it will be noted that the energies of the photons emitted by the fission products can be grouped in 4 categories:

I. Photons with energies of some 0.4 Mev: $Ba^{140}$, $Nd^{147}$, $Ru^{103}$ . . .

II. Photons with energies of some 0.75 Mev: $Zr^{95}$, $Nb^{95}$, $Sr^{91}$, $I^{132}$, $I^{133}$ . . .

III. Photons with energies of some 1.5 Mev: $La^{140}$, $I^{135}$, $I^{132}$ . . .

IV. Photons with energies of some 2.5 Mev: $La^{140}$, $I^{135}$ . . .

With the knowledge of the fission yield[2] and radioactive half-lives of the fission products,[1] it is possible to determine, for each category, the intensity of the ionization produced, and its decay as a function of time.

The curves shown on Fig. 1 give the ionization intensities and their decay for each one of the categories of the photon energies of the fission products.

Computation is made for 1 kg of uranium irradiated to the saturation point, with a power of 1 w/gm.

The curves provide a simple means of finding out the required shield thickness for the handling of the uranium slugs. According to the duration of cooling before the slugs are handled, and the nature of the material chosen for the protective shield, one of the 4 categories of photons generally predominates.

For instance, for a one day cooling, the photons of the third category would be the only ones to be considered when determining the shield thickness.

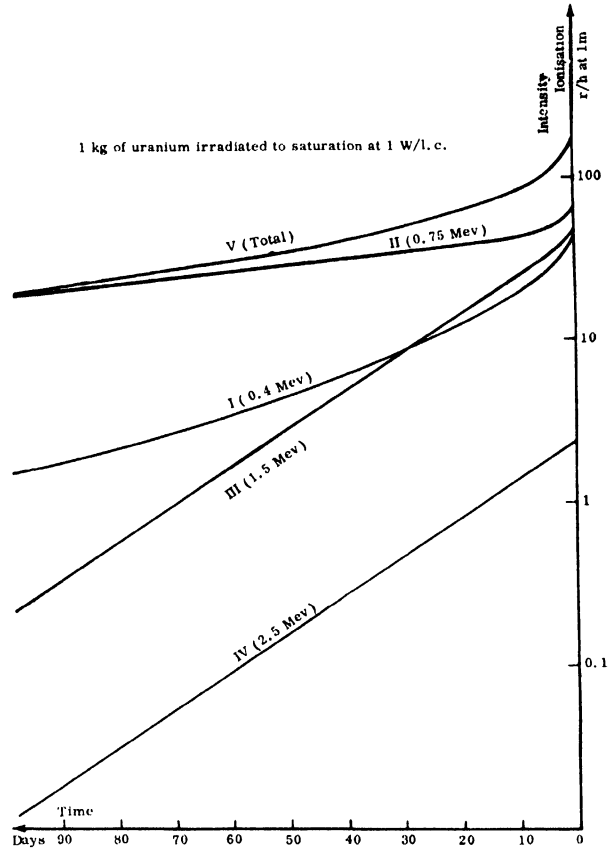



Figure 1. Decrease in the activity of the uranium

For an activation duration of 3 months, the photons of category II shall be considered.

## DANGER OF CONTAMINATION OF THE AIR ONCE THE URANIUM SLUGS ARE TAKEN OUT OF THE CLADDING

The radioactive families of the fission products which lead to substantial activities of radioactive gases are indicated in Table I.

The half-lives of these radioelements are very short. The half-lives of the rare gases, at the end of the chains, are the longest.

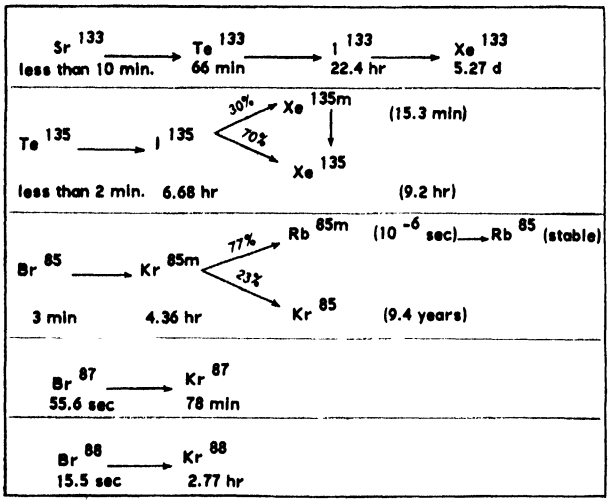
Since the fission gases are rare gases, the computation of the maximum concentrations allowable in air does not rest on the dose delivered by the radioactive element inside the body, but rather on the dose which a person supposed to be in an infinite cloud of radioactive gas will receive.

Original language: French.

* Chief, Radiation Protection Department, C.E.A.

† Radiation Protection Department, C.E.A.

Table I




Under those conditions, the limiting concentration will be reached when the energy absorbed or emitted in the air will be 0.3/1.13 rad/week (1.13 is the ratio of the stopping powers, in the tissues and in the air, for $\beta$ particles and the secondary electrons produced by X- and $\gamma$-radiations). The rad corresponds to an absorbed energy, per gram of air, of 100 ergs, or $6.3 \times 10^7$ Mev.

Thus 0.3/1.13 rad/week corresponds to an absorbed energy, per cubic meter and per second, of

$$\frac{0.3}{1.13} \times 6.3 \times 10^7 \times 1.293 \times 10^3 \times \frac{1}{0.864 \times 10^5 \times 7} = 0.355 \times 10^5 \text{ Mev}$$

The limiting concentration of the radioactive gas in air, which emits, by disintegration, an average energy of $E$(Mev) will be:

$$X = \frac{0.355 \times 10^5}{E \times 3.7 \times 10^{10}} \text{ c/m}^3$$

$$X = \frac{0.95}{E} \mu\text{c/m}^3$$

The activities of the fission gases present in a 10 kg uranium slug irradiated to saturation at a power level of 1 w/gm are indicated in Table II.

The air volume which may be contaminated by the whole of the fission gases is $4.1 \times 10^5$ m³. In the case of an intense flux reactor, giving $10^{14}$n/cm²/sec (30 w/gm), the fission gases of such a slug could contaminate a volume of $10^7$ m³.

## DANGER OF AIR POLLUTION BY BURNING URANIUM FROM AN OPERATING REACTOR RUNNING AWAY

For the population, the maximal allowable concentration of a given radioelement in the air (continually breathed in by the subject) is that which can produce, in a critical organ (the one which selectively retains the radioelement considered), an irradiation of 0.03 rem/week.

The maximal concentration allowed for the population is expressed by the formula:[3]

$$X = 0.294 \times 10^{-11} \times \frac{m}{T_E f_a \Sigma E(\text{RBE}) N} \times \frac{1}{1 - e^{-0.693\,\theta/T_E}} \qquad (1)$$

in which $X$ is maximum permissible activity in air, in c/cm³ (or c/m³); $m$ is mass of the critical organ in grams; $T_E$ is effective decay half-life of radioactivity in the critical organ in days*; $f_a$ is fraction of the total activity inhaled stored by the critical organ; $E$ is mean energy emitted by disintegration in the form of $\alpha$, $\beta$, $\gamma$, $e^-$, $n_{th}$, $n_r$, $p$ . . . emission Mev; $RBE$ is relative biological effectiveness; $N$ is a heterogeneity coefficient for accumulation in the critical organ. When the radioelement held in the critical organ is not homogeneously distributed, the doses received also are stronger at some points; now, at those points of maximal activity, the irradiation must not exceed 0.03 rem/week, hence the need for this factor $N$;

* $\lambda_E = \lambda_R + \lambda_B$ or $\frac{1}{T_E} = \frac{1}{T_R} + \frac{1}{T_B}$ where $T_R$ is radioactive half-life and $T_B$ is biological half-life.

Table II

| *Nuclide* | *Half-life* | *Fission yield* % | *4* c* | *5* mc* | *6* mc/m³* | *7* × 10⁴ m³* |
|---|---|---|---|---|---|---|
| $Xe^{138}$ | 5.3 d | 6.6 | 562 | 242 | $5 \times 10^{-3}$ | 4.8 |
| $Xe^{135}$ | 9.2 h | 5.9 | 317 | 136 | $2 \times 10^{-3}$ | 6.8 |
| $Kr^{85m}$ | 4.36 h | 1.65 | 140 | 60 | $3 \times 10^{-3}$ | 2.0 |
| $Kr^{85}$ | 9.4 y | 0.38 | 33 | 13 | $4 \times 10^{-3}$ | .3 |
| $Kr^{87}$ | 1.3 h | 3.5 | 296 | 127 | $6 \times 10^{-4}$ | 21.0 |
| $Kr^{88}$ | 2.8 h | 5 | 425 | 181 | $3 \times 10^{-3}$ | 6.0 |
| Total | | | | | | 41 |

* Column 4 is total activity of the fission gases present in the slug. Column 5 is activity of the fission gases present between the uranium and the cladding, which may be released in the case of a cladding break. This activity is due to the surface fissions, *through a thickness of 3μ of uranium* (experimental determination made at Saclay). Cartridge diameter 26 mm. Column 6 is maximal concentrations permitted for durations of exposure of 24 hr mc/m³. Column 7 is air volume which may be contaminated at the maximal concentration allowed by the fission gases released by a cladding break × $10^4$ m³.

and $\theta$ is duration of presence in the polluted atmosphere, in days.

### First Evaluation of the Danger

Equation 1 makes it possible to compute the maximal concentrations permitted for expositions which may last throughout the life of the subject.

In such a case, for all the important fission products, and for the uranium

$$1 - e^{-0.693\,\theta/T_E} = 1$$

So that we can write

$$X_{inf} = 0.294 \times 10^{-11} \frac{m}{T_E\, f_a \,\Sigma\, E(\mathrm{RBE})N}$$

For plutonium

$$1 - e^{-0.693\,\theta/T_E} = 0.34 \text{ (for } \theta = 70 \text{ years)}$$

We take for plutonium

$$X_{70\ years} = X_{inf}/0.34$$

A first evaluation of the danger of air pollution by hot uranium can be made, if it be assumed that air pollution may not exceed the permissible maximal concentrations allowed: $X_{inf}$ for the fission products and uranium, $X_{70years}$ for plutonium.

From the knowledge of the maximum concentrations allowed, the fission yield,[2] and the radioactive periods,[1] one can determine the extent of the danger of inhaling air contaminated by uranium, plutonium and fission products. The critical organs, for this type of contamination, are essentially the bones and thyroid.

Figure 2 shows the extent of the danger, and its decrease in the case of the bones.

The unit used for the ordinates is the maximal concentration permitted for the population, in the case of prolonged exposition.

The computation is carried out for the case of the contamination caused by one gram of uranium to 1 $m^3$ of air; the uranium having been irradiated for one year at 30 w/gm.

Plutonium, and the fission products as a whole, present dangers of the same order of magnitude.

One gram of hot uranium may pollute, to the permissible maximum, a volume of $10^{10}$ $m^3$ of air.

Figure 3 shows the importance of the danger, in the case of the thyroid gland.

The danger here is four times less than for the bone and its decrease is much more rapid.

### Second Evaluation of the Danger

In the case where the exposure is limited to $\theta$ = 1 day, irradiation of the critical organ will not exceed 0.03 rem/week, namely: $5 \times 10^{-8}$ rem/sec, if air contamination does not exceed

$$X_1 = \frac{X_{inf}}{1 - e^{-0.693/T_B}}$$

for fission products, plutonium and uranium ($T_B$, in those formulae, is shown in days).

A comparison between the dangers of uranium, plutonium and the fission products, for the bones,

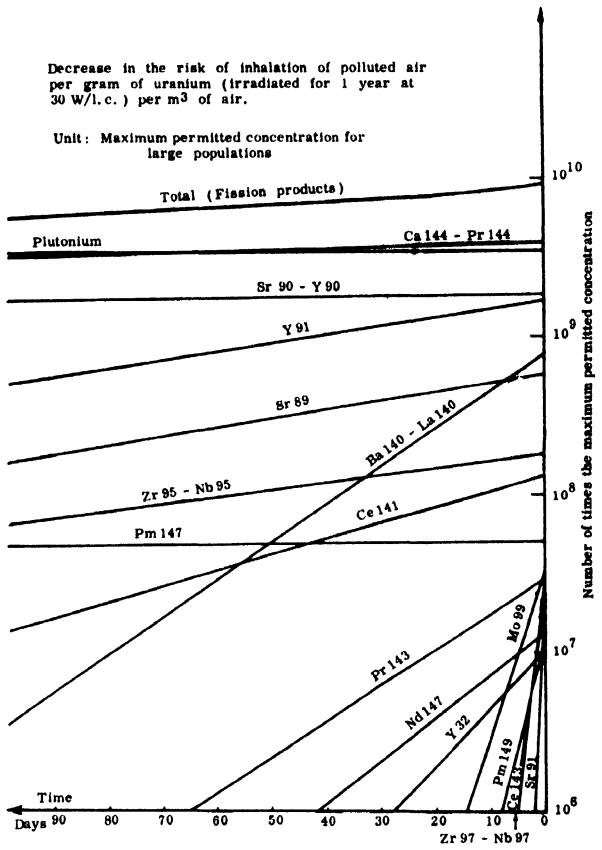



Figure 2. Bone (fission products and plutonium)

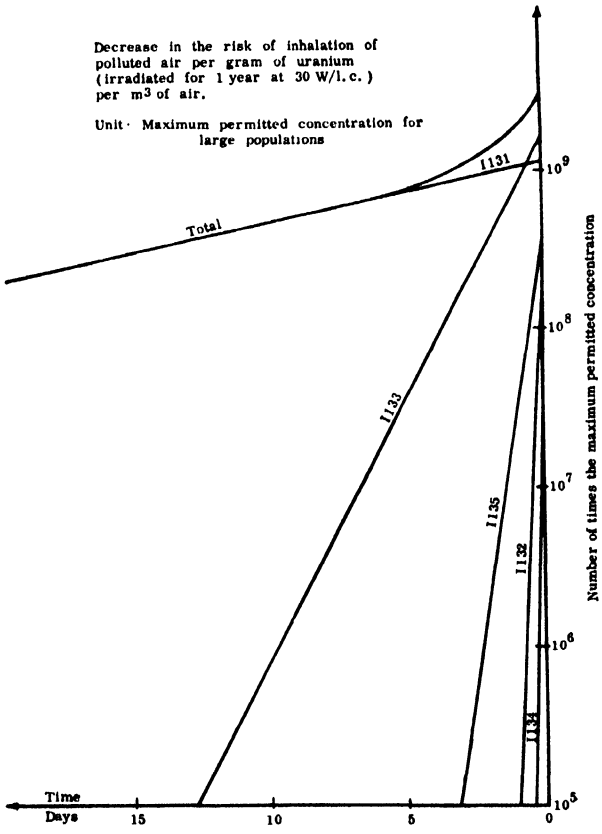



Figure 3. Thyroid gland (iodine)

**Table III**

| | Comparative dangers of: | | | |
|---|---|---|---|---|
| | *U* | *Pu* | *FP, bone* | *FP, thyroid gland* |
| 1st evaluation | 1 (unit) | 15,500 | 42,000 | 15,700 |
| 2nd evaluation | 0.023 | 0.73 | 660 | 7,100 |

and of the fission products, for the thyroid gland, are shown in Table III.

### Third Evaluation of the Danger

A third manner of evaluating the danger consists of determining the integrated dose received by the critical organ for a given duration of presence in a contaminated atmosphere ($\theta$), followed by an additional duration of presence ($t$) in an unpolluted atmosphere.

#### Intensity of Irradiation of the Critical Organ During its Presence in a Contaminated Atmosphere

If the atmosphere is polluted to a concentration $X_{inf}$ (see first evaluation), the dosage of irradiation to the critical organ is $5 \times 10^{-8}$ rem/sec, at saturation $I_\theta = 5 \times 10^{-8} \times (1-e^{-\lambda_R \theta})$ rem/sec, following a duration of presence $e$ in a polluted atmosphere.

If the atmosphere is contaminated to a concentration $x = x_o\, e^{-\lambda_R \theta}$ ($x_0$: contamination at the moment of the accident), the irradiation dose to the critical organ is:

$$I_\theta = \frac{x}{X_{inf}} \times 5 \times 10^{-8} \times (1-e^{-\lambda_R \theta}) \text{ rem/sec}$$

in which

$$I_\theta = \frac{x_o}{X_{inf}} \times 5 \times 10^{-8}.\ (1-e^{-\lambda_R \theta}) \times e^{-\lambda_R \theta} \qquad (2)$$

#### Integrated Dose Applied to the Critical Organ During Its Presence in a Polluted Atmosphere

The integrated dose to which the critical organ has been subjected is:

$$D_e = \int_0^\theta I_\theta\, d\theta, \text{ from which with Equation 2}$$

$$D_\theta = \frac{x_o}{X_{inf}} \times 5 \times 10^{-8} \times \frac{1}{\lambda_R}(1-e^{-\lambda_R \theta}) - \frac{1}{\lambda_E + \lambda_R}(1-e^{-(\lambda_E+\lambda_R)\theta}) \qquad (3)$$

This formula is general ($\lambda$ in $sec^{-1}$)

#### Integrated Dose to which the Critical Organ Has Been Subjected for a time *t*, Counted from the Moment when the Person Is Taken Out from the Polluted Atmosphere

Following contamination for a time $\theta$, the intensity of irradiation in the critical organ decreases according to the law

$$I_t = I_\theta\, e^{-\lambda_E t}$$

and the dose to which the subject has been subjected is

$$D_t = I_\theta \frac{(1-e^{-\lambda_E t})}{\lambda_E}$$

1. For $T_R$ under 10 days

$$D_t = \frac{x_o}{X_{inf}} \times 5 \times 10^{-8}\ (1-e^{-\lambda\theta}) \times e^{-\lambda\theta} \times \frac{(1-e^{-\lambda t})}{\lambda}$$

or, for $\theta = 1$ day

$$D_t = \frac{x_o}{X_{inf}} \times 6.2 \times 10^{-3} \times T \times (1-e^{-0.7/T}) \times e^{-0.7/T} \times (1-e^{-0.7\, t/T}) \qquad (4)$$

in which $T$ is expressed in days, and $D_t$ in rem.

2. For $T_R$ over 10 days

$$D_t = \frac{x_o}{X_{inf}} \times 5 \times 10^{-8} \times \theta \times (1-e^{-\lambda_E t})$$

or, for $\theta = 1$ day

$$D_t = \frac{x_o}{X_{inf}} \times 4.3 \times 10^{-3} \times (1-e^{-0.7\, t/T_E}) \qquad (5)$$

Observation: one can write

$$D_t = D_{inf}\ (1-e^{-0.7\, t/T_E}) \qquad (6)$$

In Table IV, the results of the computation of integrated doses are shown for uranium, plutonium and the fission products which give largest integrated doses.

The contamination per cubic meter of air is supposed to be produced by one gram of uranium, irradiated at 30 w/gm for 1 year.

### Application

Let us assume that, following an accident which takes place in a reactor which is in operation and has been running for 1 year at a power of 30 w/gm, the air of the building has been polluted by 1 gm of uranium per $m^3$ of air.

#### Case 1. The Neighboring Population Is Evacuated 24 hours After the Accident

Irradiation can be considered massive, only in case the dose is applied over some 50 days.

The computations indicated above give a value of $1.2 \times 10^7$ rem for the dose received in 50 days in the critical organ (bone), in case the person would stay for 24 hours in the building where the accident took place.

Let us compute the volume of contaminated air which can, in one day, be allowed to get out of the building for the dose received by the population to the leeward of the reactor (breeze blows from the latter) not to exceed, 50 days after evacuation, a value of 50 rem in the critical organ.

The dilution required then is

$$\frac{1.2 \times 10^7}{50} = 2.4 \times 10^5$$

If it be assumed that the contaminated air escaping from the building is distributed homogeneously over a cross section of 1000 $m^2$, and that it is carried by a

Table IV

| Elements | $T_B$ day | $D_\theta$ $\theta = 1$ day $10^3$ rem | $D_\theta$ $\theta = 70$ years $10^6$ rem | $D_t$ for $\theta = 1$ day $t = inf$ $10^3$ rem | $t = 10$ d $10^3$ rem | $t = 50$ d $10^3$ rem | $t = 1$ yr $10^3$ rem | $t = 10$ yr $10^3$ rem | $t = 70$ yr $10^{-3}$ rem |
|---|---|---|---|---|---|---|---|---|---|
| $Sr^{89}$ | 52 | 165 | 95 | 2450 | 300 | 1175 | 2450 | ibid | ibid |
| $Sr^{90}$ + $Y^{90}$ | $2.7 \times 10^8$ | 1 | 64,000 | 7750 | 20 | 100 | 700 | 4650 | 7750 |
| $Y^{91}$ | 51 | 48 | 310 | 7100 | 920 | 3500 | 7100 | ibid | ibid |
| $Ba^{140} + La^{140}$ | 12 | 94 | 30 | 3200 | 1375 | 3100 | 3200 | ibid | ibid |
| $Ce^{144} + Pr^{144}$ | 180 | 32 | 4100 | 16,750 | 750 | 2850 | 12,600 | 16,750 | ibid |
| $Pu^{239}$ | $4.3 \times 10^4$ | 0.35 | 162,000 | 43,000 | 7 | 37 | 260 | 2600 | 14,500 |
| Total (Bones) | | 283 | 230,000 | | 3900 | 12,000 | 28,300 | 38,800 | 53,700 |
| $I^{131}$ | 7.7 | 225 | 32 | 4600 | 2700 | 4500 | 4600 | ibid | ibid |
| $I^{133}$ | 0.92 | 1300 | 4.8 | 2350 | ibid | ibid | ibid | ibid | ibid |
| Total (thyroid gland) | | 1840 | 37 | | 5100 | 6900 | 7000 | 7000 | 7000 |
| U. (kidneys) | 30 | 0.011 | 240 | 1.0 | 0.3 | 0.7 | 1.0 | ibid | ibid |

wind having an average speed of 1 m/sec, the air volume which can be allowed to escape is then:

$$\frac{0.864 \times 10^5 \times 10^3}{2.4 \times 10^5} = 360 \text{ m}^3$$

**Case 2. The Population Is Not Evacuated**

From the computations outlined, the irradiation of the critical organ, in the theoretical case of 70 years of presence in the contaminated building, is $2.3 \times 10^{11}$ rem.

In order that the average dose of 300 mr/week over 70 years not be exceeded, the dilution factor required is:

$$\frac{2.3 \times 10^{11}}{1.1 \times 10^3} = 2 \times 10^8$$

The volume of air which can be allowed to escape from the building in one day will be:

$$\frac{0.864 \times 10^5 \times 10^3}{2 \times 10^8} = 0.43 \text{ m}^3$$

## CONCLUSION

Our study of the ordinary dangers offered by the manipulation and uncladding of the uranium slugs after their activation in nuclear reactors has enabled us to get accurate values of their nature and amplitude. The curves shown in the tables presented offer a solution for the protection problems which appear, as a function of the activation of uranium in the reactor, and the subsequent deactivation following removal of the slugs.

The study of the accidental dangers which may present themselves in the eventuality of quick combustion of uranium from overheating in an operating reactor, has been pursued along lines quite different from the rules used for common industrial radiological protection. From the maximal concentrations tolerable for the surrounding population, it has been possible to compute intensities and radiation doses as a function of the initial rate of pollution in the atmosphere, and of the time spent in the contaminated medium. The formulae presented would make it possible to solve the protection problems which would require quick action, in case of uranium combustion in the reactor: volume of air that might be allowed to get out of the building each day, time and duration of evacuation of the plant, etc.

The results brought out in our studies reveal important differences in the relative toxicities of the fission products of uranium and plutonium, in cases of accidents, with respect to the figures set by international recommendations for normal work and long lasting projects. For instance, in case of quick combustion of uranium the Ba-$La^{140}$ and Ce-$Pr^{144}$ couples, and $Y^{91}$, become the most toxic substances due to their immediate effects.

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# Health Protection in Chemical Processing Plants

**By H. M. Parker and J. M. Smith,* USA**

Large-scale radiochemical processing plants have been operated at the Hanford Works for ten years. The experience has included the design and operation of improved facilities incorporating what has been learned about hazard control as well as general process improvements. This experience should be of direct value in the advancement of the peaceful application of atomic energy.

One plausible line of development would be to consider a number of heterogeneous reactor power sites having spent fuel elements reprocessed at a central facility. Such a facility would be broadly similar to the plants on which the local hazard experience is based.

Radiological health protection programs for such units have two separate phases: (1) control of radiation hazards to the employees in the operation; and (2) elimination of deleterious effects on the environs.

This survey paper sketches the findings in these two phases. The ramifications of the environmental hazards are described in more detail elsewhere.

As an administrative tool, and to guide future development, it is convenient to divide the field into two aspects: (1) external radiation, as in the form of beams of penetrating radiation; and (2) radioactive contamination, defined as actual radioactive material in the wrong place.

The local experience demonstrates clearly that there are essentially no unresolved problems in the first aspect. The major uncertainties and the major expense and time-consuming effort reside in the control of contamination, both in the plant and in the environs.

## CONTROL OF EXTERNAL RADIATION

Within the plant, the control of radiation from all sources must be formally limited to a permissible exposure of 300 mrem per week. It is highly desirable to operate with a substantial safety margin beyond this. In local practice, it is found convenient to operate to an annual target of 3 rem, with not more than the conventional limit in any one week. This introduces a long-term safety factor of 5.

The economic justification of this plan is that in a high-level facility, the over-all cost of radiation protection is high, but the incremental cost of additional safety factors is low. In typical plants, a shielding improvement by a factor of 10 adds only 0.5–1% to the total facility cost, and greatly simplifies auxiliary control methods. In general, design to a transmission limit of 1 mr per hour for fixed installations is sound.

Of the three elementary concepts of radiation control — shielding, distance, and time — the first two are economically designed into the initial structure wherever possible. Resort to time controls, though still necessary in special operations, should be considered a secondary control method to be avoided. The large-scale facility is therefore visualized as a heavily shielded, remotely operated, and generally remotely maintained plant. External radiation in such a case presents no problem. It is conventional to record the exposure of operators by personnel meters, such as pocket ion chambers and film badges, but these will repetitively show low readings in the main operations.

Substantially greater risk of external exposure arises in auxiliary operations such as routine sampling, or contact maintenance. These steps are well controlled by written procedures supplemented by continuous radiation monitoring.

In the occasional task of removal of a major equipment piece from such a plant, radiation fields in the range dangerous to health may arise. Such tasks require temporary shielding, temporary remote hauling facilities and rigorous time limits on personnel exposure. No task has arisen at this site which could not be accomplished within the standard weekly exposure limit. Relaxation of limits to some such value as 3 r in 13 weeks is not recommended in a production plant. It discourages progressive design of improved maintenance equipment, which is more economical in the long run, and promotes unbalanced work schedules for the exposed men.

While attention on external radiation control is mainly focussed on whole-body radiation, some operations such as associated laboratory sampling lead to situations in which hand exposure may be significantly higher than the general exposure. Control is achieved by separate measurement of hand exposure, say by rings containing film. For these cases, a safety factor of 2 beyond conventional limits is considered adequate.

Operating experience has shown that where radioactive parts can be handled there will be incidents in which the exposure considerably exceeds the normal level. This directs future design to eliminate all possibilities of contact handling even in the laboratory.

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## IN-PLANT CONTAMINATION HAZARDS

The maintenance of control of radioactive contamination in radiochemical processing plants is a much more troublesome problem than external radiation control. Contamination spread can create problems of deposition within the body by various routes and also add to external radiation exposure. Significant problems are chronic in this field as the basic requirement to contain all radioactive material within process equipment has not yet proved amenable to complete engineering solution. The fundamental problem of maintaining a liquid-tight chemical processing system over a period of many years of operation is difficult to attain. This is particularly true in facilities processing highly radioactive solutions where close inspection of equipment repair and replacement work performed by remotely operated tools is not possible. Thus it is necessary to assume escape of some process material from the equipment and to design for containment within enclosures housing segments of the chemical system. However, as the enclosures must be opened periodically for material movement and remote maintenance, carefully controlled ventilation and equipment movement procedures are required to minimize contamination of the portion of the process building containing the equipment enclosures. Suitable decontamination procedures are required in this process zone to prevent contamination movement to the normally occupied non-process portions of the building. More intimate contact with process material is encountered in auxiliary operations such as in radiochemical laboratories. Operating problems of this type lead to the possibility of contamination gaining entry into the body by inhalation, ingestion, and absorption or penetration through the skin.

The potential health problems encountered resulting from necessary employee exposure are dependent on the radioisotopes involved and their biological hazard. In a separations plant the availability in the form of contamination of plutonium and the full spectrum of the long-lived uranium fission products presents a variety of hazards of a magnitude not previously encountered in large scale industrial operations. The problem is complicated by permissible limits for many of the materials present which are most restrictive in comparison with many industrial chemical toxicants.

To exemplify the problem of air contamination it is revealing to compare (Table I) the permissible concentrations of various non-radioactive dusts, fumes, and mists with those of some of radioactive materials encountered in radiochemical processing.

Comparison of permissible concentrations of toxic chemical gases with certain gaseous radioactive fission products shows the figures in Table II.

Personnel access to process zones containing air contaminated with radioactive materials in the form of dusts, mists, and gases is necessary from time to time for inspection, maintenance and processing work. A routine air-monitoring program in a separations plant is essential to determine the extent of the health hazard as it is probable that a significant fraction of the cases of body deposition of long-lived radioisotopes originate from chronic exposure to contaminated air. Respiratory protection is prescribed for entry into such areas based on the air contamination history and the probability of air contamination expected to result from the work in progress. The presence in variable quantities of natural short-lived alpha- and beta-emitting radioactive materials in the air makes it difficult to employ instrumentation for the instantaneous evaluation of process contaminants; therefore it is usually necessary to determine air sample results after the short-lived materials also collected have decayed to insignificance. This problem leads to conservatism in prescription of respiratory protection.

Health hazards from air contamination depend on the physical and chemical form of the contaminant, and it is evident that the maximum permissible concentration of a particular radioisotope may vary, dependent on the specific form encountered. The hazard of radioactive noble gases may be limited to direct irradiation of the lung, but there is possibility of dissolution in lung fluids, or adsorption on lung surfaces as indication of adsorption on air sample filters has been noted. The hazard of radioiodine is primarily that of deposition in the thyroid gland. The extent of solubility in the lung of radioactive dusts also causes hazards variations. Readily soluble material will move from the lung and be deposited in other organs and excreted in accordance with expected metabolic patterns, while insoluble or slowly soluble material may remain in the lung for extended periods of time. Hazards variations may also be a function of the particle size of the contaminant, particularly in the insoluble forms as local irradiation of small lung areas from the larger and more active particles may be limiting. Studies currently underway are aimed at answering the unknowns in this field, in-

**Table I**

| *Nonradioactive* | | *Radioactive* | |
|---|---|---|---|
| *Material* | *Permissible conc. μg/cm³ of air* | *Radioisotope* | *Permissible conc. μg/cm³ of air* |
| ZnO | $1.5 \times 10^{-2}$ | $Cs^{137} + Ba^{137}$ | $3 \times 10^{-9}$ |
| Cyanide as CN | $5 \times 10^{-3}$ | $Pu^{239}$ (sol) | $3 \times 10^{-11}$ |
| $H_2SO_4$ | $1 \times 10^{-3}$ | $Ru^{106} + Rh^{106}$ | $9 \times 10^{-12}$ |
| As | $5 \times 10^{-4}$ | $Sr^{90} + Y^{90}$ | $1 \times 10^{-12}$ |
| Pb | $1.5 \times 10^{-4}$ | $Ba^{140} + La^{140}$ | $8 \times 10^{-12}$ |
| Hg | $1 \times 10^{-4}$ | $Sr^{89}$ | $7 \times 10^{-12}$ |

**Table II**

| *Nonradioactive* | | *Radioactive* | |
|---|---|---|---|
| *Gas* | *Permissible conc. PPM by vol in air* | *Gas* | *Permissible conc. PPM by vol in air* |
| CO | 100 | $Xe^{135}$ | $5 \times 10^{-9}$ |
| $SO_2$ | 10 | $I_2^{131}$ | $1.7 \times 10^{-12}$ |
| $Cl_2$ | 1 | | |
| $O_3$ | $10^{-1}$ | | |
| $Ni(CO)_4$ | $10^{-3}$ | | |

cluding the significance of various particle sizes and activities, solubility *in vivo*, and possible physical movement following initial lung deposition.

Ingestion of radioactive material is minimized by sanitary work habits while in process areas, restricting eating and smoking to contamination-free nonprocess areas, and by prohibition of the use of the mouth in operating radiochemical laboratory equipment. In the event of accidental intake some protection is afforded by a relatively low fraction transferred from the gastrointestinal tract to the critical organ, particularly in the case of the heavy metals.

Absorption of radioactive materials through intact skin and direct entry through a skin break is hazardous because of the high efficiency transfer of many radioelements from the bloodstream to critical body organs. In addition, retention of material in the skin could possibly result in tumor production. Uncontrolled skin contamination can also lead to intake by ingestion, etc.

The spread of radioactive materials produces deposits on process area surfaces which will add significantly to the problem of external radiation control. Such deposits result from high-level air contamination and movement of inadequately contained process equipment. During the course of radiation zone work, contamination of tools, skin, and protective clothing leads to uncontrolled exposure of portions of the body. It has been found that in many cases of clothing and skin contamination, the radioactive material is deposited on many small discrete areas rather than in a quasi-uniform manner. Measurement of the surface dosage rate from such a pattern is not readily accomplished on a routine basis, and oversight of this probable distribution can lead to high, localized skin dosages.

## EFFECTIVE CONTROL MEASURES

Effective contamination control is dependent on definite delineation of radiation zones from the noncontaminated portions of the facility. Periodic surveys of the latter areas are essential to insure maintenance of a contamination-free status. When entering radiation zones protective clothing and equipment commensurate with the probable hazard is donned; in some cases complete body and respiratory protection is appropriate. During work in such areas frequent contamination surveys of protective clothing and any exposed skin areas are necessary to control additional external radiation from contamination spread to these surfaces. Upon exit from the controlled area, survey and removal of protective apparel is performed and potentially contaminated body surfaces are checked with decontamination procedures instituted immediately for removal of any skin contamination found. The establishment of permanent personnel survey and decontamination facilities at the entry points aids greatly in the routine protection of employees at the termination of radiation zone work, and prevention of contamination spread beyond the controlled zones.

In a well-regulated plant, the ultimate health hazard is probably the slow accumulation of body deposits of long-lived radioisotopes over the years, plus the problems of occasional radiation accidents. Considering the uncertainties of accurately determining the maximum permissible body burden for each of the many forms of numerous radioisotopes present in a fuel-element separations plant, conservative approaches in the design and operating bases to limit contamination spread to the practical minimum have proven desirable.

## CONTAMINATION ACCIDENTS

Personnel contamination accidents principally include incidents of severe skin contamination, injury involving radioactive material in the wound, and inhalation of air-borne radioactive material. Skin decontamination procedures are aimed at maximum contaminant removal in a minimum of time to reduce the external dose to the skin and to reduce the probability of absorption. It is important to use reagents which will not cause skin damage and to apply them only to actually contaminated skin as determined by careful survey.

When an injury occurs in a contaminated zone, immediate action is taken to institute a profuse water flush of the injury and to promote bleeding. If the injury is on an extremity, tourniquets are applied to control venous return blood flow. If surveys of the injury, blood smears, or the offending object indicate the presence of a contaminant, decontamination is performed under medical direction. In only one case has a puncture wound apparently free enough from contamination been later found to be contributing to a gradual body deposition. In serious cases, ultimate protection by excision of the wound area may be advisable.

When an incident of acute inhalation of radioactive material occurs, nasal passage irrigation is employed to remove any material deposited in the upper respiratory passages. Qualitative evidence of exposure may be obtained by survey of nasal swabs and analysis of the irrigation solution. Correlation of the probable exposure time and air monitoring results provides a quantitative estimate of intake. If the radioisotopes involved permit detection by external survey methods, monitoring of the thyroid area for radioiodine, and of the chest cavity for lung deposition is performed.

## INTERNAL DEPOSITION EVALUATION

Employees in radiochemical processing plants routinely submit urine samples for radioisotopic analysis on a frequency based on the exposure potential of their jobs. In addition, when a contamination accident occurs appropriate biological samples are taken for analysis; subsequent sampling is performed as needed based on initial findings. This bio-assay program provides the information necessary to determine whether any limitations on future work assignments of the individual are required because of deposition of radioisotopes in the body.

There has been no instance at this location of a body deposition in excess of the formal limits. However, 2% or 3% of the exposed work force have in the ten year period developed depositions in the range of 1% to 40% of the limit. The need for improved engineering as the industry becomes a long-range operation is evident.

## PROTECTION OF COMMUNITY AND ENVIRONS

While nuclear power reactors may of necessity be located near large population and industrial centers consuming the electrical energy, the associated spent fuel element separations plants could still be located in semi-isolated regions to take advantage of greater economy in radioactive waste disposal methods.

Regardless of the proximity of the separations plants to its neighbors, poor in-plant contamination control and shipment of radioactive material may cause radiation problems of concern to the community. Inadequate surveys of personnel when leaving plant radiation zones can result in spread of possibly hazardous quantities of radioactive material to public and private vehicles, employees' residences, and public places. Such situations are complicated by the probable length of time before the off-standard condition is recognized as there is little chance of detection unless residual contamination on the employee's person is discovered upon subsequent return to work. Similar problems are caused by the inadvertent release of contaminated material and equipment for off-plant use. The hazard to future handlers and users of such equipment is very rarely known.

