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Environmental Health Criteria 25

SELECTED RADIONUCLIDES

TRITIUM
CARBON-14
KRYPTON-85
STRONTIUM-90
IODINE
CAESIUM-137
RADON
PLUTONIUM

Published under the joint sponsorship of
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The International Programme on Chemical Safety (IPCS) is a joint venture of the United Nations Environment Programme, the International Labour Organisation, and the World Health Organization. The main objective of the IPCS is to carry out and disseminate evaluations of the effects of chemicals on human health and the quality of the environment. Supporting activities include the development of epidemiological, experimental laboratory, and risk-assessment methods that could produce internationally comparable results, and the development of manpower in the field of toxicology. Other activities carried out by IPCS include the development of know-how for coping with chemical accidents, coordination of laboratory testing and epidemiological studies, and promotion of research on the mechanisms of the biological action of chemicals.

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NOTE TO READERS OF THE CRITERIA DOCUMENTS

While every effort has been made to present information in the criteria documents as accurately as possible without unduly delaying their publication, mistakes might have occurred and are likely to occur in the future. In the interest of all users of the environmental health criteria documents, readers are kindly requested to communicate any errors found to the Division of Environmental Health, World Health Organization, Geneva, Switzerland, in order that they may be included in corrigenda which will appear in subsequent volumes.

In addition, experts in any particular field dealt with in the criteria documents are kindly requested to make available to the WHO Secretariat any important published information that may have inadvertently been omitted and which may change the evaluation of health risks from exposure to the environmental agent under examination, so that the information may be considered in the event of updating and re-evaluation of the conclusions contained in the criteria documents.

ENVIRONMENTAL HEALTH CRITERIA FOR SELECTED RADIONUCLIDES

At the request of the United Nations Environment Programme (UNEP), the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) prepared a paper on the Environmental Behaviour and Dosimetry of Radionuclides. In accordance with the UNEP proposal, the paper, which was prepared during the 27th - 29th sessions of the Committee and was completed and approved at the 30th session in 1981, is now being published in the WHO/UNEP Environmental Health Criteria series. The EHC document, which is entitled "Selected Radionuclides", comprises the integral report prepared and edited by UNSCEAR, together with an annex consisting of excerpts taken from "Basic Safety Standards for Radiation Protection 1982 Edition", Safety Series No 9, a document prepared jointly by IAEA/ILO/NEA(OECD)/WHO, and published by the International Atomic Energy Agency, to give guidance to the appropriate national authorities on the establishment of limits for radionuclides. The selected radionuclides discussed in the Environmental Health Criteria document are those of environmental importance for the general population and radiation workers.

Dr E. Komarov, Environmental Health Division, World Health Organization, was responsible for the final layout of the Environmental Health Criteria document.

The assistance of Dr B.G. Bennett (Monitoring and Assessment Research Centre, MARC) in the scientific editing of the Environmental Health Criteria document is gratefully acknowledged.

The contents of the 1982 UNSCEAR report to the General Assembly of the United Nations were taken into account during the preparation of the paper on the Environmental Behaviour and Dosimetry of Radionuclides, but the report was not quoted as it had not been issued at that time.

ENVIRONMENTAL BEHAVIOUR AND DOSIMETRY OF RADIONUCLIDES

1. PREFACE

1. The release of radioactive materials to the environment potentially exposes populations to ionizing radiation and increases the risk of incurring deleterious health effects. The associations of released amounts to effects establish the health criteria for radionuclides, i.e., the quantitative relationships that would be required to establish release limits governing the management of radioactive materials used by man.

2. This report has been prepared by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) for the United Nations Environment Programme (UNEP) to provide background information in establishing such health criteria. In this report the more general considerations of environmental behaviour of several radionuclides are discussed, including sources, transport to man and dosimetry. The radionuclides discussed are those most frequently released from natural and man-made sources and the greatest contributors to population radiation exposure under normal circumstances.

3. The compilation of the relevant information is based largely on the detailed presentations and evaluations of the sources of ionizing radiation by UNSCEAR in its reports to the United Nations General Assembly. The reader is referred to these reports for general concepts and for assessments of the dose commitments to man from exposures to sources such as natural radioactivity, fallout from atmospheric nuclear testing, releases from nuclear power production, occupational and medical irradiations.

4. Further information to be considered in establishing health criteria for radionuclides is that on health effects of irradiations. The relationships between radiation dose and risks of health effects in man have recently been re-evaluated based on the available data. This information can be found in the 1977 report of UNSCEAR. Only a brief summary of the general aspects of radiation effects and of radiation protection considerations is presented here.

5. The establishment of release limits for radionuclides in particular situations cannot be accomplished without rather more detailed considerations of the local and regional

environment and the special pathways of transfer to man. With this in mind, it is recognized that the material given here can only serve as background guidance.

6. The following scientists have contributed in the preparation of this report: Dr. W.J. Bair, Dr. D. Beninson, Dr. B.G. Bennett, Dr. A. Bouville, Dr. P. Patek, Dr. G. Silini and Dr. J.O. Snihs.

I. INTRODUCTION

7. Radionuclides are a special class of environmental substances. They are the unstable configurations of chemical elements which undergo radioactive decay, emitting radiation in the form of alpha or beta particles and x or gamma rays. The interaction of radiation with biological materials causes energy to be released to these materials which may result in a variety of harmful effects. Radiation is thus a potential hazard to man, although it may also be used in many beneficial ways, as in medical diagnosis and treatment, in industrial and consumer products and in the generation of electricity with nuclear reactors.

8. The realization of the harmful potential of ionizing radiation, which was dramatically brought to the attention of the public by the atomic bombing of Hiroshima and Nagasaki in 1945, was the cause of considerable attention that has been paid throughout the years to the effects of radiation. As a result of these studies, a great deal is now known about radionuclide behaviour in the environment and in man and about the somatic and genetic consequences of irradiation. This information surpasses by far that relating to any other class of environmental pollutants.

9. Considerable experience has been gained in environmental radiation measurements, particularly in tracing the movement of fallout radionuclides produced in atmospheric testing of nuclear weapons. Much of this information has in turn contributed to the general knowledge of atmospheric and oceanic transport processes and of bio-geochemical cycles of elements. Extensive studies of radiation effects in animals and numerous epidemiological surveys of exposed population groups have by now been conducted. They have considerably enlarged our understanding of the biological effects of radiation on man and the environment, although uncertainties still remain, particularly regarding the basic mechanisms of action and the risk evaluations at low doses and dose rates [U1-U7].

10. A few definitions and general concepts should be introduced before the detailed presentation of radionuclide assessments. The basic unit of radioactivity is the becquerel (Bq), corresponding to one disintegration per second. The previously used unit was the curie (Ci), one Ci corresponding to $3.7 \cdot 10^{10}$ Bq.

11. The basic measure of radiation interaction in irradiated materials is the absorbed dose (D). This quantity is also the basis of health risk estimates, under the assumption of a linear relationship between dose and risk. The absorbed dose is defined as the mean energy (joules) imparted to the irradiated material per unit mass (kg) at the point of interest. The unit of absorbed dose is called the gray (Gy) which corresponds to 1 J/kg. The unit of absorbed dose previously in use, the rad, is one hundred times smaller than the Gy.

12. Radiations of different types and energies have different effectiveness for producing effects, depending on the amount of energy transferred per unit length (LET) along the path of the charged particles. In order to quantify this differing effectiveness, use is made of a normalizing quantity called the quality factor (Q). For general purposes of radiological protection the assumed values of Q are: 1 for x and gamma rays and for electrons; 10 for neutrons and protons; 20 for alpha and multiply charged particles.

13. The product of the absorbed dose, D, and the quality factor, Q, is termed the dose equivalent (H). The unit of dose equivalent is the sievert (Sv). The previously used unit was the rem (1 rem = 0.01 Sv). Use of the dose equivalent allows the summation of doses from all types of radiation of different biological effectiveness.

14. The exposure of an individual to a source of radiation may be expressed in terms of the absorbed dose or dose equivalent during the period of exposure. In the natural radiation environment the exposure is continuous and it is sufficient to give the annual average dose or dose rate. There are important spatial variations to be considered, for example, as a function of the altitude in case of exposure to cosmic radiation or as a function of the geographical location due to the different radionuclides present in soil.

15. For specific releases of radioactive materials into the environment (atmospheric nuclear tests, operation of nuclear reactors) there are also important temporal variations in the exposure. In order to account for the exposures which will occur in the future from specific sources, use is made of the dose commitment (D^c). This quantity is the infinite time integral of the average individual dose rate. Dose commitments may not represent doses to specific individuals. For example, if the radionuclide released has a very long half-life, the dose commitment is derived from the doses to successive generations in the population.

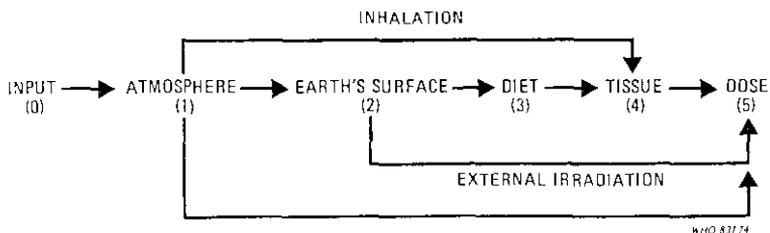
16. The collective harm to a population resulting from the exposure of all individuals is related to the collective dose in the population, particularly if the linearity of the relationships between dose and effects may be assumed for the exposures involved. The collective dose (S) in a given population is the summation of products of the average individual doses and the number of individuals in each range of doses. The summation may become an integral for continuous variations over the entire range of doses. The unit of the collective dose is man Gy and the corresponding unit of collective dose equivalent is man Sv.

17. The measure of the total exposure of a population from a specified source or release practice is the collective dose commitment (S^c), defined as the infinite time integral of the collective dose rate. The relevant units are man Gy, or man Sv in case of the collective dose equivalent commitment.

18. In radiation exposure assessments, it is often necessary to account for the different sensitivity of individual organs of the body with respect to each other or to irradiation of the whole body, particularly in the case of internally deposited radionuclides. Weighting factors for the relevant organs may be derived for this purpose from relative risk estimates. These factors will be listed in the section on radiation effects with some additional discussion.

19. The summation of the products of the weighting factors and the dose equivalents for individual organs gives a single measure to be used as an index of health detriment, called the effective dose equivalent (H_E). The concepts of collective and committed doses may also be used with this quantity. Thus a final quantity for health assessments may be the collective effective dose equivalent commitment, (S_E^c) which is a collective dose, weighted for the effects of doses within the body and dose distributions within the population.

20. The chain of events leading from the release of radioactive materials into the environment to the irradiation of human tissues may be expressed schematically as a series of compartments connected by transfer pathways. Such models are necessarily simplifications of the actual transfer pathways. The following diagram illustrates the transfer stages most usually considered in assessments by UNSCEAR.



21. The basic task in the assessment process is to evaluate the transfer factors ($P_{i,j}$) which relate the appropriate quantity of radioactivity amount or dose in step i of the sequence to the appropriate quantity in the subsequent step j . Since the desired quantity in the final step is the time integrated dose rate, the dose commitment from a specific source, the quantities in the other steps are the time integrated activity concentrations. The transfer factor is the quotient of time integrated quantities in successive compartments. The total transfer factor for steps in series is the product of the transfer factors involved. The total transfer factor of several parallel pathways is the sum of the transfer factors of the individual pathways.