The shipment of radioactive material, if in accordance with regulations currently in effect in the United States which give adequate protection in the control of radiation and the containment of radioactive material under normal conditions, should be of little concern in passage through communities. However, accidents en route may create public health problems. Spillage of radioactive solids, such as irradiated fuel elements, would produce potentially dangerous radiation levels for distances of several hundred feet from the source, and retrieving of the material should be undertaken only under carefully controlled conditions. Leakage of radioactive liquids would result in spread of contamination possibly accompanied by significant radiation levels. Excavating operations may be required to recover valuable materials and to remove the hazard. Particular attention should be devoted to the possibility of material entering sewage systems. Communities should be prepared for such incidents to the extent that public officials are aware of this unusual type of problem, and that police would be instructed to clear the accident area of all personnel until the extent of hazard can be evaluated by radiation control experts.

## RADIOACTIVE WASTE DISPOSAL

A principal responsibility of the radiochemical industry is the use of radioactive waste-disposal practices of assured safety. In a separations plant, the hazardous material in waste material includes the products of nuclear fission plus the processing losses of fissionable materials. Wastes containing mixed fission products include long-lived radioisotopes such as strontium-90 and cesium-137 which decay to negligible quantities only after several centuries, while wastes containing highly toxic plutonium-239 with a half-life of 24,000 years are essentially a permanent problem. Potential health hazards exist for each of the two general approaches to waste handling, "dilution and dispersion" into the environs, and "concentration and containment" in storage facilities.

The disposal of radioactive liquid wastes generated in a separations plant presents some of the most significant potential hazards with regard to long-term public health in the plant environs. Disposal to public waterways may be of concern to downstream users for domestic, agricultural, and industrial purposes, and may affect aquatic life. When this method is used, a continued monitoring program to assure safe concentrations at usage points and in aquatic organisms is essential. The degree of control necessary in the release of such wastes is indicated by a comparison of maximum permissible concentrations in drinking water of radioelements possibly present in separations plant wastes with those of some chemical poisons (Table III).

**Table III**

| *Nonradioactive* | | *Radioactive* | |
|---|---|---|---|
| *Material* | *MPC, in PPM by wt, in $H_2O$* | *Radioisotope* | *MPC, in PPM by wt, in $H_2O$* |
| Sulfate | 250 | $Cs^{137} + Ba^{137}$ | $3 \times 10^{-5}$ |
| Copper | 3 | $Pu^{239}$ | $2 \times 10^{-5}$ |
| Fluoride | 1.5 | $Sr^{90} + Y^{90}$ | $4 \times 10^{-9}$ |
| Lead | 0.1 | $Sr^{89}$ | $3 \times 10^{-9}$ |
| Arsenic | 0.05 | $I^{131}$ | $2 \times 10^{-10}$ |

In some locations advantage is taken of the retentive and adsorptive qualities of natural soils for the sub-surface disposal of waste streams contaminated with radioisotopes of concentrations exceeding permissible drinking water limits. In general this practice has been limited to semi-isolated regions of low precipitation, terrain high above ground water, slow ground-water movement, and significant distances to public waterways. Health hazards may arise in this practice if long-lived radioisotopes eventually migrate down the soil column and move to a river at a rate greater than anticipated. Disposal of even slightly contaminated liquid wastes on the ground surface is to be avoided as concentration of the radioactive material on the surface sediments leads to windborne contamination spread and hazards to wildlife making use of such water sources.

The storage of highly radioactive liquid wastes in underground tanks presents no public health problems if tanks of assured long-term integrity can be provided. Tank failure allowing even partial leakage of contents may lead to significant ground water con-

tamination and eventual escape to rivers. In areas used for ground storage or disposal, the need for ground water usage control is evident.

The disposal of radioactive solids by ground burial may also cause migration of contaminants to areas of public concern unless adequate containment methods are employed to resist dissemination by natural forces. When liquids or solids containing long-lived radioisotopes are placed in ground storage or disposal units, permanent land control of the site is indicated to prevent inadvertant disturbance in the future. Alternate disposal of such waste materials to deep sea locations may be required for operations located in an environment unfavorable for waste disposal; such methods add to potential public health difficulties in transport and may prove economically unattractive for large radiochemical enterprises.

The release of process effluents to the atmosphere is a continual operational problem in large-scale radiochemical separations. Such releases may involve gases or intrinsically volatile substances or fine mists producing particulate contamination. Decontamination of process off-gases and process equipment enclosure ventilation air by means of high efficiency filtration and chemical treatment before release to the atmosphere through tall stacks is usually required. Control of the release of iodine-131 generated in the dissolution of fuel elements has proved particularly important in maintaining environs vegetation contamination at a level safe for human and animal consumption. The release of long-lived noble gases may be of increasing importance as atomic energy utilization expands. The deposition of radioactive material on inert carrier particles, such as ventilation system corrosion products, can cause potential hazards from inhalation of small particles and external skin exposure from body surface deposition of large particles. Fallout of such material in the environs may necessitate institution of radiation control procedures for outside work and personnel movement for distances of several miles from the source. Secondary windborne movement of ground deposited particles may add to the area of potential hazard.

Considerations of potential health problems in the environs of a radiochemical processing plant from planned or accidental release of radioactive material play an important part in the design and operation of such a facility.

# The Radioactive Survey of the Area Surrounding an Atomic Energy Factory

By D. R. R. Fair and A. S. McLean,* UK

The first of the British production piles and the associated chemical separation and purification plants are located at the Windscale Works at Sellafield in Cumberland. Before radioactive operations commenced it was necessary to measure the levels of the naturally occurring radioactivity in the area surrounding the factory. The reasons for so doing included technical, managerial and public relations aspects.

The main technical reason for the survey was that the amounts of naturally occurring radiation and radioactive substances were not known in detail and had not been measured in the area before. If an assessment were to be made of the effect of the factory's operations on its environment, the levels prior to the commencement of operations would have to be known. Additionally the measurement of the naturally occurring radiation and radioactive materials provided an opportunity to establish techniques and to train both scientific and semi-skilled employees in the methods of radioactive survey. The factory management decided, at an early stage, that it would be wasteful of scientific man-power to use trained scientists to make routine radioactive surveys. The surveys, which will be described later, were directed by scientists but were carried out by locally recruited and trained semi-skilled labour.

The importance of good public relations was given proper consideration from the beginning. The industry was associated in the public mind with atomic bombs and therefore with danger and it was necessary to reassure the people who lived in the surrounding countryside that there would be no danger from the operation of the new factory. The fact that a natural background of radioactivity existed at all soon became known in the Cumberland area because of the activities of the Atomic Energy Authority's mobile laboratories. These surveys and their subsequent discussion by the local people and the local press helped to bring some of the features concerning radiation and radioactive materials, which were previously not known generally, into better focus in the public mind.

## THE SURVEY AREA

The area surrounding the Windscale Works within a radius of twenty miles was surveyed. A total of 240 survey points was established and arranged in 21 routes. Each route was planned so that a mobile laboratory team could cover the necessary distance and make the required readings and observations during the course of a normal shift of eight hours. The survey points were arranged with most points near to the factory (Table I).

**Table I**

| *Distance from factory* | *Number of survey points* |
|---|---|
| 0–5 miles | 70 |
| 5–10 miles | 70 |
| 10–15 miles | 60 |
| 15–20 miles | 40 |

The points had to be accessible to a mobile laboratory and therefore certain areas, where suitable roads were not available, did not contain survey points. In these void areas there are no centres of population.

## THE MOBILE LABORATORIES

The mobile laboratories derived their electric power either from a 24-volt direct current to 230-volt alternating current rotary converter working from a large bank of batteries or from a petrol-driven generator producing 230-volt alternating current. The laboratories were equipped with both morse and speech radio which provided two-way communication to a control room in the factory. The radioactivity measuring equipment consisted of: (1) gamma-radiation measurement—standard type ionisation chamber and head amplifier, vibrating reed electrometer; (2) beta air-activity measurement—air sampler complete with anemometer, Geiger-Müller probe in lead castle—power unit and scalar unit, and (3) alpha air-activity measurement—air sampler complete with anemometer, scintillation counter unit, power unit and scalar unit.

## THE GAMMA-RADIATION SURVEY

The natural gamma radiation arises partly from the uranium and thorium and their radioactive decay products in the soil, partly from the effect of cosmic rays on the earth's atmosphere and to a very small extent from the airborne radioactive gases radon and thoron in the atmosphere.

The gamma radiation was measured by the ionisation current produced in a 4½ litre ionisation chamber

* UK Atomic Energy Authority, Risley.

containing argon at 20-atmospheres pressure. From cosmic-ray measurements it was expected that the natural gamma-radiation intensity at ground level would be of the order of 10 microroentgens per hour. This radiation level produces an ionic charge of either sign of $5 \times 10^{-18}$ coulombs/millilitre/second in air at 0°C and 760-millimetres pressure. Consequently even with a 4½ litre, high-pressure ionisation chamber, the total ionisation current expected would be only of the order of $10^{-13}$ amperes. The ionisation current produced was used to generate a voltage across a high resistance. The voltage was then amplified by a vibrating reed electrometer. It was found that the readings obtained were dependent on the ambient temperature. The changes in temperature experienced were sufficient to alter significantly the value of the resistor. A series of experiments to measure this temperature effect was made and the following empirical relationship was established.

$$\theta_T = \theta_{60}\, e^{-\frac{(T-60)}{100}}$$

where $\theta_T$ is the instrument reading for a particular gamma-radiation level at a temperature of $T$°F, and $\theta_{60}$ is the instrument reading at the same gamma-radiation level at a temperature of 60°F. This relationship was used to correct all observations to a standard temperature of 60°F.

The instrument was calibrated by placing a standard radium source at varying distances from 40 feet to 90 feet from the ionisation chamber. The source was 0.985 milligram of radium in a platinum capsule. This calibration, in effect, defined the "roentgen" as used in this survey and the natural gamma-radiation levels were expressed in terms of the radium gamma level which would produce the same ionisation current in the ionisation chamber. The calibrations were made with the mobile laboratories in a fixed position on an open space, in order to avoid scattered gamma radiation from buildings and other vehicles. The standard radium source was fixed in a source holder mounted on a tripod, and held at the height of the centre of the ionisation chamber. The source was located by a plumb bob to be directly above permanently marked positions on the ground. The calibration procedure was carried out every eight hours and the instruments were, as far as possible, operated continuously to reduce the possibility of zero drifts and transient effects due to switching.

The field measurements of the naturally occurring gamma-radiation levels were made by driving the mobile laboratory to each of the selected survey points. The vehicle was parked so that the ionisation chamber was precisely located. This permitted exact comparisons to be made with other measurements at the same survey point. The zero setting of the instrument was checked and then five readings were taken at half-minute intervals and the temperature near the instrument was recorded. After applying the temperature correction and calibration factor, the value of the gamma-radiation field at the point was obtained in microroentgens per hour.

The gamma-radiation level in the area varies between four and ten microroentgens per hour (Table II). The mean level over the whole of West Cumberland is seven microroentgens per hour with a mean variation of approximately two microroentgens per hour at any one location. The cosmic component of this gamma-radiation level is calculated to correspond to a radiation level of approximately one microroentgen per hour. Due to the particle showers, caused by the cosmic radiation on the walls of the ionisation chamber, the measured ionisation from the cosmic rays is greater than this calculated value for free air. From experimental work the effect of cosmic rays on the ionisation chamber was estimated to be the same as three microroentgens per hour from the standard radium source. It was not found possible to correlate the measured radiation levels with any physical factor, such as height above sea level, position or weather conditions.

## THE RADIOACTIVE SURVEY OF SOIL, ROOTS AND VEGETATION

Samples of turf, complete with roots and adherent soil were collected from thirty survey points in the area around the factory in West Cumberland. The mobile laboratories were used to collect the samples but all subsequent work was carried out in the factory's radiochemical laboratories. The operations concerned in the radioassay of the samples were: (1) the preparation of the samples for presentation to the counting equipment; (2) the calibration of the counting equipment; and (3) the determination of the radioactivity in the prepared sample.

Each sample was divided into three parts, soil, roots, and aerial vegetation. The vegetation and root samples were weighed, dried and ashed at 500°C and then reweighed, thus providing a wet weight to ash weight ratio. The soil was dried and ashed to destroy organic matter. All samples were ground until they would pass through a number 60 British standard sieve which has an aperture of approximately 250 microns. The sample was finally thoroughly homogenised and spread evenly on a clean counting tray. The depth of sample was five millimetres for the alpha-activity determinations and 18 millimetres for the beta-activity determinations.

The direct determination of alpha activity in a sample of low activity is difficult since a large quantity of material is required to give a count significantly above the background count rate of the instrument. The alpha count rate cannot be increased indefinitely by increasing the quantity of material since the alpha particles have a very short range. The thicker the sample the more alpha particles are absorbed in the lower layers of the sample and if the depth of a soil sample is greater than one millimetre then an "infinite thickness" sample is obtained. For such a sample the only non-instrument factors which affect the counting rate are the specific activity and the surface area of the sample. Thus if successive samples are spread to a depth greater than

**Table II. Natural Gamma-Radiation Levels for Some Selected Locations in the West Cumberland Area**

| Location: National Grid Reference | Gamma radiation level in microroentgens per hour | | | Range of levels at fixed locations in microroentgens per hour |
|---|---|---|---|---|
| | Mean | Maximum | Minimum | |
| 220244 | 6.65 | 7.85 | 6.00 | 1.85 |
| 192177 | 7.15 | 7.55 | 6.48 | 1.07 |
| 176171 | 7.39 | 8.00 | 6.95 | 1.05 |
| 325129 | 7.62 | 8.93 | 6.10 | 2.83 |
| 038272 | 5.68 | 6.87 | 4.99 | 1.88 |
| 155227 | 6.85 | 8.55 | 6.26 | 2.29 |
| 319974 | 7.71 | 8.65 | 6.09 | 2.56 |
| 203009 | 7.20 | 7.80 | 6.24 | 1.56 |
| 197883 | 6.41 | 8.29 | 5.65 | 2.64 |
| 106930 | 6.65 | 8.07 | 5.37 | 2.70 |
| 162066 | 6.65 | 7.27 | 5.95 | 1.32 |
| 044162 | 6.87 | 7.56 | 5.64 | 1.92 |
| 083993 | 6.07 | 6.75 | 5.28 | 1.47 |
| 075013 | 5.78 | 6.45 | 5.31 | 1.14 |
| 064060 | 7.17 | 7.47 | 6.87 | 0.60 |
| 013108 | 7.13 | 8.12 | 5.87 | 2.25 |
| 948148 | 5.77 | 6.52 | 5.62 | 0.09 |
| 058043 | 6 60 | 7.52 | 5.27 | 2.25 |
| 014061 | 6.06 | 6.70 | 5.53 | 1.17 |
| 052986 | 5.86 | 6.50 | 5.44 | 1.06 |
| 031329 | 6.06 | 7.25 | 5.30 | 1.95 |
| 129336 | 5.73 | 6.87 | 5.08 | 1.79 |
| 248914 | 6.99 | 8.25 | 5.86 | 2.39 |
| 141899 | 6.84 | 7.20 | 6.37 | 0.83 |
| 257143 | 6.67 | 7.33 | 5.01 | 2.32 |
| 225136 | 6.81 | 8.04 | 5.45 | 2.59 |

one millimetre in identical type counting trays the counting rates in excess of the instrument background count will be directly proportional to the specific alpha activity of the sample. However, unless the precise alpha emitters and their relative proportions in the sample are known it is not possible to obtain the specific radioactivity by direct calculation from the count rate of the sample. To overcome this difficulty calibration sources were prepared by adding known quantities of chemically pure uranium as uranyl nitrate to acid-washed silver sand which had passed through a number 60 British standard sieve. The counters were calibrated by the calibration sources at intervals during the counting period. Consequently the alpha activity of the samples was assessed in terms of the uranium alpha activity required to give the same count rate as the sample.

The determination of the beta activity of the sample was carried out in a manner similar to that for alpha activity. The "infinite depth" sample was 18 millimetres in depth and the calibration source was prepared by using a known amount of potassium as potassium chloride. Consequently the beta activity of the sample was assessed in terms of the potassium beta activity required to give the same count rate as the sample. The measurement of the beta activity of vegetation and roots in terms of the equivalent potassium activity is quite good since the natural activity is mainly due to the radioactive isotope of potassium. The measurement of the alpha activity of soil in terms of the equivalent uranium activity is not so good since only 25% will be due to uranium. The remainder is due to thorium and the decay products of uranium and thorium.

Typical values of the naturally occurring alpha and beta activity in soil, roots and vegetation are given in Table III.

## THE DETERMINATION OF THE ALPHA AND BETA AIRBORNE RADIOACTIVITY

The earth's atmosphere contains small quantities of the radioactive gases radon and thoron formed by the radioactive decay of radium and thorium. The daughter products of radon and thoron are themselves radioactive but are not gases. When they are formed from their parent atoms they are ionised and consequently attach themselves to any atmospheric dust or solid matter.

One method, therefore, of measuring the atmospheric radioactivity was to assess the radioactivity found on atmospheric dust. The operations con-

**Table III**

| | Alpha activity | Beta activity |
|---|---|---|
| Soil | 10 micromicrocuries/gm dry weight uranium equivalent | 24 micromicrocuries/gm dry weight potassium equivalent |
| Roots | 0.3 micromicrocuries/gm wet weight uranium equivalent | 1.7 micromicrocuries/gm wet weight potassium equivalent |
| Vegetation | 0.3 micromicrocuries/gm wet weight uranium equivalent | 3.1 micromicrocuries/gm wet weight potassium equivalent |

cerned in the radioassay of atmospheric dust were: (1) the collection of the dust sample in such a form that its assessment could begin immediately after collection; (2) the counting of the radioactivity of the sample; and (3) the determination of the percentages of radon and thoron decay product and the calculation of the airborne activity.

The air was drawn through a filter paper by a standard domestic type vacuum cleaner. An anemometer attached to the air outlet of the cleaner recorded the amount of air in linear feet which passed through the filter paper. From the result of calibration experiments this figure was converted to the volume of air sampled in cubic metres. The filter paper holder was a hollow brass perforated cylinder closed at one end with a screw cap. The filter paper was cut to the required size, sealed with a cement and eased into position in the holder. The ends of the cylinder of filter paper were then sealed to the inside of the holder with a cement and the whole placed in a dessicator until the cement hardened. The filter paper had to collect dust efficiently without reducing the air flow sufficiently to cause overheating of the vacuum cleaner motor. The paper chosen for this duty permitted an air sample of approximately 150 cubic metres to be drawn through it in three hours. The assessment of the radioactivity on the filter paper commenced in the mobile laboratory as soon as the sample was collected. The filter paper in the holder was placed over a Geiger-Müller tube in a lead castle. The decay curve of the activity was determined by a series of five-minute counts taken over a period of two hours with one minute's interval between each count.

This was followed for the next two hours by nine-minute counts with one minute's interval between each count and finally two half-hour counts were taken. The measured instrument background count was subtracted from these counts and the results graphed. The shape of the decay curve depends upon the percentage of radon to thoron decay products. Theoretical curves for a range of 0 to 30 per cent of thoron decay products were scribed on "Perspex" and the sample decay curve was matched to one of the theoretical curves, thereby obtaining the percentage of thoron decay products present in the sample. From a knowledge of the proportion of the count rate due to radon decay products and that due to thoron decay products it was possible, using the known counting efficiency of the equipment, to turn the measured count rate to microcuries of activity. The determination of the airborne alpha activity was similar to that for the beta determination, including the matching of the measured decay curve to a theoretical curve to obtain the proportion of radon to thoron decay products. The counting apparatus differed in that a portion of the filter paper was presented to a scintillation counter and a factor of 2 which had been determined experimentally was allowed for the absorption of the alpha particles in the filter paper itself.

The survey showed that local variations in both the alpha and beta atmospheric activity at any one survey point were so great that no one value could be assigned to the point. Meteorological conditions, such as wind speed and general atmospheric turbulence, affect the amount of the alpha and beta activity. It was found that in the West Cumberland area:

1. The atmospheric beta activity varies between 7 and $350 \times 10^{-6}$ microcuries/millilitre of air sampled with a mean value of $54 \times 10^{-6}$ microcuries/millilitre.

2. The atmospheric alpha activity varies between 6 and $630 \times 10^{-6}$ microcuries/millilitre of air sampled with a mean value of $51 \times 10^{-6}$ microcuries/millilitre.

# Atomic Energy and Meteorology

**By H. Wexler, L. Machta, D. H. Pack and F. D. White,* USA**

The development of atomic energy from its infant laboratory stage of only a few years ago to a major technological activity presents an important challenge to the ingenuity of meteorologists. Radioactivity, when confined and properly shielded, is of direct biological concern only to persons in the immediate area. It is not until it escapes or is released into the air, water or ground that it can affect living organisms at any considerable distances. Since the atmosphere is the most mobile of all geophysical media and can transport radioactivity swiftly over large areas the meteorological problems assume a particular importance in hazards analysis of atomic energy installations. Since health and safety aspects have been one of the paramount considerations in the development of atomic energy it was decided early in the program not to wait until after the release of material into the atmosphere to call on the meteorologist for aid but rather to ask him beforehand to estimate what would be the behavior of radioactivity introduced into the highly variable atmosphere. Since it is evident to the meteorologist that certain locations, because of their geographical or physical location, could result in greater concentrations of activity, it is recommended practice to evaluate the air pollution potential, or the dilution efficiency of the atmosphere at prospective locations before making a final decision on just where the plant will be located.

Thus the consideration of meteorology enters into the atomic energy industry at an early stage and should play an important role in site selection and plant design even before the architect and reactor designer put their early concepts on paper.

## DIFFUSION BY THE ATMOSPHERE

### Physical State of Effluent

It is the task of the meteorologist working with atomic energy to determine the behavior of radioactive material released into the atmosphere. This material may be in the form of a gas, or a finely divided solid with a large range of particle size. It may enter the air continually in small increments from a controlled release or from a leak. Or possibly it may be injected abruptly in relatively large quantities by a major failure of a containing structure. The material may be released at the ground with near ambient temperatures and diffuse along the surface, or it may have highly elevated temperatures and rise rapidly to several hundred meters before appreciable diffusion begins. Knowing the physical state of the material when it is released into the atmosphere, it is necessary to consider the parameters that influence the turbulent diffusion of this material through an ever increasing volume. Account must be taken of the particle size distribution of the material in order to compute the "fallout" of the radioactive material and the deposition at the ground.

### Geographical Effects

A second factor which must be considered in any diffusion evaluation is the geographical location of the source, including topography. The meteorological regimes of the middle latitudes are usually so variable that at one time they may possess wind and temperature patterns characteristic of the tropics and at other times of the polar regions. Hills and mountains superimpose wind and temperature perturbations on the general flow. Since it is usually desired to locate atomic energy installations downwind from population centers these differing patterns must be considered. Also the vertical temperature gradients will vary through wide ranges depending on the latitude, air mass, and time of the day. The diffusion of material under the steep inversion of temperature typical of air over the snow cover of northern latitudes will differ greatly from the diffusion patterns in the daytime superadiabatic lapse rates over the tropical continents. These extreme conditions are also approached by the change from typical night to day conditions.

The pattern of wind and temperature gradient will also be very much altered by the location of an area in relation to large bodies of water. The mid-continent area will have much larger variations in daily lapse rates, as well as larger seasonal variations.

Figure 1 illustrates the diurnal changes that may occur at a mid-continent station. In contrast, certain combinations of hemispheric wind and ocean currents and ocean-continent relationships may result in persistent temperature inversions—called "trade" inversions—found in some areas.

The west coast of North America from southern California southward to central Mexico, the Peruvian and Chilean coasts of South America, and the African coast from Capetown to near Sao Paulo and from Dakar to near Gibraltar are all subject to these inversions and as a result have entirely different diffusion regimes from the one illustrated at Oak Ridge National Laboratory in the mid-United States.

* US Weather Bureau.

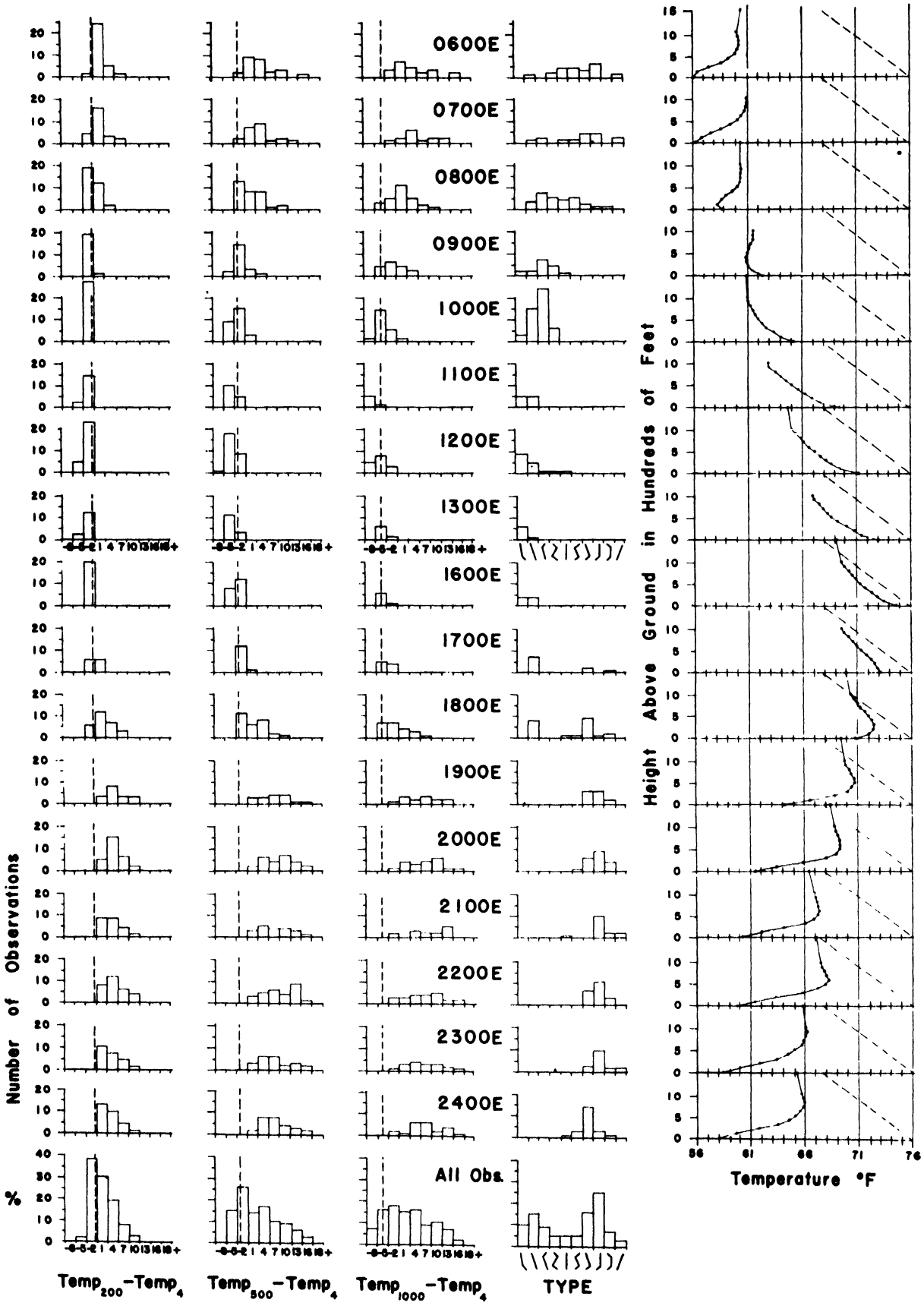



Figure 1. Temperature gradient frequencies (Oak Ridge, Tennessee)

### Topographical Effects

On a much smaller scale but no less important, are the effects on the dilution, of the terrain within 50 to 150 kilometers of the prospective installation. It is most difficult, if not impossible to make a quantitative determination of the effects of local terrain on diffusion patterns without a considerable program of meteorological measurements. However, a qualitative estimate of preferred wind patterns or anomalous eddy currents can usually be made from the geographic features and consideration of the general meteorology of the location. This may suffice for the preliminary evaluation of a site particularly where the terrain is relatively flat for considerable distances as in the middle United States. Where the topography is quite irregular even qualitative estimates may err considerably. In any event a program of meteorological measurements will be required to determine quantitative data that will permit diffusion computations of the required accuracy. Figure 2 shows the marked channeling effect of the terrain on winds near the United States National Reactor Testing Station.

### Model Studies

Before leaving the subject of terrain influences on atmospheric flow, mention should be made of the possibilities of model studies[1] as a precursor to, and perhaps eventually a substitute for, full scale meteorological measuring programs. Experiments of this type have been made in England and at New York University. The use of model techniques in meteorology is still very much in its infancy but offers great promise.

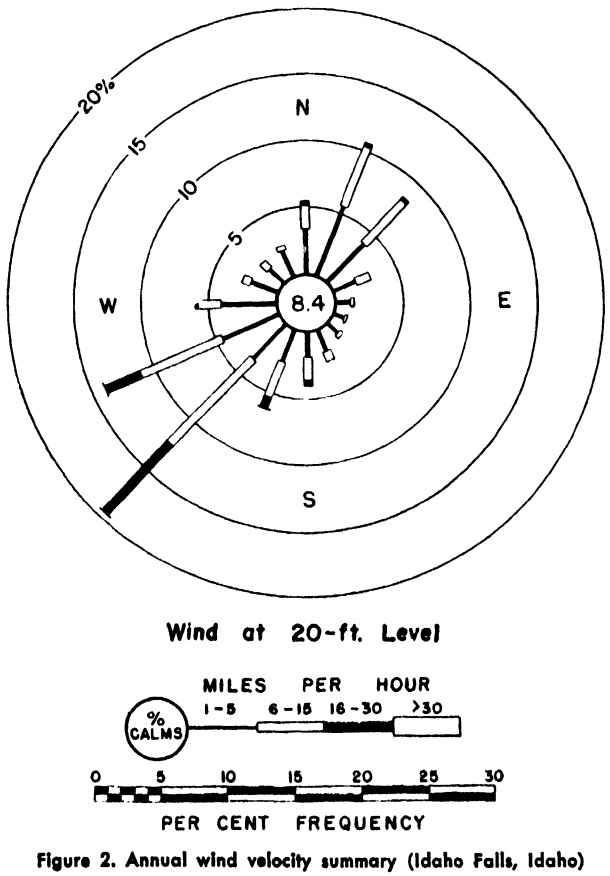



Figure 2. Annual wind velocity summary (Idaho Falls, Idaho)

## METEOROLOGICAL PARAMETERS

While geographical location and local terrain are prime factors governing the dilution efficiency of the atmosphere they are not meteorological parameters per se, nor are they amenable to measurement in terms that can be used to determine atmospheric diffusion. In the practical application of diffusion estimates to atomic energy we must postulate a minimum number of meteorological elements that can be measured routinely and that permit an adequate description of the transport and spread of material released into the atmosphere. In essence, this requires choosing parameters which themselves may be determined by several variables, but which represent the integrated action of these variables. In research investigations into turbulence or turbulent diffusion this approach would be crude indeed but at operational or commercial installations the pressure of time and the difficulty of maintaining complicated and delicate experimental apparatus precludes the measurement and consideration of such items as the flux of heat and moisture or direct determination of the vertical motions of the air. Instead it is possible to measure only the space and time gradients of horizontal wind velocity and of temperature and arrive at estimates of the diffusion activity of the atmosphere. The theoretical considerations of these relationships will be mentioned later but let us now look at these parameters qualitatively.

### Wind

Other factors being equal, dispersion of material in the atmosphere will be enhanced by high wind velocities. At higher velocities the effect of obstructions, both natural and man-made, will be increased and will result not only in vertical motions whose amplitude increases with increasing velocity, but also in increased lateral extent of turbulent eddies. The end result is not unlike the result obtained by moving the nozzle of a hose from side to side and up and down, thus distributing the material through a larger volume of air with the resulting lower concentrations. In addition where radioactivity is concerned the speed of the wind may be very important. If the half-life of the released material is long, and it is moving as a single "puff" cloud, higher wind speeds will carry it over and past any receptor in less time with resulting smaller exposures. If however the decay scheme of the material is relatively short as is the case for fission products produced by a nuclear power excursion, the higher speeds may be less favorable since the material will be carried rapidly to a receptor with less opportunity for radioactive decay to reduce the source strength. It would not be appropriate to detail the various combinations of wind velocity, radioactivity, and population dis-

tribution that might result in differing exposures. Each location must be evaluated separately on its own merit.

### Temperature Gradient

In addition to the effect of mechanical turbulence caused solely by wind speeds, thermal turbulence as represented by the vertical gradient of temperature, has at least an equal influence on the dispersal of material in the air. The well-known relationships of the density and buoyancy of a gas can be interpreted in terms of the vertical temperature gradient, and the potential vertical motions of the air estimated. Since, with the so-called "neutral" or dry adiabatic lapse rate the air has neutral buoyancy it is possible to compare actual lapse rates with this and if the decrease in temperature with height exceeds the adiabatic rate we know that vertical motions will be more vigorous. If the actual lapse rate is less than the adiabatic then vertical motions will be suppressed. It is worth commenting further on the inversion of temperature with height. Experiments have shown that under strong inversions vertical and horizontal motions are so small as to be almost negligible. Plumes of visible material have been tracked under inversion conditions for as much as 20 miles and have shown little or no dispersion with distance. The simultaneous occurrence of a situation of this type with a large release of radioactive material might result in the maintenance of high concentrations of radioactivity to large distances from the source. It should be noted however that if the half-life of the material released were short, and if the material were released at a sufficient altitude, a receptor at the ground might receive less radiation than in other circumstances. Here again each facet of the situation must be considered. It is difficult to make a categorical statement that a particular meteorological situation represents a maximum hazard without also considering non-meteorological parrameters such as the physical nature (gaseous or particles) of the effluent and its radioactive half-life.

### Precipitation

When the action of precipitation on a traveling cloud of radioactive material is considered it is almost always found that a maximum hazard can be assumed if sufficient of the radioactive material can be washed out of the traveling cloud and deposited on the ground. Consideration of previous work by numerous individuals[2-6] on the collection efficiency of precipitation does indeed show that with reasonable assumption of rainfall rates, drop sizes, and collection efficiencies high ground concentration of radioactivity may result.

## THEORETICAL DIFFUSION EQUATIONS

### Fickian Theories

We have mentioned that a necessary requirement for the application of meteorological measurements to the problems of diffusion is an appropriate formulation which will use available data and describe the changing concentration of material released into the atmosphere. Historically the earliest meteorological theory of diffusion was given independently by G. I. Taylor[7] and W. Schmidt[8] who derived the following differential equation of the problem:

$$\frac{dX}{dt} = \frac{\partial}{\partial x}\left(K_x \frac{\partial X}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial X}{\partial y}\right) + \frac{\partial}{\partial z}\left(K_z \frac{\partial X}{\partial z}\right) \tag{1}$$

where $X$ is the concentration of some quantity expressed, for example in grams per cubic centimeter; $t$ is time and $K_x$, $K_y$, and $K_z$ are coefficients of diffusion in the $x$-, $y$-, and $z$-directions.

The diffusion equation in this form is not directly applicable to computations and O. F. T. Roberts[9] gave the following equation for the concentration distribution due to the instantaneous release of a quantity, $Q$, of material released at a time $t = 0$ from a point

$$X_{(x,y,z,t)} = \frac{Q}{(4\pi x)^{3/2}(K_x K_y K_z)^{1/4}} \exp\left\{-\frac{1}{4t}\left(\frac{x^2}{K_x} + \frac{y^2}{K_y} + \frac{z^2}{K_z}\right)\right\} \tag{2}$$

where the parameters are in the conventional coordinate notation with the $z$-direction positive upward and the $x$- and $y$-directions chosen to be along and across the mean wind. It should be noted that the concentration in this formulation decreases as the $\frac{3}{2}$ power of time and that the concentration through the cloud follows a Gaussian distribution. Before these results can be applied in a working theory of diffusion the $K$-values must be determined. However, other considerations and modifications must be made to permit the $K$-theory to more nearly represent the real atmosphere. The variation of wind with height, surface roughness, and the effect of changing atmospheric stability must be included if the $K$-theory is to be useful. Calder[10], Deacon[11] and others have extended this theory to account for many of these elements. The mathematical achievements of the $K$-theory are impressive, but the practical utility is limited. A fundamental shortcoming is that the necessary $K$-values obtained by fitting observations to Equation 1 vary according to the scale of the meteorological processes acting and range from about 0.2 $cm^2$ $sec^{-1}$ for molecular diffusion to $10^{11}$ $cm^2$ $sec^{-1}$ for diffusion due to large-scale cyclonic storms.

### Statistical Theory

This circumstance led Sutton[12,13,14] to develop his "statistical" theory of turbulent diffusion, following Taylor's statistical theory of turbulence. Sutton's formulation for an instantaneous point source with non-isotropic diffusion is:

$$X_{(x,y,z,t)} = \frac{Q}{\pi^{3/2} C_x C_y C_z (\bar{u}t)^{3(2-n)/2}} \exp\left\{-(\bar{u}t)^{2-n}\left(\frac{x^2}{C_x^2} + \frac{y^2}{C_y^2} + \frac{z^2}{C_z^2}\right)\right\} \tag{3}$$

where $X$ = concentration, $Q$ = source strength (doubled to account for ground reflection), $C_x$, $C_y$, $C_z$ are "virtual diffusion coefficients" in the $x$-, $y$-, and $z$-directions respectively, $\bar{u}$ is the mean wind speed, and $n$ is a dimensionless stability parameter presumably accounting for thermal stability.

Lettau[15] and Bosanquet and Pearson[16] have also developed non-Fickian diffusion formulations which will not be discussed here.

### Choice of Equations

Each theory has its place in current meteorological practice. It may be said, arbitrarily but with some justification, that essentially in the atmosphere one faces diffusion problems on three characteristic scales. One scale, extending in length up to perhaps a kilometer, is exemplified by certain aspects of the application of chemical agents such as insecticides, where very precise information on the concentrations close to the source or near the ground is required. The second, reaching out to perhaps tens of kilometers, is that of air pollution, and of stack and reactor hazard meteorology. A third, involving horizontal distances up to continental dimensions, is important in the treatment of the diffusion of volcanic dust. For certain source types and for relatively small distances, Calder's extensions of the $K$-theory seem to be most appropriate. For the very large-scale problems, an adaptation of Robert's theory has been used. Sutton's theory, on the other hand, has been the one most generally and most easily applied to the intermediate scale and the subsequent discussion refers to applications and modifications of his work.

As can be seen from the previous discussion we owe much to our British colleagues both for leadership in this analytical approach and for having supplied many of the theoretical tools which we use.

### Application of Equations

The instantaneous point-source equation represents a fundamental solution from which, by integration with respect to one or more of the coordinate directions and time, other formulae for more complex sources may be built up. Equations for the continuous point source may be used to evaluate the emission from stacks or vents; the continuous line source formula can be used with an array of stacks or vents; and the instantaneous volume-source formulation may be used for clouds resulting from explosions. Certain geometrical properties of these formulae, namely the maximum concentration and its location, and the plume width or height are often extremely useful.

The distance of the maximum concentration from the source (represented by $d_{max}$) for an instantaneous point source is

$$d_{max} = \left(\frac{2h^2}{3C^2}\right)^{1/(2-n)} \tag{4}$$

and for a continuous point source is

$$d_{max} = \left(\frac{h^2}{C^2}\right)^{1/(2-n)} \tag{5}$$

while the maximum concentration at the point $d_{max}$ is, for the instantaneous point source

$$X_{max} = \frac{2Q}{(2/3\,e\pi)^{3/2}h^3} \tag{6}$$

and for the continuous point source

$$X_{max} = \frac{2Q}{e\pi\bar{u}h^2} \tag{7}$$

The cloud width, $2Y_0$, and the cloud height $Z_0$ are easily obtained from

$$2Y_0 = 2\left(ln\frac{100}{P}\right)^{1/2} C_y(x)^{(2-n)/2} \tag{8}$$

$$Z_0 = \left(ln\frac{100}{P}\right)^{1/2} C_z(x)^{(2-n)/2} \tag{9}$$

where $p$ is any desired percentage of the axial concentration.

However these formulae together with the parameters obtained by Sutton were developed under conditions which may differ greatly from those where the computations must be made. First and foremost it must be remembered that the parameters were based on three minute-average concentrations. Secondly, the terrain over which the experiment was conducted was relatively smooth. And third, the equations were developed for neutral temperature gradient conditions. If these factors are always considered the equations may be used with a fair degree of success, and the meteorological uncertainties are seldom greater than other factors such as the rate and amount of the effluent released.

It has been shown by Holland[17] that the diffusion coefficients may be modified experimentally to correspond to the site over which gaseous effluent is released. Figures 3 and 4 illustrate some of these data obtained at the Oak Ridge National Laboratory.

Finally a group of special results and extensions involving corrections for radioactive decay, ground surface depositions, diffusion in very stable atmospheres can be deduced. All of these modifications or extensions have their uses in application to atomic energy problems. Airborne concentrations are required for the evaluation of the external gamma and beta hazards from a moving cloud and for the computation of internal exposure from ingestion or inhalation of this cloud. Ground deposition from fallout and precipitation scavenging may result in external gamma and beta dosages and knowledge of the plume width and height would be required for estimation of the lateral area of hazard.

### Limitation of Present Theory

While the present theoretical approaches give reasonable estimates of the radiation to be expected from a moving cloud of radioactive debris they have limitations. Comparison of theory with careful ob-

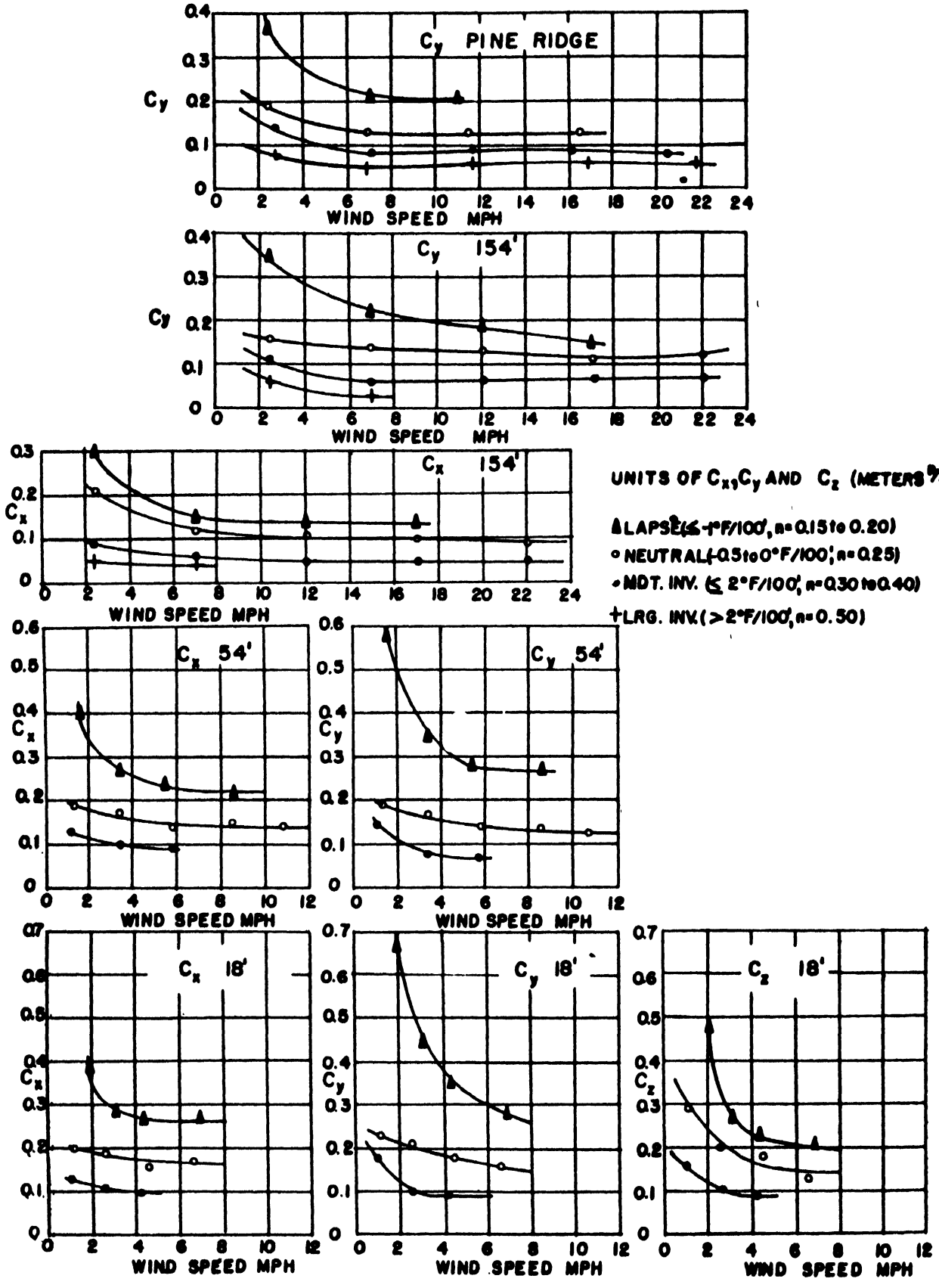



Figure 3. Observed $C_x$, $C_y$, and $C_z$ vs wind speed and stability

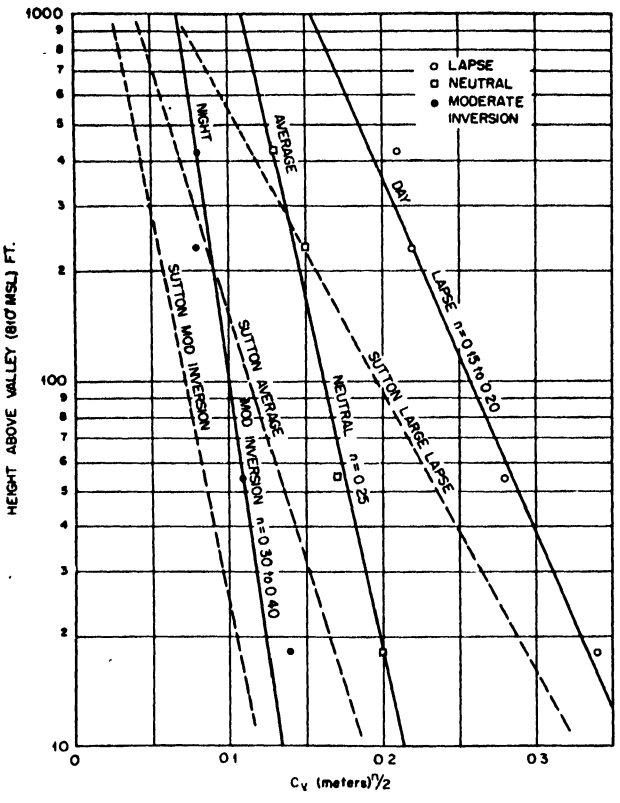



Figure 4. Variation of $C_y$ with height

servations indicates that the computed values are within an order of magnitude about 95 per cent of the time. For conditions of neutral stability, the condition under which the majority of formulae were developed, an analysis by D. J. Holland[18] shows that the accuracy improves by a factor between two and five.

None of the present methods permit us to evaluate turbulence directly by instrument, only by the indirect effect on stationary meteorological equipment, that is the time variation of wind and temperature gradient at fixed locations. The various formulae have been extended to unstable and stable atmospheric conditions by expedients which leave much to be desired. This is particularly true of stable atmospheric situations where the plume concentrations aloft may be several orders of magnitude higher than predicted, while the ground concentrations are essentially zero. This situation is one of great concern since under the proper conditions radioactivity may travel for tens of kilometers with little decrease in concentration.

Many of the gaps in our knowledge can be filled if properly precise field experiments in diffusion can be obtained. Measurements of air and ground concentrations at various distances from the point of release at times when meteorological conditions are accurately known would permit the determination of the parameters of the equations under varying conditions and thus enable one to use them as interpolation formula. Measurements at various locations would give insight into the effect of varying terrain, and indeed this has been attempted for many of the atomic energy sites in the United States. Even more important than refining the present theories is the fact that from the analysis of appropriate data we may logically expect to develop diffusion theories that will answer many of the problems that plague us today.

## RADIOACTIVE METEOROLOGICAL TRACERS

### Suitability of Radioactive Tracers

Even without a complete theoretical explanation of the spread of material through the atmosphere the meteorologist is obtaining a better understanding of the action of the atmosphere by the use of radioactivity as a tracer.

Radioactivity may be used as a means of identifying tracer substances for meteorological experiments. Many of those which man may produce occur in nature in such small amounts as to reduce the background problems. Specific identification of radioisotopes is further enhanced through half-life, type and strength of the radiation discrimination. Instruments for measuring the radioactivity are capable of workable accuracy through very wide ranges of activities. Further, by proper selection of the isotope one may avoid chemical interaction with the environment and one may select the scale and nature of the meteorological phenomena to be studied.

### Artificial Radioisotopes

Radioactive tracers may be conveniently classed into two categories: man-made and natural.

Man-made radioisotopes as meteorological tracers are especially well suited for long-range atmospheric travel in comparison with most other available tagging agents. In principle one may design an experiment for the safe release of large quantities of radioactive materials. Man-made radioactive tracers may be either in a particulate or gaseous state and, in fact, the simultaneous release of both states will provide information concerning the separation of particles of various characteristics from the air mass with which they were initially embedded. Size discrimination between particles may be simplified by assigning a different type of radiation or life to each size range. If desired some substances may be made radioactive after sampling so as to eliminate any hazard from an initial intense source of radioactivity.

The two main phenomena which can be studied by such a program are meteorological trajectory verification and the intensity of atmospheric diffusion. Release of the tracer in regions of special meteorological concern such as jet streams, calm winds, monsoon areas and so forth may reveal clues to many unsolved problems.

The main difficulties in such a tracer experiment would be financial. The cost of the huge quantities of necessary isotopes will be sizeable. The sampling, to be effective, must be performed in the free air requiring expensive aircraft observations or as yet undeveloped balloon-borne equipment.

Since the background of tritium is relatively small, it represents a logical choice for a tracer to follow the hydrological cycle. For example, by introducing tritium into the atmosphere, one may determine whether the given air mass contributed to the formation of precipitation. One may also use radioactive forms of many of the chemicals currently used to stimulate artificial precipitation as further and easier identification of the substances in the precipitation elements.

### Natural Radioisotopes

Naturally produced tracers may also be either gaseous or particulate. One may further distinguish between those which originate from the earth's surface and from cosmic ray showers.

Radon and thoron and their daughter products are produced at the earth's surface—primarily over land masses. The short half-life of the measured thoron daughter (Thorium B—10.6 hours) limits its value since it is largely decayed before it moves through a meteorologically significant distance. It probably has a certain value in micrometeorological studies. Radon (3.8 days) is more suited to weather investigations.

Free air measurements of radon or its daughter products are not numerous. In 1949, two aircraft flights to measure radon were made in the United States. These are shown in Figs. 5 and 6 as a plot of the relative activity per unit mass against altitude. The Ohio curve exhibits the expected vertical distribution from a ground source while the California profile, taken offshore, illustrates the absence of a radon source near the water. The higher values aloft on the California profile can, it is believed, be traced back to air coming from Asia. The relative high wind speeds across the Pacific prevented the decay from depleting the values below that observed. Proper interpretation of many such profiles would yield a reliable estimate of the intensity of vertical atmospheric mixing.

Radioisotopes originating from the interaction of cosmic rays with the atmosphere which have been noted as being useful to meteorology are tritium, $Be^7$ and $A^{37}$. Libby[19] has reported the values of many precipitation samples and the values of water from lakes, rivers and oceans. His interpretation in terms of the time since the water originated from the ocean where it is diluted with old ocean water yields interesting information concerning the hydrologic cycle. Arnold and Al-Salik[20] reported on the natural occurrence of $Be^7$ (half-life = 53 days) and its possible meaning for the meteorologist. Professor Lyle Borst of New York University called our attention several years ago to the possibility of using $A^{37}$ (half-life = 34 days) as a tracer for vertical diffusion experiments.

In general, the drawbacks in the use of natural radioactive tracers formed by cosmic rays is the small quantities in which they are present in the atmosphere. Extensive chemical refinement is necessary. Further, they do not originate at a fixed height but through a deep layer being peaked near 30,000 feet. Until the source strength at each altitude is known with some confidence, the interpretation of ground values in terms of vertical mixing is uncertain.

## APPLICATION OF METEOROLOGY TO ATOMIC ENERGY

### Site Selection

When the development of a nuclear energy installation enters the planning stage the question of choosing an appropriate site is of immediate concern. Examination of meteorological records and consideration of terrain effects will permit first approximations of the probable patterns of air flow. At a few locations temperature gradient measurements will be available and the variation of the vertical temperature structure can be examined. At many locations however, no meteorological data will be readily available. Still however a survey of meteorological records from surrounding locations together with a few representative measurements at the actual site may serve to permit a determination as to whether the dispersion of atmospheric contaminants is favored or hindered by the location, plus informa-

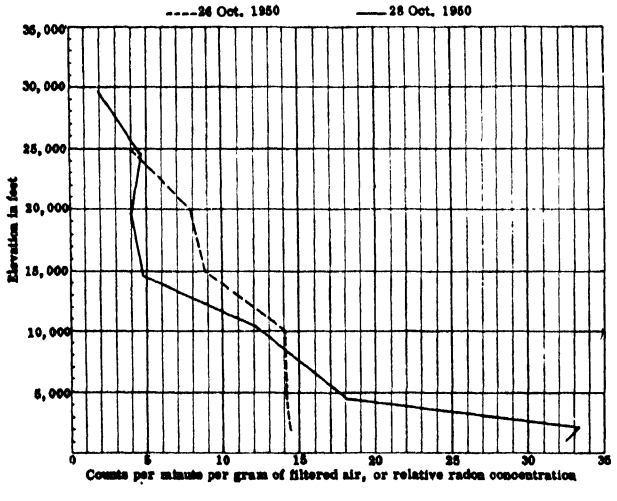



Figure 5. Vertical distribution of radon (Ohio)

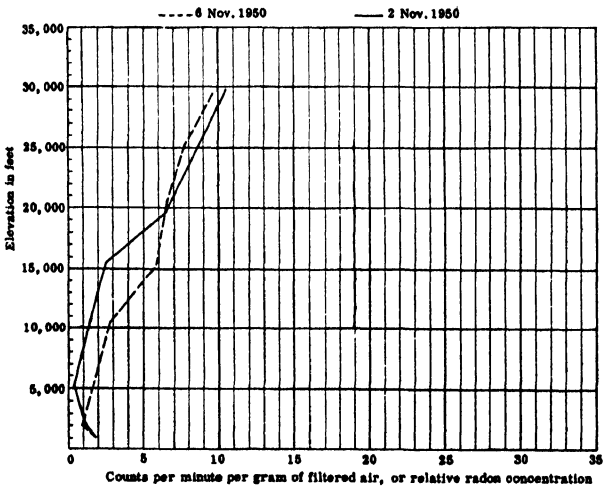



Figure 6. Vertical distribution of radon (California)

tion on whether the most likely route of travel of any released material will be towards areas of population. Often indirect data may be used to estimate the existence of frequent inversions. Persistent low visibility, numerous ground frosts, and even a distinctive ecology may indicate that a location may frequently be subject to stagnant conditions. These first estimates, although likely to be quite general, will usually serve to separate several possible sites into those where the meteorology is favorable to dispersion of airborne material, and others where diffusion may more often be restricted.

An actual site survey was recently made for a nuclear reactor to produce commercial power at Shippingport, Pennsylvania. This location is along the Ohio River and about 30 miles from Pittsburgh, Pennsylvania. The nearest meteorological station is more than 12 miles distant. Examination of the area indicated that the gross features of the precipitation regime, the temperature gradients above 500 to 1000 feet, and the general wind flow could be obtained from existing records and this was done. However, the valley location and the irregularity of the terrain indicated that the wind patterns in the immediate vicinity of the site and the temperature gradients in the lowest 500 feet would differ markedly from existing records which are made on a plateau several hundred feet higher than the reactor site. A meteorological program to measure wind, temperature gradient and humidity has been inaugurated and the first observations do indeed show significant local differences in wind and temperature gradient at the site. Advantage was taken of the irregular terrain in obtaining temperature gradients by placing one meteorological station in the valley and another on a nearby hill 395 feet above the valley. Thus the temperature gradient is obtained over a considerable height without the problem of erecting a meteorological tower. A program of collection and summarization of meteorological data will be continued until sufficient data is available to describe the micrometeorological patterns for this location.

### Meteorological Observations

After a definite location has been chosen more specific information can be obtained by the installation of a program of meteorological measurements at the site. This program will usually consist of records of wind temperature and precipitation and, if required, temperature lapse rates. The scope of such a site meteorological survey will depend on several factors, namely, the magnitude of the potential pollution sources, the size of the site, and the roughness of the terrain. A large site with many nuclear installations, or one in very irregular terrain, will require more elaborate meteorological programs to define the diffusion patterns than would a single laboratory located in a large level plain.

### Application of Data

Having instituted such a program it is necessary to convert the meteorological data into a form that can be applied to the nuclear operations. It has been found that joint correlations of wind direction and stability, of wind direction and precipitation, and data on the frequency of the durations of inversions can all be used to estimate the times that are favorable for the release of gaseous wastes. Data showing the diurnal variation in stability will provide additional information for the most favorable time for releasing material to the atmosphere. As an example we may take the problem of a chemical processing plant that generates quantities of waste gases while dissolving radioactive materials. These gases must be disposed of. One successful method of release has been to determine from both climatological data and weather forecasts when the atmospheric conditions will enhance diffusion. The dissolving process is then timed so that the maximum gaseous release occurs during favorable conditions. Another application of meteorology to this same type of process is the determination of the stack heights that will be required to obtain sufficient atmospheric dilution. A study of average inversion heights may show that they are mostly very shallow and that a two or three hundred foot stack will vent gases above the inversion a large percentage of the time, thus minimizing interruptions due to unfavorable weather.

Another application has been to compute radiation dosages on the basis of typical conditions of wind and stability for a unit emission of radioactivity. Isolines of resulting ground concentrations are then drawn. From these and knowledge of the amount of radioactivity to be released a good idea of the resulting dosages can be obtained.

Although the application of the diffusion equations to the release of material from stacks and vents is the most common, it may often be necessary to provide information on the dispersion of gases due to the evaporation of liquid wastes. In this instance knowledge of rainfall and evaporation records and consideration of the evaporating surface as a finite volume source are required to give satisfactory answers to this problem.

## ANALYSIS OF ENVIRONMENTAL HAZARDS FROM REACTORS

### Postulated Accidents

One of the most important applications of meteorology to atomic energy has been the evaluation of environmental hazards resulting from nuclear reactors. A nuclear reactor will develop highly radioactive fission products in proportion to its operating power level and time of operation. These products, if released into the air in sufficient quantity, may result in concentrations of radioactivity sufficient to injure living organisms. It is necessary therefore to examine each proposed nuclear reactor to see if a hazard could result in the operation of a specific reactor at a specific location. This individual approach has been made necessary not only because of the desire to minimize any possible hazards but also because the features of reactor behavior were at first

not too well known. With proven reactor types such determinations become much easier.

The current approach is to examine the reactor from the nuclear and chemical aspects and to see if there are any plausible mechanisms which may result in an accident. The magnitude of the accident is then examined to see if it would result in the contained fission products being released. If reasonable postulation shows that an accident releasing fission products may occur, then this accident is taken as the starting point for the meteorological portion of the hazard analysis. An assumption is made of the proportion of fission products released and of the initial heat energy resulting from the event. It is then necessary to compute what action this available heat will have on the cloud of fission products. It is usually assumed that this excess heat is distributed uniformly through the initial volume of air within the original container, which may have been an ordinary building or perhaps a reinforced barrier to contain the fission products. If the secondary barrier can withstand the heat and pressures of the incident well and good. If it cannot, then it must be assumed that the heated mixture of air, fission products and perhaps other gaseous materials will, upon escaping from the container, rise until expansion and entrainment of cooler air halt the rise.

### Height of Rise of Reactor Clouds

The logical determination of the height of rise is important since the increase in attenuation of the gamma radiation with increasing distance from the ground will rapidly reduce the radiation intensity. The possible inhalation hazard near the release is reduced to negligible proportions by any appreciable height of rise and in addition the added distance and time required for an elevated cloud to diffuse to the ground will result in lower ground concentrations at all distances.

Sutton[21] uses his theory for diffusion from a point source for computation of the diffusion of heat in the cloud as it rises and determines the height at which the cloud is no longer warmer than its environment. The formula is

$$H = \left[\frac{2\,(3m + 2p)\,Q}{9C_p\rho\pi^{3/2}\,C^3 a}\right]^{1/(p\,+\,3m/2)} \qquad (10)$$

where $m = 2-n$, $\rho$ is air density, $Q$ represents the amount of heat released in the explosion, $C_p$ is the specific heat at constant pressure for the gases of the cloud, $C$ is the Sutton generalized coefficient of diffusion and $a$ and $p$ are defined below. Sutton suggests that $C$ might be about 0.45 (meters$^{1/8}$) and $m$ about $\frac{1}{4}$, values which apply to ordinary atmospheric turbulence for a cloud which moves horizontally. The applicability of the same turbulence factors for a cloud which moves relative to the air is questionable. The numbers $a$ and $p$ are derived from the relationship, $\theta = \theta_0 - az^p$ where $\theta$ stands for potential temperature, $z$ for height and subscript "0" denotes the potential temperature at the place where $z = 0$. Figure 7 is a graph of Equation 10 for the conditions listed above, and $p = 1$, $a = 10^{-3}$°C/meter.

Machta[22] has developed a formula for the maximum rise of a gaseous cloud based on a constant rate of entrainment of environmental air. It is written

$$H = \frac{1}{\frac{1}{M}\frac{\partial M}{\partial z}} \ln\left\{\frac{\frac{1}{M}\frac{\partial M}{\partial z}}{\frac{\partial \theta'}{\partial z}}\left[(\Delta\theta)_0 + \frac{\frac{\partial \theta'}{\partial z}}{\frac{1}{M}\frac{\partial M}{\partial z}}\right]\right\} \qquad (11)$$

where $H$ is the maximum height of the cloud, $M$ stands for the mass of the cloud and $(1/M)\partial M/\partial z$ is the constant percentage rate of entrainment per unit increase in height, $(\Delta\theta)_0$ is the initial excess in potential temperature of the cloud over the environment, and $\partial\theta'/\partial z$ is the lapse rate of potential temperature.

The atmosphere plays a role in two terms; first, in the rate of entrainment (which for cumulus clouds is estimated to be about $0.5 \times 10^{-5}$ cm$^{-1}$ and which is greater with increased turbulence) and second, in the potential temperature lapse rate of the air. For a standard atmosphere, this lapse rate is about 3.5°C per kilometer and for an isothermal atmosphere in 10°C per kilometer. Equation 11 also is graphed in Fig. 7. A standard atmosphere is assumed and the constant entrainment rate is used that was given above.

Machta's formula gives a rise of several kilometers for cloud temperatures only a few hundred degrees warmer than the environment, and therefore may be less applicable to clouds from low-order explosions. It is easily seen that cloud size is an important factor. Machta's formula would be more applicable to a very large cloud (say as large as an average cumulus congestus cloud, whose bases may be some three to five miles across) in which a temperature excess of 100°C would represent an enormous amount of heat energy. Also the entrainment rate assumed would be more likely to hold for a large cloud.

One may criticize these formulae as being very crude, but then the initial cloud temperature (or heat release) will not be accurately known in an actual

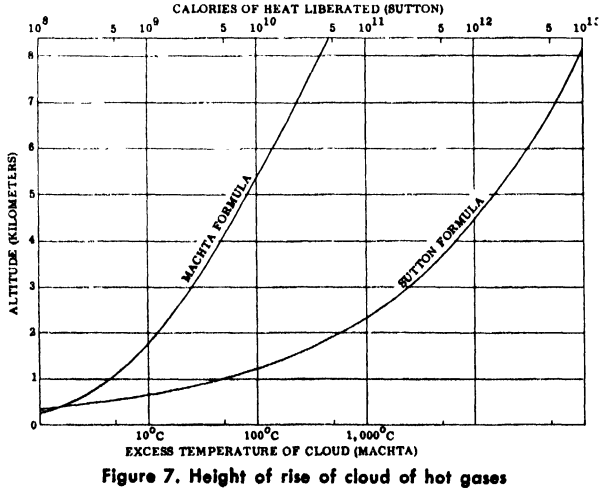



Figure 7. Height of rise of cloud of hot gases

explosion. The formulae may be used to obtain an order of magnitude approximation of cloud rise, and perhaps a qualitative notion of the sense and magnitude of the meteorological factors.

The two theories are similar in that they deal with a gaseous cloud which rises because of its temperature excess over the environment, and the cloud cools by mixing with the environment air and by adiabatic expansion. In both theories, cooling due to radiation is omitted—a factor which may be highly important in the first few seconds after an explosion. Further, the density of the cloud is undoubtedly affected by the presence of solid matter.

Additional factors which may influence strongly the height of rise, but which also cannot be dealt with mathematically, are initial cloud shape, cloud circulation, and the effects of inertia. For example, in a sharp explosion smoke clouds or puffs may be ejected as smoke rings, or toroidal circulations, that may rise higher than if only buoyancy forces were operating. Having estimated a height of rise it is also necessary to determine the final volume of the cloud at its stabilization point. This may be done conveniently by the gas law.

### Finite Volume Sources

In order to apply the Sutton "point source" equations to this finite volume Holland[23] has suggested the idea of a "virtual point source." This is an imaginary source located upward of the real source just far enough to produce the required volume source at the actual point of origin. By adding the virtual source distance to each actual distance downwind from the real source the required concentrations may be obtained. The initial distribution of material in a volume source obtained in this manner is of course Gaussian. Real distributions in a volume source may be neither uniform nor Gaussian. This problem has been studied by Gifford[24] and his results may be applied if further refinement is desired.

Not all actual or postulated reactor incidents will result in an energy release sufficient to breach the building and immediately release the fission products. A lesser event may only crack the reactor vessel and the fission products leak out slowly. In this instance the source is essentially a continuous one and the appropriate formulation must be used. Radioactive decay assumes considerable importance in this case, even for the longer lived products.

### Radiation Exposure

After the radioactivity reaches the atmosphere it is usually desirable to examine the hazards that may result from varying meteorological conditions. These will not only include variations in wind direction and speed so that the travel time to populated areas is minimized but also the concentrations which may result from diffusion in stable and unstable conditions. Additional factors are introduced when the irradiation processes are considered. A receptor may receive beta radiation from a traveling cloud, by direct fallout on the skin, or by inhalation. Gamma irradiation may result from the traveling cloud or from deposition on the ground or from inhalation. The beta, gamma deposition and beta and gamma inhalation hazards are essentially functions of the local concentration of activity and are easily obtained from the diffusion equations. Gamma irradiation from a traveling cloud is more complicated, involving attenuation, multiple scattering buildup and integration of activity over a considerable volume of space. A most useful solution to this problem has been obtained by J. Z. Holland and is included elsewhere in these present Proceedings.

### Deposition by "Washout"

The "washout" of radioactivity by precipitation has been mentioned previously as usually constituting the major hazard. Two treatments are available to obtain quantitative estimates of radiation dosages due to this mechanism. A. C. Chamberlain[25] has adapted Sutton's equations and introduced a factor representing the proportion of the material removed and deposited by precipitation. This modification may be maximized for each distance and the resulting equation is

$$\omega_R = \frac{Q}{e\pi^{1/2} C_y x^{(2-n)/2}} \tag{12}$$

where $Q$ = source strength, $\omega_R$ = maximum washout deposition, $C_y$ = lateral diffusion coefficient, $x$ = distance from source, and $n$ = stability parameter.

For the limiting case of instantaneous deposition of an entire cloud or plume of airborne material such as might occur in a sudden heavy rain shower, Holland[26] gives these formulae:

Instantaneous point source:

$$\text{Deposition} = \frac{Q}{\pi C^2 (\bar{u}t)^{2-n}} \tag{13}$$

Continuous point source:

$$\text{Deposition} = \frac{Q}{(2\pi)^{1/2} \bar{u} x^{(2-n)/2}} \tag{14}$$

where $C$ = isotropic diffusion coefficient, $\bar{u}$ = mean wind speed in the $x$-direction, and other symbols have their usual meaning.

## CONCLUSIONS

It can be seen that the most vigorous nuclear incident may not result in the highest possible ground radiation dosages since the cloud of debris may rise enough to reduce ground effects. In fact, it is almost impossible to determine what combination of all the factors, nuclear, meteorological, population movement, etc. would result in the maximum hazard. It is usually wise to consider several pessimistic combinations and discuss them with candor.

The application of meteorology to atomic energy begins in the first phases of site selection and continues through the operational evaluation of the devices at this site. Meteorology can provide data to aid

in planning routine operations, to insure safety of potentially hazardous processes and give post facto assistance if radioactivity is released into the air.

Meteorological information is being successfully used in the United States to promote the safe, economical and expeditious introduction of atomic energy into everyday use.

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# Radioactivity in Rainwater and the Air Observed in Japan, 1954-1955

**By Yasuo Miyake,* Japan**

## THE RADIOACTIVITY IN RAIN-WATER IN SUMMER, 1954[1]

M. Kamada at Kagoshima University, situated at the southernmost part of Japan, detected first the artificial radioactivity in rain-water on 14 May (0.02-$\mu$c/l) and on 16 May (0.08 $\mu$c/l). Thereafter, more or less, artificial radioactive rains began to fall incessantly all over the Japanese Islands throughout the summer rainy season.

The maximum activity was 1 $\mu$c/l observed by members of Kyoto University on 16 May.[2] F. Yamazaki at the Scientific Research Institute, Tokyo, also measured 0.16 $\mu$c/l at Tokyo on 17 May.

The presence of fission products was also confirmed in several laboratories. The radioactive nuclides identified in rain-water were $Ba^{140}$, $La^{140}$, $Sr^{89}$, $Zr^{95}$, $Nb^{95}$, $Y^{91}$, $I^{131}$, $I^{132}$, $Te^{129m}$ and a group of rare-earth elements. Y. Yokoyama at Tokyo University assumed the presence of fairly larger amount of $Np^{239}$ (45%) and $U^{237}$ (20%) in rain-water precipitated on 17 May in Tokyo area which might be induced from $U^{238}$. It is to be noticed that the composition of fission products in rain-water seems to vary secondly in a comparatively wide range as decay curves are considerably different for each rainfall.

Members at the Central Meteorological Observatory studied the air trajectory at 700 mb and suggested that air which had been over Bikini Atoll came near the Japanese Islands in a week passing by the Philippine Islands and Formosa (Fig. 1).

On the other hand, from the anomalous vibrations recorded on barographs at many weather stations in Japan, it could be supposed that the tests would have been done on 1 March, 27 March, 26 April and 5 May; thus, radioactivities in rain-water in Japan since mid-May seem mainly due to the last experiment.

Radioactive dust particles were also collected during the 14th to 18th of May by T. Shiokawa at Shizuoka. He collected the airborne dust on a sheet of paper (30 × 30 $cm^2$) coated with a thin film of vaseline. The sheets were ignited afterward and radioactivity measurements as well as chemical analysis were performed.

The method adopted for measuring the artificial radioactivity is as follows: Rain-water collected on a polyvinyl sheet or in a vessel made of plastic is evaporated in a planchet made of stainless steel. To avoid the effect of the natural radioactivity due to RaB and RaC, etc., counting is done after 5–6 hours with a $\beta$-ray G-M counter keeping the distance between a mica window and a bottom of a planchet of about 10 mm. As to the reference activity, a standard ($Sr^{90}$ + $Y^{90}$) source supervised by the Special Committee in Science Council of Japan is used.

Some of the results of observation from the beginning of May to the end of December, 1954, are given in Fig. 2. The ordinate in the figure indicates $\beta$-ray counts per minute per litre of water brought about by the artificial radioactivity in a logarithmic scale. The value of counts in Fig. 2 is not corrected. However, we can see relative changes and time variations.

The general trends in the radioactivity in rain-water are observable at every place, which is probably due to the alteration of the air mass. It is interesting to note that the radioactivity in rain-water was always stronger on the Pacific side of Japan than on the side of the Japan Sea during the period from May to the middle of September. The intensity in the latter area was not more than 4 m$\mu$c/l. Such geographical distribution of activity suggests that the source of radioactive materials was located in the Pacific Ocean.

The active material is concentrated at the beginning of rainfall like other chemical constituents as well as post-emanation elements. This means that the artificial radioactive particles are being suspended not only in the stratosphere but also in the troposphere and they are washed down by water drops. As a matter of fact, the present author detected by means

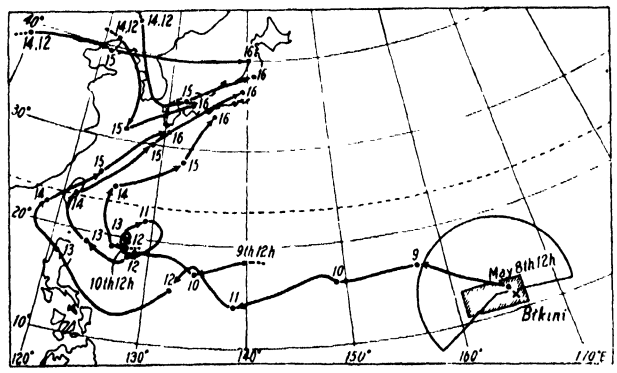



Figure 1. Trajectories of the air in the Pacific, May, 1954

* Meteorological Research Institute, Tokyo.