22. There are many common features of the behaviour of different radionuclides in the environment and their transfer to man. For example, the physical dispersion of radionuclides in the environment following release from a source is largely the same for broad classes of material, such as particulates and gases. Several models used to describe the transfer of radioactive material within an environmental medium or from one medium to the next have general applicability. A review of such general behaviour and modelling procedures can be found in the 1982 report of UNSCEAR [U8]. Therefore, in the presentations which follow only the rather more specific aspects of environmental behaviour and dosimetry of the radionuclides are considered.

REFERENCES

- U1 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Thirteenth Session, Supplement No. 17 (A/3838). New York, 1958.
- U2 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Seventeenth Session, Supplement No. 16 (A/5216). New York, 1962.
- U3 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Nineteenth Session, Supplement No. 14 (A/5814). New York, 1964.
- U4 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-first Session, Supplement No. 14 (A/6314). New York, 1966.
- U5 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-fourth Session, Supplement No. 13 (A/7613). New York, 1969.
- U6 United Nations. Ionizing Radiation: Levels and Effects. A report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, with annexes. United Nations sales publication, No. E.72.IX.17 and 18. New York, 1972.
- U7 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General Assembly, with annexes. United Nations sales publication No. E.77.IX.I. New York, 1977.
- U8 United Nations. Ionizing Radiation: Sources and Biological Effects. United Nations Scientific Committee on the Effects of Atomic Radiation 1982 report to the General Assembly, with annexes. United Nations sales publication No. E.82.IX.8. New York, 1982.

II. TRITIUM

A. INTRODUCTION

23. Tritium, ^3H , is a radioactive isotope of hydrogen which decays into the stable nuclide ^3He . Tritium is a pure beta-emitter with a half-life of 12.3 a, a maximum energy of 18 keV and an average energy of 5.7 keV. Tritium is produced naturally in the atmosphere, where it results from the interaction of cosmic ray protons and neutrons with nitrogen, oxygen, and argon. Man-made tritium, in amounts substantially larger than the natural inventory, has been injected into the stratosphere by thermonuclear explosions. In addition, tritium is produced during the operation of nuclear reactors.

24. There are many applications of tritium in industry. It is widely used in consumer products, such as radioluminous timepieces and also as a tracer in biomedical research. Environmental tritium is mainly found as tritiated water. As such, it follows the hydrological cycle and penetrates into all components of the biosphere, including man.

25. This document is mainly based on the 1977 UNSCEAR report [U1], but makes also extensive use of the contents of recent reviews or symposia on tritium [I4, J1, M7, N1, N2].

B. SOURCES

1. Natural tritium

26. Natural tritium is produced by nuclear reactions in the atmosphere and, to a much smaller extent, in the hydrosphere and in the lithosphere. In addition, some tritium may be created in the extra-terrestrial environment and enter the atmosphere along with cosmic rays. Most of the natural tritium is found in the environment as tritiated water, generally designated as HTO.

27. In the atmosphere, natural tritium is produced by the interaction of high energy cosmic rays with atmospheric nitrogen and oxygen. The estimates of the number of atoms of tritium produced per unit time and per unit area of the earth's surface range from 0.1 to $1.3 \text{ cm}^{-2} \text{ s}^{-1}$ [U1]. In the UNSCEAR 1977 report [U1], a production rate of $0.25 \text{ cm}^{-2} \text{ s}^{-1}$ was adopted; this corresponds to a production rate of $3.6 \cdot 10^{16} \text{ Bq a}^{-1}$ in each hemisphere and to a global inventory of $1.3 \cdot 10^{18} \text{ Bq}$ at equilibrium.

28. It has been suggested that tritium might be ejected from the sun during solar flares [L1] and from stars [F1]. Flamm et al. [F2] estimated that the solar flares could account for an additional production rate, averaged over the solar cycle, of $0.1 \text{ cm}^{-2} \text{ s}^{-1}$.

29. In the lithosphere and in the hydrosphere, tritium is produced by interaction of neutrons with ${}^6\text{Li}$ nuclides. The production rates have been assessed at $10^{-3} \text{ cm}^{-2} \text{ s}^{-1}$ in the lithosphere and at $10^{-6} \text{ cm}^{-2} \text{ s}^{-1}$ in the hydrosphere [F1, K1].

2. Nuclear explosions

30. Nuclear tests have been conducted in the atmosphere since 1945 and have produced tritium in amounts that greatly exceed the global natural activity. The tritium activity arising from atmospheric nuclear tests can be estimated from the fission and fusion yields or from environmental measurements.

31. Bennett [B1] has published an estimate of the total and fission yields for each reported atmospheric test from 1945 to 1978; according to that compilation, 422 nuclear tests were conducted in the atmosphere up to 1979, with cumulative yields of 217 Mt and 328 Mt for fission and fusion, respectively. The tritium activity produced per unit yield depends on the characteristics of the device, as well as on those of the explosion site, but is in any case much greater for fusion than for fission [N1]. Miskel [M1] estimated the yield for fission explosion to be $2.6 \cdot 10^{13} \text{ Bq Mt}^{-1}$ and that for fusion to be typically $7.4 \cdot 10^{17} \text{ Bq Mt}^{-1}$. The total tritium activity produced by atmospheric tests is thus assessed at

$$328 \text{ Mt (fusion)} \times 7.4 \cdot 10^{17} \text{ Bq Mt}^{-1} = 2.4 \cdot 10^{20} \text{ Bq}$$

Most of this activity was produced during the large yield test series which took place during 1954-1958 and 1961-1962; the contribution of the nuclear tests carried out since 1964 is less than 5% of the total.

32. Almost all the tritium produced by fallout occurs as HTO in the atmosphere and the hydrosphere, and thus follows the hydrological cycle. The total activity injected can therefore be conceivably derived from measured concentrations in water samples. From the study of Schell et al. [S1] on the tritium concentrations in precipitation at stations in the IAEA network, it can be estimated [U1] that the total production

was about $1.7 \cdot 10^{20}$ Bq. Other estimates, using vertical profiles of ^3H in the oceans as a basis, lead to injections of $1.2 \cdot 10^{20}$ Bq (in the oceans only) [O1], $1.3 \cdot 10^{20}$ Bq [B2, U1], and $2.0 \cdot 10^{20}$ Bq [M2].

33. All the estimates presented above are in fairly good agreement, as they lie in the limited range from $1.2 \cdot 10^{20}$ to $2.4 \cdot 10^{20}$ Bq. In its 1977 report UNSCEAR adopted a value of $1.7 \cdot 10^{20}$ Bq for the total globally dispersed activity of tritium produced in atmospheric tests up to 1976 [U1].

3. Nuclear fuel cycle

34. Tritium occurs in nuclear reactors by ternary fission in the fuel and also by neutron activation reactions with lithium and boron isotopes dissolved in, or in contact with, the primary coolant as well as with naturally-occurring deuterium in the primary coolant (Figure II.I).

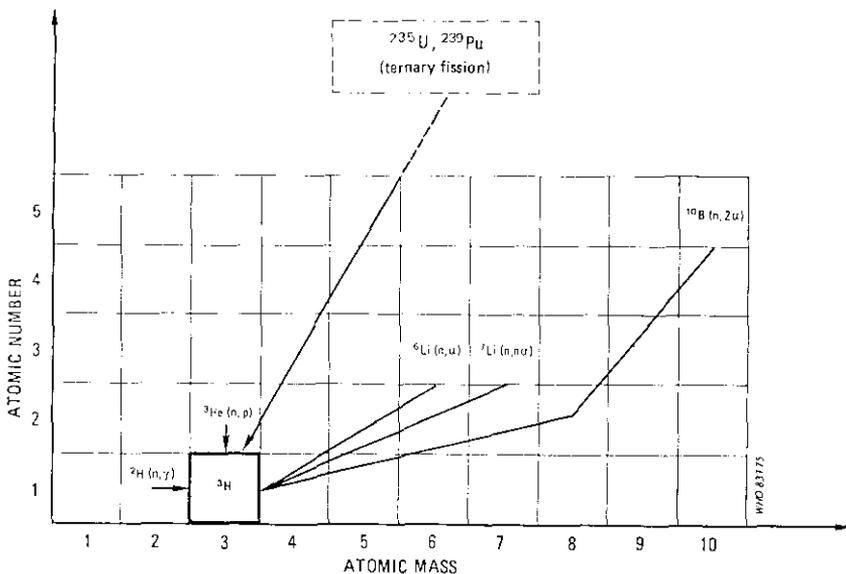


Figure II.I Main nuclear reactions giving rise to ^3H in reactors.

35. Most of the fission product tritium produced in the fuel rods is usually retained within the fuel and is not released into the environment at the reactor site; it is instead released during fuel reprocessing, if that practice is carried out. The activity produced in the coolant is partly or entirely released in the effluent streams according to the waste management practices at the plant.

36. Releases into the environment are mainly in the form of HTO in reactors that use water as primary coolant, as well as in fuel reprocessing plants.

(a) Nuclear reactors

37. Four types of reactors have been considered (PWR, BWR, HWR, GCR), the emphasis being on PWRs and BWRs which currently represent the largest share of nuclear capacity. Estimated generation rates and appearance of tritium in effluent streams of reactors are summarized in Table II.1.

38. The annual production of fission product tritium in the fuel rods of a pressurized water reactor (PWR) is in the range of 6 to $9 \cdot 10^{11}$ Bq per MW(e)a [N1]. A small percentage, 1% or less, is expected to be released into the coolant through defects in the cladding, currently made of zirconium alloy. In contrast, the use of stainless steel cladding in earlier PWRs resulted in the release to the coolant of most of the tritium produced in the fuel.

39. Tritium generation in the primary coolant (water) of a PWR is mainly due to reactions with boron ($2.6 \cdot 10^{10}$ Bq per MW(e)a) which is dissolved as boric acid to control reactivity; in addition, the maintenance of 2 ppm lithium hydroxide for pH control [L2] results in the formation of about $7 \cdot 10^8$ Bq per MW(e)a.

40. Environmental tritium discharges from PWRs depend on waste management practices as well as on the materials used in the reactor. Average normalized releases of tritium were shown in the UNSCEAR 1977 report [U1] to be about $7 \cdot 10^{10}$ Bq per MW(e)a in liquid effluents and $7 \cdot 10^9$ Bq per MW(e)a in airborne effluents for the reactors in operation in 1973-1974. However, large differences between PWRs are due to the type of fuel cladding. For an old reactor using stainless steel Kahn et al. [K3] measured ^3H releases of about $4 \cdot 10^{11}$ Bq per MW(e)a in liquid effluents and $4 \cdot 10^{10}$ Bq per MW(e)a in airborne effluents, whereas the combined releases of

Table II.1
Estimated rates of generation of tritium and of its release
in effluent streams of different types of reactors
 (10¹⁰ Bq per MW(e)a)
 {G1, K2, S2, T1, U1}

Source	PWR		BWR		HWR		CCR	
	Generation	Effluent stream						
<u>Fission</u>	75	< 0.7	75	< 0.7	55	< 0.6	75	< 0.7
Activation								
Deuterium	0.004	0.004	0.04	0.04	2000	75 <u>a/</u>	2	0.4
Lithium	0.07	0.07	30	0				
Boron	2.6	2.6						
Rounded total	80	3	110	0.5	2000	75	80	1

a/ Depending on the irradiation time and on the net leakage of heavy water.

9 PWRs with zirconium alloy clad fuel (current practice) were reported by NCRP [N1] to be about $3 \cdot 10^{10}$ and 10^9 Bq per MW(e)a in liquid and airborne effluents, respectively.

41. In boiling water reactors (BWRs) tritium is produced by ternary fission in the fuel at about the same rate as in PWRs (6 to $9 \cdot 10^{11}$ Bq per MW(e)a). The generalized use of zirconium alloy cladding limits the tritium release into the coolant to less than $7 \cdot 10^9$ Bq per MW(e)a.