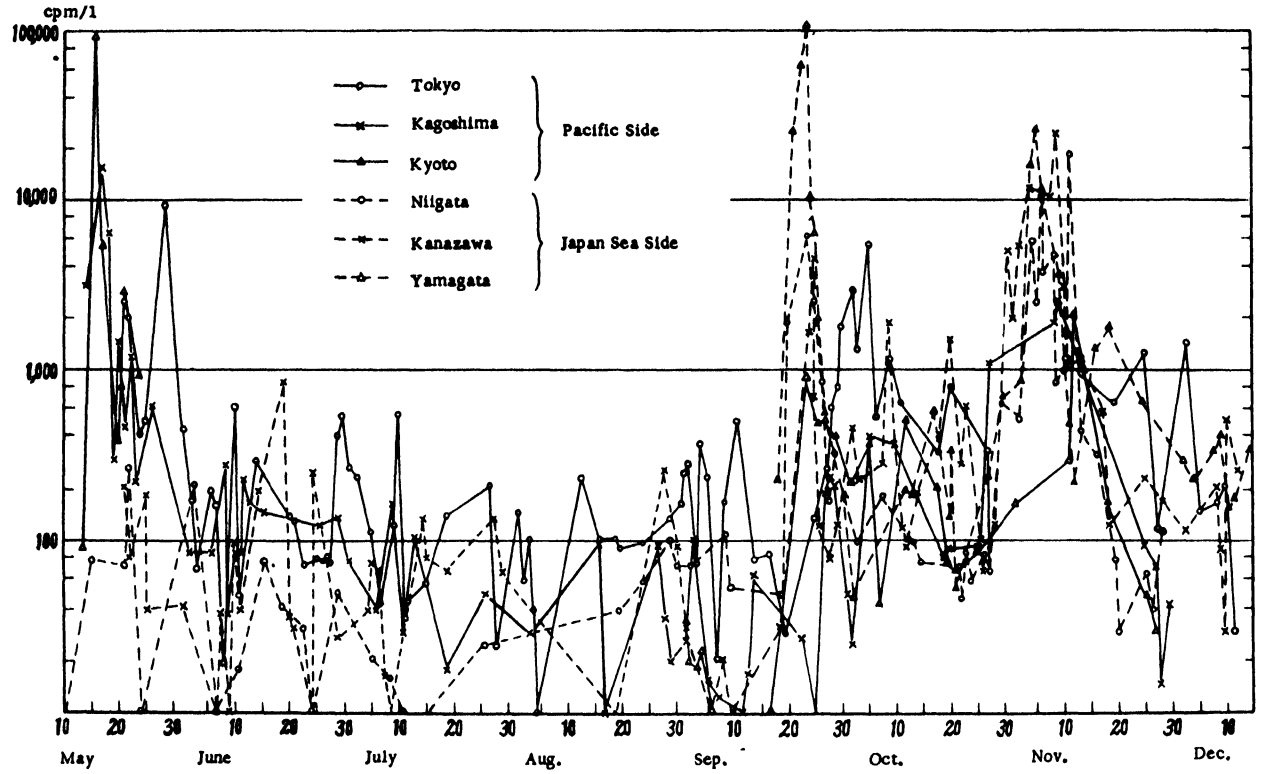



Figure 2. Artificial radioactivities observed in Japan, May–December, 1954

of the impinging dust collector on an aeroplane an activity of ~0.8 to 2.0 × $10^{-18}$ curie/cm³ on an average in the air from 1000 m to 3000 m above the ground in Tokyo and its surroundings (3 August).

## THE RADIOACTIVITY IN RAIN-WATER FROM SEPTEMBER TO NOVEMBER, 1954

At about midnight of 18 September, 1954, a typhoon (No. 14, 1954) ran away toward the Pacific Ocean after attacking Japan. In the wake of the typhoon a colder and less moist air flowed in from the north (Fig. 3). Just at the time, new activity of artificial origin was detected in rain-water by H. Watanabe at Niigata University and S. Suzuki at Hirosaki University, both situated along the Japan Sea coast in the northern part of the Main Island.

It seems that the rain which had a higher level of activity fell mostly along the cold front as shown in the weather chart (Fig. 4). From 22 to 24 September, the activity increased rapidly spreading over a larger area and finally the activity as strong as 0.3 μc/l was counted on 22 September by T. Terazaki at Yamagata University. The activity decreased when the warmer air from the south flowed in accompanied by the Typhoon No. 15 (1954) on 25 September and increased again at the beginning of October along with the incoming colder air mass from the North (Fig. 3).

At the end of October and the beginning of November, another new activity, almost comparable to those in May and September, visited Japan again from the North (Fig. 2).

As in the case of September, it was recognized at first at only a few places in the northeastern part, especially along the Japan Sea coast. But, on the 4th–5th of November, peaks in the activity appeared at many places. For example, T. Kiba of Kanazawa Univ. detected the activity of 36 mμc/l and 72 mμc/l respectively on 4 and 9 November. The activity of the order of 10 μc/l was measured also at many other stations during the period from the 4th to about the 10th of November.

At Tokyo there was no rain at that time. However, the radioactivity in falling dust of about 5 mμc/m²/-day was observed as the value for 24 hours from the 4th to the 5th of the month. In the same period K. Ito and N. Yano at our institute also measured the activity of the order $10^{-12}$–$10^{-10}$ curie/m³ in the air near the ground by means of the electric precipitator.

The dates of explosion were estimated respectively on the 13th–16th of September and the 28th–30th of October, assuming that the gross activity decreases proportionally to the negative power of about 1–2 of the time. The sites of explosion were unknown, but in consideration of air trajectories in the upper layer which extended from Siberia to Japan (Figs. 5 and 6), and also of the flow rate of airborne dust and the results of chemical analysis, it might be supposed that the radioactivities were caused by the tests which would have taken place somewhere in USSR as reported by Tass on 16 September, assuming that there were no other source of airborne activities.

Presence of fission materials was also confirmed in both cases in autumn by several researchers. It is

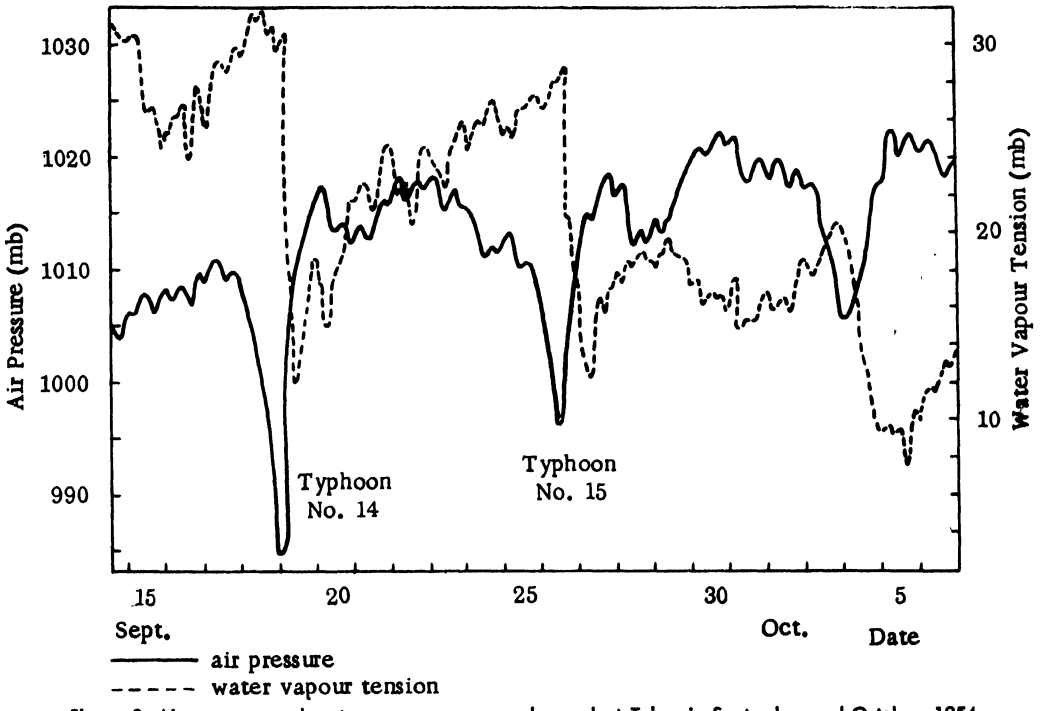



Figure 3. Air pressure and water vapour pressure observed at Tokyo in September and October, 1954

to be noticed that as in the case of rainfalls in May, S. Koyama assumed the presence of $Np^{239}$ and $U^{237}$ in rain-water precipitated on 24 September.

Contrary to the case in May, the activity in rain or snowfall was stronger along the coast of the Japan Sea than that on the Pacific in autumn. This means that the source of the activity lay north of Japan.

## THE RADIOACTIVITY IN RAIN-WATER IN SPRING, 1955

Since the 5th of March, 1955, the radioactivities which might probably be attributed to Nevada experiment, assuming that it was the only source, were detected several times both in rain-water and the air at Tokyo and other stations. They were presumably due to the experiments performed respectively in February and March, 1955. In these cases, dust would have been transferred by the westerly winds in the upper air.

The highest activity in rain-water was of the order of 3–4 mμc/l. E. Tajima at St. Paul's University, Tokyo, confirmed the radioactivity in the air near the ground by means of the filtration method. The maximum value of about $2 \times 10^{-12}$ curie/$m^3$ observed on 12 April was probably attributable to the experiment at the end of March. J. Nemoto found the good correlation between sites of the centre of anti-cyclones moving from the north (mean speed 14 m/sec) and variation in activity of the air observed by E. Tajima. According to Nemoto the activity of the air at Tokyo increases when the centre of high is located in the area above the Japan Sea or of the northeastern coast of the Pacific side.

It is interesting to note that comparatively dense, radioactive dust containing fragments of quartz, calcite, albite, clay mineral, etc. fell on several places in spring, among which those fell on the 13th of April at Asahikawa, Hokkaido Island were strongest (2660 cpm/gm as the value on 22 April). It is supposed from the decay curve that the activity might also be attributed to the experiment made some time around 29 March.

## DISCUSSIONS

The maximum activity of artificial radioactivity in rain-water measured in Japan in 1954 was 1 μc/l

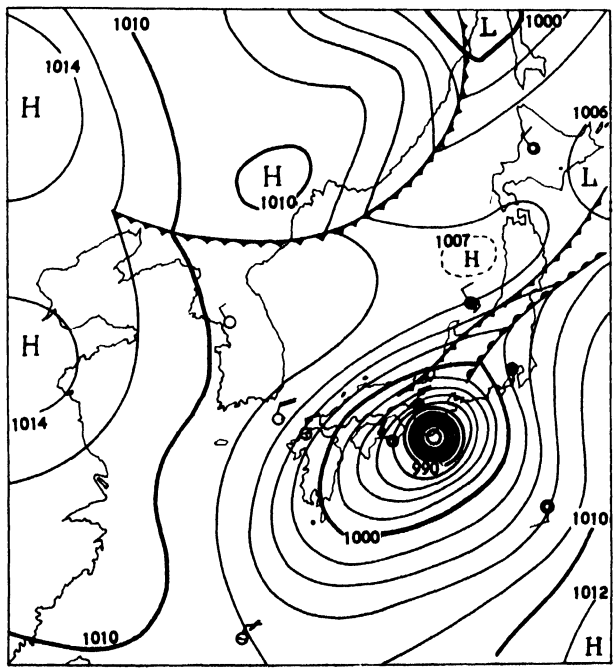



Figure 4. Weather chart in the neighbourhood of Japan at 16 h, on September 18th, 1954

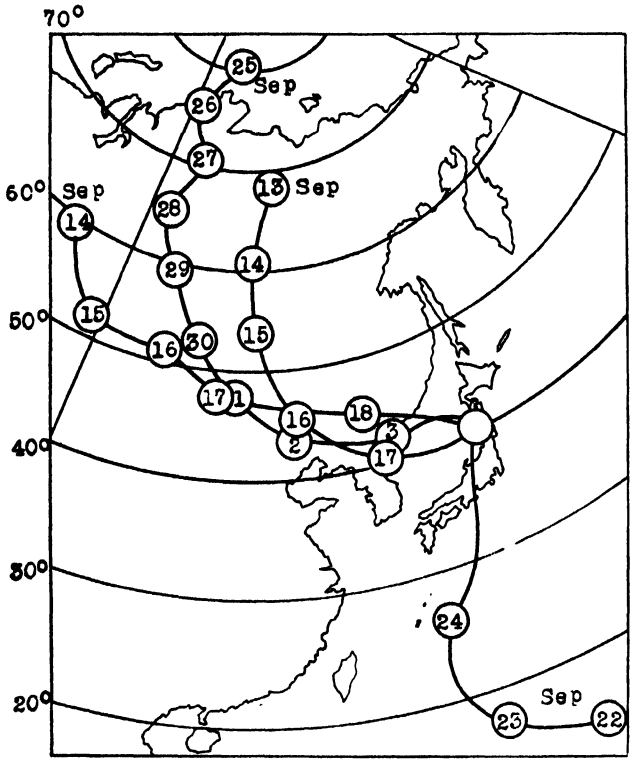



Figure 5. Air trajectories at 700 mb during the period from September 14th to October 4th, 1954

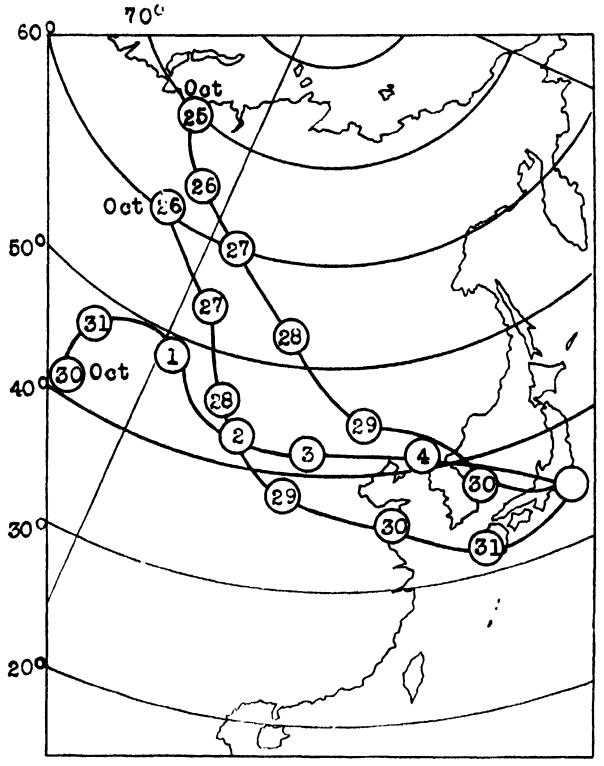



Figure 6. Air trajectories at 700 mb during the period from October 25th to November 5th, 1954

detected at Kyoto on 16 May.[1] The second was 0.3 $\mu$c/l found at Yamagata on 22 September.

In April, 1953, the radioactive rain-out was observed in eastern Massachusetts, USA, two days after detonation at Nevada of which maximum activity was 1.9 $\mu$c/l as the 3-day activity.[3] G. Jaffe, W. Wittmann and C. C. Bates[4] detected the radioactivity of about 6200 cpm/l (as the value 78 hours after the explosion) in hailstones which attacked the District of Columbia area on 26 May 1953, only 29 hours after the test at Nevada.[5]

On the other hand, assuming that the last Bikini experiment in 1954 was carried out on 5 May, the 3-day activity in rain-water precipitated in Kyoto area would have been 5 $\mu$c/l, which was much stronger than the values observed in the USA mentioned above. According to P. E. Damon and P. K. Kuroda[6] the artificial radioactivity in rain-water observed in Arkansas, USA, in the period from June to July 1953, was of the order of $10^{-6}$–$10^{-11}$ curie/l, which was considerably weaker than that observed in Japan. It is rather amazing to find that the activity in rain-water observed in Japan caused by Bikini tests in 1954 was stronger than those observed in the USA in 1953 as a result of continental tests.

In the case of the second maximum of 0.3 $\mu$c/l at Yamagata in September 1954, the date and the site of explosion were unknown, but, supposing that the date was 14 September, the 3-day value would have been about 1.2 $\mu$c/l: by no means a weaker activity.

These facts seem to indicate that the radioactivity of rain-out would not decrease appreciably beyond a certain distance from the explosion site. This is in agreement with the observed relation between activity in fallout and the distance from the zero site.[7] However, the topographic effect on the activity was clearly seen in Japan. Owing to the central mountain range along the Main Island of Japan, the activity was always much weaker on the opposite side of the mountain range (Fig. 2).

In the case of May rainfall, the abrupt rise in radioactivity was observed on 14th of the month only at Kagoshima, the southernmost city of Japan, though rain fell over a large area in Japan on the day. Likewise, the activity coming from the north was found at first only at Niigata and Hirosaki on the 18th–19th of September. Similar facts were observed also at the end of October when the third maximum began. In each case, the activity spread out after a few days over a larger area and in the same period the maximum activities in rain-water were measured at many places.

Such a situation has been also observed in the USA. Generally, the rain-out arrives at places in a certain limited area far from the explosion site, on occasion, more rapidly than the falling dust from the air. It is not unusual that activities in rain in the eastern part of USA, about 4000 km apart from the experimental site, have been detected only about two days after detonation.[3,4,5]

It has been found further that a remarkably uneven distribution of radioactivity may occur[3] even in a limited area, which is mainly due to its unevenness in the air. On the other hand, falling dust spread over

the whole area of USA in about 4–5 days after detonation in rather complicated forms of distribution.[7,8] These facts mean that the airborne dust which is thrown up into the upper atmosphere is transferred first by the rapid flow such as of a jet stream without any appreciable horizontal diffusion (probably less than 10 deg) and when they meet by chance with rainy clouds the radioactive rain-out may fall on a comparatively limited area. Later on, falling dust settling from a higher level to the ground falls on a larger area.

It is interesting to note that, as mentioned above, the radioactive materials presumably caused by Nevada experiments carried out in the spring, 1955, were also detected at many places in Japan. If such had been possible, radioactive dust would have arrived in Japan in 10–20 days after detonation.

M. Abribat *et al.*[9] and G. Garrigue[10] observed also the artificial radioactivity in rain-water precipitated in France. According to them, the radioactive rain-out due to Bikini tests in June to July in 1946 and Nevada experiments in the fall of 1951 reached Paris respectively in about 20 days and 8–15 days after detonations.

In these experiences including those in the USA and in Japan, the dust might have been transferred by speedy, westerly winds in the upper atmosphere except in the case observed in Japan in May, 1954. As a result of a rough estimation of the flow rate of dust particles it was found to be of the order of 1000–2000 km/day on an average. The maximum speed ever known was 3000 km/day in the case of hail storms in Troy area, N. Y., USA in April, 1953.[5] It has become obvious now that the effect of radioactive fall-out due to atomic tests is occurring on a global scale.

## DECONTAMINATION OF RAIN-WATER

Decontamination of rain-water was studied by using ion-exchange resins, sand filters and other filtering materials. Among them, charcoal for daily use in Japanese homes is convenient as a decontaminant. It is possible to remove more than 90% of radioactivity from rain-water by passing it through a filter consisting of charcoal and sand in which the latter is spread at the top and the bottom. Diatomaceous earth is also effective as a filtering material. For example, 15 litres of rain-water was passed once through a layer of 300-gm diatomite earth and the activity in the filtrate was reduced to less than 5% of the original intensity.

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# Consideration of the Total Environment in Power Reactor Waste Disposal

**By Eugene P. Odum,* USA**

The effects of radiations due to fission products on cells, tissues and individual organisms are being intensively studied. However, little has been learned concerning radiation effects on the higher levels of biological organization as represented by the population, the biotic community and the biogeochemical cycles vital to the functioning of world ecological systems. Safe disposal of radioactive waste products resulting from large scale uses of atomic energy for power or other peaceful purposes involves not only the protection of individuals from direct effects, but also an understanding of the long-term influences of low-level radiations on aquatic and terrestrial environments into which the by-products may be released.

Levels of ecological organization as listed above (that is, populations, communities, and ecological systems or ecosystems) have unique characteristics which are additional to those of the individual organisms or units making up the functional entity. For example, natality rate ("birth rate") and mortality rate are attributes pertaining to the population level, while food chain or *trophic structure* and rate of organic fixation or *productivity* are two characteristics which have meaning only at the community or ecosystem level. Furthermore, it is possible to treat quantitatively many of these features. Thus, since the structure and metabolism of the individual organism can be measured, often by indirect means, it should be possible to measure *community structure* and *metabolism.*

Studies of the effect of radiations on isolated organisms, mice in cages, for example, are valuable, but we must also accelerate the study of the function of intact biotic communities in order that *total* radiation effects can be evaluated. By analogy, we could point out that the effect of a drug on the muscular system of an intact human body could not be determined solely on the basis of a study of an isolated frog muscle. The result of injection into the intact body must also be observed. Therefore, in addition to the excellent work now being done by the radiation geneticist, the radiation physiologist and the health physicist, there is great need for progress in another area which we may call *radiation ecology.*

It is conceivable that every large atomic power plant of the future will need a radiation ecologist to work with environmental problems outside of the plant. We need to begin training young men simultaneously in the fundamentals of modern ecology and radiation biology to meet this inevitable need.

Before the total effects of radiations can be assayed and practical methods developed for determining the tolerance levels for entire ecological systems, it is necessary that we find methods for the measurement of total community structure and function. At present, there appear to be at least three feasible approaches to the problem. These possibilities are outlined and each illustrated with an example from an actual study.

## MEASUREMENT OF PRODUCTIVITY

The rate of organic fixation by "producer" organisms (i.e., primary productivity) and by "consumer" organisms (i.e., secondary productivity) provides an important index to the functioning of the whole system in situations where measurement is feasible. If the pattern of "basal community metabolism" is once established, significant changes or deviations from this pattern would signify that an important effect has been produced. It is conceivable, for example, that low-level radiations might reduce the total photosynthetic rate (and thus the food-making potential) of a community without causing mortality of individual organisms.

The rate of organic decomposition by microorganisms and other "decomposer" organisms as well as the ratio of primary productivity to average standing crop of organisms, which we may call *community turnover,* are two other measurable aspects of total function. These aspects in turn have an important bearing on what happens to radioactive elements released into the system.

Productivity may be measured by determining the amount of rare material used, the amount of by-product produced, or by weighing the growth produced in a given time interval in situations where it is not being immediately "harvested."[1] We may use as an example the determination of productivity of a coral reef by the measurement of a by-product of photosynthesis, namely, oxygen.

During the summer of 1954 we made a realistic test of certain theories concerning the assay of total function by selecting for study a windward coral reef on a Pacific atoll. The results have been prepared for publication.[2] The coral reef is an ideal natural unit for the study of total function because it is located

* Including work by Eugene P. Odum, University of Georgia; and Howard T. Odum, Duke University.

in a relatively constant environment and is, from the energetic standpoint, quite stable or "climax." It is also an isolated system with definite boundaries. The reef selected for study has been little disturbed by man since the object was to establish indices of community function in the normal situation in order to provide a basis for comparison with disturbed areas.

Productivity of the reef was estimated by what may be termed the flow method. The oxygen content of the water upstream and downstream is measured simultaneously. The oxygen increase between stations during the day is the net photosynthetic production of the community. The oxygen decrease between stations during the night is the total respiration of the community. By taking a series of measurements over the daily cycle, one obtains the course of production during the day. Measurement of the current transport permits calculation of total reef metabolism. The respiration at night plus the net production during the day gives the total production. By comparing the area of the graph between the day curve and the zero line with areas of the graph under the zero line at night, one can obtain an indication of what part of the excess production during the day is used up by respiration during the night.

A series of such measurements was made on several different days of similar and typical cloud cover of from $\frac{1}{10}$ to $\frac{3}{10}$ small cumulus and $\frac{1}{10}$ to $\frac{4}{10}$ high and middle cloudiness and at night. These values are expressed on an area basis in Fig. 1 following their conversion from depth and current measurements. When areas above and below the zero line are compared, it is evident that production approximately balances respiration.

Besides small errors due to inherent fluctuations and variability of oxygen samples and methods, current and depth measurements, and cloud cover changes, there is a major source of error that tends to cause values of production and respiration to be too small. This is the diffusion of oxygen from and to the atmosphere. This error is greater when the displacement of the gaseous content of the water from equilibrium with the atmosphere is greatest. Thus the error in decreasing the production estimate is greater than that in the respiration since greater displacement from equilibrium occurs. Furthermore, during the day's production a carpet of bubbles of oxygen is observed to form over the algal mat surfaces particularly on the front reef. These bubbles are continually breaking off and reach the surface without dissolving so that some of the oxygen is lost, further lowering the estimate of production below the correct figure. Further study should make it possible to correct for these errors.

Even though the flow method may result in underestimation, the productivity of the reef is certainly quite high, greater than 24 gm/m²/day which is equivalent to about 74,000 pounds/acre/year. This is equal to man's best agricultural efforts. The reef maintains a high productivity despite the very low productivity of the surrounding ocean waters (about

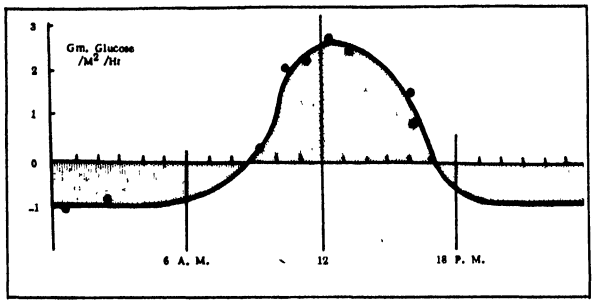



Figure 1

0.2 gm/m²/day or less than $\frac{1}{120}$ as productive). Marked symbiosis between plant and animal components results in the hoarding or cycling of scarce nutrients (nitrogen, phosphorus) within the reef with little loss as the water passes over the reef. Corals, for example, have large amounts of symbiotic algae in their skeletons, which, as shown by radioautographs, are not in direct contact with the outside environment. Therefore, it is concluded that coral polyps and the algae exchange food and nutrients at close range in an efficient manner. Any disturbance which interferes with these intricate relationships would most certainly have a marked effect on the metabolism of the reef as a whole.

Since the average standing crop, that is, the dry weight (biomass) of living algae present at any one time, was estimated as about 700 gm/m², and the productivity as 24 gm/m²/day or 8760 gm/m²/year, the *annual community turnover* is 12.5. This is another figure which is believed to represent an important measure of total community function.

Although more study is needed, it would appear that assaying productivity by the flow method, or a modification of it, is practical for certain aquatic situations. Only simple equipment and a short period of time are required. Oxygen measurement, of course, is not practical in many situations but other raw materials or by-products may be substituted. For certain terrestrial environments the author has found that harvesting the standing crop from sample areas at short intervals during the growing season provides an assay method for productivity (unpublished data).

## MEASUREMENT OF TROPHIC STRUCTURE

The over-all food chain (trophic) structure of a community can best be shown for the purposes of comparison by a graphic device called a *biomass pyramid*. The total weight (on an area basis) of the standing crop of living organisms (biomass) is determined for basic ecological groups as classified according to their food and energy roles in the community complex. In the simplest form, food-making organisms (producers), herbivorous organisms (primary consumers) and carnivorous organisms (secondary consumers) are distinguished. These data are then arranged in the form of a pyramid with producers representing the base of the pyramid, as shown in Fig. 2. Where a species population is omnivorous, its biomass is divided between herbivore and car-

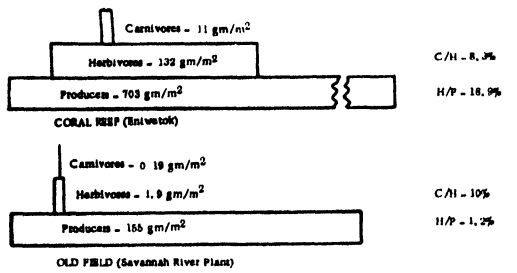



**Figure 2**

nivore level in proportion to the amount of activity at each level. Decomposer organisms should also be included in the pyramid, but at this time there are almost no data on total biomass of this ecological group.

In Fig. 2 a biomass pyramid is diagrammed for the same coral reef as described in the preceding section, and this is compared with the trophic structure of a cultivated field abandoned for one year. The field is one of a series being studied at the site of the Savannah River Atomic Energy Plant. Because green plants (algae) are so intimately interwoven with animal and dead skeletal material on the coral reef, the chlorophyll extraction method was used to estimate producer biomass. Algal dry weight was determined by relating spectrophotomically chlorophyll content of corals and other reef substrates with known dry weights of a reference species, *Codium edule*. Animal biomass was determined from samples drawn from quadrats of appropriate size for sessile and motile groups. The data represent the average for the entire reef. For the old field separation of producer and consumer protoplasm involved fewer technical difficulties, although the precise trophic relations of some species are not adequately known. The data represent the average for the growing season.

Much work remains to be done before the pyramids can be considered more than crude estimates. However, comparison of the pyramids in Fig. 2 reveals what would appear to be a fundamental difference in the ratio between food-making plants ($P$) and herbivorous animals ($H$). The $H/P$ ratio for the coral reef was about 20% and for the old field only about 1.2%. The coral reef, as indicated above, is a stable community with production and consumption (growth and decay) approximately balanced, while the old field is unstable; it will undergo marked changes from year to year (i.e., ecological succession). Thus, organic matter is accumulating faster than it is "consumed" or "decomposed" in the unstable community (the reverse would result in a "senile" situation—another kind of unstable community). Theoretically, should the stable coral reef be subjected to a large-scale disturbance, the trophic structure would tend, at least for a time, to shift to that of the unstable community. Thus over-all trophic structure gives important clues as to the condition of the community.

It will be noted from Fig. 2 that the standing crop of producers is greater in the coral reef as compared with the old field ecosystem. This is correlated with the greater productivity of the reef. The field community was growing on rather sterile, sandy soil. It does not follow, therefore, that unstable communities necessarily exhibit low productivity; actually, such communities may be quite productive, even though they have a low $H/P$ percentage. Actually, the relation of trophic structure and productivity has been studied in but few situations. At present we are inclined to believe that there is a more definite relation between the two in stable than in unstable communities. In other words, it may be possible in the future to determine productivity from data on standing crop biomass in the former community type.

## MEASUREMENT OF SPECIES STRUCTURE

Changes in the competitive structure due to changes in environment often may be detected by determining the percentage of sensitive and insensitive species. When oxygen in a stream, for example, is reduced as a result of pollution, the number of species in groups which are not tolerant is greatly reduced. On the other hand, the number of species of tolerant groups may increase as a result of the absence of competition from the eliminated species. Richardson[3] worked out a system for pollution assay in the Illinois River based on changes in the ratio of tolerant to intolerant species. Recently Patrick[4] has successfully used this method in extensive studies of industrial pollution. The method is illustrated in Fig. 3 using data of the latter author. In Fig. 3, the Roman numerals I–VII represent successively less tolerant groups of organisms. In the unpolluted stream there is a moderate number of species in all seven groups (number of species indicated at the top of each bar), while in the polluted section of the same stream the number of tolerant species is increased and the number of intolerant forms decreased. While this assay method has not been tested in connection with radioactive wastes, there is no reason to believe that it would not be useful as a measure of "atomic pollution" as for other types. In general, it is better to consider the entire complex of species rather than a few "indicator" species. Considerable experience in the study of pollution has shown that the situation is rarely so simple that the presence or absence of one or two species can be relied upon, especially where the effect is only moderate. The method is most effective where the taxonomy of all major organisms is well known. Incomplete taxonomic information is more of a handicap with this method than with the other two methods discussed in this paper.

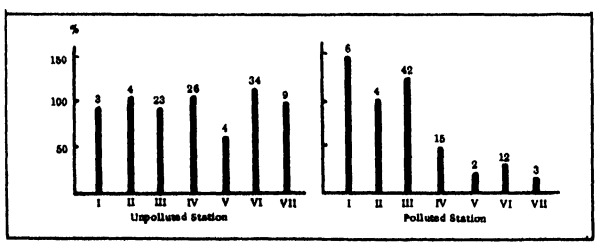



**Figure 3**

The "species structure" assay method is especially useful in situations where the discharge of pollutants is periodic, or the effect occurs during a brief period. In other words, changes in community structure often persist long after the presence of pollutants is undetectable chemically.

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# The Absorption of Fission Products by Plants

**By J. H. Rediske and F. P. Hungate,* USA**

Since the fission product elements enter the biotic cycle in largest part through plant life, an evaluation of the quantities which plants may contain is necessary. The two methods whereby fission products may contaminate crops are, first, by the direct deposition on a maturing crop, and, second, by the contamination of an agricultural soil with subsequent absorption into a crop.

The first method of contamination would occur on the external surface of the plants, could happen only during limited periods, and the hazard could be directly evaluated by radio-assay.

The second method of contamination would result from fission product deposition on the soil. Each year that crops were raised on that soil fission products would be absorbed into the plant tissues. This would continue until both radioactive decay and plant absorption reduced the contamination to levels of non-significance. The absorption of fission products from soil is the concern of this paper. Measurements of plant absorption were made on a variety of fission products to evaluate this indirect hazard of soil contamination.

## METHODS

To compare the uptake of the various fission products a simple reproducible method such as the Neubauer seedling test[1] was required. This test consists of growing 100 barley seeds in 100 gm of soil which was previously contaminated with a fission product. Cultures were maintained for 20 days, when the leaves were harvested. It was previously determined that the fission product concentration in the leaves reaches a maximum by this time.

* Including work by J. F. Cline, J. H. Rediske, and A. A. Selders, General Electric Company.

Other studies were conducted by standard nutrient solution culture techniques using Hoaglands solutions[2] of the essential salts. Plants were cultured in six-liter containers, one plant per liter of solution.[3]

The absorption of the fission product by the plant is expressed on the dry weight basis as a concentration factor:

$$\frac{\text{fission product concentration in leaves}}{\text{fission product concentration in root environment}}$$

Because of the highly controlled conditions of nutrient culture, concentration factors obtained in this manner are not directly comparable to those obtained from soils. They serve, however, in a relative sense to determine how single substrate factors may affect plant absorption.

## RESULTS

### Relative Fission Product Absorption

Table I contains the results of the Neubauer tests for the important individual fission products as determined on some local agricultural soils. The soils were all slightly alkaline, but varied widely in texture and cation-exchange capacity. It is apparent from these data that the barley plant concentrates strontium and iodine in its leaves in greater amounts than any of the other fission products. Barium, which is chemically similar to strontium, is somewhat less concentrated by the plant than is strontium. Cesium may be moderately important in some soils, but for the most part is absorbed in small amounts. All other fission products tested, and plutonium, have concentration factors less by two or three orders of magnitude than does strontium.

**Table I. The Concentration Factors (conc. of isotope in leaves/conc. of isotope in soil) of Several Fission Products as Determined by the Neubauer Seedling Test Using Barley**

| Soil | pH | Organic matter, % | Cation exchange capacity, me/100gm | Isotope | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | $Sr^{90}$ | $Ba^{140}$ | $I^{131}$ | $Cs^{137}$ | $Y^{91}$ | $Ce^{144}$ | $Pm^{147}$ | $Zr^{95}$-$Nb^{95}$ | $Ru^{106}$ |
| Ringold silty clay | 7.6 | 7.9 | 34.1 | 0.96 | 0.13 | | | | | | | 0.008 |
| Wheeler silt loam | 7.7 | 4.3 | 4.8 | 1.6 | | | 0.04 | | 0.004 | | 0.02 | 0.012 |
| Ephrata loamy sand | 7.3 | 1.8 | 8.1 | 1.7 | | 1.1 | 0.10 | 0.006 | | | | 0.015 |
| Winchester fine sand | 7.4 | 2.4 | 3.4 | 3.1 | 0.32 | | 0.08 | | 0.004 | 0.001 | 0.004 | 0.012 |

### Factors Affecting Absorption

The concentration factors for three fission products as tested on four different plant types are shown in Table II. The difference in strontium and iodine absorption among these species of plants is small. With cesium the variation is greater and may be as much as a factor of ten.

Table II likewise contains the concentration factors for the various plant organs. In nearly all cases the leaves have a concentration factor as great or greater than any other tissues. The fruiting organs generally have the smallest concentration factor. The stem usually has slightly less fission product than do the leaves. No values are included for roots because of the difficulty in measuring the amount absorbed into the root. It appears that the roots contain about the same amount of activity as do the stem and leaves. If externally absorbed activity is included, the root value often surpasses all others.

The effect of nutrient pH on the uptake of strontium, barium, cesium and iodine is illustrated in Fig. 1. As the hydrogen ion concentration increases the uptake of these fission products also increases. The effect of pH on absorption is not uniform for the four elements. The absorption of other fission product elements, such as yttrium,[4] is more pH sensitive and may vary by as much as a factor of ten over this same pH range.

Figure 2 illustrates the effect which addition of the stable isotope has on the uptake of four radioactive fission products. In general, by increasing the carrier concentration of strontium, cesium, and iodine, the fission product concentration factor increases. This increase is pronounced with iodine and slight with cesium and strontium. Barium exhibits an initial increase to a point near 10 μg/gm and then the concentration factor decreases.

**Table II. The Concentration Factors for the Important Fission Products by Representative Plants Grown in Nutrient Solution Culture***

| Plant | Concentration factor | | |
|---|---|---|---|
| | $Sr^{90}$ | $Cs^{137}$ | $I^{131}$ |
| Bean | | | |
| Leaves | 0.3 | 0.7 | 0.3 |
| Pod and seeds | 0.02 | 0.2 | 0.04 |
| Primary leaves | 0.3 | 0.5 | 0.1 |
| Stem | 0.3 | 0.3 | 0.2 |
| Tomato | | | |
| Leaves | 0.4 | 0.3 | 0.2 |
| Fruit | 0.009 | 0.2 | 0.02 |
| Stem | 1.6 | 0.2 | 0.2 |
| Russian thistle | | | |
| Leaves and stem | 0.1 | 0.05 | 0.5 |
| Fruit | 0.008 | | 0.4 |
| Barley | | | |
| Leaves | 0.3 | 0.02 | 0.5 |
| Head | | 0.01 | |

* The pH of the nutrient environment was 6.0, with 1.0 μg/ml of carrier present.