42. Tritium can be generated by neutron activation in the coolant and in the control rods. Prior to 1971, control rods of boron carbide were used in BWRs [S2]; the production of tritium by activation of these control rods has been estimated to be about $3 \cdot 10^{11}$ Bq per MW(e)a. However, tritium has not been shown to diffuse through the boron carbide matrix [T1]. In the coolant itself, tritium is generated by activation of deuterium at a rate of about $4 \cdot 10^8$ Bq per MW(e)a.

43. Tritium activities discharged from BWRs into the environment are lower than those of PWRs because less tritium is produced in or diffuses into the primary coolant. UNSCEAR [U1] reported the average discharge rates to be $4 \cdot 10^9$ and $2 \cdot 10^9$ Bq MW(e)a in liquid and airborne effluents, respectively.

44. The amount of tritium generated in fuel of heavy water reactors (HWR) by ternary fission is approximately the same as in light water reactors, but it is largely exceeded by the production in the D_2O coolant and moderator by neutron activation, which has been estimated to be about $2 \cdot 10^{13}$ Bq per MW(e)a [K2].

45. Environmental discharges depend upon the D_2O leakage which is kept as small as possible for economical and radiological reasons, and upon the tritium activity in the coolant and moderator, which builds up with the irradiation time. Annual losses of from 0.5% to 3% are anticipated in HWRs [U1]. For the optimal loss of 0.5% per year, the normalized tritium release rate ranges from 10^{11} Bq per MW(e)a in the first year of operation to about $7 \cdot 10^{11}$ Bq per MW(e)a in the tenth year. Based on the latter value as representative of the reactor life, the normalized 3H release rates are estimated to be $6 \cdot 10^{11}$ and $1.5 \cdot 10^{11}$ Bq per MW(e)a in airborne and liquid effluents, respectively [G1]. Reported releases roughly agree with these estimates: they are $6.3 \cdot 10^{11}$ and $2.6 \cdot 10^{11}$ Bq per MW(e)a for the Pickering A station in Canada, in airborne and liquid effluents, respectively whereas the Atucha reactor in

Argentina releases about $8 \cdot 10^{11}$ Bq per MW(e)a both in airborne and in liquid effluents.

46. In gas-cooled reactors (GCR), tritium is produced by ternary fission (about $7 \cdot 10^{11}$ Bq per MW(e)a) and by activation of lithium in the graphite moderator. Based on the experience with UK reactors (mainly Magnox reactors), the tritium release is about $7 \cdot 10^9$ Bq per MW(e)a in liquid effluents and ranges from 10^9 to 10^{10} Bq per MW(e)a in airborne effluents [U].

(b) Fuel reprocessing plants

47. At the fuel reprocessing stage of the nuclear fuel cycle (if it is undertaken) the elements uranium and plutonium in the irradiated nuclear fuel are recovered for reuse in fission reactors. When the fuel elements are reprocessed, the uranium is first taken out of its cladding material and then dissolved in nitric acid. Most of the tritium released from fuel during dissolution appears in the liquid waste stream while some is carried out in the dissolver off-gas stream and a portion is immobilized as a solid zirconium compound in the cladding.

48. In 1980, the only reprocessing plants operating commercially in the world were at Windscale (U.K.) and La Hague and Marcoule (France); their combined capacity was much lower than the amount of fuel discharged from reactors worldwide. Luykx and Fraser [L3] have expressed the reported releases from the three reprocessing plants during the 1974-1978 time period in terms of activity discharged per unit of electricity generated. The average figures for each plant are given in Table II.2.

Table II.2

Average normalized tritium activities discharged into
the environment by fuel reprocessing plants
(10^{10} Bq per MW(e)a) [L3]

Plant location	Airborne effluents	Liquid effluents	Total
Windscale	17	55	72
La Hague	0.4	28.5	29
Marcoule	5.2	41	46

49. As there is no retention system for tritium in the currently operating reprocessing plants, the activity released corresponds to that which is contained in the fuel elements (with the exclusion of cladding) at the time of reprocessing. The production rate of tritium in reactors being about $75 \cdot 10^{10}$ Bq per MW(e)a (Table II.1), approximately half of the theoretical fuel content seems to be unaccounted for at the La Hague and Marcoule plants.

(c) Summary

50. In 1980, the installed nuclear capacity was $1.25 \cdot 10^5$ MW(e) on a worldwide scale [11]. Assuming an average load factor of 0.6, the energy produced was $7.5 \cdot 10^4$ MW(e)a. Using the average figures given previously for production and release in the types of reactors considered, the global production and release of tritium at the reactor sites in 1980 are estimated to be about $1.5 \cdot 10^{17}$ Bq and $4 \cdot 10^{15}$ Bq, respectively. Table II.3 provides a breakdown of the environmental discharges from reactors according to reactor type.

Table II.3

Estimated global discharge of tritium
from nuclear power stations in 1980

Reactor type	Number	Capacity [MW(e)]	Estimated tritium discharges in 1980 (Bq)		
			Airborne effluents	Liquid effluents	Total
PWR	96	64239	$3.9 \cdot 10^{13}$	$1.2 \cdot 10^{15}$	$1.2 \cdot 10^{15}$
BWR	62	35170	$4.2 \cdot 10^{13}$	$8.4 \cdot 10^{13}$	$1.3 \cdot 10^{14}$
HWR	14	5963	$5.4 \cdot 10^{14}$	$2.1 \cdot 10^{15}$	$2.6 \cdot 10^{15}$
GCR	36	7086	$1.3 \cdot 10^{13}$	$3.0 \cdot 10^{13}$	$4.3 \cdot 10^{13}$
Other	33	12527	-	-	-
Total	241	124985	$6.3 \cdot 10^{14}$	$3.4 \cdot 10^{15}$	$4.0 \cdot 10^{15}$

51. In comparison, the tritium releases reported for the three currently operating commercial fuel reprocessing plants were about $2 \cdot 10^{15}$ Bq in 1978. All together, the current tritium production rate in the nuclear fuel cycle is comparable to the natural production rate, whereas the release rate is about 20 times less.

4. Tritium production plants

52. Artificial production of tritium on an industrial scale is necessary to provide an essential component of thermonuclear weapons. In addition, relatively small amounts of tritium are used for other industrial and scientific applications. The most economical way to produce tritium is the irradiation of lithium metal, alloys or salts in a nuclear reactor [J1]. The tritium is isotopically separated from other hydrogen isotopes and is processed in tritium-handling plants [C1].

53. Tritium airborne release rates from Savannah River Plant, which is the primary production source of tritium in the U.S.A., have ranged from $1.4 \cdot 10^{16}$ Bq a^{-1} to $9.9 \cdot 10^{16}$ Bq a^{-1} from 1974 to 1977 with an average of $4.1 \cdot 10^{16}$ Bq a^{-1} [M4]. Under normal operating conditions, the releases are about 20% HT and 80% HTO. However, accidental airborne releases, which seem to be essentially in the gaseous HT form, have raised the contribution of HT to the total activity released to 60% in 1974 and 57% in 1975 [M4]. The activity of tritium released in the liquid effluents appears to be about 10% of that in the airborne effluents [N1].

54. Data on releases from other tritium production plants have not been found in the literature. However, an indirect estimate of $7 \cdot 10^{16}$ Bq for the worldwide release of HT in 1977 has been made by Mason and Östlund [M3] on the basis of their measurements of the atmospheric HT content.

5. Consumer products

55. Tritium has been used extensively in the dial-painting industry for the illumination of timepieces, the radiation emitted by ^3H being converted into light by a scintillator which is usually zinc sulfide containing small amounts of copper or silver. In recent years, this illumination system has been in competition with the tritium gas-filled glass tubes, coated internally with phosphor, which are used to illuminate some types of LCD (liquid crystal display) watches. Exit signs and electronic tubes are other types of consumer products containing tritium [C2, K4, U1, W1].

56. In luminous compounds, the fractional release rate of tritium, in the form of HTO, HT and short-chain organic radicals of the styrene type, is about 5% annually [K4, K5] while it is negligible from gas-filled glass tubes. It has been estimated that about $7 \cdot 10^{16}$ Bq was processed in 1978 in the worldwide production of timepieces and that the activity released is probably under 10^{14} Bq a⁻¹ for luminous compounds and $2 \cdot 10^{12}$ Bq a⁻¹ for gas-filled glass tubes [K5]. Environmental releases due to breakage through accident or disposal could be more important [C2, W1].

6. Controlled thermonuclear reactors

57. Large-scale use of controlled thermonuclear reactors for heat or power generation seems quite unlikely in the next 25 years. However, if thermonuclear reactors come into use, they will contain substantial inventories of tritium and will pose considerable tritium management problems [N1]. The production of tritium in a nominal 1000 MW(e) controlled thermonuclear reactor is anticipated to be about $5 \cdot 10^{17}$ Bq d⁻¹ and the inventory of the order of 10^{19} Bq [C1, C3, H1]. In order to prevent massive releases of tritium into the environment, an extraordinary degree of control will be required. However, conceptual designs for fusion power plants show that the effluent release rate can be limited to $4 \cdot 10^{13}$ Bq a⁻¹ by applying present-day tritium technology [C3].

C. BEHAVIOUR IN THE ENVIRONMENT

1. Natural and fallout tritium

58. Natural and fallout tritium are mainly produced in the stratosphere where they are essentially found in the HTO form. Tritiated water vapour is transferred from the stratosphere to the troposphere with a half-time of about one year, then from troposphere to the earth's surface through rainfall and molecular exchange with a half-time of about ten days. Tritiated water then follows the hydrological cycle. Water deposited on the ocean surfaces is diluted in the mixed layer. Part of it evaporates back to the atmosphere, with a much lower concentration, while a smaller fraction is transferred to the deep ocean. Tritiated water deposited on land surfaces is partitioned partly to surface run-off (leading to a pond, a lake, a stream, or an ocean) and partly to infiltration in the soil from where it can be absorbed by plants, evaporate, or move with groundwater to a surface stream or to an ocean.

59. Part of the tritiated water deposited on soils finds its way into vegetable and animal products and thus contaminates dietary foodstuffs. Tritium incorporated into those biological materials, and in soil and sediments as well, is found to be present in at least two separable fractions, one easily exchangeable, that is available by freeze-drying (free water tritium fraction) and one less easily exchangeable, available by combustion ("organically bound" fraction) [B6]. The analysis of soil, water, and various components of the diet in the New York area in 1978 [B6] revealed that water, soil and diet were in equilibrium with respect to free water tritium; however, the specific activities (activity concentration per unit mass of hydrogen) of the "organically bound" tritium in various foodstuffs were higher by a factor of 2 to 4 than those of the water tritium. It is suggested that tritium was incorporated uniformly into biological materials during the period of highest deposition rates in the early 1960s and that differences in specific activities developed due to longer biological residence half-time of the "organically bound" fraction compared to the free water tritium fraction [B6].

2. Industrial releases

60. Industrial releases consist mainly of HTO and HT, and probably tritiated methane, CH₃T [B7]. The residence times of HT and CH₃T in the atmosphere are not known with certainty but the estimates point to average values of 5 to 10 years [B7]. The main removal processes are bacterial action and photochemical oxidation for HT and photochemical oxidation alone for CH₃T [B7]. In both cases, the resulting product is presumably HTO. As HTO is much more biologically active than HT and CH₃T, it is this tritium compound that is of most concern in the case of industrial releases.