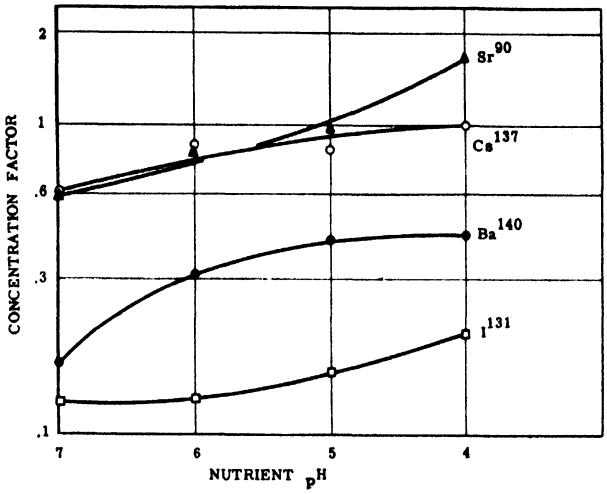



**Figure 1. The effect of pH on the concentration factors of four important fission products for bean plants grown in nutrient solution. (The concentration of the stable isotope was 1.0 μg/ml)**

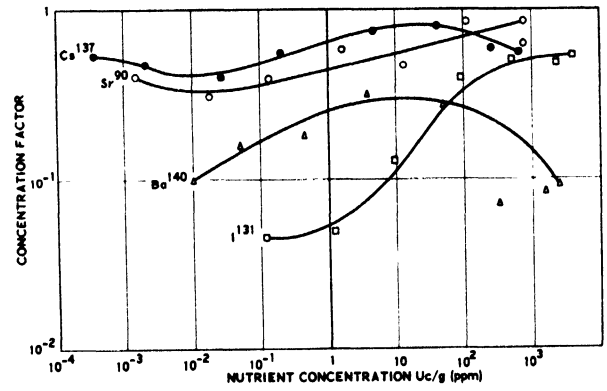



**Figure 2. The effect of carrier concentration on the concentration factors of four important fission products for bean plants grown in nutrient solution. (The culture solutions were maintained at pH 6.0 with various concentrations of the stable isotope)**

## DISCUSSION

From our experience at an atomic energy installation it appears that $I^{131}$ by its deposition on vegetation may be the limiting fission product isotope. In more general situations $Sr^{90}$ may be more critical from ecological considerations. At ten years post fission the activity from this element constitutes over 40 per cent of the total activity from fission products. The significance of strontium in plants results largely from the ability of the plant to concentrate this element and the possibility of direct or indirect transfer of this strontium-containing plant matter to man.

The ineffectiveness of the addition of carrier in suppressing the absorption of radioactivity into plants may appear unusual. In fact, some of the elements show a significantly increased absorption with increased carrier. Two possible mechanisms for the stimulation of absorption by increased concentration have been considered. One of these, a toxic effect from high concentrations of the element, has been ruled out through the simultaneous use of two elements. Toxicity in this case appears to depress ab-

sorption. The second possibility is that of a competition between ions adsorbed to colloids in the root environment and the absorptive processes of the root. With differing degrees of adsorption to the colloid this would result in an increased availability to the plant as the concentration of element increases. The second possibility has not as yet been tested.

If $Sr^{90}$ should enter the biotic cycle in quantities sufficient to be of concern, it would be difficult to establish a mechanism of removal. One possible mechanism is to shunt the $Sr^{90}$ into the bones of animals from which release occurs very slowly and which could be stored if necessary.

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# Biological Cycles of Fission Products in Agriculture in Japan

**By R. Sasaki,* Japan**

The nuclear detonation test explosion at Bikini Atoll exerted a great influence upon the agriculture of Japan. A number of surveys and investigations were made on the radioactive contamination and its effects upon rainwater, soil, cereals, vegetables, tea, fruits, silkworms, farm animals and their products. These surveys and investigations are summarized below.

## SOIL, RAINWATER AND IRRIGATION WATER

The measurement of radioactivity in the soil of uncultivated land, dirt roads and cultivated fields showed that radioactivity[1] normally accumulated on the hard surface of uncultivated ground or dirt roads, or in low-lying areas. Where the soil was soft and had good drainage, as in cultivated fields, surface radioactivity was weak. Little difference in the amount of radioactivity was noticed between the upper and the lower levels of the soil.

After the middle of May, 1954, strong artificial radioactivity in rainwater at Tokyo was detected. Semi-quantitative analysis of radionuclides in the soil showed that the fractions of rare earth elements and alkali earth elements were appreciably radioactive.[2]

The analysis of the radioactive elements, in mulberry field soil in a suburb of Tokyo, conducted in June 1954, confirmed the fact[3] that, where the soil was left uncultivated, most of the contaminating radioactivity stayed within 0.5 cm of the surface, while the radioactive substances included in the second group were translocated into the depth of 1.5 cm.

In a survey made in October 1954,[4] a little radioactivity was detected in the surface soil and subsoil of a tea garden in Shizuoka Prefecture. However, it was shown that the surface soil was normally apt to contain a larger amount of radioactivity.

A report on rainwater[5,6] shows the following. The rainwater that fell during the period of May and August 1954 at a lighthouse area[5] had a maximum of 8.0 cpm and a minimum of 0.6 cpm radioactivity per litre of solid matter after evaporation. A survey made in June 1954 on river water and canal water for irrigation in Kanagawa Prefecture,[6] showed that the water of a small river and a canal contained less than 7 cpm/1 radioactivity, while the Banyu River—a fairly large one—contained 12–28 cpm radioactivity per litre.

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## CEREAL CROPS

A number of surveys were conducted on the radioactive contamination of rice, wheat and other cereal.[1,5,7,8] The radioactivity of unhulled rice, hulls, unpolished rice, polished rice and bran, harvested in Niigata Prefecture was measured in 1953 and 1954.[5] This survey disclosed obvious contamination by artificial radioactivity of unhulled rice and hulls harvested in 1954, while each sample of the rice produced in 1953 and unpolished rice, polished rice and bran of 1954 showed no contamination at all. According to a survey on rice crops in Kanto Region,[7] artificial radioactivity was detected not only in leaves and stems but also in the ears. In a survey[7] of 52 samples collected from all parts of the country, no artificial radioactivity was found in polished rice and bran harvested in 1952 and 1953, while the hulls and bran of 1954 had artificial radioactivity, and some contamination was also detected in polished rice.

Radioactivity was also observed in the soybeans and upland rice planted between tea bushes, and in rice and wheat straws spread under the bushes.[9]

In regard to the absorption of radioactive elements by plants, a water-culture experiment with paddy rice, using the ashes of the "Haenawa" rope of the No. 5 Fukuryu Maru, produced the following result.[1,10,11] A majority of the fission products added to the water-culture solution accumulated in the roots. The alkaline-earth group, in particular, was selectively absorbed and translocated into the shoots. According to a white-sand culture experiment utilizing wheat seedlings, most of the fission products accumulated in the roots, and only 10% of them were translocated to the aerial portion. The absorption of radioactive substances was slightly increased when ammonium salts, such as ammonium chloride or ammonium sulphate, were added. Absorption was negligible when phosphate was added.

## VEGETABLES

According to a number of surveys made on radioactive contamination of various kinds of vegetables,[1,5,7,8,12,13] it was noted that radioactivity was generally found in vegetables with large leaves and rough surfaces. In a survey during May and September, 1954,[5] radioactivity was noticed only on the surfaces, and not inside the plants. A survey of November, 1954, showed the existence of considerable radioactivity in the leaves and stems of taro, burdock, sweet

potato and garden radish planted between tea bushes in the tea gardens of Shizuoka Prefecture.[9] The ashes of those vegetables[7] contain most of the radioactivity in elements of the rare earth group.

## TEA

Most of the tea produced during late spring through early summer of 1954 was more or less contaminated.[8] The tea harvested in May 1954 had strong radioactivity.[8] The radioactivity of tea infusion[9] varies according to the temperature of the water used in its preparation. Tea made with boiling water showed as high as 6–7 cpm, while that made with hot water at about 98°C had less radioactivity. Tea made with water at 60° had far less radioactivity than the above, and scarcely any radioactivity was found in tea made with cold water. Further, the difference was also noticed in the cropping time. The 2nd crop had 2–4 cpm the 3rd and 4th crops showed 1–3 cpm and 6–15 cpm respectively.

The analysis of radioactive elements in tea leaves[5,7] showed that the radioactivity originated mostly from rare and alkaline earths, but was not contributed by $Sr^{90}$.

## FRUIT TREES

An investigation of samples collected during August and November, 1954 from the whole country[1,4] showed that radioactivity was highest in the leaves of fruit trees, less in the pericarp, and almost nil in fruit flesh.

## MEADOW GRASS, WILD GRASS AND FLOWERS

In meadow grass and wild grass, the quantity of radioactivity differs according to the form of their leaves and stems, regardless of the species. The same also applies to flowers.[1] Analysis of the ashes of contaminated clover[11] showed that alkaline earths, especially $Sr^{89}$ and $Sr^{90}$, are selectively absorbed, and accumulate in the plant.

## SERICULTURE

An experiment on feeding silkworms with contaminated mulberry leaves[14] disclosed the following fact: A comparatively large amount of radioactivity was excreted in the silkworm faeces, while little radioactivity was found in the body and cocoon layers. Hardly any radioactivity was detected in the silkworm pupae and reeling water. A number of unfertilized eggs were discovered among silkworms fed with contaminated mulberry leaves. Their hatching results were inferior to that of normally fed silkworms.[14]

A maximum of 1674 cpm and a minimum of 33 cpm radioactivity[15] was measured per gram of ashes of mulberry leaves in Tokyo, Kyoto, Yamagata and Niigata Prefectures. Most of the radioactive elements concerned belonged to the third group.

## FARM ANIMALS

Radiation injury[16] was studied in hens treated with ashes of heavily contaminated rope from the No. 5 Fukuryu Maru. An experiment was also made on the influence of radioactivity on the polymerization of desoxyribosenucleic acid in the hen. The fodder mixed with $P^{32}$ and $I^{131}$ was administered to the hen in order to observe the effect of the radioactivity.

The accumulation of radioactive isotopes in the body of farm animals was surveyed[17] by measuring radioactivity in the faeces of sheep chained to graze. In the ashes of the faeces, no artificial radioactivity was detected.

## MILK AND MILK PRODUCTS

Milk powder processed in Japan before and after the H-bomb explosion at Bikini Atoll has 30.6 to 38.6 cpm radioactivity per gram of the ashes,[18,19] which is considered to be the natural radioactivity of $K^{40}$. No artificial radioactivity was detected in the ashes.[5] However, since $Sr^{89}$ and $K^{40}$ have almost same $\beta$-radiation, it was suspected that $Sr^{89}$ might be contained in milk. Therefore, the ashes of the milk were administered to a white rat in order to study the distribution of radioactivity in each part of the rat's body. By analyzing the ashes, non-existence of $Sr^{89}$ was confirmed.[20]

The translocation of radioactive substances was made a subject of an experiment by administration of contaminated shark skin to a white rat, and the ashes of "Haenawa" rope from the No. 5 Fukuryu Maru to a goat.[20] The radioactivity of the goat's milk was high after 6–48 hours, but disappeared in 96 hours. Artificial radioactivity was detected in the faeces, but hardly any in the urine.[20]

## HEN'S EGGS

Artificially radioactive substances were excreted in the faeces within a few days after the administration of the ashes of contaminated shark's skin to a laying hen.[21] The radioactivity stayed in the hen's body no longer than 5 days. The egg laid on the day following the administration was only slightly radioactive, but most of this activity was detected in the shell.

The ashes of the "Haenawa" rope from the No. 5 Fukuryu Maru were administered to a laying hen in order to observe the translocation of fission products in the hen's egg.[21] The egg's shell obviously contained $Sr^{89}$ and $Ca^{45}$, and $Sr^{90}$, $Y^{90}$, $Rh^{106}$, and $Ru^{106}$ may also have been present. Sr and Ca were detected in the egg white and yolk, but the amount of artificial radioactivity in the white and yolk was extremely small.

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# The Behaviour of $I^{131}$, $Sr^{89}$ and $Sr^{90}$ in Certain Agricultural Food Chains

**By A. C. Chamberlain, J. F. Loutit, R. P. Martin and R. Scott Russell,* UK**

The contamination of the earth's surface which can result from industrial development has been apparent for over a century. The blackened areas which surround some industrial towns, the occurrence of smog, the release of gaseous effluent from cement factories and the pollution of rivers have led to localised though sometimes disastrous consequences. Remedial measures have too frequently not been contemplated until contamination has reached major proportions. Now when we can look forward to the large scale use of atomic energy for industrial purposes the importance of assessing potential hazards in advance of their occurrence is emphasised both by our past experience and by the insidious effects of radiation.

The International Commission on Radiation Protection[1] has made the first contribution by assessing permissible doses of radiation and radioactive materials for those who are occupationally exposed. Their figures do not take into account genetic hazards which need to be considered when large sections of a population are involved.

Nevertheless they provide a useful baseline when we considered environmental as opposed to internal problems.

In the laboratory and in industry, work with radiation and radioactive materials usually involves primarily exposure of the body to radiations from without —external radiation. The risk of inhaling or ingesting radioactive material is in general smaller. It is envisaged that if fission products escape into the atmosphere, the same will apply as far as the human population is concerned. Part of man's diet is of animal origin which will not be directly contaminated by atmospheric "fall-out". Part is of vegetable origin which is not only stored for long periods, during which physical decay of radioactivity will occur, but is protected from direct contamination by husk or peel which is discarded.

Fresh leafy vegetables such as spring cabbage and lettuce which are open to direct contamination form only a very small fraction of the diet and in some cases this contamination may be minimised by washing and cooking.

Grazing animals such as cattle, sheep and horses on the other hand have no such protection. When not supplied with supplementary foodstuffs they derive their food entirely from pasture. Close grass swards, which are the best pasture, are also the best physical trap for particles deposited from the air. This coupled with the fact that the daily intake of food by large farm animals may represent the yield of several hundreds of square metres means that such animals ingest daily the scourings of thousands of cubic metres of air.

Fortunately mixed fission products are composed largely of elements which, whatever their chemical form, are not readily absorbed from the gastro-intestinal tract. As exceptions to this general rule are iodine, and the alkaline earths, strontium and barium. Molecular iodine and iodides are almost wholly absorbed and as such are highly concentrated in the thyroid gland. Oxides of the alkaline earths are readily soluble in the acid stomach juice and on absorption are largely deposited in bone. Both iodide and the alkaline earth ions are secreted in milk in significant quantities. Thus as well as hazard to the grazing animal itself from radioactive $I^{131}$, $Sr^{89}$ and $Sr^{90}$ there is the secondary hazard to those human beings whose diet consists largely of milk, notably infants, who because of their youth may be considered super-susceptible to the effects of radiation.

It is established that the physical solubility and the biological availability of individual and mixed fission products depend on their physical and chemical form. For the purposes of calculation, to which we now proceed, of permissible levels of environmental contamination for grazing animals—and consumers of milk, it is well to assume that the important fission products will be in fully available form.

The duration of exposure is a major factor—determining the tolerable deposition per unit time. If frequent release of fission products occurs, the permissible levels in herbage can be related to those accepted in occupational exposure. If fission products are released only at infrequent intervals, relatively higher levels should be acceptable temporarily. In a preliminary assessment it is convenient to consider first the maximum continuous deposition. The atmospheric content which would give rise to these levels can then be compared with those which cause hazards of other types.

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## TOLERABLE LEVELS OF $I^{131}$, $Sr^{89}$ AND $Sr^{90}$ IN HERBAGE CONSUMED BY GRAZING ANIMALS ASSUMING CONTINUOUS DEPOSITION

### Assumptions

1. Maximum permissible daily intakes in $\mu c$ are:

| | *Dairy cows* | *Infants* |
|---|---|---|
| $I^{131}$ | 2.3 | 0.01 |
| $Sr^{89}$ | 12 | 0.016 |
| $Sr^{90}$ | 0.14 | 0.00019 |

Continuous exposure is assumed and the figures are based on values for occupational exposure in the Recommendations of the International Commission for Radiological Protection.[1] Allowance has been made for differences in weight of the critical organ from that of the standard man as shown:

| | *Man* | *Infant* | *Dairy cow* |
|---|---|---|---|
| Thyroid wt | 20 gm | 1.5 gm | 35 gm |
| Skeleton wt | 7 kg | 0.75 kg | 55 kg |

For cows it has been assumed that the resultant figures can be increased by a factor of 10 on account of their life span being shorter than that of man. No more direct estimate of tolerance levels for animals is available.

2. Weight of herbage ingested by dairy cattle per day—14 kg (30 lb) dry weight (this and other agricultural data here are within range for Great Britain).

3. Yield of herbage dry weight per unit area—56 gm/m² (i.e., 500 lb acre).

4. Fraction of fission products deposited per unit area which is retained in edible herbage—0.25. There are unfortunately no direct experimental data. This figure is based on laboratory reports and on the examination of pastures.

5. Fraction of the daily milk production of a cow consumed by an infant—0.13, it being assumed that a cow yields 8 litres of milk and a child consumes 1 litre.

6. Fraction of fission products ingested by cows which is present in milk consumed by children—$I^{131}$, 0.03 (from Glascock[2]); $Sr^{89}$ and $Sr^{90}$, 0.02 (from Comar[3]). In deriving the $I^{131}$ figure allowance has been made for the interval between ingestion by the cow and consumption by the infant 24 hours after the secretion of milk.

### Maximum Permissible Levels in Pastures

Maximum permissible levels (MPL) have been obtained by substituting the appropriate values for the assumptions in the following equations:

$$\text{MPL/gm herbage} = \frac{1.}{2.} \begin{matrix}\text{for hazards to cows}\\ \text{(figures shown in}\\ \text{paragraphs above).}\end{matrix}$$

$$= \frac{1.}{2. \times 3. \times 4.} \text{ for hazards to infants.}$$

$$\text{MPL/m}^2 \text{ ground surface} = \text{MPL/gm} \times \frac{3.}{4.}$$

The values thus derived are shown in Table I, columns 3 and 4.

### Daily Deposition per Unit Area To Give Rise to Maximum Permissible Level

Fourteen days appears to be a median figure for the period which elapses between successive grazing of the same area of herbage. Thus at the time of grazing the herbage will have been contaminated for 14 days. In column 5 of Table I figures are shown for amounts of $I^{131}$, $Sr^{89}$ and $Sr^{90}$ deposited daily which would lead to the proposed MPL. Allowance has been made for decay.

### Limitations to the Calculations

The majority of the assumptions used in the calculations are based on unfortunately limited evidence. However even if this limitation did not exist the proposed figures could not be universally applicable because the wide differences in the grazing pattern, which occur from region to region, will greatly affect the tolerance levels. The use of tolerance levels for infants which assume continuous exposure is open to question because of the rapid reduction in milk consumption relative to body weight which occurs with their growth. In effect therefore an additional safety factor has been introduced. From the viewpoint of assessing hazards from the isotopes of iodine this does not however affect the general conclusion as the MPL for cattle and infants appear to be in the same general order.

### Application of these Calculations to Intermittent Releases of Fission Products

Since the effects of radiation are largely cumulative it is permissible on a single isolated occasion to receive a total dose equivalent to that acceptable in a considerable period of continuous exposure. Thus if deposition is intermittent it may be possible to allow deposition on a single occasion equivalent to the deposition normally permitted in 100 days continuous exposure. In such circumstances it would be necessary to ensure that the daily deposition in the intervals between these large releases was correspondingly lowered.

## TOLERABLE DEPOSITION OF FISSION PRODUCTS IN SOIL IN WHICH CROPS ARE SUBSEQUENTLY GROWN

In addition to fission products being deposited on edible herbage significant quantities will enter the soil. Since on the average several months are likely to elapse between the entry of fission products into soil and their absorption by plants $I^{131}$ will from this viewpoint be a small source of hazard and relative to $Sr^{89}$ the hazard for $Sr^{90}$ will be increased. Fission products other than these are known to be absorbed markedly more slowly by plants. Thus the hazard for $Sr^{90}$ will be limiting.

The quantity of calcium in soil is a major factor affecting the absorption of strontium. It has been

suggested[4] that plants grown in soil discriminate against strontium as compared with calcium by a factor of 0.4. This conclusion is however not considered acceptable in the light of observations of the absorption of calcium and stable strontium under natural conditions.[5] Moreover water-culture studies provide no evidence that the absorption of the two elements is markedly different.[6,7] The elucidation of the interaction of strontium and calcium in soil is complicated by the fact that little is known regarding the equilibration of fission product Sr with soil calcium fractions. In the absence of information on this question it is simplest to consider the steady state which will be attained when $Sr^{90}$ has been deposited at a uniform rate for long periods; the deposition of $Sr^{90}$ from the atmosphere will then equal its removal by plant absorption, in drainage or through decay. So the fullest equilibration will have been attained between $Sr^{90}$ and the labile calcium in the soil.

It is suggested therefore that the general magnitude of the quantity of $Sr^{90}$ tolerable in soil at equilibrium may be gauged from the equation:

$$\frac{Sr^{90}}{Ca} \text{ tolerable in man} = 0.5 \frac{\text{MPL } Sr^{90} \text{ in soil}}{\text{labile Ca}}$$

The factor 0.5 is introduced since animals in contrast to plants appear to discriminate against strontium relative to calcium[3,8] and part of the intake by man will be in animal produce.

Assuming (1) that the ratio of $Sr^{90}$ to Ca tolerable in man is $10^{-3}$ (maximum permissible body burden 1 $\mu$c; Ca content 1050 gm)[1]; (2) that the labile calcium in soil is 10 milliequivalents/100 gm† and that the density of soil is 1.4; and (3) that plants derive their calcium from the upper 25 cm of soil, the MPL of $Sr^{90}$ per $m^2$ ground surface calculated by the above equation is 2 $\mu$c.

Radioactive decay will reduce the $Sr^{90}$ in soil by approximately 3% per annum while absorption by plants and drainage may remove up to 2% per annum in some cases. Thus an annual addition of $10^{-1}$ $\mu$c $Sr^{90}/m^2$, or a daily addition of $3 \times 10^{-4}$ $\mu$c, would after the passage of years cause the proposed MPL to be attained. This value is in the same order as the deposition of $Sr^{90}$ which appears to give rise to the MPL through deposition on edible herbage (Table I).

The comments made earlier regarding the imprecise nature of present assessments, and the variation between different localities, apply also to this calculation.

## COMPARISON OF LEVELS OF CONTAMINATION WHICH WOULD LEAD TO AGRICULTURAL AND OTHER HAZARDS

The most complete assessments of tolerable levels of fission products for man in direct exposure have been based on the content of air or water[1] and the best basis for the comparison of these levels with the present agricultural assessment is in terms of atmospheric content. Chamberlain and Chadwick[9] have shown that $I^{131}$ gas is deposited on grass by diffusion. Defining the velocity of deposition as follows: Velocity of deposition = Deposition per square cm of ground per second/Volumetric concentration per $cm^3$ of air, they found that on average the velocity of deposition of $I^{131}$ onto grass-covered ground was 2.5 cm/sec.

Table I. Provisional Maximum Permissible Levels of Fission Products in Herbage Grazed Continuously by Dairy Cattle

| (1) Fission product | (2) Hazard to | (3) $\mu$c/gm herbage | (4) $\mu c/m^2$ | (5) Deposition $\mu c/m^2/day$ |
|---|---|---|---|---|
| $I^{131}$ | Thyroids of cattle | $2 \times 10^{-4}$ | $4 \times 10^{-2}$ | $4 \times 10^{-2}$ |
| | Thyroids of infants consuming milk | $2 \times 10^{-4}$ | $4 \times 10^{-3}$ | $5 \times 10^{-3}$ |
| $Sr^{89}$ | Skeletons of cattle | $9 \times 10^{-4}$ | $2 \times 10^{-1}$ | $1 \times 10^{-2}$ |
| | Skeletons of infants consuming milk | $4 \times 10^{-4}$ | $1 \times 10^{-1}$ | $8 \times 10^{-3}$ |
| $Sr^{90} + Y^{90}$ | Skeletons of cattle | $1 \times 10^{-5}$ | $2 \times 10^{-3}$ | $2 \times 10^{-4}$ |
| | Skeletons of infants consuming milk | $5 \times 10^{-6}$ | $1 \times 10^{-3}$ | $9 \times 10^{-5}$ |

Applying this value the atmospheric content which would maintain the MPL of $I^{131}$ in Table I is $2 \times 10^{-12}$ $\mu$c $I^{131}/cm^3$ air. This is lower by over $10^3$ than the accepted MPL for man through breathing which is $6 \times 10^9$ $\mu c/m^3$ air.[1]

With regard to the rate of deposition of $Sr^{89}$ and $Sr^{90} + Y^{90}$ from the atmosphere less information is available. Unlike $I^{131}$, which is expected to be present in the atmosphere in a gaseous form, $Sr^{89}$ and $Sr^{90}$ are expected to occur as particulates which may vary in size from $10^2$ to $10^{-2}$ microns. However, by the comparison of the permissible rate of deposition of $Sr^{89}$ from Table I ($8 \times 10^{-3}$ $\mu c/m^2/day$) and the permissible level in air ($2 \times 10^{-8}$ $\mu c/cm^3$) for the inhalation hazard,[1] it can be deduced that the hazard from deposition will be limiting unless the velocity of deposition is less than $4 \times 10^{-4}$ cm/sec. There is evidence that the velocity of the deposition of aerosols produced under either experimental or natural conditions exceeds this value by two orders of magnitude. Hence the deposition of $Sr^{89}$ as opposed to its presence in the atmosphere appears to be the limiting hazard. The same is true for $Sr^{90}$.

## DISCUSSION

The present assessment suggests that levels of $I^{131}$ in the atmosphere which are lower by a factor of 1000 than those which create a breathing hazard to man may be dangerous if the fission products are deposited on herbage which is subsequently consumed by grazing animals. Similarly the major source of hazard from $Sr^{89}$ and $Sr^{90}$ may be from its deposition on agricultural land.

It is apparent from Table I that the quantities of $I^{131}$ and $Sr^{89}$ which may give rise to hazards are in

† It is assumed the agricultural estimates of "available" calcium are a measure of this quantity.

the same general order. In practice the relative activities of $I^{131}$ and $Sr^{89}$ deposited may vary by a considerable factor, depending on whether the fission products released have been freshly produced or whether they are in an equilibrium mixture. It may be noted that when fresh fission products are released the iodine hazard will be increased since the short lived isotopes $I^{132}$, $I^{133}$ and $I^{135}$ will make a considerable contribution. The hazard to cattle will thereby be increased relative to that of infants because of the shorter time in which iodine will reach the thyroid. It would appear that $Sr^{90}$ is likely to become the limiting hazard only when fission products have been incorporated in the soil for prolonged periods.

These results are not regarded as justifying any alarm since it should be possible to prevent contamination approaching the proposed MPL by a considerable factor. However, in the determination of precautionary measures, the possible occurrence of agricultural hazards should be a major consideration. The inadequacy of the data on which the present assessment is based has already been stressed as also have been the wide variations in tolerable levels which will result from different agricultural systems. It is apparent therefore that a large amount of investigation must be undertaken before an adequate assessment can be made. Fortunately much of the necessary work is likely to serve a dual purpose as it should add considerably to our knowledge of many aspects of plant nutrition.

In Europe there is a special need for the examination of these questions since, unlike the more happily situated inhabitants of the New World, we must envisage atomic reactors being located relatively close to areas of intensive agricultural production on which large populations depend for their survival. The sole justification for discussing the present meagre conclusions is the hope that they may encourage the examination of this subject.

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# The Accumulation of Radioactive Substances in Aquatic Forms

By R. F. Foster and J. J. Davis,* USA

Aquatic organisms living in rivers or lakes downstream from atomic energy installations have been found to accumulate radioactive materials from the water. This paper explains how the aquatic forms become radioactive and describes conditions found in the Columbia River below the Hanford reactors.

## NATURAL SOURCES

Waters of the earth's surface commonly contain trace amounts of naturally occurring radioisotopes which have been dissolved from radioactive ores. Typically, $U^{238}$, $Ra^{226}$ and $Th^{232}$ are present in measurable amounts; but other natural radioisotopes, such as $K^{40}$, also occur. Love[1] lists background levels ranging from $3.6 \times 10^{-10}$ μc/ml to $3.4 \times 10^{-9}$ μc/ml for normal surface water. Values in excess of $10^{-4}$ μc/ml are reported for certain hot springs of high radium content. The heavy metals are not taken up in appreciable amounts by aquatic organisms, but most forms contain enough $K^{40}$ to give concentrations of about $5 \times 10^{-7}$ μc/gm.

## SOURCES FROM ATOMIC REACTORS

Artificial radioisotopes are produced in large quantities by nuclear reactors. The fuel of typical reactors is well isolated, however, and the fission products which build up do not escape under normal operating conditions. Nevertheless, reactors which are cooled directly by water, such as the plutonium-producing units at Hanford, may be responsible for appreciable quantities of water-borne radioactive materials. Such radioisotopes result from dissolved solids present in the cooling water which become neutron activated as they travel through the reactor. Corrosion products, forming on heat exchange surfaces, also become radioactive and are picked up by the water. The effluent from the Hanford reactors contains a complex mixture of radioisotopes. Relatively large amounts of short-lived materials are initially present, but most of them quickly decay in large retention basins. Although the radioactivity of the effluent is well below permissible levels before release to the Columbia River, appreciable amounts of such isotopes as $Mn^{56}$, $Na^{24}$, $Cu^{64}$, $As^{76}$ and $Si^{31}$ are present, together with smaller amounts of longer-lived isotopes, including $P^{32}$, rare earths, and $Cr^{51}$. These are emitters of beta and gamma radiation; no significant amounts of alpha emitters, such as uranium or plutonium, escape from the reactors under normal circumstances. Comparatively little amounts of fission products may occur in effluent from reactors owing to uranium present in the cooling water.

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## RADIOACTIVITY IN AQUATIC LIFE

The organisms living in the Columbia River which have picked up radioactive substances from the reactor effluent may be utilized as a large-scale experiment in which the isotopes serve as tracers. In this way, studies designed primarily to monitor the level of radioactivity, also provide information on nutrient cycles, metabolic rates, and ecological relationships.

Sampling of the water was started in 1944, before the reactors began producing plutonium and routine sampling of the organisms was initiated in 1946. The relative activity densities of the different types of aquatic life vary over a broad range as shown in Fig. 1. The gross activity density of the plankton (mainly diatoms) is about 2000 times that of the water. This value is of strictly local application, however, since concentrations of individual isotopes differ by several orders of magnitude. In spite of the large variety of isotopes present in the Hanford effluent, 30 to 50 per cent of the radioactivity in the plankton is from $P^{32}$, 25 to 50 per cent from $Cu^{64}$, 5 to 15 per cent from $Na^{24}$, and less than 10 per cent from mixtures of the rare earths and iron groups. Although $P^{32}$ is responsible for less than one per cent of the radioactivity in the water, it makes up 70 to 95 per cent of the radioactivity in most invertebrates and fish. During the seasons when the animals are most radioactive, the $P^{32}$ content of small fish, *Richardsonius balteatus* (Richardson), is about 150,000 times that of the water; in caddis fly larvae, *Hydropsyche cockerelli* Banks, the concentration factor is about 350,000.

Differences in activity density of the various aquatic forms (Fig. 1) also result from dissimilarities in chemical composition or physiological demands for specific elements. The great concentration of radiophosphorus reflects the paucity of phosphate ion in Columbia River water (about 0.03 ppm) and the high demand for this ion by the organisms. The isotopes are likewise deposited in particular tissues according to need. Since $P^{32}$ is responsible for nearly all of the radioactivity in fish, the phosphorus-rich structures, such as bone and scales, are most radioactive, while muscle and fat are least active (Fig. 2).

Similar biological concentration of phosphorus was found by Krumholz[2] at Oak Ridge. Radiostrontium, radiocesium and some rare earths were also selectively concentrated from the Oak Ridge waste, which contained fission products. Obviously the radioactivity of aquatic life is dependent upon both the chemical nature of the radioactive materials and the physiological characteristics of the individual species.

## MECHANISMS OF ACCUMULATION

Adsorption to biological surfaces undoubtedly plays an important role in the uptake of radioisotopes by aquatic organisms. The high activities found in Columbia River plankton and sponge (Fig. 1) are associated in part with their extensive surface areas available for adsorption. Surface textures, such as gelatinous coverings, and surface flora of bacteria may further modify adsorption characteristics.

Radioactive materials which readily diffuse through living membranes are picked up in substantial quantity directly from the water. In aquatic plants, which build up protoplasm by photosynthesis, this includes all of the essential inorganic ions and certain organic compounds. Since the animal membranes are more selective, only a few radioisotopes are absorbed directly in significant amounts. Fish immersed in Hanford reactor effluent, for example, concentrated the $Na^{24}$ about 130 fold, but direct absorption of other isotopes from the water was inconsequential. Adsorption of $Na^{24}$ through the anal gills of mosquito larvae was demonstrated by Treherne.[3] Metabolic processes are thus of greater importance than adsorption in the uptake of isotopes by most aquatic forms.

Since the animal forms must obtain the bulk of their essential elements from their food, originally from the green plants, food chains are of paramount importance in the accumulation of radioisotopes. Each organism acts as a reservoir, which decreases the rate of transfer of radioactive substances to higher

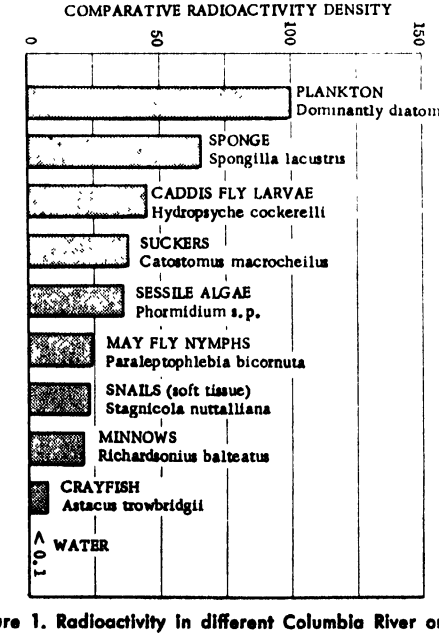



**Figure 1. Radioactivity in different Columbia River organisms**

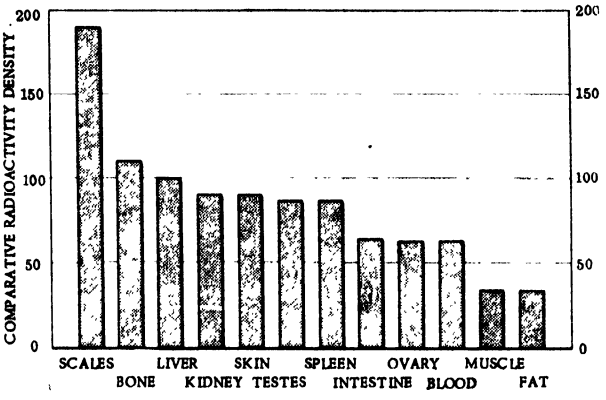



**Figure 2. Radioactivity in different tissues of Columbia river fish**

forms. Our laboratory studies[4] indicate that plankton requires about 15 hours to reach maximum concentration after introduction of radiophosphorus, although more than one-half of this is taken up within the first hour. Accumulation of $P^{32}$ in sessile algae and bottom organisms is considerably slower and small fish reach maximum concentration after about two weeks. Radiophosphorus introduced into natural ponds by Coffin *et al.*[5] did not appear in fish until about two days later. In the Columbia River, where shorter-lived isotopes predominate, the plankton seems to reach maximum activity density about one hour after floating into the zone containing the reactor effluent. The time lags inherent in the transfer of radioactive materials from one form to another allow decay of the shorter-lived isotopes and consequently a general reduction in activity density along food chains.

## SEASONAL VARIATIONS

The radioactivity in Columbia River water downstream from the Hanford reactors is directly related to the dilution factor. As shown in Fig. 3, it is highest during the fall and winter, when river flow is low, while during the spring freshet period, minimum activities occur. The activity density of the plankton (Fig. 3) follows that of the water since direct absorption and adsorption are involved. Substratal algae follow a similar pattern. With animals, which acquire most of their radioactive substances from their food, the pattern follows the feeding rate. Food consumption is influenced by metabolic rate and is largely temperature dependent. The seasonal change in radioactivity of a common Columbia River minnow (*Richardsonius balteatus*) is compared to temperature in Fig. 3. Activity density of the fish is low during the cold months when only a limited amount of radioisotope-bearing food is eaten; in the late summer, when temperatures and food consumption are at a maximum, the activity density level is high. This general pattern is also characteristic of all other aquatic animals studied, including insects, crustaceans and molluscs. Some deviation may occur, however, where complex life-cycles are involved. This is particularly true of immature insects which are less

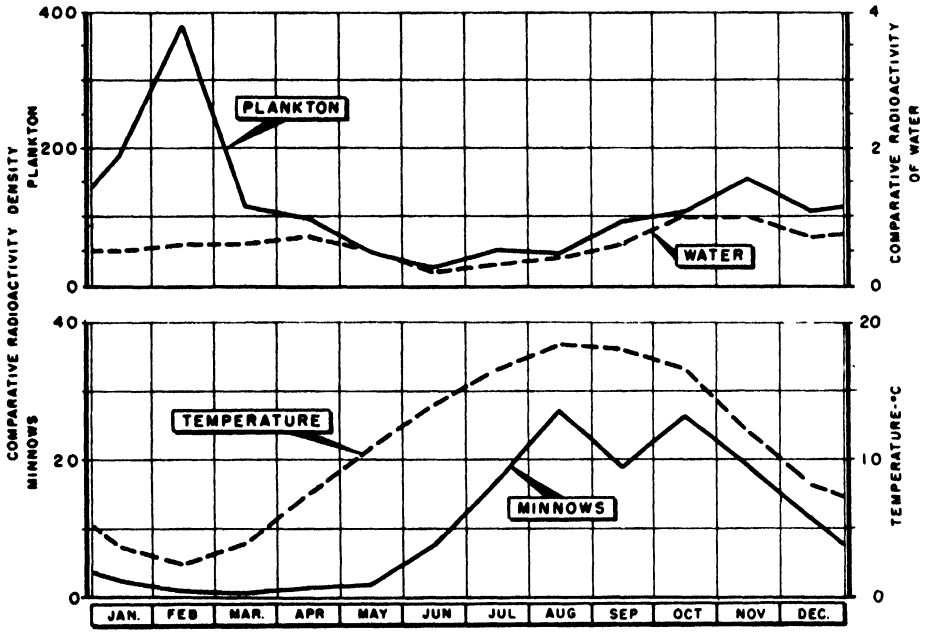



Figure 3. Seasonal fluctuations in radioactivity of Columbia river water and organisms

radioactive during resting stages than when the larvae or nymphs are feeding. The accumulation of radioisotopes is most rapid during periods of growth when mineral deposition occurs. Young, rapidly growing fish are thus more radioactive than old, mature forms.