61. Industrial releases may be to the atmosphere or to water (river or sea). In addition, releases to ground water have taken place but they are of little consequence as the movement of water in suitable aquifers is very slow. The environmental behaviour of HTO released by industry is not different from that from natural or fallout sources.

D. TRANSFER TO MAN

62. Transfer to man of environmental HTO takes place via inhalation, diffusion through skin and ingestion of beverages and foodstuffs; in the case of HT, inhalation is the only meaningful pathway to man. Exposure to an atmosphere

contaminated with tritiated water vapour results in total absorption of the inhaled activity through the lungs and absorption of about 50% of that amount through the intact skin [I2]. Ingested tritiated water is completely absorbed from the gastro-intestinal tract.

63. Absorbed tritiated water is rapidly distributed throughout the body via the blood. Tritiated water in blood equilibrates with extracellular fluid in about 12 minutes. However, in poorly vascularized tissues, such as bone and fat, equilibrium with plasma water may take days to weeks [N2, W2]. The biological half-life of tritium in the body following intake of tritiated water has been found to range from 2.4 to 18 days among 300 individuals [B3, W3]. The experience from observations of human cases of accidental tritium exposures with intakes large enough to allow relatively long-term monitoring shows that the excretion rate can be represented as the sum of three exponentials with half-times of residence of the order of 10 days, one month, and one year [L4, M5, M6, S3]. The first component is believed to reflect the turnover of body water while the second and the third components are likely to represent the turnover of tritium incorporated into organic compounds.

E. DOSIMETRY

1. Dose per unit intake

(a) Tritiated water

64. External irradiation from tritium does not need to be considered as the range of the electrons emitted by decay (at most 6 μm in soft tissue) is shorter than the depth of the basal cells in the epidermis. Following a chronic intake of 1 Bq l^{-1} of tritium (as HTO) in air, water and food the equilibrium dose rate in active wet tissue (the totality of soft tissues with the exclusion of fat) is $2.6 \cdot 10^{-8}$ Gy a^{-1} . Of that dose, 16% is calculated to be due to tritium contained in organic pools of the body. These results were derived by Bennett [B4] based on human retention data.

65. When all the sources of intake (air, water and food) are assumed to be contaminated at the same level, use can be made of the specific activity model which consists in assuming that the specific activity of tritium (activity concentration per unit mass of hydrogen) in the body is the same as that in the intake. A chronic intake of tritium at a concentration of 1 Bq per litre of water would thus give rise to an absorbed

dose averaged over the whole body of

$$\frac{1 \text{ Bq}}{1 \text{ H}_2\text{O}} \times \frac{10^{-3} \text{ l H}_2\text{O}}{\text{g H}_2\text{O}} \times 18 \frac{\text{g H}_2\text{O}}{\text{g H}} \times 0.1 \frac{\text{g H}}{\text{g body}}$$

$$\times 5.7 \cdot 10^{-3} \frac{\text{MeV}}{\text{Bq s}} \times 3.16 \cdot 10^7 \frac{\text{s}}{\text{a}} \times 1.6 \cdot 10^{-10} \frac{\text{Gy g body}}{\text{MeV}}$$

$$= 2.6 \cdot 10^{-9} \text{ Gy a}^{-1} \text{ per Bq l}^{-1}$$

This result is numerically equal to that of Bennett [B4]. The doses in individual tissues depend on their hydrogen concentrations. According to the values adopted for the Reference Man of ICRP [I2], the hydrogen concentration per unit mass is the same (10%) in total body and in total soft tissues and is, as a first approximation, uniform in the soft tissues. Hydrogen content is lowest in mineral bone (about 4%) and highest in adipose tissue (12%). Since the range of the beta-particles emitted by tritium decay is very small, it can be assumed that all the energy emitted in a given tissue is absorbed in the same tissue. The effective dose equivalent is therefore numerically equal to the absorbed dose averaged over the whole body and is $2.6 \cdot 10^{-9} \text{ Sv a}^{-1}$ per Bq l^{-1} . Assuming a rate of intake of 3 litres of water (in beverages and in food) per day and a water vapour atmosphere concentration of 8 g m^{-3} , the effective dose equivalent per unit intake is found to be $2.2 \cdot 10^{-11} \text{ Sv Bq}^{-1}$ while the effective dose equivalent rate per unit atmospheric concentration would be $2.1 \cdot 10^{-9} \text{ Sv a}^{-1}$ per Bq m^{-3} .

(b) Tritiated hydrogen

66. The doses from inhalation of HT are much lower than those from HTO for a given atmospheric concentration of tritium. The dose rate to the lungs per unit concentration of HT in air is about $10^{-14} \text{ Gy h}^{-1}$ per Bq m^{-3} [I3], while the doses in tissues from the absorbed gas are 60 to 150 times smaller [I3]. The corresponding effective dose equivalent rate per unit concentration in air is therefore $1.1 \cdot 10^{-11} \text{ Sv a}^{-1}$ per Bq m^{-3} .

2. Dose per unit release

(a) Natural tritium

67. Doses from natural tritium can be estimated from the few tritium measurements in environmental materials that were

carried out before the contamination with fallout (or that had been preserved from contamination). Activity concentrations of continental surface waters were then found to be in the range from 0.2 to 0.9 Bq l⁻¹ [K1]. The production rate of natural tritium being constant in time and relatively uniform on the global scale, the concentrations in all the components of human intake (air, water and food) of natural tritium are in steady-state equilibrium with the concentrations in continental surface waters. Using the specific activity approach, it is assumed that the specific activity of natural tritium is the same in the continental surface waters, in all the components of human intake and in the body. The effective dose equivalent rate is thus found to range from 0.2 Bq l⁻¹ x 2.6 10⁻⁸ Sv a⁻¹ per Bq l⁻¹ = 5.2 10⁻⁹ Sv a⁻¹ to 0.9 Bq l⁻¹ x 2.6 10⁻⁸ Sv a⁻¹ per Bq l⁻¹ = 2.3 10⁻⁸ Sv a⁻¹, being therefore of the order of 10⁻⁸ Sv a⁻¹. The effective dose equivalent commitment per unit release would then be

$$\frac{10^{-8} \text{ Sv a}^{-1}}{7.2 \cdot 10^{16} \text{ Bq a}^{-1}} = 1.4 \cdot 10^{-25} \text{ Sv per Bq}$$

Taking the world's population to be 4 10⁹ people, the global collective effective dose equivalent commitment per unit of activity produced is about 5 10⁻¹⁶ man Sv per Bq.

(b) Nuclear explosions

68. The doses from fallout tritium can be estimated in the same way as those from natural tritium. On the basis of the variation with time of the tritium activity concentration in surface waters [B5] and of the latitudinal distribution of the fallout deposition [S1], UNSCEAR [U1] estimated the effective dose equivalent commitments to the populations of the northern and southern hemispheres to be 2 10⁻⁵ and 2 10⁻⁶ Sv respectively.

69. The effective dose equivalent commitment from fallout tritium was also estimated indirectly, using the relationship obtained for natural tritium between the production rate and the dose rate

$$H^C = \gamma_0 \frac{W}{B}$$

where H^C is the effective dose equivalent commitment (Sv) from production of fallout tritium in a given hemisphere; γ_0 is the effective dose equivalent rate from natural tritium ($\gamma_0 = 10^{-8}$ Sv a⁻¹); W is the activity of tritium released by nuclear explosions (1.5 10²⁰ Bq in the northern hemisphere and 0.2 10²⁰ Bq in the southern

hemisphere); and B is the natural rate of production ($3.6 \cdot 10^{16}$ Bq a^{-1} in each hemisphere). The effective dose equivalent commitments thus derived are $4.2 \cdot 10^{-5}$ Sv for the population of the northern hemisphere and $5.6 \cdot 10^{-6}$ Sv for the population of the southern hemisphere. These results are higher than the direct estimates by a factor of 2 to 3. The global collective effective dose equivalent commitments per unit activity released are estimated to be $9 \cdot 10^{-16}$ and $4 \cdot 10^{-16}$ man Sv Bq $^{-1}$ using the latter and the former method, respectively. UNSCEAR [U1] used an intermediate value of $8 \cdot 10^{-16}$ man Sv per Bq.

(c) Nuclear installations

70. While the production of natural and fallout tritium brings about a relatively uniform contamination of the whole biosphere, the releases from nuclear installations occur at discrete points on the earth's surface giving a highly heterogeneous spatial distribution of concentrations.

71. UNSCEAR'S practice is to divide the collective doses into two components: the local and regional collective doses, which are due to the first passage of the plume, over distances of 100 to 1000 km from the point of release, and the global collective doses, which arise from the mixing of tritium in the whole biosphere. As the doses per unit concentration of tritium in air are much higher for HTO than for HT, tritiated water will be the only compound considered in the estimate of the local and regional collective doses.

(i) Local and regional collective dose

72. A distinction is made between airborne and liquid effluents. Tritium present in airborne effluents can contribute to the local and regional collective doses through inhalation, absorption through skin and ingestion. As the contribution from the ingestion pathway is quite variable from site to site owing to differences in local hydrology and water usage, UNSCEAR [U1] has not taken this pathway into account in its assessment of the local and regional collective doses. Assuming an atmospheric dispersion factor of $5 \cdot 10^{-7}$ s m^{-3} at 1 km from the release and a reduction in concentration inversely proportional to the 1.5 power of the distance expressed in kilometres, the local collective dose per unit activity released can be assessed by integration over the local area. Integrating from 1 to 100 km for a population density of 100 km^{-2} , UNSCEAR [U1] estimated the local collective dose from airborne tritium per unit activity released to be about $5 \cdot 10^{-17}$ man Sv per Bq.

73. The collective dose commitment from the input of ^3H to water bodies, normalized per unit activity released, can be estimated [U1], using the expression

$$S_1^c = \frac{\sum_k N_k I_k f_k \varphi}{V(\lambda + 1/\tau)}$$

where V is the volume of the receiving waters, τ is the turnover time of receiving waters, λ is the decay constant of ^3H , N_k is the number of individuals exposed by pathway k , I_k is the individual consumption rate of pathway item k , f_k is the concentration factor for the consumed item in pathway k , and φ is the collective dose per unit activity ingested collectively by the exposed group.

74. The quantity $\frac{1}{V(\lambda + 1/\tau)}$ is the infinite time integral of the water concentration per unit of activity released, while the quantity multiplied by f_k is the infinite time integral of the concentration in the consumed item k . For radionuclide inputs into small volumes of water, the concentrations in water and in fish will be high, but the population which can be served with drinking water or by fish consumption will be limited. For inputs into larger volumes of water, the concentrations will be smaller, but the populations involved will be correspondingly larger. It is reasonable, therefore, to assume as a first approximation that the quantities N_k/V are relatively constant, independent of V . The values for these quantities as well as values for the other parameters of the above expression have been extensively discussed [U1].

75. A summary of the values used in the assessment, based on UNSCEAR [U1], and the evaluation of the collective dose commitments for a release of 1 Bq of ^3H in liquid effluents are given in Table II.4.

(ii) Global collective dose

76. For HTO releases, the global collective effective dose equivalent commitment established for fallout tritium ($8 \cdot 10^{-16}$ man Sv per Bq) can be applied without change. With respect to HT releases, if it is assumed that the conversion to HTO takes place on average 5 years after the discharges, the global collective effective dose equivalent commitment is estimated to be

$$8 \cdot 10^{-16} e^{-0.693 \times 5/12.3} = 6 \cdot 10^{-16} \text{ man Sv per Bq.}$$

Table II.4

Collective dose factors for ³H in liquid effluents

	Fresh water	Salt water
Activity released, A	1 Bq	1 Bq
Turnover time of receiving water,	10 a	1.0 a
Sediment removal correction factor, s	1.0	1.0
Time integral of activity in water,		
$W = \frac{As}{1/\tau + \lambda}$	6.36 Bq a	0.946 Bq a
Water utilization, V/N	$3 \cdot 10^7$ l/man	$3 \cdot 10^9$ l/man
FRESHWATER PATHWAYS		
1. <u>Drinking water</u>		
Treatment removal factor, f ₁	1.0	
Consumption, I ₁	438 l a ⁻¹	
Collective dose commitment		
$S_1^c = W f_1 \left(\frac{NI}{V}\right)_1 D$		$2 \cdot 10^{-15}$ man Sv
2. <u>Fish</u>		
Concentration factor, f ₂	1.0	
Consumption, I ₂	1 kg a ⁻¹	
Collective dose commitment		
$S_2^c = W f_2 \left(\frac{NI}{V}\right)_2 D$		$5 \cdot 10^{-18}$ man Sv
SALT WATER PATHWAYS		
3. <u>Fish</u>		
Concentration factor, f ₃	1.0	
Consumption, I ₃	6 kg a ⁻¹	
Collective dose commitment		
$S_3^c = W f_3 \left(\frac{NI}{V}\right)_3 D$		$4 \cdot 10^{-20}$ man Sv
4. <u>Shellfish</u>		
Concentration factor, f ₄	1.0	
Consumption, I ₄	1 kg a ⁻¹	
Collective dose commitment		
$S_4^c = W f_4 \left(\frac{NI}{V}\right)_4 D$		$7 \cdot 10^{-21}$ man Sv

(iii) Summary of collective dose commitments per unit activity released

77. Table II.5 summarizes the values obtained above for the collective effective dose equivalent commitments per unit of ^3H activity released. With respect to the local and regional component due to industrial releases, the largest collective effective dose equivalent commitment per unit activity released is obtained for a river discharge and the smallest for a sea discharge while an intermediate value is found for the airborne discharge.

Table II.5

Summary of collective effective dose equivalent commitments per unit tritium activity released (man Sv per Bq)

Origin	Local and regional component	Global component
Natural		$5 \cdot 10^{-16}$
Nuclear tests		$8 \cdot 10^{-16}$
<u>Industry</u>		
Airborne discharge	$5 \cdot 10^{-17}$ (HTO)) $8 \cdot 10^{-16}$ (HTO)
River discharge	$2 \cdot 10^{-15}$	
Sea charge	$5 \cdot 10^{-20}$	

F. REFERENCES

- B1 Bennett, B.G. Environmental aspects of americium. EML-348 (1978).
- B2 Bowen, V.T. and W. Roether. Vertical distributions of strontium-90, caesium-137 and tritium near 45° north in the Atlantic. J. Geophys. Res. 78: 6277-6285 (1973).
- B3 Butler, H.L. and J.H. LeRoy. Observations of biological half-life of tritium. Health Phys. 11: 283-285 (1965).
- B4 Bennett, B.C. Environmental tritium and the dose to man. p. 1047-1053 in Proceedings of the Third International Congress of IRPA. CONF-730907 (1973).
- B5 Bennett, B.C. Fallout tritium in the environment and the dose commitments. HASL-268 (1973).
- B6 Bogen, D.C., G.A. Welford and C.G. White. Tritium distribution in man and his environment. p. 567-574 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- B7 Burger, L.L. Distribution and reactions of tritiated hydrogen and methane. p. 47-64 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- C1 Crowson, D.L. Man-made tritium. p. 23-27 in Tritium (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics, Las Vegas, Nevada, 1973.
- C2 Comps, F. and R.J. Doda. Large-scale distribution of tritium in a commercial product. p. 93-99 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- C3 Coyle, P.E. Laser fusion. Status, future and tritium control. p. 139-153 in Behaviour of Tritium in the Environment. IAEA, Vienna 1979.
- F1 Fireman, E.L. Measurement of the (n, H^3) cross section in nitrogen and its relationship to the tritium produced in the atmosphere. Phys. Rev. 91: 922-926 (1953).
- F2 Flamm, E., R.E. Lingenfelter, J. F. MacDonald et al. Tritium and helium-3 in solar flares and loss of helium from the earth's atmosphere. Science 138: 48-49 (1962).

- G1 Gratwohl, G. Erzeugung und Freisetzung von Tritium durch Reaktoren und Wiederaufarbeitungsanlagen und die voraussichtliche radiologische Belastung bis zum Jahr 2000. Kernforschungszentrum Karlsruhe report KFF-Ext. 4/73-36 (1973).
- H1 Häfele, W., J.P. Holdren, G. Kessler et al. Fusion and fast breeder reactors. IIASA RR-77-8 (1976) (revised July 1977).
- I1 International Atomic Energy Agency. Power reactors in member states. IAEA, Vienna, 1980.
- I2 International Commission on Radiological Protection. Report of the task group on reference man. ICRP publication 23. Pergamon Press, 1975.
- I3 International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. ICRP publication 30. Annals of the ICRP 2: 3/4 (1979)
- I4 International Atomic Energy Agency. Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- J1 Jacobs, D.G. Sources of tritium and its behaviour upon release to the environment. AEC Critical Review Series. TID-24635 (1968).
- K1 Kaufmann, S. and W.F. Libby. The natural distribution of tritium. Phys. Rev. 93: 1337-1344 (1954).
- K2 Kouts, H. and J. Long. Tritium production in nuclear reactors. p.38-45 in Tritium (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics, Las Vegas, Nevada, 1973.
- K3 Kahn, B., R.L. Blanchard, W.L. Brinck et al. Radiological surveillance study at the Haddam Neck PWR nuclear power station. EPA-520/374-007, Washington, 1974.
- K4 Krejčí, K. and A. Zeller, Jr. Tritium pollution in the Swiss luminous compound industry. p. 65-77 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- K5 Krejčí, K. Discussion. p. 101 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- L1 Lal, D. and B. Peters. Cosmic ray produced radioactivity on the earth. p. 551-612 in Encyclopaedia of Physics, Vol.

- XLVI/2 on Cosmic Rays (K. Sitte, ed.). Springer-Verlag, New York, 1967.
- L2 Locante, J. and D.D. Malinowski. Tritium in pressurized water reactors. p. 45-57 in Tritium (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics, Las Vegas, Nevada, 1973.
- L3 Luykx, F. and G. Fraser. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European community. Discharge data 1974-1978. Radiological aspects. Commission of the European Communities. V/4116/80 (1980).
- L4 Lambert, B.E., H.B.A. Sharpe and K.B. Dawson. Am. Ind. Hyg. Assoc. J. 32: 682 (1971).
- M1 Miskell, J.A. Production of tritium by nuclear weapons. p. 79-85 in Tritium (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics, Phoenix and Las Vegas, 1973.
- M2 Michel, L. Tritium inventories of the world oceans and their implications. Nature 263: 103-106 (1976).
- M3 Mason, A.S. and H.G. Ostlund. Atmospheric HT and HTO: V. Distribution and large-scale circulation. p. 3-16 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- M4 Murphy, C.E. Jr. and M.M. Pendergast. Environmental transport and cycling of tritium in the vicinity of atmospheric releases. p. 361-372 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- M5 Minder, W. Strahlentherapie. 137: 700 (1969).
- M6 Moghissi, A.A., M.W. Carter and E.W. Bretthauer. Further studies on the long-term evaluation of the biological half-life of tritium. Health Phys. 23: 805-806 (1972).
- M7 Moghissi, A.A. and M.W. Carter, eds. Tritium. Messenger Graphics, Las Vegas, Nevada, 1973.
- N1 National Council on Radiation Protection and Measurements. Tritium in the environment. NCRP No. 62 (1979).
- N2 National Council on Radiation Protection and Measurements. Tritium and other radionuclide labelled

- organic compounds incorporated in genetic material. NCRP No. 63 (1979).
- O1 Ostlund, H.G. and R. A. Fine. Oceanic distribution and transport of tritium. p. 303-314 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- S1 Schell, W.R., S. Sauzay and B.R. Payne. World distribution of environmental tritium. p. 374-385 in Physical Behaviour of Radioactive Contaminants in the Atmosphere. IAEA, Vienna, 1974.
- S2 Smith, J.M. and R.S. Gilbert. Tritium experience in boiling water reactors. p. 57-68 in Tritium (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics, Las Vegas, Nevada, 1973.
- S3 Sanders, S.M. Hr. and W. C. Reinig. Assessment of tritium in man. p. 534-542 in Diagnosis and Treatment of Deposited Radionuclides (H.A. Kornberg and W.D. Norwood, eds.). Excerpta Medica Foundation, Amsterdam, 1968.
- T1 Trevorrow, L.E., B.J. Kullen, R.L. Jarry et al. Tritium and noble gas fission products in the nuclear fuel cycle. I. Reactors. ANL-8102 (1974).
- U1 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General Assembly, with annexes. United Nations sales publication no. E.77.IX.I. New York, 1977.
- W1 Wehner, G. Discharges of tritium to the environment from unrestricted use of consumer products containing this radionuclide. p. 79-91 in Behaviour of Tritium in the Environment. IAEA, Vienna, 1979.
- W2 Woodard, H.Q. The biological effects of tritium. United States Atomic Energy Commission. HASL-229 (1970).
- W3 Wylie, K. F., W. A. Bigler and G.R. Grove. Biological half-life of tritium. Health Phys. 9: 911-914 (1963).

III. CARBON-14

A. INTRODUCTION

78. Carbon-14 has always been present on the earth. It is produced by cosmic ray interactions in the atmosphere. This nuclide is a pure beta-emitter, with a half-life of 5730 years, a maximum energy of 185 keV and an average energy of 49.47 keV [N1].

79. Carbon is one of the elements that are essential to all forms of life and is involved in most biological and geochemical processes on the earth. Associated with the stable isotopes of carbon (^{12}C and about 1.1% ^{13}C), there is a very small amount of ^{14}C formed in the atmosphere and which has subsequently entered in the carbon cycle. The specific activity of biological carbon, as measured in wood samples grown in the nineteenth century, was 0.227 ± 0.001 Bq per gram of carbon [T1], corresponding to an atmospheric inventory of $1.4 \cdot 10^{17}$ Bq. During the present century the specific activity of ^{14}C has decreased due to the diluting effect of releases into the atmosphere of carbon dioxide from the combustion of fossil fuels. This effect (the Suess effect) accounts for a reduction of a few percent.

80. In addition to its natural production, carbon-14 is also produced by the detonation of nuclear explosives and by the operation of nuclear reactors. The assessment of the collective dose commitments from the releases of man-made carbon-14 is facilitated by knowledge of the production rate of natural carbon-14.

B. SOURCES

1. Natural carbon-14

81. Carbon-14 is produced by the action of cosmic ray neutrons on nitrogen atoms, both in the stratosphere and in the upper troposphere. UNSCEAR [U3] has estimated the natural production rate to be about 10^{15} Bq per year, a value which has been derived from assessments of the natural ^{14}C inventory. The production rate has also been estimated directly from assessments of cosmic ray neutrons and the values obtained by different authors range from 1 to $1.4 \cdot 10^{15}$ Bq per year [U3]. Considering the uncertainties involved in determining both the direct production rate and also the total ^{14}C inventory of the earth, the estimates are in reasonable agreement.

2. Nuclear explosions

82. Carbon-14 is formed in nuclear explosions through the capture of excess neutrons by atmospheric nitrogen. After large atmospheric nuclear explosions, most of the ^{14}C is transported into the stratosphere, from where it equilibrates with the troposphere with a half-time of 1 to 2 years [U3].