## REDUCTION IN RADIOACTIVITY

A major factor in reducing activity density is decay of the short-lived isotopes. Since a large part of the radioactivity of Columbia River plankton originates from the very short-lived materials, its activity drops rapidly with travel time below the reactors as shown in Fig. 4. In higher forms, where the activity arises mainly from $P^{32}$ (14.3 day half-life) the reduction with distance is much less abrupt. There is some evidence that an appreciable fraction of the radiophosphorus is retained in the biomass of the river and thus is quite slowly transported downstream. Such retention permits additional radioactive decay so that downriver forms are less active than might be expected if only the time lapse for river flow is considered.

In certain unique situations, the establishment of aquatic communities to act as biological retention reservoirs for radioactive materials might alleviate undesirable conditions elsewhere and thus best serve the public welfare. Biological fixation of soluble radioactive materials may also help decontaminate drinking water since the radioactive materials bound to plankton can be filtered out in conventional water treatment plants.

Ultimately the radioisotopes still in solution or bound to suspended materials are swept into the sea. Some fraction of the radioactive material must also deposit with sediments, however. This would include the ions adsorbed on inanimate planktonic particles and dead organisms which settle to the bottom and excreta of larger forms. Although considerable deposition of silt occurs in the forebay of a dam fifty miles below the Hanford reactors, no long-term accumulation of radioisotopes is evident in the sediments. Small amounts of the radioactive materials are removed when insects emerge from the river, by terrestrial animals (including man) which feed on the aquatic life, and by use of river water for agricultural irrigation.

## RADIATION HAZARDS

Extensive studies have been made of the radioactivity in river organisms below the Hanford reactors since the radiation levels could not accurately be predicted. Results indicate that concentration of very short-lived isotopes is of limited consequence in higher organisms such as fish.

Radiophosphorus, on the other hand, is concentrated more than one hundred thousand times. Although the $P^{32}$ is highly concentrated, existing

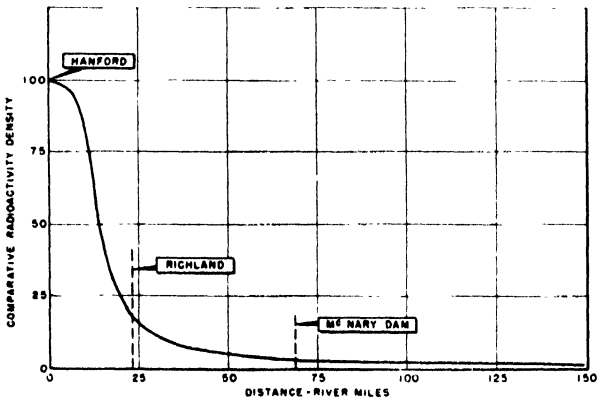



Figure 4. Diminishing radioactivity in plankton with distance downstream

amounts in the Columbia River are well below dangerous levels. Even in the most radioactive section, the young fish receive only about 0.1 rad per day from beta emitters—far less than the amount which would produce discernible damage. Both laboratory and field studies of river forms have shown no injurious effects from the presence of the reactor effluent. It is questionable that widespread decimation of aquatic populations will occur from radiation damage in situations where contamination levels in fish must remain below maximum permissible levels for human food. The difference in the activity density of Columbia River fish over that of the water, owing to the tremendous power of aquatic forms to concentrate some radioisotopes, illustrates the need for careful consideration of potential hazards prior to disposal of liquid wastes to public waters. If radiophosphorus were allowed to reach the maximum level permitted for drinking water, organisms living in the water would suffer radiation damage and the fish would be unsafe for human food.

The seriousness of radioactive contamination in an aquatic environment depends not only upon the quantities of individual isotopes which may be released, but also upon the physical, chemical, and biological properties of the water. Where contamination of a river or lake may be significant, careful investigation of each particular case is essential since complex biological processes may introduce hazards not included in such conventional limits as permissible concentrations for drinking water.

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# Biological Cycle of Fission Products Considered from Viewpoint of Contamination of Marine Organisms

**By Yoshio Hiyama,* Japan**

I

On the first of April 1954, the Fukuryu-Maru No. 5, a Japanese tuna long-line boat, operating long-line fishing in the Marshall Islands, was exposed to the fall-out dust of a hydrogen-bomb test at Bikini Atoll. Her crew was ignorant of the character of the dust falling on the deck, their clothing etc. The ship arrived back at the port of Yaizu on the 14th, and unloaded her catch of tuna at the fish market for sale. The fish had been distributed to various localities before one of the crew consulted a doctor and radioactivity was found on his body and clothing. Afterwards, the tuna in Yaizu and the fish markets concerned were inspected by the Government inspectors who detected the radioactivity with Geiger-Muller counters. Almost all of the tuna brought in by the fishing boat were recovered and disposed of.

Subsequently, as the people were so much frightened by the radioactive tuna, the government started radiological inspection, at the major tuna landing ports, of all fish brought in by tuna fishing boats from the area around the Marshall Islands and continued it until about the end of that year. As a result of these inspections, all fish showing radioactivity of more than 100 cpm by Geiger-Muller counter of the common $\beta$-ray type at a distance of 10 cm from the surface of the fish body were ordered disposed of. Contaminated fish accounted for slightly less than 0.5% of the total number of tuna examined. Some of these tuna were examined more closely at the laboratories of several institutes, and the results of the examination were in agreement in the points mentioned, roughly as follows:

In the early stage, during March and April, some of the sample fish were contaminated on the surface of their bodies (probably during the processes of handling after the fish were caught) by fission products which fell on the deck of the boat.

In the later period, the radioactivity was found internally, in the bodies of the tuna. The organ where the radioactivity was detected varied according to the time elapsed since the explosion of the bomb. At first, small fish, squids and other animals found inside the stomach, and consequently the walls of the alimentary canal were especially contaminated. And then, as the next step, liver, pancreas, and gills were markedly higher in radioactivity than other organs. And in the final stage, after June or July, the kidney had the highest activity among the contaminated organs. These three periods were not distinctly separated, but overlapped one upon another. However, the tendency was clearly observed in the species of the fish investigated here. In all cases, muscles were the least contaminated among the organs and tissues examined, though the dark meat was usually more radioactive than ordinary muscle.

Various kinds of fish were caught by tuna long-line and inspected. Among them, the species found most frequently contaminated were dolphin, *Coryphaena hippurus,* sail fish, *Istiophorus orientalis* and *Tetrapturus angustirostris,* and yellow-fin tuna, *Neothunnus macropterus.* It should be noted that these species are mostly carnivorous surface dwellers. Next to these were marlins, *Makaira mazara* and *M. mitsukurii,* Spanish mackerels, *Sawara niphnia* and *Scomberomorus chinensis.* In addition, swordfish, *Xiphias gladius,* big-eyed tuna, *Parathunnus sibi,* albacore, *Germo germo,* and skip-jack, *Katsuwonus vagans* were sometimes found to be contaminated. And a few blue-fin tuna, *Thunnus orientalis,* were contaminated. Numerous species of shark were caught frequently. On inspection they proved to be uncontaminated, although some of them were contaminated externally at their surface during the early period. It should be noted in considering the biological cycle of fission products in aquatic systems, that sharks are usually considered to rank higher than tuna and other carnivorous fish mentioned above.

The fishing grounds where these contaminated fish were caught were plotted on the map with regard to the date of catch for the convenience of seeing their seasonal movement. Though some difference is shown according to the species of fish, as the migration habit would differ from one other in a certain degree, the following general pattern of the movement is shown. In the early stage, namely, during March and April, most of such spots were confined within the area around the Marshall Islands, some west and some east of the target area. From May of that year, such spots moved westward to the area east of Luzon, and then moved northward to the area south of Japan along the route of Kuroshio current. But they do not extend further to the northern tuna fishing ground lying at about 40 deg N in the Northern Pacific

* Professor, University of Tokyo. This article is based upon numerous papers, prepared by several authors, which will be published by Science Council of Japan in the near future.

Ocean, from where only a few contaminated tuna were caught in the fall. From the above it can be considered that either the tuna which had been contaminated around the target area migrated along the route of Kuroshio current or the contaminated organisms to be the food of tuna migrated or drifted through or with the current.

During the summer months, the spots where the contaminated fishes were caught appeared again in the area around the Marshall Islands, and in the fall these spots were scattered over almost the entire fishing grounds of the North Pacific.

The tissues and organs which were found to have intensive radioactivity were chemically analysed in the laboratories of several institutes. The results of these analyses tell that most of the radioactive elements belong to Group III. Group IV was found in the stomach and pyloric coecum. Analysis of the muscle of exceptionally highly active specimens showed that the contaminating material was, in most cases, $Zn^{65}$, and it was so recognised also in the light of physical characteristics. So far as the muscle is concerned (at a time supposed to be some months after the detonation), the most dangerous radioactive element for food use, $Sr^{90}$, was found to contribute less than 1% of total activity.

II

The Japanese government wanted to know the oceanographical situation of the tuna fishing grounds related with the contamination by fission products, and sent the Shunkotsu-Maru, a governmental research boat with the expert personnel and scientific apparatus to the area around the Marshall Islands. She left Tokyo on May 15 and returned to the same port on July 4. The result of this survey tells that the fission products were carried by oceanic currents (North equatorial, South equatorial and Equatorial counter current). In most of the survey points where the contaminated sea water was found, plankton and other organisms, including tuna, were also found to be contaminated. By comparing the intensity of radioactivity, in counts per minute per unit weight, it was found that micro-plankton ranked first in intensity, followed by macro-plankton. Large-sized fish were generally less radioactive than the latter. The distribution of radioactivity in the bodies of tunafish was quite similar to that found in the case of the landed contaminated fish.

Later, a similar sort of the oceanographical survey was carried out by Kagoshima University. The Kei-ten-Maru, a university research vessel, sailed through the Caroline Islands to the Coral Sea from October 28 to January 28, 1955. The results of the survey show that the activity of sea water and of marine organisms had been weakened by diffusion and decay after the survey of the Shunkotsu-Maru. But a slight activity was found in the Coral Sea, probably brought in by the South equatorial current from the Marshalls.

Beside the above, the Daifuji-Maru, a research boat belonging to the prefectural government of Shizuoka, made a similar sort of survey with the staff of Nankai Regional Fisheries Research Laboratory of Fisheries Agency, by the route passing through the west of Bikini Atoll, from November 30 to February 18, 1955. She found less activity than Shunkotsu-Maru but, even at that time, a slight activity was found in sea water and plankton at several points directly west of Bikini.

III

With the object of ascertaining the marine biological cycle of fission products by analysing the facts obtained by laboratory investigations of contaminated fish and the above-mentioned oceanographical surveys, various sorts of aquarium experiments were carried on by groups of biologists to study the physiology of aquatic organisms with regard to metabolism of fission products. The materials used were the fall-out dust collected from the deck of the Fukuryu-Maru No. 5, and radioisotopes such as $Sr^{90}$. The results were utilized in the consideration of food hygiene.

The fall-out dust dissolved in sea water is easily made to adhere to the surface of the organisms as well as that of the water container. As the body surface per unit body weight is greater for micro-organisms than for macroorganisms, the uptake of radioactivity is higher in microorganisms. For the same reason, the gills of fish are more efficient than other parts of the body in absorbing fission products.

The uptake of fission products by fish from the surrounding water is not marked unless the products are present above a certain level. This level is higher in the case of sea water than in the case of fresh water, probably because the former originally contains a larger amount of the elements similar in physiological behavior to the fission products.

When culture phyto-plankton contaminated by fission products was given to zooplankton as food, the latter took up only a small part of the radioactivity. Similar results were obtained with organisms higher in the food chain. When fall-out dust was directly inserted into the stomach of the living fish, most of it was not absorbed through the alimentary tract.

$Sr^{90}$ given to the living fish either by feeding, or by injection or through the environmental water, goes mainly to the hard tissue, such as scale, bone, fin ray, gill and others.

IV

Fresh water systems, brackish water areas and marine coasts were also surveyed, to assess the effect of airborne fission products. The result of the survey shows that fission products fall on the ground either in rain or dust, are absorbed on the surface of the ground at the beginning, and then (already weakened) are carried to the heads of streams and rivers. In the lower part of the river therefore, the water has no remarkable activity even when the intensive radioactive rain falls. In a porcelain water pot, a small concrete pond and a small drinking-water reservoir

where there were inadequate numbers of microorganisms to noticeably reduce the concentration of fission products, some radioactivity was often kept by the water until it physically decayed. In large-sized natural ponds and lakes where various organisms were present in abundance, the fission products were retained by these organisms for a considerable length of time. Under such conditions, if the water is stagnant, activity decays naturally.

## V

On the basis of data obtained through the surveys and experiments mentioned above, we can consider the general features of biological cycle of fission products in aquatic systems, though each chemical element has its own peculiar selective behavior.

When fission products reach the water, first of all, the microorganisms such as bacteria, protozoa, phytoplankton and others take these up most remarkably. Thereafter, these are transmitted to the macroorganisms of higher rank in food order through the food chain. During this process, the activity is weakened by biological and physical decay as time passes, and also by the mechanism under which only a part of the fission products are taken up by the animal in each food step.

When the concentration of the fission products in the environmental water is not high enough, the food chain would be the main course of transmission of these materials among aquatic fauna and flora. However, if it is high enough, even animals ranking high in food order would take up these materials directly through their body surfaces, such as the gills of fish, more easily in fresh water than in marine environments.

These two routes of the transmission of fission products in biological cycle often exist at the same time in nature, and this makes the results of field surveys rather complicated.

# Nuclear Science and Oceanography

**By R. Revelle, T. R. Folsom, E. D. Goldberg, and J. D. Isaacs,* USA**

Our earth is unique among the planets in its possession of a world-encircling ocean. This watery cloak is intimately related to such diverse phenomena as petroleum, climates and life. Major obstacles to understanding of these phenomena arise from the magnitude and subtlety of oceanic processes.

Many scientific problems of the ocean can be resolved into questions concerning the fluxes within its realms. Promising possibilities exist for study of these fluxes by the tracer techniques of nuclear science, but to date the possibilities have been only timidly explored. A more vigorous and imaginative application of nuclear tools in the marine sciences would certainly result in important breakthroughs.

The practical problem of oceanic disposal of nuclear wastes involves nearly all phases of oceanography. Conversely the use of radioactive materials as tools in oceanography must lead to a better understanding of the disposal problem.

## NATURAL RADIOACTIVITY IN THE OCEAN

In considering possible applications of radioactivity to the study of the ocean, two facts are of prime importance: the very low level of natural radioactivity in the sea and the enormous volume of the ocean waters. Both of these facts are well illustrated in Table I.

The amount of radiopotassium per unit volume of average igneous rocks is about 100 times the amount in the ocean. Uranium in the rocks is about 3000 times as great as in an equal volume of sea water, radium about 10,000 times, and thorium a million times. On the other hand, the carbon-14 per unit volume of sea water is 300 times that in air and the total carbon-14 in the ocean is between 50 and 100 times the total in the atmosphere. Nearly all tritium is in the ocean, chiefly in the top 50 to 100 meters.[2]

Potassium-40, with a gamma-radiation energy of 1.5 Mev, is the principal radioactive nuclide in sea water. It yields about 100 times as many disintegrations per unit volume and time as the uranium and thorium series and 50 times as many as rubidium-87. The very low thorium-uranium ratio in sea water indicates that the effective "solubility" of thorium is far less than that of uranium. We would expect that the thorium isotope, ionium, one of the intermediate products in the uranium-radium series, would also have a very low concentration, and that for this reason the amount of radium would be much lower than the equilibrium value computed from the uranium content. Table I shows that this is in fact the case. Both uranium and thorium are effectively "saturated" in the ocean and the rate of precipitation in the bottom sediments is very nearly equal to the rates at which these elements are brought to the sea by rivers.

In contrast to the low level of activity per unit volume, the total amounts of radioactive nuclides in the ocean are very large, ranging from 460,000 megacuries for potassium to 8 megacuries for thorium. Fission of all the uranium-238 and -235 in the ocean would yield $2 \times 10^{33}$ ergs of energy, equal to 30 times the total amount of solar energy falling on the earth's surface each year, or $1.2 \times 10^{6}$ times the present annual consumption of energy by human beings and their industrial civilization (about $4.5 \times 10^{10}$ megawatt hours).[30]

## USE OF ISOTOPIC TRACERS IN THE MARINE SCIENCES

### Currents and Diffusion

Among the basic questions of the marine sciences are: where, and how fast and by what mechanism do the ocean waters move? We are interested in the well-defined transport of ocean currents and in the apparently random motion called diffusion. These two processes are related by the following equation, in vector notation:

$$\frac{\partial c}{\partial t} = \nabla \cdot A \nabla c + Q - v \cdot \nabla c \qquad (1)$$

where $c$ is the amount per unit volume of something present in the water, $\partial c/\partial t$ the time rate of change of concentration at a fixed point, $v$ the vector current velocity, $A$ the effective diffusion coefficient, and $Q$ a source function. For radioactive substances that are unaffected by biological activity, $Q = \lambda c$, where $\lambda$ is the radioactive decay constant.

We may conveniently think of the ocean outside the Arctic and Antarctic as a two-layer system, consisting of (1) an upper layer, ten to several hundred meters thick, in which the temperature is constant with depth during winter months, vertical diffusion is rapid, and currents vary markedly in speed and direction over short periods of time, and (2) a lower layer, several thousand meters thick, in which temperature decreases with depth nearly all the way to the bottom. This layer is stably stratified, that is the density of the water increases and the entropy decreases with depth, and vertical turbulence appears

* Contribution (New Series, No. 794) from the Scripps Institution of Oceanography, University of California, La Jolla, California.

Table I. Radioactivity of Sea Water

| Nuclide | Concentration (gm/cm³) | Specific activity (number of disintegrations/cm³/sec) | Total amount in ocean (megatons) | Total activity in ocean (megacuries) | Energy of γ-radiation (Mev) |
|---|---|---|---|---|---|
| $K^{40}$ | $4.5 \times 10^{-6}$ | $1.2 \times 10^{-2}$ | 63,000 | 460,000 | 1.5‡ |
| $Rb^{87}$ | $8.4 \times 10^{-8}$ | $2.2 \times 10^{-4}$ | 118,000 | 8400 | no γ |
| $U^{238}$ | $2.0 \times 10^{-9}$ | $1 \times 10^{-4}$* | 2800 | 3800 | .05 – .82 |
| $U^{235}$ | $1.5 \times 10^{-11}$ | $3 \times 10^{-6}$* | 21 | 110 | .06 – .18 |
| $Th^{232}$ | $10^{-11}$ | $2 \times 10^{-7}$* | 14 | 8 | .03 – .08 |
| $Ra^{226}$ | $3.0 \times 10^{-10}$ | $3 \times 10^{-5}$* | $4.2 \times 10^{4}$ | 1100 | .18 – .60 |
| $C^{14}$ | $4 \times 10^{-17}$ | $7 \times 10^{-6}$ | $5.6 \times 10^{-5}$ | 270 | no γ |
| $H^{3}$† | $8 \times 10^{-20}$ | $2.5 \times 10^{-5}$ | $1.5 \times 10^{-9}$ | 12 | no γ |

*Activity of nuclide + daughter products. †Only in top 50–100 meters of the ocean. ‡ $\gamma/\beta = 0.1$

to be virtually non-existent. The lower layer seems to behave as if it consisted of a series of separate sheets, of great horizontal extent and extreme thinness, piled up and moving quasi-independently.

In the open ocean, current speeds in the upper layer vary from a few centimeters to one or two hundred centimeters per second. The currents in the lower layer are presumably smaller but little is known about them.

Up to the present, radioactive tracer techniques have not been much used to study the currents in the upper layer, although their application has been encouraged by the pioneer studies of Miyake and coworkers.[1] Nevertheless, they have furnished important information about the diffusion. Von Buttlar and Libby[2] measured the amounts of tritium present in rivers, ocean surface waters, and in rain. Assuming that storage occurs on land and in the ocean but not in the atmosphere, their results can be expressed approximately in the form

$$\lambda ch = P = R_s\,(T_s - 0.25) \qquad (2)$$

where $R_s$ is the average rainfall over the sea in cm/sec, $T_s$ is the average tritium content of this rain in atoms/cm³, $P$ is the rate of tritium production by cosmic rays, in atoms/cm² sec, and $c$ is the tritium content of sea water in atoms/cm³, assumed uniform from the surface to a depth $h$. Von Buttlar and Libby found a best value for $P$ of 0.14 .Taking $c$ as about $1.7 \times 10^4$, the weighted average of their measured values for surface sea water, and $\lambda$ as $1.7 \times 10^{9}$ sec$^{-1}$, $h$ becomes 50 meters. This is very roughly the average depth of the upper layer in the sea. We must conclude that over times of the order of 12 years, the half-life of tritium, there is little interchange between the upper layer and the waters at greater depth.

One of us (Folsom) has investigated the vertical mixing in the upper layer with the aid of artificially radioactive substances (fission products). These were introduced at the surface in an area where the upper layer was about 100 meters thick. The rate of downward motion of the lower boundary of the radioactive water was approximately $10^{-1}$ cm/sec$^{-1}$. This motion ceased abruptly at the bottom of the upper layer.

Horizontal diffusion in the relatively shallow waters of Bikini lagoon was studied by Munk, Ewing and Revelle[3] using the observed spreading of radioactive material (fission products) introduced more or less uniformly through a column of water 60-meters deep. The average value of the horizontal component of the effective diffusion coefficient, $A_h$, during three days in which the radius, $r$, of the radioactive area increased to about 4 kilometers, was found to be $1.5 \times 10^5$ cm²/sec$^{-1}$, and $A_{h/r}$ was close to 0.5 cm/sec$^{-1}$. This value of $A_{h/r}$ is in good agreement with estimated values of this ratio, obtained by other means elsewhere in the ocean, over a range from $10^3$ to $10^8$ cm in $r$.

The stably stratified waters below the upper layer are believed to move in a series of great current systems superimposed on one another. Little is known about any of these systems but it is generally assumed that currents in the deep and bottom waters have a significant meridional component (north-south or south-north). These waters sink from the surface in the Arctic and Antarctic, where vertical density stratification is minimal or lacking. They move slowly to lower latitudes under the overlying less dense waters, then slowly return to very high latitudes where they are again mixed with surface waters. At the surface, they come into at least partial equilibrium with the carbon dioxide in the atmosphere, and hence with the atmospheric carbon-14. After the waters have sunk they retain the carbon dioxide acquired at the surface, but the carbon-14 slowly decays. Because of the density stratification, vertical diffusion can be neglected, and if we also neglect horizontal diffusion, and assume that a steady state exists, Equation 1 is reduced to:

$$v \cdot \nabla c = \lambda c \qquad (3)$$

Analysis of carbon-14 in deep-water samples should therefore tell us something about the average meridional component of velocity from the source in high latitudes to the point at which the sample was collected. Kulp[4] has attempted measurements of this type. The preliminary results are complex but not inconsistent with a meridional velocity between 1 and 0.01 cm/sec. Estimates of this velocity component by other methods give values of the order of 0.1 cm/sec, that is, a round-trip time of several hundred years for the deep waters, from high latitudes to the tropics and back again.

The authors have recently studied the processes occurring when sea water containing mixed fission products is introduced into the lower layer. The radioactive water was observed to spread laterally in a sheet, with a thickness of the order of a meter and an area of 100 or more square kilometers, at a depth determined by the vertical density distribution, that is the radioactive water spread outward along an iso-density surface. The center of the sheet moved horizontally with about the same velocity as the waters just above and below it. Vertical mixing, at least in the upper part of the lower layer, evidently is very slow.

The extreme vertical stability of the lower layer gives promise of the use of relatively small amounts of radioactive substances as a tool for studying the horizontal diffusion and the horizontal and vertical components of the currents at depth. Because the water behaves as if it consisted of thin, independent sheets, we are concerned chiefly with the two-dimensional spreading of radioactive material. We need to follow the radioactive water from shipboard, and experience shows that to do this successfully we must be able to detect the introduced radioactivity by lowering a measuring instrument into the water from the ship.

To obtain some insight into the possibilities of such experiments, let us suppose that $N$ curies of a gamma-radiating nuclide are introduced in solution at a point in the lower layer and the solution spreads vertically through a depth $h$cm. Over what area can it spread horizontally and still be detected by *in situ* measurements with a probe immersed in the radioactive water? Let $r$ be the radius of the area over which the radioactive material can spread and yet be detected. The cosmic ray background may be virtually neglected below a few tens of meters. Let $t$ be the counting time, $V$ the equivalent volume of the detector, $e$ the counting efficiency, $B$ the specific gamma activity of natural sea water in photons/cm$^3$/sec$^1$, and $n$ the fractional error in measurement of introduced radioactivity. For 95% probability that a measured value gives the concentration of the introduced radioactivity with an error less than $n$, we have, if we neglect the decay of the radioactive material:

$$\pi r^2 = \frac{3.7 \times 10^{10} NVten^2}{2h\,[1 + (1 + 2n^2 BVte)^{1/2}]} \qquad (4)$$

This equation rests on the well-known fact that counting errors are inversely proportional to the square root of the number of counts. If the background is continuously measured, the time limitation does not apply to background measurement. The denominator of Equation 4 becomes $2h\,[1 + (1 + 2\,n^2\,BVte)^{1/2}]$. This increases $r$ (max) roughly by 1.2.

If possible, the error in measurement of the introduced radioactivity should not exceed 50%, that is $n \leq 5$. For measurements *in situ*, the counting time is limited by the difficulty of maintaining an instrument suspended at a fixed depth in the sea from a rolling and drifting ship. Other practical considerations limit the speed with which the probe can be raised or lowered to not less than 10 cm-sec$^{-1}$. The largest practical value of $h/t$ is therefore 10, and, if possible, $t$ should not exceed 5 sec.

With the instruments described below, $e = .03$ and $V$ about $10^3$ cm$^3$. From Table I, $B = 1.2 \times 10^{-2}$ photons/cm$^3$-sec$^{-1}$. A "reasonable" value for $N$ is 10 curies. Our experience suggests that $h$ will not exceed 100 cm within the duration of the experiment.

With $t = 5$ and the other stated numerical values, $\pi r^2 \leq 2.9 \times 10^{10}$ cm$^2$, and $r$ (max) is slightly less than 1 km. The instrument will record only 19 counts during the counting interval. Of these, 0.2 counts will be from the oceanic background. Unless great care is exercised, the internal background of the instrument will be a serious limiting factor.

The situation can be improved by use of a scintillation-type detector with $e$ about .25. For this value of $e$ and $t = 10$, the maximum detection area becomes 23 km$^2$ and $r = 2.7$ km. For $n = 1$, that is for a 95% probability of detecting the radioactivity but without a reliable measure of its concentration, the maximum detection area is 52 km$^2$ and $r$ is 4.1 km.

If we employ one of the nuclides listed below (Table II) such as rubidium-86 with a gamma energy of 1.1 Mev, and the instrument is capable of discriminating against the 1.5 Mev radiation from potassium-40, the permissible dilution can be increased. Gates and Eisenhauer[5] have computed the energy spectrum of the scattered radiation from single frequency sources. They show that for a source energy of 1.5 Mev the proportion of energy in a 0.2 Mev band around 1.1 Mev is about 10%, whereas for a 1.1 Mev source 50% of the energy will be in this band. With proper counting conditions it should be possible to triple or quadruple the detection area.

Thus far we have not considered the decay of the introduced radioactive material. Because of the linear relation between $A_h$ and $r$, the duration of the experiment will vary almost directly with the radius of the detection area. For the computed values of $r$, the observation time probably cannot exceed a week, so that with nuclides of suitable half-life the decay will not be very significant.

We have given some thought to a single nuclide that might be used for experiments of the type described above. The prerequisites for such a tracer include a half-life compatible with the time constant of the vertical and horizontal motion of the water and the diffusion, and yet not so long as to engender potential hazards to health through contamination of marine products of commerce or beaches. Further, the half-life must be such that delivery from the production site to the point of the experiment can be made without excessive decay. Such considerations indicate that the half-life should be of the order of a week to a month. The tracer should form a readily soluble ionic species in sea water, so that the volume of material handled can be minimal and precipitation and settling out of colloidal particles from the radioactive layer will not occur. Elements of groups IA,

Table II

| Isotope | Half-life | Cost per curie | Specific activity available | Gamma energy Mev |
|---|---|---|---|---|
| $Rb^{86}$ | 19.5 day | $1000. | 9 mc/gram | 1.1 |
| $I^{131}$ | 8.0 day | 750. | Carrier-free | 0.36 and 0.72 |
| $Ba^{140}$ | 12.8 day | 500. | Carrier-free | 0.16 and 0.54 |

IIA, VA, VIA and VIIA in the periodic table are preferable. The tracer should be cheap and readily available in multi-curie amounts. Consequently it should either be produced in a pile by neutron-gamma reactions or be a fission product that is readily separable. High specific activities are desirable. The tracer should be a gamma-ray emitter with energy between .2 Mev and 1.3 Mev so that *in situ* assays can be made readily and discrimination of the isotope from naturally occurring nuclides can be accomplished. Three promising isotopes are listed in Table II. Rubidium and iodine decay to stable daughters, while barium is parent to gamma-ray emitting $La^{140}$, which increases the total activity after a few days.

### Composition of Sea Water and Marine Organisms

The concentration of minor elements in sea water is not accurately known, in fact many elements are present in such small amounts that they are undetectable by older analytical methods.

Assay of trace elements in the concentration range of parts per billion or less has been advanced by two new analytical techniques, activation analysis and isotope dilution. Cesium, arsenic and rubidium in marine waters were determined by the former method by Smales and his co-workers[6,7] using the Harwell pile. An imaginative extension of this technique was employed by Stewart and Bentley,[8] who measured the uranium content of Pacific Ocean waters by counting the fission-fragments from uranium in sea salt irradiated in the Argonne heavy-water reactor CP-3. The isotopic dilution technique was applied by Rona and co-workers[9] to obtain the uranium content of Gulf of Mexico and North Atlantic waters. Such new developments as pile activation of samples followed by gamma-ray spectroscopy should further advance our knowledge of the chemical composition of the ocean.

Many elements known to be present in trace amounts in sea water and some, such as thallium, cadmium and zirconium, which no one has been able to detect, exist in high concentrations in marine organisms. What are the paths by which these trace metals are concentrated in living creatures?

Two general processes for the transfer are evident. The first involves direct uptake of dissolved substances. For example, vanadium exists in the oceans to the extent of 1 microgram per liter, while the blood of certain ascidians (sea squirts) can contain up to 10% vanadium, a concentration of a hundred million. By the use of cyclotron-prepared radiovanadium, Goldberg, McBlair and Taylor[10] were able to show that these animals can assimilate ionic vanadium from sea water and that the reactive site for the vanadium uptake is the mucus. Such accumulation parallels the uptake of iodine by the thyroid, and its specificity equals that of the best analytical reagents devised by man.

The mucus of marine pelecypods has been shown to be the adsorbing agent for strontium-90 by Fretter[11] and for calcium-45 by Rao and Goldberg.[12] Spooner[13] treated species of algae with carrier-free radioactive strontium and yttrium. Brown seaweeds extracted strontium upon their surfaces by factors of 10 to 40 over sea water, while red algae took virtually all the ionic yttrium from the water.

A preliminary survey[14] indicates not only that trace metals in marine organisms are retained by fairly strong chemical bonds but also that the relative degree of concentration of ions follows the order of the relative stability of these ions in organic complexes as given by Schubert.[15] Fission products consist dominantly of elements that form such relatively strong chemical bonds with organic compounds. The Noddacks[16] found for elements within the fission product spectrum the following enrichment factors for wet weight of marine animals compared to sea water: Mo, 600; Ag, 2000; Zn, 3250; Cd, 450; Ge, $> 760$; Sn, 270; As, 330; and Sb, $> 30$. Black and Mitchell[17] found the following concentration factors for algae: Zn, 400–1400; Mo. 2–15; Sr, 8–90. In preliminary analyses we have found that organisms living in waters containing mixed fission products concentrate the radioactivity by a factor of approximately 1000.

Further understanding of the intimate chemistry of these powerful concentration processes would be important not only to biologists interested in metabolism, but also to inorganic chemists seeking specific reagents for analytical methods. Knowledge of the functional groups within mucus and plant slimes may best be obtained by already familiar tracer techniques.

Marine organisms also accumulate trace elements by the assimilation of particulate matter. Iron, a necessary nutrient for plants, exists in the ocean principally in colloidal form. Goldberg,[18] utilizing radioactive iron, demonstrated that growing marine diatoms (microscopic plants) assimilate particles of hydrated iron oxide, but do not recognize ionic iron in complex form such as citrate, ascorbate or humate. Zirconium, titanium and other anions are scavenged from sea water by particles of hydrated oxides of iron and may be incorporated into plants with the iron. Such reactions could explain the high amounts of titanium and zirconium reported in "red tide" waters near the west coast of Florida in October and November, 1952, by Collier.[19]

### Organic Productivity

Organic productivity in the ocean is among the major problems of marine biology. Fundamental to

this problem is the determination of the amount of organic matter photosynthesized from inorganic substances by the marine autotrophic plants, the basis of life in the sea. Nielsen[20] developed a powerful and elegant technique for this determination, using carbon-14. He was concerned primarily with the microscopic plants that make up the plant plankton of the open sea. In Nielsen's method, tracer amounts of radiocarbon in the form of bicarbonate are added to a sample of water and plant plankton, freshly collected from a point in the sea. The sample of sea water and plants is placed in a transparent glass bottle and lowered to the depth in the ocean from which it was taken, or placed in a water bath illuminated to the light intensity at that depth. After a measured time interval, the water and the contained plants are separated by filtration. The radioactivity of the filtered organisms gives a direct measure of the amount of carbon assimilated in photosynthesis.

From measurements of this type in many parts of the ocean, Nielsen[21] estimated that the average net photosynthetic production is about $1.5 \times 10^{-10}$ gms/cm$^2$/sec or a total net production for the whole ocean of about $1.5 \times 10^{10}$ tons of carbon per year. This total production is about the same as that estimated for the lands of the earth. In this study Nielsen mapped such marine deserts as the Sargasso Sea, where his measured productivity was less than $.5 \times 10^{-10}$ gm carbon/cm$^2$/sec, and such oceanic "Edens" as the waters of the Benguela current on the west coast of Africa, where he found apparent rates of production of $25 \times 10^{-10}$ gm carbon/cm$^2$/sec. Other workers, notably Riley,[22] using quite different techniques, have arrived at total productivity estimates for the ocean that are an order of magnitude higher than Nielsen's. The discrepancy is not resolved.

### Beaches and Sedimentation

The sedimentary cycle, in which continental rocks are eroded and their components transferred via the ocean to sedimentary deposits, can be considered as a set of fluxes. Radioactive substances are naturally present or can be incorporated in these fluxes and can be used as tags.

In Santa Barbara, California, there is a destructive longshore transport of beach sand, estimated by Johnson[23] to be approximately 770 cubic yards per day. To prevent beach erosion, the path of the sand must be ascertained before remedial measures can be taken. Tracer sand grains for this purpose must be easily and rapidly distinguishable from the mass under study. Further, the size distribution, density and shape of the grains must be close as possible to those of major components of the sand. In the past, various nonradioactive tracers have been employed for problems of this type, but these have suffered from a number of inadequacies. The densities and other properties of colored glasses or rare minerals are usually so unlike the sand under study that they give erroneous pictures of the transport. Dyed or etched components of the sand itself are difficult to distinguish from the untreated grains, and the treatment is usually ephemeral.

In an attempt to obtain a more suitable tracer, Goldberg and Inman[24] irradiated quartz grains in the Oak Ridge National Laboratory slow-neutron pile. They found that the only significant induced activity is that of phosphorus-32. This nuclide is a pure beta emitter with a half-life of two weeks. By incorporating the activated quartz in natural sands, and by using radioautographic techniques, one active grain in one million inert grains is readily distinguished. In such an area as Santa Barbara, it should be possible to trace the sand transport over a 30-day period to dilutions of one in a million using 13 millicuries of irradiated material. This amount, after dispersal over the beach area, would certainly not create a health hazard. Quartz samples from various parts of the world show adequate phosphorus contents for the technique. Quartz is usually one of the principal components of sand and can be separated easily in a pure state by heavy-liquid, magnetic or electrostatic methods.