83. The inventory of ^{14}C from nuclear explosions has been estimated from measurements of excess specific activity in the troposphere and in the surface ocean waters, and models representing the exchange of ^{14}C between the atmosphere, the biosphere and the ocean. UNSCEAR [U3] has estimated that nuclear explosions up to 1972 have injected into the atmosphere $2.1 \cdot 10^{17}$ Bq, while subsequent injections have increased this amount by about 1%.

84. For the past pattern of atmospheric nuclear explosions, the production mentioned above corresponds to about $3.7 \cdot 10^{14}$ Bq per megaton. This value, however, is not representative of any given nuclear explosion, because the production of ^{14}C will depend on the type of nuclear device exploded and also on whether the explosion took place on the surface of the earth or high in the atmosphere. A "surface" test will produce approximately 50% of the ^{14}C that would be produced by the same device in an "air" test, because about one half of the escaping neutrons will be captured in the soil or water rather than in the atmosphere.

3. Nuclear fuel cycle

85. Carbon-14 is produced in nuclear reactors and is released to the environment at the reactor itself or at reprocessing plants where spent fuel is reprocessed. Only recently has attention been given to the production and release of this radionuclide at nuclear fuel cycle installations.

(a) Nuclear reactors

86. The production of carbon-14 in nuclear power reactors is due to several nuclear reactions in the fuel, core construction materials and moderator. Figure III.I summarizes the relevant reactions.

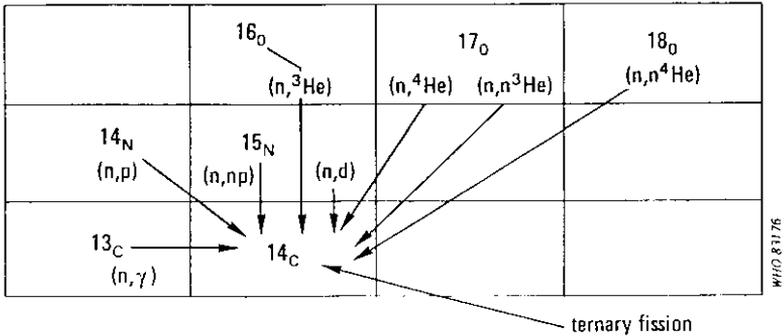


Figure III.I Nuclear reactions giving rise to the production of carbon-14.

87. Production rates depend upon the neutron flux, the shape of the respective neutron spectra and the resulting effective cross sections, on the amount of the target elements present in different reactor components and on the abundance of the target isotopes in the target elements. The target elements are uranium, nitrogen, oxygen, and also carbon in the case of graphite moderated reactors. Nitrogen is present as an impurity in the fuel, as dissolved gas in the coolant, as nitrogen compounds sometimes used for pH control in the coolant, and as an impurity in structural materials. Oxygen is present in water moderators and coolants, in CO₂ coolants, and in oxide fuels (e.g., UO₂).

88. The place of origin of ¹⁴C within a nuclear reactor has a strong influence on the discharge pathway. One can basically distinguish between three locations of ¹⁴C generation, namely, ¹⁴C in the fuel, ¹⁴C in structural materials of the core (and solid moderator, if applicable) and ¹⁴C in the reactor coolant (and liquid moderator, if applicable).

89. The ¹⁴C produced in liquid or gaseous coolants will be present in different chemical compounds (CO₂, CO, methane), depending on the chemical environment. Under the influence of intensive radiation fields several chemical reactions may occur, influencing the chemical form of carbon-14. The compounds in the coolant are released mainly together with off-gas and waste water from the coolant purification and

treatment system. Part of the carbon-14 also leaks from the primary coolant circuit into the plant ventilation system and is released with ventilation air.

90. Significant reactions for the production of ^{14}C in light water reactors (LWR) are: (n) reactions with ^{17}O present in the oxide fuel and in the moderator; (n, p) reactions with ^{14}N present in the fuel as impurities; and ternary fissions. Ternary fission production per unit electrical energy generated is practically independent of reactor design, while the normalized production of ^{14}C by the other reactions depends on the enrichment of the fuel, the relative masses of the fuel and moderator, the concentration of nitrogen impurities in the fuel and the temperature of the fuel and moderator.

91. In boiling water reactors (BWR), the gaseous ^{14}C is transported with the steam until it arrives at the turbine condenser. There the gases are continuously withdrawn over a catalytic recombiner to burn the hydrogen and oxygen produced by radiolysis of the primary water. Measurements have shown that one half or more of the total ^{14}C produced in the coolant will be discharged in the form of CO_2 together with the filtered gases from the turbine condenser. There are other pathways of release of ^{14}C , mainly caused by leakage from the primary circuit into the reactor building and the turbine hall. These releases are also mainly in the form of CO_2 . A part of the ^{14}C remains dissolved in the primary water purification and treatment systems, causing smaller sources of release, for example in the auxiliary building and finally in the waste water system.

92. The primary circuit water of a pressurized water reactor (PWR) contains hydrogen in excess to recombine the oxygen produced by radiolysis. Under such reducing conditions compounds like methane will be formed. Therefore, contrary to the BWR, a PWR will release most of the ^{14}C bound in hydrocarbons. The main release pathways for gaseous compounds of ^{14}C in PWRs are leakages of the primary water circuit into the containment air and the degasification of the primary water. The escaping or withdrawn gases may be stored in decay tanks prior to release, and the gaseous ^{14}C compounds can be oxidized to CO_2 or released through charcoal beds. Leakages may also arise in the auxiliary building from the primary water purification and treatment systems by way of degassing. Also, a part of the ^{14}C compounds stays dissolved in the water and is released at the different steps of the waste water treatment.

93. The total environmental release of carbon-14 at the reactor, expressed as a fraction of the production rate, is on the average about 50% in BWRs and 30% in PWRs, but the value is quite variable, as has been shown by several recent monitoring programmes [R1, L1]. UNSCEAR summarized the estimates of production in LWRs from several authors, the values being in the range 0.5 to $1.9 \cdot 10^9$ Bq per MW(e)a, and also derived an independent value of about $0.7 \cdot 10^9$ Bq per MW(e)a [U3].

94. Carbon-14 is generated in heavy water reactors (HWR) through reactions similar to those described for LWRs. Owing mainly to the large moderator mass, the production rate of ^{14}C in HWRs is expected to be considerably larger than in LWRs [U3]. The production rate in pressure vessel reactors is estimated to be $1.7 \cdot 10^{10}$ Bq per MW(e)a, with 90% generated in the moderator. The production of ^{14}C in CANDU reactors is estimated to be $1.6 \cdot 10^{10}$ Bq per MW(e)a, 95% being produced in the moderator.

95. In gas-cooled graphite-moderated reactors (GCR), the major source of ^{14}C production is the graphite moderator, due to $^{13}\text{C}(n, \gamma)^{14}\text{C}$ reaction and also to the $^{14}\text{N}(n, p)^{14}\text{C}$ reaction based on the incorporated nitrogen impurity. Production rates have been estimated to be about $0.7 \cdot 10^{10}$ Bq per MW(e)a in Magnox reactors and $1.1 \cdot 10^{10}$ Bq per MW(e)a in advanced gas-cooled reactors (AGR) [U3]. Production of ^{14}C in the carbon dioxide coolant, mainly from activation of nitrogen impurities and from the $^{17}\text{O}(n, \alpha)^{14}\text{C}$ reaction, is a smaller source estimated to be about 10^8 Bq per MW(e)a for Magnox reactors and $4 \cdot 10^8$ Bq per MW(e)a for AGRs.

96. Carbon-14 discharges from Magnox reactors and AGRs result from coolant leakage and include ^{14}C released to the coolant from corrosion of the moderator. The fraction released at the reactor is about 3% in Magnox reactors and about 6% in AGRs, of the total production rate of ^{14}C in these reactors [U3].

(b) Fuel reprocessing plants

97. While the ^{14}C produced in the reactor coolant and moderator has a potential for immediate release at the nuclear reactor, the ^{14}C produced in the fuel will be released only later during nuclear fuel reprocessing. Depending on reprocessing plant operation characteristics the release may be continuous or discontinuous. There are few measurements of ^{14}C releases from reprocessing installations [S1], but it seems reasonable to assume that almost all the inventory of

the fuel elements is released during the chemical dissolution of the fuel. In the case of the Purex process the ^{14}C is released in the form of CO_2 .

(c) Summary

98. A very rough estimate can be made of the total production and release of ^{14}C from nuclear fuel cycle installations, based on the average values given above. Installed nuclear capacity worldwide in 1980 was $1.25 \cdot 10^5$ GW(e) [12]. Assuming an average load factor for reactor operation of 0.6, the energy produced was $7.5 \cdot 10^4$ GW(e)a. Global production and release of ^{14}C from reactor sites are thus estimated to be about $1.4 \cdot 10^{14}$ Bq and $6 \cdot 10^{13}$ Bq, respectively. The estimated discharges by reactor types are given in Table III.1. There are no estimates of production and release from other reactor types representing 10% of the total installed capacity. The difference between production and reactor discharge estimates will largely represent the release from reprocessing plants, to the extent that the fuel is eventually reprocessed.

Table III.1

Estimated global discharge of carbon-14
from nuclear power stations in 1980

Reactor type	Reactor number	Capacity [MW(e)]	Production rate [Bq per MW(e)]	Release fraction (%)	Estimated carbon-14 discharge (Bq)
PWR	96	64239	$7 \cdot 10^8$	30	$8 \cdot 10^{12}$
BWR	62	35170	$7 \cdot 10^8$	50	$7 \cdot 10^{12}$
HWR	14	5963	$1.6 \cdot 10^{10}$	70	$4 \cdot 10^{13}$
GCR	36	7086	$9 \cdot 10^9$	5	$2 \cdot 10^{12}$
Other	33	12527	-	-	-
Total	241	124985			$6 \cdot 10^{13}$

C. BEHAVIOUR IN THE ENVIRONMENT

99. Carbon-14 is present in atmospheric carbon dioxide, in the biosphere, and in the bicarbonates dissolved in the ocean. The specific activity of natural ^{14}C in the terrestrial biosphere, as measured in wood grown in the nineteenth century, was 0.227 ± 0.001 Bq per gram of carbon. The Suess effect, accounting for a few percent decrease of specific activity at present, could reach a figure of the order of 20% in the year 2000 [U2], but is of little importance in the long range, when fossil fuel resources are exhausted.

100. Leaving aside the Suess effect, it has been suggested, however, that the present-day inventory does not correspond to the equilibrium value, but is increasing. In fact, measurements of wood samples of known age show that only cyclic variations of atmospheric ^{14}C , amounting to a few percent, have occurred in the past 6000 years [U2]. Two types of variations have been recognized; one, with a time scale of the order of 100 years, has been explained by the solar wind modulation of the cosmic-ray flux density; the other, with a time constant of more than 1000 years, may largely be due to a variation of the geomagnetic shielding of the earth.

101. Contrary to the case of natural carbon-14, the levels of man-made carbon-14 are not at steady state in the different compartments of the environment. Due to the very long mean life of carbon-14, continuing practices are not expected to last long enough to allow the environmental levels to reach the steady state. The predictions of the time-evolution of ^{14}C levels in the atmosphere, biosphere and ocean after a release into the environment require, therefore, the use of compartment models.

102. Many models describing the dispersion of released ^{14}C , and the subsequent exchange between the different compartments involved in the carbon cycle, have been proposed [C1, P1, N2, Y1, N3]. UNSCEAR [U3] also developed a dynamic model for the assessment of doses from ^{14}C released by nuclear explosions. This model includes compartments for the atmosphere and short-term biosphere, the terrestrial biosphere, the surface ocean and the deep ocean, and represents the thermocline layer in the ocean as a thick diffusion barrier.

D. TRANSFER TO MAN

103. Carbon-14 released to the environment enters the carbon cycle, giving rise eventually to increased levels in man. From measurements of fallout carbon-14, it was noted that the specific activity in human tissue comes into equilibrium with that of atmospheric CO₂ with a delay time of about 1.4 years [N5].

104. Intake of carbon by man is primarily from diet. Ingestion intake is of the order of 300 g d⁻¹ with nearly complete absorption, whereas inhalation intake is about 3 g d⁻¹ with only 1% retained in the body [U3]. The total carbon content of the body is 1.6 10⁴ [I1]. The quotient of this with the intake rate gives an estimated mean residence time of carbon in the human body of 53 days.

105. Man comes, therefore, into fairly rapid equilibrium with carbon-14 in his immediate environment. It is generally sufficient in carbon-14 dose calculations to adopt a steady-state model which assumes that the specific activity of carbon in tissues is in equilibrium with that in air and in the diet.

E. DOSIMETRY

1. Dose per unit intake

106. An intake of carbon-14 at a specific concentration of 0.23 Bq per gram of carbon, corresponding to the present value for natural carbon-14, gives rise to the following absorbed dose rate averaged over the whole body

$$0.23 \frac{\text{Bq}}{\text{g}_c} 1.6 \cdot 10^{-10} \frac{\text{Gy g}}{\text{MeV}} \frac{0.049 \text{ MeV/Bq s}}{7 \cdot 10^4 \text{ g}}$$
$$3.15 \cdot 10^7 \text{ s/a } 1.6 \cdot 10^4 \text{ g}_c = 13 \text{ } \mu\text{Gy a}^{-1}$$

The dose rates in individual tissues depend on their carbon concentrations. The carbon content per unit mass averages 23% for the whole body, but ranges from 9% in gonads and 10% in lungs to 41% in red bone marrow and 67% in adipose tissue [I1]. The annual absorbed doses are 5 μGy in gonads, 6 μGy in lungs, 20 μGy in bone-lining cells and 22 μGy in red bone marrow [U3]. The tissue-weighted annual effective dose equivalent from natural carbon-14 is 12 μSv.

107. This dose is due almost entirely to ingestion intake of carbon-14. If the carbon intake rate is 300 g d⁻¹ at the

specific activity of 0.23 Bq g^{-1} , the intake rate of ^{14}C is 69 Bq d^{-1} . The effective dose equivalent per unit ingestion intake of ^{14}C is

$$\frac{12 \cdot 10^{-6} \text{ Sv/a}}{69 \text{ Bq/d}} \cdot \frac{1 \text{ a}}{365 \text{ d}} = 5.2 \cdot 10^{-10} \text{ Gy Bq}^{-1}$$

The dose factor for inhalation intake is less by a factor of 10^{-2} , since absorption into the body is that much less by this pathway.

2. Dose per unit release

108. The doses given above for natural carbon-14 correspond to the annual global production of 10^{15} Bq . This production is essentially constant in time and uniform over the world. Therefore, equilibrium has become established. The effective dose equivalent commitment per unit release is

$$\frac{12 \cdot 10^{-6} \text{ Sv/a}}{10^{15} \text{ Bq/a}} = 1.2 \cdot 10^{-20} \text{ Sv Bq}^{-1}$$

The collective dose equivalent rate from natural carbon-14 to the world population of $4 \cdot 10^9$ people is $4.8 \cdot 10^4 \text{ man Sv a}^{-1}$.

109. The assessment of the dose commitment from a given release of man-made carbon-14 is carried out by direct analogy with natural carbon-14. Once the released carbon-14 enters the global carbon cycle, the effective dose equivalent commitment per unit release is $1.2 \cdot 10^{-20} \text{ Sv Bq}^{-1}$.

110. It is difficult to assess with precision the collective dose commitment per unit release of carbon-14, because the projected increase in the world population is very uncertain. Assuming that it will attain an equilibrium value of 10^{10} persons, in a time short compared with the mean effective life of ^{14}C [U3], the collective effective dose equivalent commitment per unit released is approximately $1.2 \cdot 10^{-10} \text{ man Sv per Bq}$.

111. In order to calculate the complete collective dose commitment [U3] required for assessments of maximum future mean annual doses from a continuing but finite practice releasing ^{14}C , it is necessary to use dynamic models predicting the time evolution of environmental levels. Assuming that power production by nuclear fission will last for a few hundred years (for example, 500 years), the

incomplete collective dose commitment can be calculated using the model with diffusion barrier already mentioned. The incomplete collective dose commitment, integrated over 500 years, is about $2.3 \cdot 10^{-11}$ man Sv per Bq released. This value is somewhat higher than a value of about $1.4 \cdot 10^{-11}$ man Sv per Bq which can be deduced from a recent assessment of the environmental significance of ^{14}C [N3], but in view of the uncertainties involved, the difference is probably insignificant.

112. The contribution of local and regional exposures to the collective dose commitment is very small, of the order of a percent, and can be neglected [N3]. The assessment of individual doses at some selected locations, however, is necessary for radiation protection purposes. Its calculations can be carried out by the use of specific activity methods. One simple model assumes that the specific activity of ^{14}C in air is equal to that in the body. A more sophisticated calculation assumes that the specific activity in the vegetation at the location of interest is equal to that of air. The dose can then be assessed from knowledge of the relative proportion of contaminated food in the diet. Both methods require the use of micrometeorological models to assess quantitatively the dispersion of ^{14}C from the release point to the locations of interest. Some publications [U4, N4, C2], present improvements to the classical formulations describing the local atmospheric dispersion.

F. REFERENCES

- C1 Craig, H. The natural distribution of radiocarbon and the exchange time of carbon dioxide between atmosphere and sea. *Tellus* 9: 1-17 (1957).
- C2 Clarke, R. A model for short and medium range dispersion of radionuclides released to the atmosphere. A first report of a working group on atmospheric dispersion. NRPB-R91 (1979).
- I1 International Commission on Radiological Protection. Report of the task group on reference man. International Commission on Radiological Protection publication 23 (1975).
- I2 International Atomic Energy Agency. Power reactors in member states. IAEA, Vienna, 1980.
- L1 Luykx, F. and G. Fraser. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European community: discharge data 1962-76. Radiological aspects. Commission of the European Communities. V/4604/78-EN (1978).
- N1 National Council on Radiation Protection and Measurements. A handbook of radioactivity measurements procedures. National Council on Radiation Protection report No. 58 (1978).
- N2 Nydal, R. Further investigation on the transfer of radiocarbon in nature. *J. Geophys. Res.* 73: 3617-3635 (1968).
- N3 Nuclear Energy Agency, OECD. Radiological significance and management of H-3, C-14, Kr-85 and I-129 arising from the nuclear fuel cycle. Report by an NEA group of experts. OECD/NEA (1980).
- N4 NRPB and CEA. Methodology for evaluation of radiological consequences of radioactive effluents released in normal operations. Commission of European Communities. V/3865/79 (1979).
- N5 Nydal, R., K. Lovseth and O. Syrstad. Bomb 14-C in the human population. *Nature* 232: 418-421 (1971).
- P1 Plesset, M. and A. Latter. Transient effects in the distribution of carbon-14 in nature. Proceeding of the

National Academy of Sciences 46: 232-241 (1960).

- R1 Riedel, H. and P. Gesewsky. Zweiter Bericht über Messungen zur Emission von Kohlenstoff-14 mit der Abluft aus Kernkraftwerken mit Leichtwasserreaktor in der Bundesrepublik Deutschland. Bundesgesundheitsamt report STH-13/77 (1978).
- S1 Schuettelkopf, H. and G. Herrman. 14-CO₂ Emissionen aus der Wiederaufarbeitungsanlage Karlsruhe. p. 189 in Report for the Commission of the European Communities. V/2266/78-D (1978).
- T1 Telegadas, K. The seasonal atmospheric distribution and inventories of excess carbon-14 from March 1955 to July 1969. HASL-243 (1971).
- U2 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, with annexes. Volume I: Levels, Volume II: Effects. United Nations sales publication No. E.72.IX.17 and 18. New York, 1972.
- U3 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General Assembly, with annexes. United Nations sales publication No. E.77.IX.I. New York, 1977.
- U4 U.S. Nuclear Regulatory Commission. Regulatory Guide 1.111 (1977).
- Y1 Young, J. and A. Fairhall. Radiocarbon from nuclear weapons test. J. Geophys. Res. 73: 1185-1200 (1968).

IV. KRYPTON-85

A. INTRODUCTION

113. Krypton is element number 36 in the periodic table. It belongs to the group of inert gases together with helium, neon, argon, xenon and radon. It occurs naturally in the atmosphere to an estimated extent of 1 to 2 10^{-6} by volume.

114. The naturally occurring stable krypton isotopes and their atom percentage abundances are: ^{76}Kr (0.35%), ^{80}Kr (2.27%), ^{82}Kr (11.56%), ^{83}Kr (11.55%), ^{84}Kr (56.9%), ^{86}Kr (17.37%) [N1]. The radioactive isotopes of krypton include mass numbers of 74-77, 79, 79m, 81, 81m, 85, 85m, 87-95 and 97. Some of these occur naturally in low trace amounts as a result of cosmic ray induced reactions with stable krypton isotopes and by spontaneous fission of natural uranium.

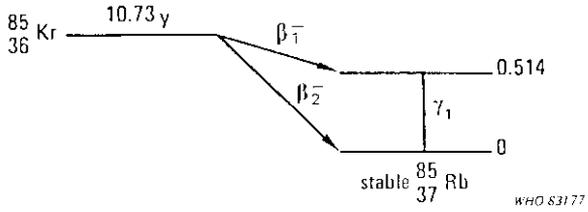
115. The radioactive isotope ^{85}Kr is produced in nuclear fission. With a half-life of 10.7 years, it can become widely dispersed in the atmosphere following release. The average fission yields differ by about a factor of 2 for ^{239}Pu and ^{235}U , being about 0.6 and 1.3 atoms per 100 fissions, respectively (Table IV.1).

Table IV.1

Fission yields of krypton-85
[C2]

Nuclide	Fission yield (%)	
	thermal	fast
^{237}Th		4.14
^{235}U	2.28	2.12
^{239}Pu	1.32	1.33
^{238}Pu		0.74
^{238}U	0.558	0.62

116. The decay scheme of ^{85}Kr is presented in Figure IV.I. Two beta particles and a single gamma photon are emitted, along with several low-energy conversion electrons and x rays.



Radiation type	Energy (keV)	Intensity (%)
β_1^-	max. 173.0 avg. 47.5	0.437
β_2^-	max. 687.0 avg. 251.4	99.563
γ_1	514.0	0.434

Figure IV.I. Decay scheme for krypton-85.

117. Being chemically inert, krypton and other inert gases are not usually involved in biological processes. They are, however, dissolved in body fluids and tissues following inhalation. Krypton is characterized by low blood solubility, high lipid solubility and rapid diffusion in tissue [K1]. The biological involvement of inert gases has been noted in numerous studies [K1].

B. SOURCES

118. Krypton-85 is produced by cosmic ray interactions in the atmosphere, in nuclear power reactors, and nuclear explosions. The main release source is the dissolution step in the reprocessing of nuclear fuel.