Natural radioactive substances have been used to determine the rates of accumulation of deep sea sediments. In these sediments, the radium content is commonly much higher than the equilibrium value for the uranium. As shown by Holland and Kulp,[25] the sedimentary particles probably extract from the sea water, by processes of base exchange and adsorption, both radium itself and its precursor, the thorium isotope, ionium. Ionium has a half-life of $8.3 \times 10^4$ years, while deep sea sediments accumulate at rates of the order of 1 cm/1000 years. The radium in equilibrium with the ionium ought therefore to decrease to about half the surface value at a depth in the sediments of the order of one meter.

In its simplest form the method of determining the rate of sedimentation depends on the assumption that migration of the radioactive elements does not take place after the sediments are deposited. Indications that migration does in fact occur were obtained by Arrhenius and Goldberg.[26] In studies of the radioactivity of cores of deep sea sediments from the Pacific Ocean, they found that sedimentary layers with a high concentration of radium also contain an abundance of the zeolite, phillipsite. This mineral forms within the sediment and as it forms, radium must be accumulated within it from the interstitial water. Apparently the resulting concentration gradients of radium in the interstitial water bring about a migration to the phillipsite-rich layer.

### Interactions between the Ocean, the Atmosphere, and the Lithosphere

The ocean acts as an aqueous reservoir for many of the components of the atmosphere. New techniques, using stable and unstable isotopes, are becoming increasingly useful in studying the interactions between the two great earth fluids. We shall select one example.

The interchange of carbon dioxide between the at-

mosphere and the ocean was recently elucidated by Suess.[27] He studied the carbon-14 content of tree rings, in an attempt to ascertain whether the carbon dioxide in the atmosphere has been increased by the combustion of the fossil fuels. The specific activity of carbon-14, corrected for decay, is apparently a few per cent less in tree rings grown in the present time than in the 19th Century. This dilution of the cosmic-ray produced activity coupled with the well-known quantities of combusted fuels leads to an average lifetime for the $CO_2$ molecules in the atmosphere and terrestrial biosphere of 20 to 50 years before solution takes place in the ocean.

The origin, evolution and geologic history of the ocean are fundamental problems in the earth sciences and biology. It is apparent from differences in abundance of the rare gases in the earth and the stars, that our ocean, as well as our atmosphere, evolved during a secondary stage of earth evolution. The elements in the present day ocean were not initially retained as an atmospheric blanket but were held chemically within the earth's interior. On the other hand, the amounts of these elements now present on the earth's surface far exceed the amounts that could have been produced by weathering of igneous rocks. When and how did these materials seep out of the earth?

Perhaps the processes giving rise to the ocean and the atmosphere are recorded in the isotopic composition of the interacting phases. Craig[28] has begun to investigate the variations in the ratio of $C^{12}$ to $C^{13}$ in all types of natural material, in an attempt to determine this ratio in primordial carbon and the rate of outflow of carbon from the earth's interior. This attempt, if successful, might tell us how the compositions of the ocean and the atmosphere have varied throughout geologic time.

Thode and his co-workers[29] studied $S^{32}/S^{34}$ ratios in a variety of marine sulphate and sulphide deposits. In reactions involving sulphate-sulphide equilibria, the sulphates become enriched in the heavier isotope. Biological processes provide the mechanism for the oxidations and reductions undergone by the sulphur isotopes. The first significant indications of isotope fractionation are found in pyrites and limestone sulphates formed circa 700–800 million years ago. Thode concludes that marine autotrophic organisms may have arisen during this era.

## INSTRUMENTS FOR SURVEY OF RADIOACTIVITY AT SEA

The present Scripps Institution equipment for surveying ocean radioactivity was designed primarily for economy and reliability under severe shipboard usage; nevertheless, it has considerable flexibility in range and sensitivity and is readily adaptable to several types of ocean measurements—above water—on the sea-surface—and under the surface to depths of about 1000 meters. The detector is designed to withstand hydrostatic pressures at 2000 meters and hence underwater measurements are limited to determination of fairly hard gamma constituents—those capable of penetrating the steel shell of the detector. This limitation is not a severe one.

Requirements peculiar to shipboard use led to the choice of halogen-filled Geiger counters. In one class of instruments, ten or more slender Geiger tubes of a type (Anton #315) designed for oil-well exploration are combined in parallel so as to feed their pulses to a simple pulse-shaper and amplifier. The detector reports activities corresponding to local gamma intensities from 100 mr/hr down to the cosmic ray background in air. This range is divided into six minor ranges by modifying the pulse-amplifier gain.

For measurements of higher activities, the conventional Geiger tubes are replaced by a group of ten or more gas discharge tubes of the "integrator" type which (1) are undamaged by high fluxes, (2) report by means of currents so high that no amplifier is needed and (3) compress their signal report of a very long range of gamma intensities into a single logarithmic response signal (four to five decades).

Either of the two discharge tube arrays can be used interchangeably in several tubular protective shells. These shells also contain batteries, cable terminals, and range selector sensitivities. They are roughly two meters long, with an outside diameter of 8 cm. The cylindrical walls are about 7-mm thick; both ends are fitted with removable plugs sealed with "O" rings. One end-plug joins a cable which brings the signal to an electrical recorder on deck.

For vertical sounding, the cylindrical detector assembly is attached to a multiconductor cable, about 1000 meters long and 7 mm in outside diameter, protected externally by two layers of steel wire armour. This cable is wound in and out on a small winch which is driven by an oil transmission system, in order to escape induced signals. The winch is provided with amalgam slip-rings permitting continuous electrical connection of the detector through four conductors to the recording devices on deck. We have called this combination a radiological sonde or probe. It permits rapid survey of water to the depth of the cable (Fig. 1a). It may also be towed aft of a ship steaming at full speed, and then records the horizontal profile of activity in the near-surface water.

The radiological sonde contains a pressure indicator (a bourdon-actuated potentiometer) so that a signal reporting the depth is sent up one of the conducting wires. This permits the radioactivity to be measured as a function of depth independent of the ship's speed, the amount of cable out, or the state of the sea and weather.

The assembly also carries a resistance-bridge temperature indicator which reports on a separate conductor to the deck.

The cable terminates in the ship's laboratory in a pair of recorders, each of which plots with a pen on chart paper the depth signal on the vertical coordinate ($Y$ axis). One recorder plots the radiation signal on its $X$ axis; the other plots the temperature signal. Consequently as the probe is lowered or

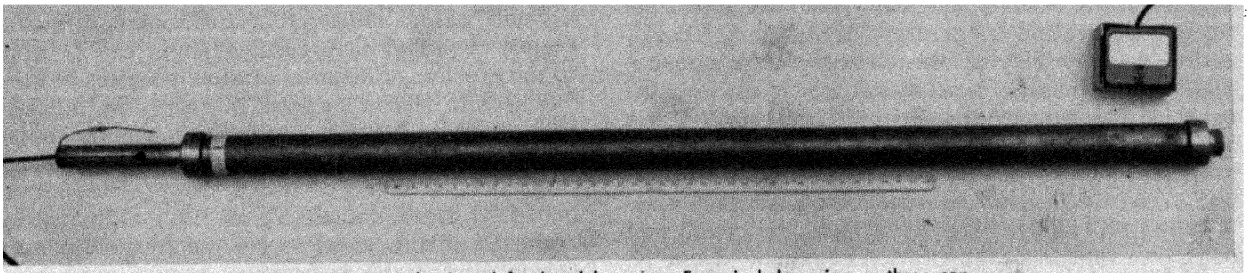


Figure 1. (A) A sonde rigged for hand lowering. For winch lowering, a three conductor armored cable is used rather than this rubber covered cable. The special winch permits deck recording of radioactivity vs depth and temperature vs depth;

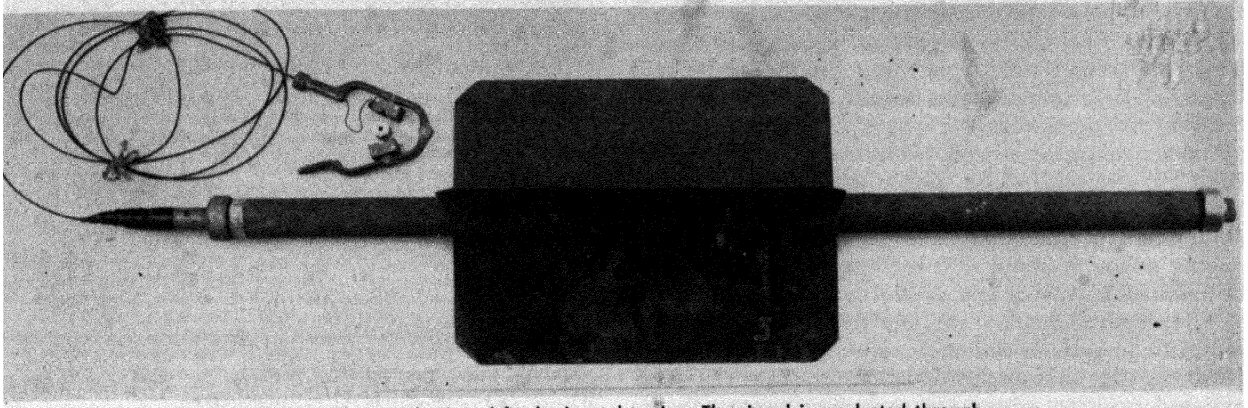


Figure 1. (B) A sonde rigged for horizontal towing. The signal is conducted through the armored pennant (on the left) through the special swivel and perforating clamp, and into the plastic coated towing cable. This arrangement permits fast towing of sondes at five or more different levels, transmitting to a single deck recorder

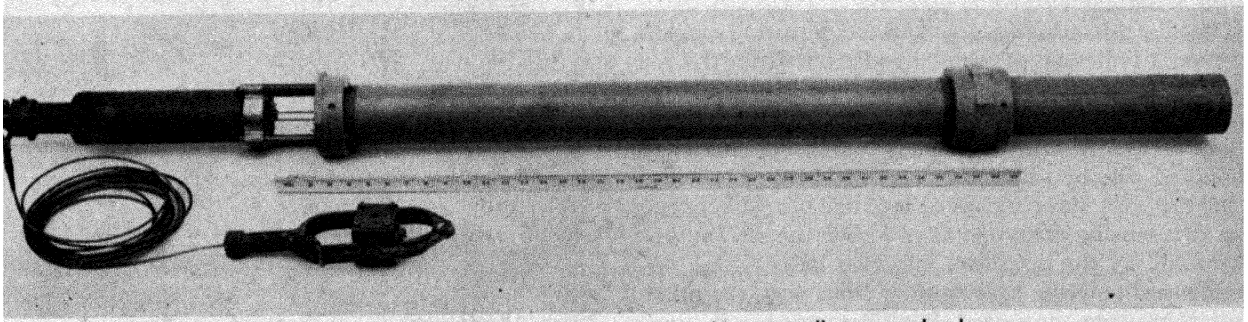


Figure 2. (A) A sampler rigged for towing. This can be set to collect a sample when a predetermined level of activity is encountered. It is towed from a depressed cable by the swivel clamp, which is rigged here for towing but not signalling;

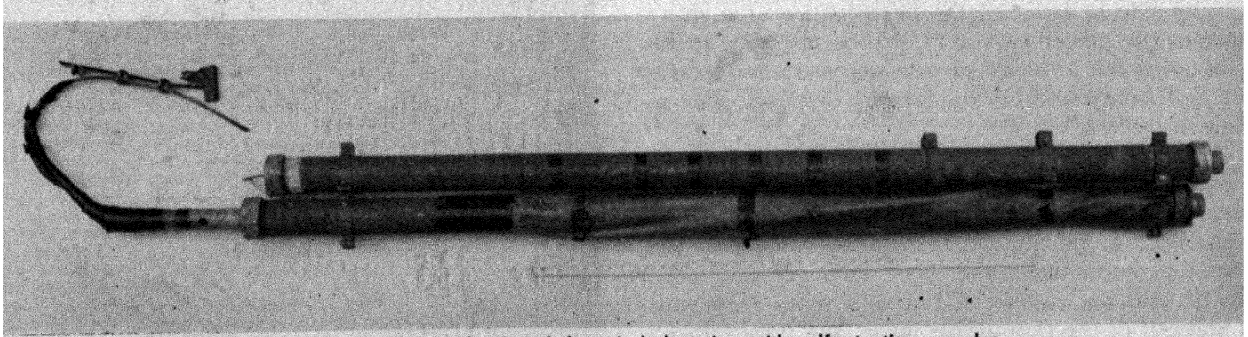


Figure 2. (B) The sonde rigged for winch lowering with self-actuating sampler attached. Interchangeable components permit versatility and improvisation in the field (as in the above) to attack special measurement problems

raised, radiation and temperature are plotted directly against depth. Range, sensitivity, and response rate of the pressure and temperature devices can be modified widely by varying the circuit parameters, but none of the precision traditional in physical oceanography has yet been attempted. The temperature and pressure plots serve mainly to round out the graphical picture of the radioactivity in the water, and to aid in rapidly locating a radioactive layer.

When provided with a set of fins, the cylindrical detector assemblies can be towed in groups of two or more behind the ship, attached (by means of special swivel clamps and short pennants) to a heavy cable kept submerged by a mechanical depressor (Fig. 1b). In this use the temperature element is replaced by a simple electronic integrator circuit which converts the Geiger tube signal into time-spaced signal "pips." These low voltage, time-spaced pips can be transmitted up an ordinary heavy towing cable to a suitable deck recorder, provided the towing cable has been covered with a thin polyvinyl or nylon coating.

This depressed-towing technique is useful when continuous measurements are desired at two or more depths down to about 400 meters. Such depths can be obtained at towing speeds of 7 to 10 knots (Fig. 5).

The vertical sonde assembly may be used as an indicator of gamma radiation coming through the air from the sea surface, provided it is mounted in an exposed position where the radiation can strike it. Our practice has been to mount the unit on the side of the ship about one meter above the sea surface, and to record its signal continuously against time with a suitable electronic recorder.

Beside this running log of surface activity, a directional indicator of gamma radiation from the sea surface is useful in detecting boundaries of active sea water. This consists of a pair of sensitive Geiger tube arrays (each having 10 tubes in parallel) mounted side by side on an elevator part of the ship near the bow. Four inches of lead are placed between the two sensing arrays so as to divide the sea surface "viewed" by the detectors into two halves. One detector sees only the rays coming from port, the other only those from starboard. Each detector has its own radiation indicator (in the form of a simple microammeter) and the two indicating meters are mounted side by side in front of the helmsman on the ship's bridge. The officer on watch obtains not only an indication of the presence of radioactive surface water but also immediate information as to which way to steer to reach it.

Ordinary water samplers may be attached to the electrical cable of the vertical sonde. However, in attempting to sample the radioactive layers at depth, it has been found difficult to trip the sampler by the usual messenger technique. The messenger moves too slowly, and the rolling of the ship jerks the probe in and out of the thin active layer. We have tripped the samplers automatically, using the radiation signal from the probe (Fig. 2).

## THE DISPOSAL PROBLEM

Some of the information and many of the techniques described in preceding pages can be applied to the problem of disposal of atomic wastes in the sea.

The rate of consumption of energy by human beings and their industrial civilization is doubling about once every 30 years. Let us assume a future energy utilization of about 2.5 times the present one, estimated by the Woytinskys[30] as $4.5 \times 10^{10}$ megawatt hours. If all this energy were obtained from nuclear fission, and if the radioactive wastes were dumped into the ocean after a month's storage, the total radio-

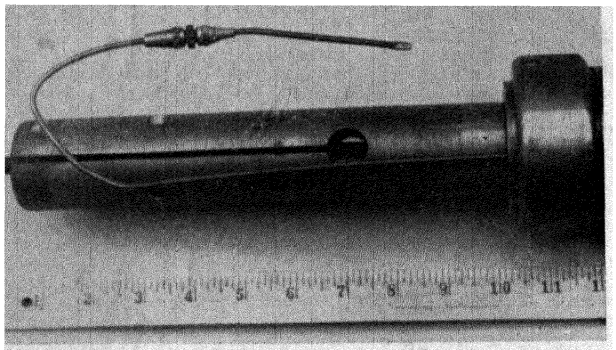


Figure 3. Cable termination of a sonde. The small tube is the pressure transmitter pickup

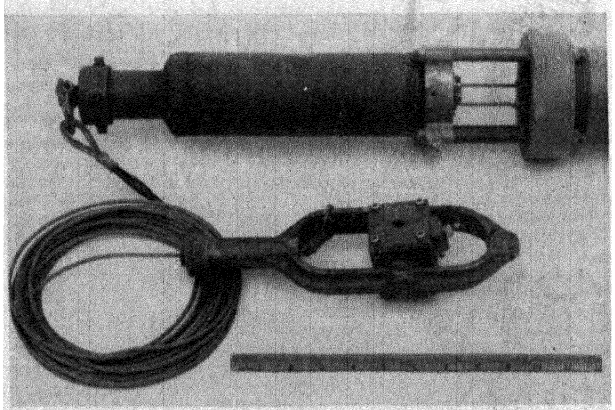


Figure 4. Termination of a sampler showing the actuator, towing pennant and swivel for attachment to the towing cable

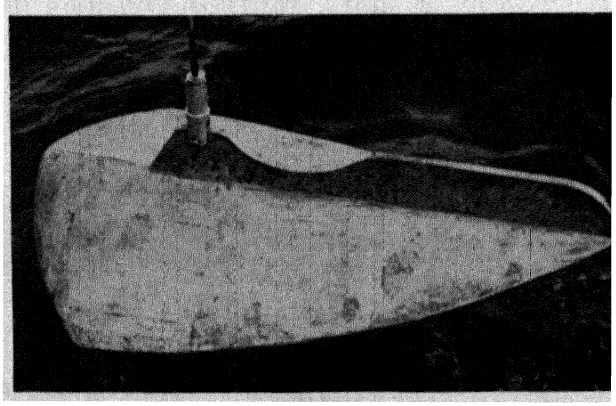


Figure 5. Aluminum depressor for the towing cable. The special insulating swivel isolates the depressor from the cable circuit

activity of the ocean would be increased by $3.7 \times 10^6$ megacuries, approximately eightfold. The total mass of fission products added per year would be 5000 tons.

The average radioactivity of sea water would still be quite low, but because of the very slow mixing, some water masses might show a thousandfold increase. For example, radioactive wastes introduced into the upper layer might remain there for many years, and would be diluted by a volume of water only a fiftieth to a hundredth the volume of the ocean. We can speak with much less certainty about the lower layer. Waste products in solution, introduced at a particular location and depth in the lower layer, might spread out over a very wide area in a thin, nearly horizontal sheet of concentrated activity. But rate of spreading and current transport of such a sheet are hard to estimate with present data.

A tenfold increase in the radioactivity of a particular water mass, if brought about by the presence of fission products, would probably result in an increase of several orders of magnitude in the radioactive content of the organisms living in the water. This would be expected because of the tendency of marine plants and animals to concentrate nuclear species having an intermediate position in the atomic table. Average activities of 100 disintegrations/sec[1] per gram of wet organic matter would be likely.

Some elements would be much more concentrated by organisms than others, and there would be significant differences between different groups of organisms and between parts of the same organism. Wastes introduced in solid inorganic phases might be concentrated by filter-feeding animals, such as shrimps and certain mollusks. Zinc would be especially concentrated in the spleen, liver, and stomach of fishes, the average enrichment factor on a wet weight basis being $10^4$, according to Vinogradov.[81] It is of interest to note that the isotope $Zn^{65}$, presumably arising from nuclear detonations, has been found concentrated in the internal organs of fish by Japanese investigators.[82] The possibility of human ingestion of radiozinc from fission products is reduced by the fact that the zinc-rich organs normally are not eaten.

The hazards to human beings from marine disposal of radioactive wastes might well be lowered, perhaps to negligible proportions, by taking advantage of the slow mixing and the stratification of the ocean waters, and of the fact that nearly all organisms of direct importance to man live in the upper one or two hundred meters.

It must be remembered, however, that unlike the waters, the animals of the sea move vertically at relatively high speeds. The possibilities of vertical transport of radioactive substances between levels by marine animals need careful investigation.

**Appendix. A comparison of fission product isotopes in the ocean with the amounts of radioactive nuclides present assuming $10^{11}$ megawatt-hours of nuclear power production and subsequent oceanic disposal**

| *Element* | *Radioactive nuclide* | *Amount of element in oceans in megatons* | *Curies of nuclide disposed to ocean* | *Nuclide half-life in years* | *Tons of nuclide* | *Specific activity, c/gm* |
|---|---|---|---|---|---|---|
| Pm | $Pm^{147}$ | – | $9.8 \times 10^{11}$ | 2.6 | – | |
| Nb | $Nb^{95}$ | 1.4 | $7.0 \times 10^{10}$ | 3.65 | 66 | $5 \times 10^{-4}$ |
| Ce | $Ce^{144}$ | 560 | $3.1 \times 10^{10}$ | 0.79 | 9.6 | $1.8 \times 10^{-5}$ |
| Ru | $Ru^{106}$ | 0.014 | $1.8 \times 10^{10}$ | 1.0 | 5.2 | 1.3 |
| Sr | $Sr^{90}$ | $10^7$ | $6.8 \times 10^{10}$ | 20. | 340 | $6.8 \times 10^{-3}$ |
| Cs | $Cs^{137}$ | 700 | $6.0 \times 10^{10}$ | 33. | 750 | $8.6 \times 10^{-4}$ |
| Kr | $Kr^{85}$ | 420 | $3.3 \times 10^9$ | 94. | 7.3 | $7.9 \times 10^{-4}$ |
| Sm | $Sm^{151}$ | 56 | $2.0 \times 10^7$ | 73. | 0.7 | $3.6 \times 10^{-4}$ |

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# On the Distribution of Radioactivity in the North Pacific Ocean in 1954-1955

**By Yasuo Miyake,* Japan**

After departure from Tokyo Harbour on May 15, 1954, observations were carried out until July 4 aboard the survey boat the "Shunkotsu-Maru" belonging to the Ministry of Agriculture and Forestry of Japan, in the sea area near Bikini Atoll, to clarify the relation of radioactive contamination to living matter, sea water, the air, etc.

As a result of the expedition, fairly strong radioactivity was detected in a wide area around Bikini Atoll.[1]

## THE METHOD OF MEASUREMENT OF RADIOACTIVITY IN SEA WATER

It is difficult to measure the radioactivity in sea water directly by a simple evaporation method, owing to its high salinity; therefore, the carrier method was adopted. After adding 2 gm of solid ammonium chloride and 1 ml each of aqueous solutions of iron alum (86.3 gr/l) and barium chloride (17.8 gr/l) to 1 litre of sea water, the water is heated to 60–70°C in a beaker, stirring well. A few drops of an alcoholic solution of phenolphthalein (1%) are added as an indicator; then ammonium hydroxide solution (1 : 1) is carefully dropped in with a pipet, until a faint pink colour appears. After about two minutes boiling, the precipitate is settled on standing for a couple of hours at room temperature. The precipitate is filtered under suction on a round filter paper (dia. 3.5 cm) laying above a glass filter. The precipitate is dried together with a filter paper in an air bath or a desiccator holding it in a small brass case to avoid the folding of the surface.

Radioactivity is measured by putting the precipitate under the mica window of a G-M counter. The G-M counter used aboard the "Shunkotsu-Maru" is Radiation Counter, Model 32 manufactured by Science Research Institute Ltd., Tokyo. The thickness of the mica window is 3.7 mg/cm$^2$ and the distance between the mica window and the surface of the precipitate is 1.5 cm. After measurement is finished, the precipitate is covered by a polyethylene film to protect the surface. In this way, at least 80% of the total activity in sea water could be transferred to the precipitate.

To obtain the counting efficiency, an aqueous solution containing $Ce^{144}$ whose activity had been referred to the standard, RaE, was treated chemically in the same way as mentioned above, and its radioactivity was measured under the same geometrical conditions. The efficiency of measurement was found to be 7.7%, that is, 1000 counts per minute is equivalent to about 5.9 m$\mu$c.

The maximum radioactivity in sea water obtained aboard by this method was 7025 cpm/l, which was found at a depth of 75 m, 450 km west of Bikini Atoll on June 21. The second was 6050 cpm/l found at a depth of 20 m, 150 km west of the atoll on June 12. It was found that most of the radioactive materials were dispersed in water as true solutions, ionic or eucolloidal, since more than 99% of the radioactivity readily passed through a filter paper (pore size: 0.5 $\mu$).

It is rather surprising that the activity in sea water in this area was often stronger than, or at least comparable with, that of White Oak Lake receiving waste effluent from nuclear reactors of Oak Ridge National Laboratory, USA. The latter was 2710–5190 cpm/l on an average.[2]

## THE DISTRIBUTION OF RADIOACTIVITY IN SEA WATER AROUND BIKINI ATOLL

### Horizontal Distribution

Figure 1 shows the horizontal distribution of the activity in the surface water near Bikini Atoll. There may have been considerable changes in the activity due to variation in sea conditions, natural decay and loss by precipitation, etc. during the course of observation. However, causes of such changes were so complicated that corrections were almost impossible. Therefore, the distribution was drawn by using directly the observational data without giving any corrections.

As shown in Fig. 1 the radioactive water extended in a WNW direction leaving near Bikini Atoll, while a branch spread out to the WSW. The cause of this branching is not clear. But, it seems that there was a weak discontinuous boundary along a line passing Bikini and Eniwetok Atolls. The water system $B_1$ (Fig. 2) on the northern side of the line had, on an average, a temperature of 27.1°C, chlorinity of 19.23%, and $\delta_t$ of 22.51 at 50 m depth.

On the other hand, the water system $B_2$ on the southern side of the line had $T$, 27.0°C; Cl, 19.04%; and $\delta_t$, 22.29. Thus, it may be said that there was a slight but definite difference in density between the two water systems, which might be one of the causes of the branching.

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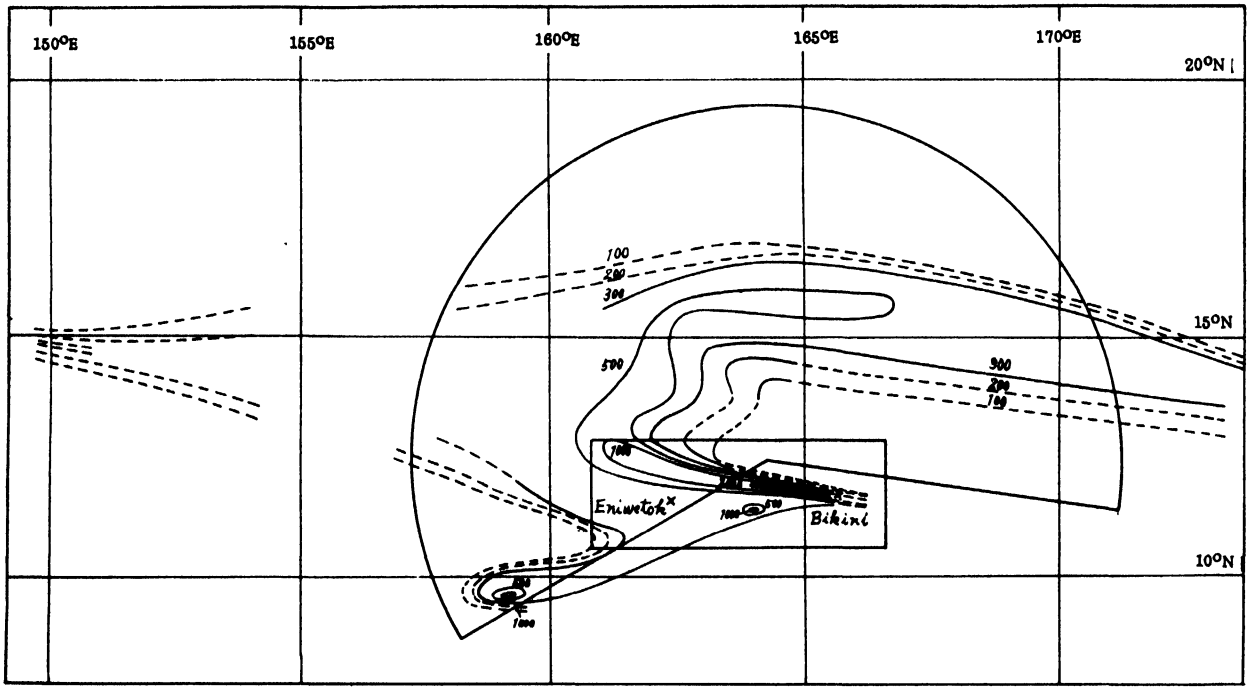



Figure 1. The horizontal distribution of radioactivity in the surface water near Bikini Atoll (cpm/l)

The radioactive water flowing in the main direction of the North Equatorial Current extended sooner toward the west than in the southern part. In the latter part the flow rate decreased because it was near the boundary between the North Equatorial Current and the Equatorial Counter Current. Thus, in the WNW direction, water with an activity of 100 cpm/l reached as far as 2000 km from Bikini Atoll, while in the WSW direction the extension was only 1000 km.

It is to be noticed that radioactivity was also detected at a position 700–800 km from Bikini in the ENE direction. This is the end part of the radioactivity distribution turning toward the east at a distance of about 350 km north of Bikini Atoll. At present, however, it is not clear whether it was the extension of the easterly current along the northern boundary of the North Equatorial Current which had been confirmed recently between 180 deg E 140 deg W[8,4] or whether it was only a local eddy.

When we investigate the horizontal distribution of water system near Bikini Atoll by using the *T-S* diagrams of water at a depth of 50 m below the surface, we find that there are at least four different types of water (Fig. 2). The first one, *A*, is highly saline water (chlorinity, 19.3‰, water temperature, 27.5°C, $\delta_t$, 22.77) covering the area further north than 15 deg N.

In the southern part of the atoll there is a second type of water, *C*, which belongs to the Equatorial Counter Current with lower chlorinity (Cl, 18.9%) and higher temperature (*T*, 28.0°C). The third type, *B*, belongs to the North Equatorial Current representing an intermediate condition between *A* and *C*. In the southern part of the water *C*, there extends the water *D* of the South Equatorial Current with higher chlorinity (Cl, 19.6%) and higher temperature (*T*, 28.0°C).

The water *B* may be classified into two sub-groups $B_1$ and $B_2$ as mentioned above. As shown in Fig. 2 the east part of *B* was comparatively broad and it became narrower on approaching to the west. It was probably due to the fact that the boundary between water *B* and *C* somewhat inclined to the north at the time. Therefore, the North Equatorial Current was pressed by the Equatorial Counter Current and the current flowing backward toward the north-east direction of Bikini Atoll would have resulted.

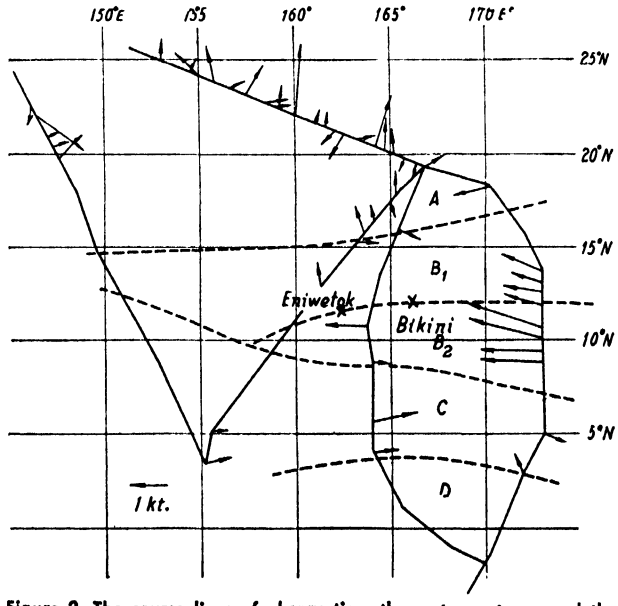



Figure 2. The course lines of observation, the water systems, and the current speed and its direction

The observation of the current direction obtained in site also suggests the presence of counter or eddy current in the same area. It is, however, not sure whether this current would be the origin of the general easterly current.

### Vertical Distribution of Radioactivity in Sea Water

Figures 3, 4 and 5 show the distributions of radioactivity of sea water on the vertical section perpendicular to the North Equatorial Current along the lines respectively at 150 km, 570 km and 1300 km west of Bikini Atoll. The profile at 150 km west of Bikini shows already the branching of the activity in the direction of N-S. The dotted lines in the figures represent respectively the latitudes of the sites of Bikini and Eniwetok Atolls. The strong activity is separated on both sides of these dotted lines and the distance of two maxima is about 100 km. The depth which shows the activity of 100 cpm/l is at about 80 m below the surface in the northern and southern parts and in the middle part it sings down to about 120 m. It is noteworthy that the activity decreases rapidly to less than 30 cpm/l at 200 m depth. This corresponds to the depth of thermocline which was about 150 m in this area.

The estimated amount of flow of radioactivity was about $1 \times 10^5$ curie/hour passing through this section. On the vertical section of 570 km west of Bikini the distance of branching became broader and also the depth of 100 cpm/l activity was 200 m below the sea surface on an average. The flow of radioactivity was about $0.6 \times 10^5$ curie/hour. The distance between the former and the latter section was about 400 km. Assuming that the speed of the current was about 0.7 knots in the direction perpendicular to the sections it would have taken about 2 weeks to flow between them. Thus, the activity passing through both sections would have been about the same when the rate of decay during two weeks was taken into consideration.

## THE RELATION BETWEEN THE RADIOACTIVITY IN SEA WATER AND THE RADIOACTIVE FALLOUT

The fishing boat "Fukuryu-Maru" was situated at a position 150 km east of Bikini Atoll on the early morning of 1st March when the crewman recognized small, white dust particles falling on the deck. The density of dust covering the surface of the deck was not clearly known; however, it may be supposed that it was about 0.5 gm/m² or so. On the other hand, the radioactivity of dust particles was estimated as about 1.4 curie/gm at the time of falling (3 hours after detonation).[5]

Assuming that the mean diameter of the dust particles was 0.1 mm, the rate of sinking of such particles in sea water is about 30 m/h. Thus, it takes about 1–1.5 sec to pass through a water layer of 1 cm. Assuming also that at most 0.01% of the radioactive substances in the dust could dissolve into sea water in one second, most of the radioactive materials would have been brought into solution before dust particles

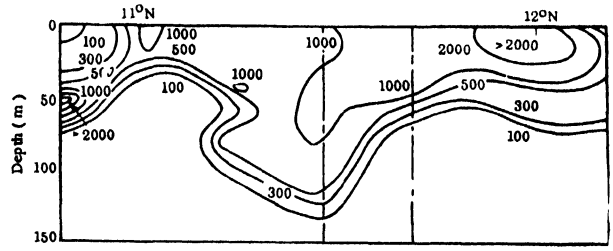



Figure 3. The vertical distribution of the radioactivity in sea water in the section perpendicular to the North Equatorial Current along the line at 150 km west of Bikini Atoll

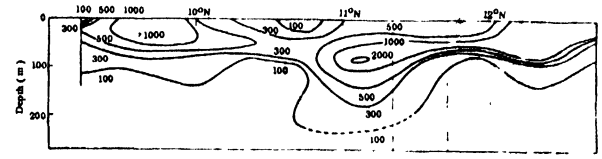



Figure 4. The vertical distribution of the radioactivity in sea water in the section perpendicular to the North Equatorial Current along the line at 570 km west of Bikini Atoll

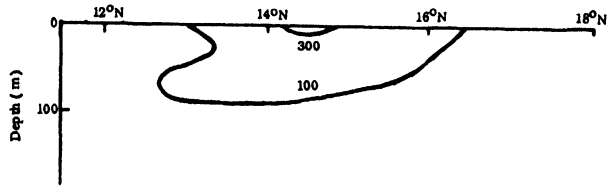



Figure 5. The vertical distribution of the radioactivity of sea water in the section perpendicular to the North Equatorial Current along the line at 1300 km west of Bikini Atoll

reached 100 m depth below the sea surface. Thus, the mean activity in water near the sea surface around there would have been 3.1 $\mu$c/l at the time five or six hours after detonation.

Needless to say, this was the activity of sea water in the area about 150 km away from Bikini Atoll; therefore, the activity near and in the atoll would have been much more stronger. On the other hand, it was confirmed by the records of micro-barographs and tide gauges obtained in Japan[6,7] that tests had been carried out at least four times—March 27, Apr. 26, and May 5. It was therefore not surprising that the large area far from Bikini Atoll had been contaminated by fission materials as strongly as it was observed.

## THE NATURE OF THE RADIOACTIVE SUBSTANCES IN SEA WATER

The relation between the gross radioactivity $A_t$ of the fission products and the time after detonation may be simply expressed by the next equation,

$$A_t = ct^{-a}$$

where $c$ and $a$ are constants. From the decay curve of the radioactivity of the precipitates, it has been found that $a$ is confined within the comparatively narrow limits of 1.3–1.6 (mean, 1.5) assuming the date of the last detonation was May 5. In normal cases, $a$ is about 1.2,[8] whereas it was 1.37 for the

radioactive dust that fell on the deck of the "Fukuryu-Maru."

In sea water some of the radioactive elements contained in the original fission products might be preferentially precipitated owing to the pH of sea water. Therefore, the chemical composition should be somewhat different from the original fission materials, which might be the cause of the change in the value of $\alpha$.

A radiochemical analysis of the fission materials in sea water was performed also by the present authors. Results[9] showed the presence of $Sr^{89}$, $Y^{91}$, $Zr^{95}$, $Nb^{95}$, $Ru^{103}$, $Ru^{106}$, $Rh^{106}$, $Ce^{141}$ and $Ce^{144}$. Among them in the contributions from Y and Ce were strongest in December, 1954. The scheme of radiochemical analysis of sea water adopted by the present authors is given in Fig. 6.

After "Shunkotsu-Maru" expedition was over, the radioactivity in the North Pacific Ocean was investigated several times through the cooperation of several marine laboratories.[10] Even in the area of Kuroshio flowing off the Japanese Islands faint activity appeared in July and August, 1954. From the end of November, 1954 to February, 1955 another survey was carried out by Regional Fisheries Research Laboratory at Kochi, Shikoku Island aboard the "Daifuji-Maru." The results showed that the radioactivity had already decreased in the region of Bikini owing to radioactive decay and water movement; however, weaker activity had spread out northwestward over a larger area in the North Pacific.

## DECONTAMINATION OF SEA WATER

Decontamination of sea water was studied at our laboratory. Activated charcoal, alumina and silica gel were used as decontaminants, among which activated charcoal was found to be most effective. The radioactivity in a sample of sea water was measured first by the method mentioned above.

To each one litre of the sample, respectively 0.06 gm, 0.12 gm and 0.4 gm of activated charcoal was added and, after standing for one hour with thorough stirring, it was filtered and the radioactivity adsorbed in the charcoal was measured. The efficiencies of decontamination were respectively 56%, 67% and 96%.

Next, sea water was passed through a glass tube filled with alumina. In this case, almost all of the radioactive material was adsorbed within a layer of 2 cm from the top of the column. A glass tube of inner dia. 8 mm and 20 cm length was used for the tests. One of the results is given in Table I.

**Table I. Decontamination of Sea Water by Alumina**

| Distance from top of column, cm | 0–1 | 1–2 | 2–3 | 3–4 | 4–5 |
|---|---|---|---|---|---|
| Radioactivity, cpm | 316 | 68 | 20 | 20 | 0 |

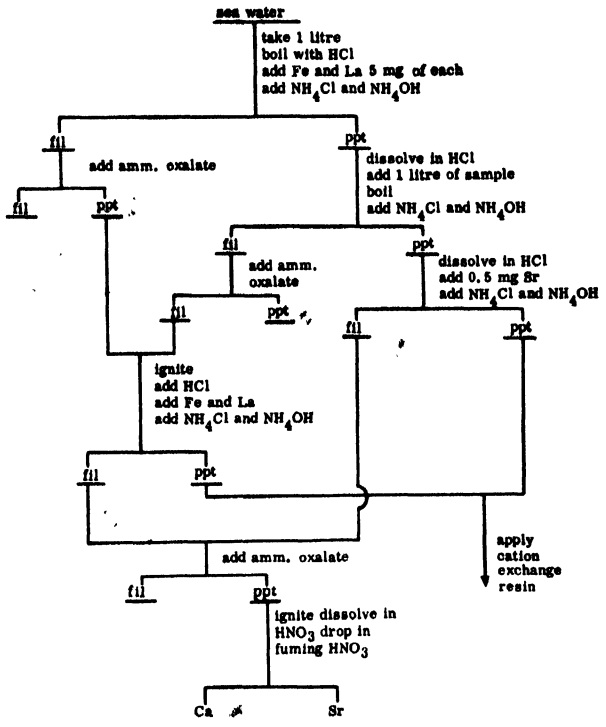



**Figure 6. The scheme of radiochemical analysis of sea water**

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# Radioactivity in Terrestrial Animals Near an Atomic Energy Site

By W. C. Hanson and H. A. Kornberg,* USA

The nature of radioactive contamination encountered in terrestrial animals and waterfowl near an atomic energy installation is dependent primarily upon the type of operation. Results presented herein are derived from a program of biological monitoring at the Hanford Works, which is engaged in the large scale production of plutonium.

As by-products of the manufacturing process, radioactive elements in controlled amounts are released to the Columbia River, to the ground, and to the atmosphere. The source of radioactive contamination of the river is neutron activated minerals in the river water used to cool the reactors. Other environmental contaminants are fission products resulting from the chemical separation of plutonium and unreacted uranium.

## TRANSFER OF RADIOACTIVE SUBSTANCES FROM THE RIVER TO WATERFOWL

A fraction of the group of contaminants released to the river enters aquatic biological chains. Of the radioactive substances present, $P^{32}$ is concentrated most significantly among members of the biological community. This occurs primarily because of the avidity of local aquatic forms for phosphorus. Samples of waterfowl and insectivorous birds nesting and feeding along the shores of the river are regularly taken and the concentrations of $P^{32}$ in their tissues determined. Table I lists the ratios of $P^{32}$ concentrations in birds, and in eggs of one species, to that in water and the common food of the birds.

The surprisingly high $P^{32}$ concentration in egg yolk relative to that in water is presumably due to the biological demands of phosphate-rich egg yolk, which obtains a significant portion of its phosphate through a biological chain from the river, which is low in phosphate content (0.03 ppm).

The relatively low ratios of $P^{32}$ concentrations in river ducks and geese relative to that in water are caused by their migratory status and preference for cereal grains. The even lower ratios noted in Table I for scavenger and piscivorous birds illustrate the effects of decay of the $P^{32}$ as it is transferred through biological chains to the birds. These observations are parallel to findings in fish,[1] wherein carnivorous species usually contained a lower concentration of radioactive substances than those which feed on bottom organisms.

In young birds the greatest concentration of $P^{32}$ occurred in the skeleton and pancreas. In adult birds, muscle achieved similar status, probably because of rapid phosphate turnover in pectoral muscles.

During a year of observations, deviations from the averages shown may vary over a tenfold range. These are the result of the natural mobility of avian populations and of selective feeding upon organisms in areas exposed to other than average $P^{32}$ concentrations.

No deleterious effect on waterfowl reproduction as a result of exposure to radioactivity in or around the reservation has been noted. Hatchability of eggs was 94 per cent, and fertility of adults developed from the eggs was 98 per cent, as compared to 87 per cent hatchability and 98 per cent fertility of adults in areas of no radioactive contamination.[2] Radiophosphorus deposits in waterfowl around the Hanford Works are typically such as to generate dose-rates up to about one rad/day.

**Table I. Ratio of $P^{32}$ Concentrations in Adults, Eggs and Young of Birds, to Those in Water and the Birds' Food, Respectively**

| *Specimen* | *Bird/water* | *Common food* | *Bird/food* |
|---|---|---|---|
| Swallows* (*Petrochiledon* sp.) | | | |
| Adults | 75,000 | Insects | 0.5 |
| Young | 500,000 | Insects | 3.5 |
| Diving ducks (*Aythya* spp.) | | Insects, | |
| Adults | 50,000 | vegetation | 0.5 |
| River ducks and geese (*Anas* spp. & *Branta* sp.) | | | |
| Adults | 7500 | Insects, | |
| Young | 40,000 | vegetation, and | 0.1 |
| Egg yolk | 1,500,000 | crustaceans | 0.5 |
| Sea gulls (*Larus* spp.) | | Fish, crustaceans, | |
| Adults | 5000 | vegetation | 0.1 |
| Piscivorous ducks (*Mergus* spp.) | | | |
| Adults | 2500 | Fish, crustaceans | 0.05 |
| Young | 15,000 | Fish, crustaceans | 0.25 |

* Although not a waterfowl, swallows are included here because their food is principally aquatic insects.

* General Electric Company. Including work by R. L. Browning, and W. C. Hanson, General Electric Company.

## TRANSFER OF FISSION PRODUCTS FROM SWAMPS TO WATERFOWL

Fission products released into the atmosphere in gaseous or particulate form, or released as process waste water enter terrestrial biological chains principally by deposition on vegetation, absorption into plants from substrata, and ingestion or inhalation by animals. Of particular interest to contamination potential is the ability of waterfowl to effectively concentrate several of the fission products when chronically exposed to a source of the material.

When aqueous solutions of industrial wastes containing relatively minor amounts of fission products are impounded, plant succession may make the resulting swamps attractive to waterfowl. Of the fission mixture, radioisotopes of strontium, cesium, ruthenium, and several of the rare earths have sufficiently long half-lives to make them available to plants and animals for extended periods. Strontium is usually the most important member of this group because it is significantly absorbed by plants and animals and because of its low permissible body burden. Although the concentration of strontium in vascular plants is less than 20 per cent of the concentration in the soil in which they are grown,[8] the feeding habits of waterfowl utilizing such a swamp will effect transfer of appreciable amounts of radiostrontium to the bird.

Coots (genus *Fulica*) that were allowed to inhabit a contaminated waste swamp for about four months were sampled and their tissues analyzed for the important radioelements. The uptake ability of the organisms is summarized in Fig. 1 and Table II.

Obviously the gross concentration factors shown in Table II apply only to the specific conditions around this particular swamp. Animal tissues differ significantly in their uptake selectivity for the various fission products. The over-all transmission is conditioned by the uptake coefficients of the intermediate food chain member, the emergent vegetation.

The principal component of muscle is radiocesium. In bone, it is radiostrontium, with significant contributions of the mixed rare earths (lanthanide elements) and cesium. Figure 1 illustrates the change in composition of the fission product mixture as it is selectively metabolized through this biological chain.

Cesium-137 accounted for 90 per cent of the activity in coot muscle in this case, and this represents a concentration factor relative to the water of about

**Table II. Relative Gross Uptake of Fission Products in Vegetation and the Tissues of Coots Inhabiting a Contaminated Swamp**

| *Organism* | *Concentration factor** |
|---|---|
| Swamp water | 1 |
| Emergent vegetation | 3 |
| Coot muscle | 250 |
| Coot bone | 500 |

* The concentration factor is defined as: concentration in subject/concentration in reference material.

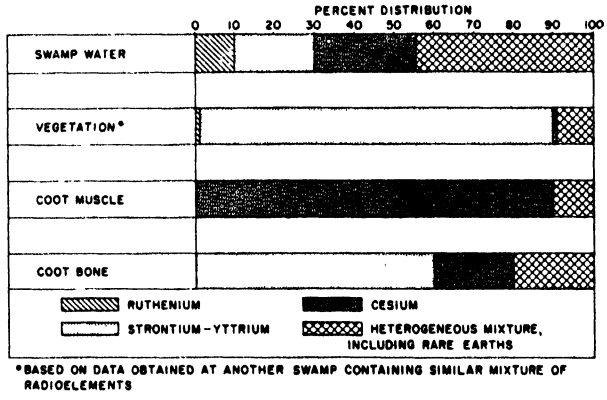



**Figure 1. Component fractions of selected fission products in water, vegetation, and tissue of coots inhabiting a contaminated swamp**

900. In bone, about 60 per cent of the radioactivity was due to $Sr^{90}$–$Y^{90}$, representing a concentration factor relative to the water of about 1500.

Ducks having access to the same swamp as the coots during this period accumulated considerably less radioactive material, in part as their transient status limited exposure to the contamination.

## TRANSFER OF $I^{131}$ FROM THE ATMOSPHERE TO ANIMALS

Seventeen terrestrial animal monitoring stations have been established at distances of from one to thirty kilometers from the chemical separations facilities, which routinely discharge $I^{131}$ and other fission products into the atmosphere. Tissue samples of jack rabbits (genus *Lepus*) from these locations were analyzed for radioactive substances monthly. Other mammals, birds, reptiles, and insects were occasionally sampled to provide comparison of animal types and food habits. Concentrations of $I^{131}$ in the thyroids of these animals were primarily dependent on their distances from the source of $I^{131}$, the rate of $I^{131}$ emission, the animal's age, and their feeding habits. Other factors that markedly affected the thyroidal $I^{131}$ concentration, but in a less predictable manner, were climatic conditions and the availability of inert iodine.

Uniformity in biological monitoring of this type is not to be expected. In an area of generalized $I^{131}$ contamination, the units of vegetation do not necessarily have the same $I^{131}$ concentration. In addition, the $I^{131}$ burden in an animal's thyroid gland at any one time is a partial historical picture of the contamination within the animal's feeding range over the preceding few weeks.

Figure 2 shows the relationships of $I^{131}$ concentrations in the thyroid glands of various animals to that of their food. At any one location the average radioiodine concentration in rabbits' thyroids was usually 500 times that of the vegetation. This factor, which may vary from 100 to 1000, is based on thyroid gland samples from 20 rabbits, collected monthly in areas from which weekly vegetation samples were analyzed. The effect of predation of several forms,

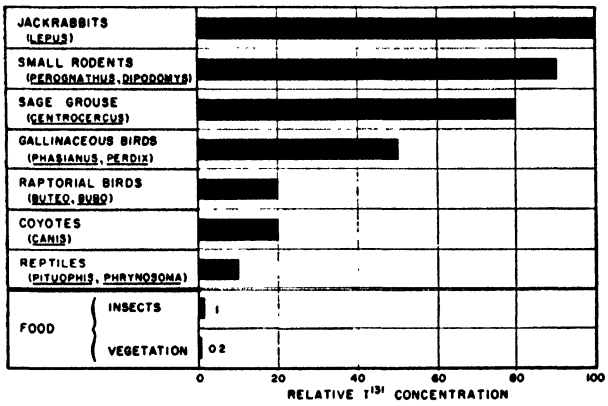



**Figure 2. Comparative $I^{131}$ concentrations in food items and thyroid glands of various animals at a given distance from radioiodine source**

one upon another, to form a biological chain for the transfer of radioiodine to larger animals is apparent in non-herbivorous animals, such as coyotes.

The left-hand portion of Fig. 3 indicates how the age of rabbits influences the $I^{131}$ uptake. The augmented uptake in immature animals has been demonstrated previously in controlled studies.[4]

The right-hand portion of Fig. 3 presents data on the relative $I^{131}$ concentration in rabbit thyroid glands as a function of distance from a high stack emitting $I^{131}$. These data cannot be reconciled precisely with the relative depositions on vegetation computed, for example, from the Sutton equations. This is to be expected from a combination of factors, such as the sample variance in biological monitoring, possible variations in succulence of vegetation at the different locations, and the distance over which the animals may have ranged from the point of collection.

Figure 4 shows ratios of average $I^{131}$ concentrations in thyroids of rabbits from five collection sites, ranging from two to twenty kilometers from the $I^{131}$ source, to average daily $I^{131}$ emission rates as a function of time. The very wide fluctuations with time appear to reflect, for the most part, the characteristic seasonal changes in the pattern of ground deposition of radioiodine. Added to this is the influence of the low inert iodine content of rabbits' thyroids during winter months. The quantity of inert thyroidal iodine in winter was about one-third of that in summer.

## TRANSFER OF RADIOACTIVE PARTICLES TO RABBITS

A familiar hazard in a radiochemical processing operation is the release of large numbers of small radioactive particles of median diameter of the order of 0.3 to 0.4 micron. The available quantity is such that in man, the lung burden of these particles cannot approach the conventional permissible limit. Biological monitoring provides the opportunity to have direct access to the animal lung for autoradiography and histological examination. In some work of this type with specimens chosen from areas of approximately maximum ground deposition of particles, rabbit lungs show a few hundred active particles per lung. The maximum activity of a single particle was

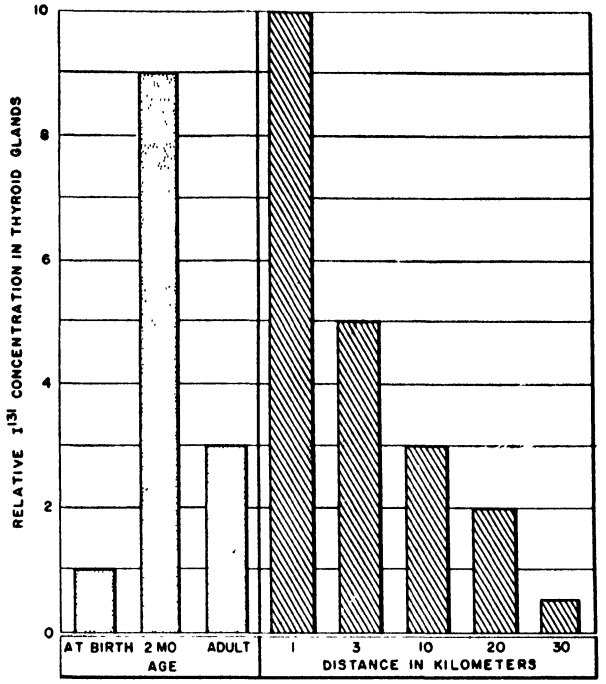



**Figure 3. Influence of age of rabbits and distance from radioiodine source on $I^{131}$ concentration of thyroid glands**

$3 \times 10^{-5}$ $\mu$c. No histological damage to lung tissue was observed. To some extent, these findings support the expectation of insignificant hazard in man from inhalation of like particles.

It may be inferred that one of the potential hazards arising from the production of atomic energy occurs via the transfer of released radioactive wastes among various forms of terrestrial life. Method and magni-

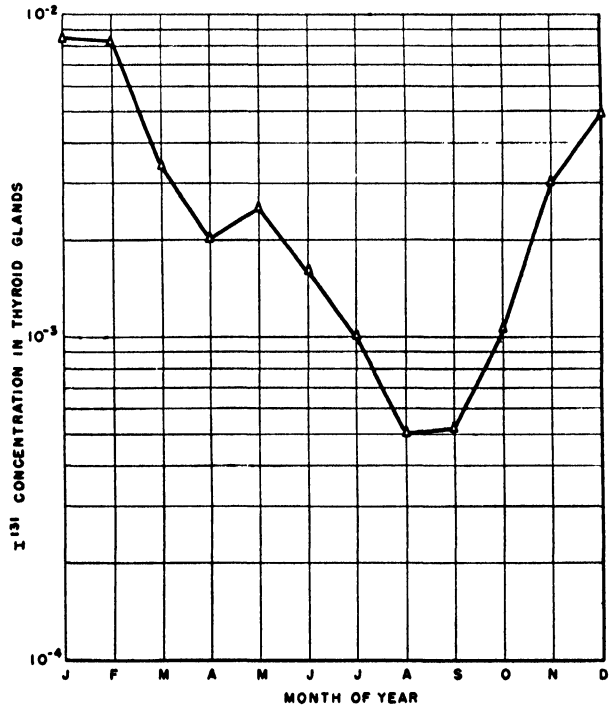



**Figure 4. $I^{131}$ concentration in rabbit thyroid glands per curie of $I^{131}$ emitted from process stacks. Monthly values represent average of three years observation**

tude of production, waste disposal practices, and environmental conditions will cause variation in the kind and extent of radioactive contamination in living forms. General conclusions drawn from this paper should therefore be supplemented by specific observations at the production site.

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# Record of Proceedings of Session 18C

## WEDNESDAY AFTERNOON, 17 AUGUST 1955

*Chairman:* Mr. J. C. Bugher (USA)

*Vice-Chairman:* Mr. J. C. Jacobsen (Denmark)

*Scientific Secretaries:* Messrs. L. F. Lamerton, E. O. Hughes and C. Polvani

### PROGRAMME

**18C.1. Hazards related to reactor and chemical processing**

P/8 Health and safety activities in reactor operations and chemical processing plants ..... A. J. Cipriani

P/240 Radiation exposure experience in a major atomic energy facility ..... H. M. Parker

P/452 Control of radiation hazards in the operation of medium-powered experimental reactors .... W. G. Marley and B. S. Smith

P/845 Approaches to treatment of poisoning by both radioactive and non-radioactive elements encountered in atomic energy operations ..... J. Schubert

Discussion

**18C.2. Ecological problems related to reactor operation**

P/278 The absorption of fission products by plants.. J. H. Rediske and F. P. Hungate

P/281 Radioactivity in terrestrial animals near an atomic energy site .... W. C. Hanson and H. A. Kornberg

P/280 The accumulation of radioactive substances in aquatic forms ..... R. F. Foster and J. J. Davis

P/393 The behaviour of $I^{131}$, $Sr^{89}$ and $Sr^{90}$ in certain agricultural food chains ..... A. C. Chamberlain *et al.*

Discussion

The Chairman: All life has evolved in a radioactive world. There is no sharp threshold at which radioactivity has an effect on biological things, as was brought out so clearly this morning. We presume that in general the effects of radioactivity on living cells and tissues are not in themselves beneficial, save perhaps in very rare genetic instances. Because of this continuity of radiation energies and intensities with respect to the biological effects, we ultimately come to the fundamental problems which are quantitative. We cannot speak in broad generalizations entirely, or even to a very large degree. In the final analysis we are concerned with how much of what kind of radiation is experienced by the living organism. It is one of the fundamental aspects of the conduct of an atomic energy program on a national and world-wide scale, and is the source of a very large part of the preoccupation with radiation safety and general matters of public health.

This afternoon, therefore, we come more closely to grips with the immediate quantitative problems of living and working in connection with atomic energy operations and installations.

Mr. A. J. Cipriani (Canada) presented paper P/8.

Mr. H. M. Parker (USA) presented paper P/240.

Mr. W. G. Marley (UK) presented paper P/452.

Mr. J. Schubert (USA) presented paper P/845.

### DISCUSSION OF P/8, P/240, P/452 AND P/845

Mr. F. G. Krotkov (USSR): I have a question for Mr. Cipriani. In your paper, you mentioned the conservative treatment of patients suffering from the effects of irradiation. May I ask you to tell us what methods of treatment you used and what results you obtained?

Mr. Cipriani (Canada): There were no specific methods of treatment used. The individuals were taken out of work at an early stage, put into another job where further exposure was unlikely, and they returned to work when the urinary excretion had dropped.

The Chairman: In these papers to which we have just listened, we have seen that it is entirely satis-

factory and practical to operate, over prolonged periods of time, elaborate atomic energy installations dealing with enormous amounts of radioactive materials. The plant personnel need not be seriously exposed; in fact under normal operating conditions no great difficulty has been encountered in restricting the levels of exposure to those well below the maximum permissible limit. When we turn to the ecological aspects of atomic energy operation, and especially with respect to the reactors, which undoubtedly would have to include the chemical processing plants also, we come into a different area of concern, one in which radioactive materials may become more or less widely dispersed through the environment and thus have the potentiality of being presented to large sections of a population. Under these conditions naturally somewhat different criteria must apply. These criteria have been mentioned and developed in the earlier sessions. Especially important is the fact that many isotopes which are important to man when taken internally go through rather complicated biological cycles in nature and thus may be presented to the human being along with his food or water, but particularly we have reference to the first category. We have become very much concerned then with the quantitative aspects of biological cycles, and the series of papers we are about to audit deal with some of these aspects.

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Mr. H. M. PARKER (USA) presented papers P/278 and P/281.

Mr. R. F. FOSTER (USA) presented paper P/280.

Messrs. B. SCOTT RUSSELL and A. C. CHAMBERLAIN (UK) presented paper P/393.

### DISCUSSION OF P/278, P/281, P/280 AND P/393

The CHAIRMAN: Before we come to the numerous questions on this very interesting and intriguing series of fundamental papers, the Chair wishes to recognize Mr. Klechkovsky who has a brief comment to make on the first paper presented by Mr. Parker.

Mr. KLECHKOVSKY (USSR): I listened with great interest to Mr. Parker's paper on the results of studies of the absorption of radioactive fission products by plants.

In the Soviet Union, experiments on the absorption of radioactive fission products by plants have been carried out by Gulyakin and others.

The accumulation in plants of radioactive fission products is not always directly proportional to their concentration in the external medium.

One reason for this is radiation damage to the root system. Hence, for instance, when the concentration of the radioisotope increases, its absorption falls, not only relatively but sometimes absolutely.

Sensitivity to radiation damage is not identical in all plants, and in a given species it depends on age; it is higher at the beginning of the vegetative period. This also affects the accumulation in plants of radioactive fission products.

Soils have a great capacity for the fixation (absorption) of fission products, which in turn has a significant influence on their absorption by plants. Hence the reaction must be more important in the case of a soil medium than in solution cultures. The control of the soil reaction may therefore be one of the factors affecting the accumulation of radioactive fission products on absorption from the soil.

Plant absorption of these radioisotopes from the soil is highly dependent on the way in which they are distributed in the soil. As a rule, their absorption by plants is less when they occur in pockets than that observed when they are distributed uniformly throughout the whole body of soil. Evidently, therefore, we must reckon with the possibility that with the passage of time the more even distribution of long-lived isotopes such as strontium-90 and cesium-137 through soil cultivation may bring about a relative increase in their absorption by plants.

With reference to the extremely interesting data given by Mr. Parker, I should like to ask the following two questions:

1. Does the concentration factor remain constant when the concentration of the radioisotope in the medium varies?

2. How do you explain the fact that the concentration factor for strontium-90 given in Mr. Parker's paper proved to be higher for soil cultures than for solution cultures?

Mr. PARKER (USA): I am sure that the authors of this paper would agree with many of the points put forward, particularly as to variations due to factors such as radiotoxicity in addition to the factor of chemical toxicity which was included in their contribution. I think it is understood that this paper was presented as showing broadly the type of things that one should look at in investigating the local problems around the site which may be releasing materials to the environment. I believe we pointed out that there were experimental variations in our own data.

As to the second question as to why the actual reported number was higher for the experiment in soil than in water when one might instinctively anticipate the reverse, I cannot answer this specifically. I believe a contributing factor is that, of the two first slides that we showed, the one which gave the breakdown into plant parts, the leaves and stems and so on, was early work which has not been repeated in the more refined laboratory arrangements which we have had lately. The other slide, on the contrary, I think was reliable data. Under the specific conditions with the specific solutions and soils available, we would take this data to be reliable. I think this accounts for the crossing over. I believe I said that if we report a concentration factor of 1 it is immaterial if we come in again and report a concentration factor of 2 or of 0.5. It is only significant that we can here discard a majority of the fission products as being hazardous by seeing that the number would be 0.001 or less, and we need only refine our experiments for specific

things, notably the strontium, iodine and possibly cesium.

Mr. KLECHKOVSKY (USSR): Mr. Chairman, I should be glad if you would permit me to thank Mr. Parker for his very detailed reply.

Mr. C. A. MAWSON (Canada): I wonder if Mr. Foster has considered the question of concentration of $P^{32}$ by the bacteria. The reason why I ask this is that in experiences of our own with lake water we have found that, when $P^{32}$ is put into lake water, the soluble $P^{32}$ disappears within minutes, in the sense that if you filter the water through a bacterial filter there is no $P^{32}$ in the liquids which comes through; it all appears to be absorbed by the bacteria within a very short time indeed.

Mr. FOSTER (USA): Yes, we have considered this very rapid uptake by bacteria. Actually in our laboratory we have not been working with the microorganisms, but undoubtedly much of the surface adsorption which we observe, not only perhaps on such things as sponge but also on the surfaces of laboratory containers and sterile versus unsterile containers, is due to bacterial pickup. Certainly the bacterial fixation of the phosphorus must be as equally important as, if not more important than, the initial fixation of the phosphorus by the algae, by the green plants.

Mr. A. R. GOPAL-AYENGAR (India): Mr. Foster mentioned, among other things, the uptake of fission products by aquatic vegetation, particularly plankton and the blue and green algae. I wonder if he has any information about the actual distribution of these fission products in the plant body. I mean, whether there is any sort of preferential uptake in particular parts of the plant—whether it is in the nucleus or cytoplasm, or whether in the rhizoidal portions or other specific parts of the plants.

Mr. FOSTER (USA): No, we do not have any information on the actual distribution within the plant cells. We would very much like to know how much of the radioactivity which we measure in these plant cells is on the outside and how much is actually incorporated in the cell protoplasm. We have no information on that at the present time, and I should certainly be interested to hear from anyone who can elucidate on that particular problem.

Mr. E. PORA (Romania): It is known that mineral substances are accumulated in animal organisms in two ways, by feeding and by direct exchange between the animal's internal system and its external environment through its tegumentary tissues and branchiae.

I would like to ask Mr. Foster whether he has specially studied osmotic exchange in the animals he has examined and to learn his views on the part played by osmotic phenomena in the accumulation of radioactive substances.

Mr. FOSTER (USA): All of our work has been done in a fresh water environment. We have no test in our laboratory where the osmotic environment has been used, so that we have only the one set of information. Also, most of our work has been done with radiophosphorus, with some work also on sodium. We find that with a mobile ion, such as sodium, we get a very rapid transfer across membranes, particularly gill membranes of fish. The accumulation of phosphorus across membranes is very slow. We get actually no concentration factor by immersing fish in a solution of radiophosphorus. The fish come into equilibrium with the radiophosphorus ion in the surrounding media, but we get no concentration this way. Some other people (Mr. Chipman, at one of the fisheries laboratories on the East coast) are working with certain other mineral ions, such as calcium, in this regard.

Mrs. P. CAMBEL (Turkey): I would like to ask Mr. Foster about further studies in fish. I read recently in one of the German papers that studies of fish were made in Hamburg for the detection of radioactivity after their migration to Hamburg. Were such studies made during fish migrations in different areas of surrounding oceanic water? Furthermore, I would like to know whether studies on laying of eggs and plankton migration were made.

Mr. FOSTER (USA): The accumulation of radioactivity in fish particularly does serve as a method of tagging these particular individuals, and perhaps the aquatic contamination which has resulted in several instances throughout the world offers a mechanism by which this can be measured. On the Columbia river itself we have very definitely been able to trace the movement of fish which have been in a contaminated area into another area. This is most marked where species of a resident nature, such as the white fish, feed in an area below the reactors and then during the spawning period will move upstream, so that we can very easily measure radioactive fish in what would normally be an uncontaminated area several miles up river from the reactors.

I do not quite recall your question in regard to the laying of eggs. Perhaps you would repeat it for me, please.

Mrs. P. CAMBEL (Turkey): I know that there are certain places to which fish migrate just for laying their eggs. Were any studies made of the eggs laid by such contaminated fish; and also on plankton migration?

Mr. FOSTER (USA): The plankton migration in the Columbia river (I will come back to eggs in a moment) is of course nothing compared to that which you would get in a lake or in the ocean. The plankton in the Columbia river is almost entirely due to the diatoms, and these are swept on down the river. We do get a displacement, shall we say, of contamination downriver; that is, the spot where we would notice maximum contamination in the Columbia river water is not quite the same place that we notice the maximum contamination in the plankton, because the plankton have picked up the radioactivity slightly slower and their maximum contamination is a bit down. In terms of vertical migration of plankton, this

has been observed in marine situations where plankton have become radioactive, and the activity of plankton has been noticed to be very small during daylight hours, but in the night time when the vertical movement of the plankton occurs, the activity of the water in the marine situation will very definitely rise because of the vertical movement of the plankton.

In terms of eggs, the radioactivity of eggs in fish according to their place of deposition will depend almost entirely on the biology of the individual species. If this is a species of fresh-water fish living during its entire life-cycle in this vicinity, then we expect to find a certain amount of radioactivity in the fish eggs which will, of course, give some indication of the exposure to the young embryo that it is maturing. I think it might be interesting to comment, however, on a species such as salmon, which is extremely important in the Columbia river, which migrate upstream. The salmon lays its eggs in fresh water and we have some spawning in the immediate vicinity of our reactors. However, the salmon mature not in fresh water but in salt water. They mature in an area which is uncontaminated and for this reason, in spite of the fact that they are spawning in the immediate vicinity of the reactors, the contamination level in the eggs is barely above background.

Mr. E. P. ODUM (USA): I am afraid, indeed, that we are overworking Mr. Foster, but I believe my question will be a sort of summary one. I get the impression from a broad comparison of your work and similar work at Oak Ridge National Laboratory that the concentration of radioisotopes is likely to be less in a river food chain than in a pond or lake system. Since the Oak Ridge work is not to be reported at this Conference, perhaps you would venture a brief comparison of the ecological fate, so to speak, of the effluents in streams as compared to ponds. I might also add that from the last two papers on this program it would seem that the most striking concentration occurred in terrestrial food chains or perhaps on transfer of materials from aquatic to terrestrial food chains. Would you care to make a comment on this?

Mr. FOSTER (USA): In regard to the pond situation, where you have a very slow exchange, you are dependent almost entirely on radioactive decay or the transfer of the radioisotopes through food chains and into a terrestrial situation in order to eliminate the contamination which has initially gained entrance to the pond. At the entrance to a river you have a situation where material is being continually swept down to the ocean. However, this sweeping action is not as complete as one might guess with a highly essential element such as radiophosphorus, an element which has been picked up very quickly by them, including definitely the plankton, the bacteria and also the larger forms. We feel that there is a significant fraction of this radiophosphorus which may be held up in the food chains of the river and transported downstream much more slowly than if it was entirely in solution and merely being swept down river. So you do actually have in some situations, with essential elements, an accumulation in the organisms of the stream which might be compared with that of a lake.

Mr. H. JAMMET (France): I would like to ask Mr. Foster or Mr. Parker two questions: 1. What are the average concentrations of radioactive products in the Columbia river after discharge of the Hanford effluents? 2. Has any increase in mutations or in teratological malformations been established among aquatic plants or animals?

Mr. PARKER (USA): The first question has to do with the concentrations of radioactive substances in the Columbia river. I think such information as that might perhaps be more readily given privately, Mr. Chairman. It would mean reference to notes. I do not have the figures immediately at hand.

As to the second question, our studies in this area are extremely limited and such as have been made have shown no effects of this nature; but the work does not purport to be exhaustive in any way.

Mr. E. C. WASSINK (Netherlands): I would like to ask Mr. Parker the following question: Are the concentration factors shown simply the result of enrichment of the elements by the metabolic chains or is there also a preferential increase in the concentration of the isotope over that of the normal form of the element?

Mr. PARKER (USA): I think we can categorically say that for the elements discussed today there is no isotopic effect. This would not be true if one considered isotopes of very low atomic weight so that there was a signficant difference in atomic weight between isotopes of the same element.

The CHAIRMAN: Mr. Parker of the United States has three questions to direct to Mr. Scott Russell.

Mr. PARKER (USA): Our experimental work on sheep shows damage at five microcuries of radioiodine per day intake and no damage whatsoever at 0.15 microcuries per day intake. In dose, this is respectively about 150 rads per week and 3 or 4 rads per week in the two cases. This lower level corresponds to about $2 \times 10^{-5}$ microcuries of iodine-131 per gram of vegetation. We feel that currently the accepted level for iodine contamination should not substantially exceed this limit. Would Mr. Scott Russell care to comment on a comparison of this with his figure of $2 \times 10^{-4}$ microcuries per gram?

Mr. SCOTT RUSSELL (UK): At the present stage, I do not feel that anyone is in a position to comment in any great detail on the experimental evidence from Hanford, which is in fact the only place where such detailed experiments have been done. However, there is a point which has been discussed elsewhere, namely, that the herbage intake per animal under those desert conditions is extremely high. In fact, on a rough calculation, the ratio of the weight of herbage consumed per weight of thyroid in sheep under Hanford conditions, is up by about 3 to 4 in relation to the cow under our conditions. Therefore, if one makes an ecological

adjustment, I suspect that there is only a factor of 2 to 3 between the two sets of calculations. I suggest that we may not be very far out.

Mr. PARKER (USA): Mr. Scott Russell emphasized that iodine-131 and the radiostrontiums are the isotopes of predominant interest for contamination of grazing land. In our experience, iodine-131 is a real and pertinent hazard. Radiostrontium contamination is entirely insignificant. Would Mr. Scott Russell know of any peaceful application to the contrary?

Mr. SCOTT RUSSELL (UK): I think two points may be made there. In the first place, we have considered strontium to quite an extent because of the milk cycle, which of course is not a matter of great importance in the neighbourhood of Hanford, I gather. Again, this is a point which could be taken up far better by Mr. Chamberlain. Depending on the method of operation, the ratio of iodine to strontium may vary by a very considerable factor. So we felt that we ought to put the general case. As Mr. Parker has seen we put iodine first, though we did not feel that strontium could be eliminated from a general discussion.

Mr. CHAMBERLAIN (UK): I should like to add just a little more to that. I think that the answer to the question of whether we have had practical experience with strontium contamination is no, we have not. However, when you are dealing with mixed fission products, you find that it is not always possible to identify the particular species you have. At times, you are reduced to seeing what would be the hazard or what would be the effect if strontium constituted an appreciable proportion of the products.

Mr. PARKER (USA): I am interested in Mr. Scott Russell's assumption that plants derive their calcium from the upper 25 centimeters of soil. Such deep-rooted plants as alfalfa and Russian thistle can recover strontium from much greater depths and can be so used as underground leak detectors. In point of fact, in connection with Mr. Chamberlain's comment on the previous question, if one had an underground source of mixed fission products with either of these two plants growing in the soil above, they would become radioactive essentially with strontium, and strontium only. Would this uptake which is applicable to strontium not also apply to calcium for the greater depths?

Mr. SCOTT RUSSELL (UK): Mr. Parker will realize that to take a single figure, as was necessary for this assessment as to depth, is necessarily a very wild guess. It is true that many of the plants which we considered also send roots much lower than 25 centimetres. On the other hand, they do not usually tap the zone below more than 20 centimetres at all fully. What we have in fact done is to say: let us assume the depth of 25 centimetres to be tapped completely and that will be equivalent to the total zone tapped. These assumptions in preliminary calculations can, I think, be justified, though they do not contest the fact that one can get absorption from greater depths.

I should like to put this question to Mr. Parker, if I may. If a calculation of this sort is done under Hanford conditions for deep-rooted desert vegetation, what figure would Mr. Parker consider more suitable?

Mr. PARKER (USA): I think I would get away from the era of calculation and try to get into the era of practical measurement. This is what happens to all of our ecological and related problems: I believe that calculation is demonstrably an ineffective tool since this afternoon we have had mention of maximum permissible limits that were palpably wrong by at least a factor of 1000 because the calculation applied to the wrong thing.

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