119. Concentrations of ^{85}Kr in the atmosphere increased sharply after 1955 due to the production and testing of nuclear weapons and the development of the nuclear power industry. More recently the input rates of ^{85}Kr into air have decreased [H2]. There have been reductions in plutonium production for military purposes and in nuclear fuel reprocessing.

120. A review of ^{85}Kr measurement data for 1950-77 has been prepared by Rozanski [R1]. The most recent data indicate that concentrations in air have stabilized at about 0.6 Bq/m^3 in the northern hemisphere and 0.4 Bq/m^3 in the southern hemisphere [R1]. The major sources are in the northern hemisphere, accounting for the higher levels in that hemisphere.

1. Natural krypton-85

121. Krypton-85 is present in small amounts in the environment as a result of spontaneous fission of natural uranium and interactions of cosmic ray neutrons with atmospheric ^{84}Kr . The steady state environmental inventories of ^{85}Kr from these sources have been calculated: $7.4 \cdot 10^{10} \text{ Bq}$ in the upper 3 m of the total land and water surface due to spontaneous fission of natural uranium, $3.7 \cdot 10^{11} \text{ Bq}$ in the atmosphere from cosmic ray production and $3.7 \cdot 10^5 \text{ Bq}$ in the oceans from the atmospheric source [D1]. These estimates, in comparison with the estimates of man-made sources of ^{85}Kr to follow, are negligible in contributing to the world's total ^{85}Kr inventory.

2. Nuclear explosions

122. Since ^{85}Kr is produced during fission, it has been generated by nuclear weapon tests. The total amount of ^{85}Kr produced in nuclear testing can be calculated from the ratio of $^{85}\text{Kr}/^{90}\text{Sr}$ fission yield of 0.08, giving an activity ratio of 0.22 [C2]. Measurements of ^{90}Sr activity have been reported and discussed in the reports of UNSCEAR [U1-U7]. There have been $6 \cdot 10^{17} \text{ Bq}$ of ^{90}Sr produced in weapon testing through 1976 [U7], corresponding to about $1.3 \cdot 10^{17} \text{ Bq}$ of ^{85}Kr .

123. Another source of ^{85}Kr associated with nuclear weapons is in the production of plutonium in military reactors. The amount of ^{85}Kr released from this source is estimated to be two times higher than that from the weapon tests [D1]. Naval propulsion reactors also contribute to the ^{85}Kr inventory with an annual production in the region of 1.1 to $1.9 \cdot 10^{16} \text{ Bq}$ [B1]. Including all sources, the total amount of ^{85}Kr produced in operations for military purposes is still rather small in comparison to the prospective generation of ^{85}Kr by the nuclear power industry.

3. Nuclear fuel cycle

124. Krypton-85 is produced by fission in the fuel of nuclear reactors and in very low trace amounts in the moderator or coolant, due to contamination with fissile material. The rates of ^{85}Kr production are related to the type of fuel and degree of burn-up. Production and emission rates may be conveniently normalized to unit electrical energy generated (for power reactors) or to the electrical energy generated by the reactors serviced (for fuel reprocessing plants).

125. The amounts of ^{85}Kr produced vary according to reactor type. For thermal reactors, the range of estimated production is about 1.1 to $1.5 \cdot 10^{13}$ Bq/MW(e)a. For FBRs the values are about 25% smaller [E1, M1], for HTGRs 50% higher [B3]. A production rate of $1.4 \cdot 10^{13}$ Bq/MW(e)a has been correlated with some measurements from reprocessing plants [U7] and this value can be taken for general evaluations.

126. An estimate of ^{85}Kr annual generation from reactor operation can be obtained from the installed capacity of nuclear reactors of $1.25 \cdot 10^5$ MW(e) worldwide in 1980 [I1], with the assumptions of 60% utilization and average ^{85}Kr generation rate of $1.4 \cdot 10^{13}$ Bq/MW(e)a:

$$1.25 \cdot 10^5 \text{ MW(e)a} \times 0.6 \times 1.4 \cdot 10^{13} \text{ Bq/MW(e)a} = 1 \cdot 10^{16} \text{ Bq/a}$$

The actual release rate is less, since delays occur before reprocessing and not all fuel is reprocessed.

127. Reported releases of ^{85}Kr and other fission noble gases were listed in the 1977 report of UNSCEAR [U7]. There are large differences in the release values of the various plants. Although the relevant data are not very extensive, there are indications of improved retention of ^{85}Kr at reactors in recent years due to the installation of additional hold-up tanks or adsorption columns.

128. In the reprocessing plant the spent fuel elements are dismantled and the nuclear material dissolved. Procedures to separate ^{85}Kr from gaseous effluents and to provide long-term retention are under study, but current practice is to allow controlled release to the atmosphere.

C. BEHAVIOUR IN THE ENVIRONMENT

129. Krypton-85 discharged to the environment disperses in the atmosphere and largely remains there until decay. It can

become washed out by rain and diffuse into surface layers of soil and oceans, but these processes account for very little transfer of ^{85}Kr from the atmosphere.

1. Dispersion in the atmosphere

130. Materials released to the atmosphere are transported downwind and dispersed according to atmospheric mixing processes. The estimation of this dispersion is commonly approached by using a diffusion-transport equation. Several models have been developed for this purpose, using a variety of boundary conditions and simplifying assumptions. Most of them are based on the Gaussian plume diffusion model [S1, I2], which has been shown to be adequate in many practical situations. The krypton concentrations in air at various distances for a release from a 30 m high stack are shown in Table IV.2 [C5].

Table IV.2

Krypton-85 concentration in air for a release of 1 Bq/s
(stack height 30 m, Pasquill category D)
{C5}

Distance (km)	Concentration (Bq/m ³)
1	$4.8 \cdot 10^{-7}$
10	$1.3 \cdot 10^{-8}$
100	$4.4 \cdot 10^{-10}$
1000	$3.2 \cdot 10^{-11}$

131. For estimation of dispersion at greater distances, some shortcomings in the Gaussian model are evident in the assumptions that the meteorologic conditions and the direction of the wind remain constant throughout the transit of the plume. To overcome these difficulties, long-range models have been developed [A1, D3, M2], which follow the trajectories of masses of air passing over the release point and take into account the changing meteorologic conditions with time. A survey of several diffusion models and of their applications is given in [C5].

132. The global circulation of ^{85}Kr can be approximated by a simple compartment model, consisting of single compartments representing the atmosphere in the northern and in the southern hemispheres. Following a single release, equilibrium concentrations in the atmosphere are achieved after about two years. Further decrease in concentrations is due to radioactive decay. In applying this model, Kelly et al. [K3] determined that the integral concentration in air would be $5.3 \cdot 10^{-10} \text{ Bq a m}^{-3}$ per Bq released. The atmospheric mass was assumed to be $3.8 \cdot 10^{21} \text{ g}$, equivalent to $3.1 \cdot 10^{18} \text{ m}^3$ at STP.

133. The dispersion calculations of Machta et al. [M2] are based on detailed meteorological considerations and allow population-weighted exposures to be determined. Table IV.3 lists the average surface air concentrations of ^{85}Kr in latitude bands following release of 1 Bq in the $30-50^\circ \text{ N}$ latitude band. Uniform concentrations are achieved after two years, after which the integral concentration until complete decay is

$$22 \cdot 10^{-20} \text{ Bq/m}^3 \frac{10.73 \text{ a}}{\ln 2} = 3.4 \cdot 10^{-18} \text{ Bq a/m}^3$$

Adding the contributions from the first two years gives $3.9 \cdot 10^{-18} \text{ Bq a/m}^3$ for the population weighted integral concentration of ^{85}Kr in air from a release of 1 Bq.

2. Removal from the atmosphere

134. There is very little removal of ^{85}Kr from the atmosphere, except by radio-active decay. The low solubility of krypton in water limits the accumulation of ^{85}Kr in rainwater. Adsorption of ^{85}Kr on particulate matter in air and subsequent deposition of the particles provides a removal means of very low efficiency [N1].

135. The transfer of ^{85}Kr to soil can occur by diffusion processes; however, estimates of this transfer can account for only about 0.05% of the total krypton in the atmosphere [N1]. Therefore, soil in general is not an important removal sink for ^{85}Kr .

Table IV.3

Average surface air concentration of krypton-85
 (1 Bq emitted uniformly over one year in 30-50° N latitude band)
 [M2]

Latitude band	Krypton-85 concentration (10^{-10} Bq/m ³)			Population distribution %
	Year 1	Year 2	Year 3	
70 - 90° N	23	32	22	-
50 - 70° N	25	31	22	12.6
30 - 50° N	23	30	22	32.0
10 - 30° N	19	27	22	39.0
10° N - 10° S	11	22	22	11.5
10 - 30° S	6.3	22	22	3.4
30 - 50° S	5.1	20	22	1.5
50 - 70° S	4.3	19	22	0.05
70 - 90° S	3.8	19	22	-
Population weighted integral concentration (10^{-2} Bq a/m ³)	19.5	27.6	22.0	

136. The efficiency of the oceans as a sink for ⁸⁵Kr can be determined from the natural krypton content of the atmosphere and of the mixed layer of the ocean. From estimates of the krypton concentration in air, the atmospheric volume and the density krypton (STP), a total mass of about $1.64 \cdot 10^{16}$ g of krypton in the atmosphere is calculated [N1]. Assuming that the mixed layer of the ocean extends to 100 m depth and an area of $3.6 \cdot 10^{18}$ cm², and using the measured average krypton concentration in this layer of seawater of $5 \cdot 10^{-8}$ by volume [B4], a total mass of $6.7 \cdot 10^{12}$ g of krypton in the mixed layer of the ocean is obtained. This corresponds approximately to 0.04% of the atmospheric mass of krypton.

137. An estimate of the total mass of krypton in the oceans as a whole is obtained using an average concentration by volume of krypton in the oceans of $9 \cdot 10^{-8}$ [B4], a total ocean volume of $1.4 \cdot 10^{24}$ cm³, and a krypton density of $3.73 \cdot 10^{-3}$ g/cm³ at STP. This calculation results in a total ocean inventory of about $4.7 \cdot 10^{14}$ g of krypton, or approximately 3% of the total atmospheric krypton [N1]. These

figures clearly indicate that the oceans can serve only as a minor sink for ^{85}Kr discharged into the atmosphere.

D. TRANSFER TO MAN

138. Following release to the atmosphere ^{85}Kr becomes widely dispersed. Exposure of man occurs by external irradiation from the passing cloud or the dispersed gas and by internal irradiation following inhalation of ^{85}Kr and absorption in tissues.

139. After intake, ^{85}Kr is distributed in the body by blood and lymph fluids and is absorbed in the various tissues. A person immersed in an atmosphere of ^{85}Kr at low concentration would rather quickly come into equilibrium with it. The concentrations in body tissues are determined by multiplying the concentration in air by a partitioning factor, called the Ostwald's coefficient. The relevant values reflect the rate at which tissues are perfused with blood, the solubility of the gas in the several tissues and the velocity of diffusion of krypton across anatomical boundaries. The concentration of ^{85}Kr in the body is not uniform, the concentration in the adipose tissue being nearly 50 times higher than that in other parts of the body.

140. As a first approximation, one may only account for a difference in the absorption behaviour of krypton in fat and non-fat tissues, with values of the Ostwald coefficient of 0.45 for fat and 0.07 for non-fat tissue [N1]. Other more elaborate models use weight-related coefficients, where the density of the absorbing tissue is taken into account [S2].

141. The total body retention of ^{85}Kr has been subjected to exponential analysis. Several clearance rates have been recognized. Recent work has suggested a model for krypton in the body consisting of five compartments [C6]. The fastest component probably represents the clearance from circulating blood, particularly blood plasma ($T_{1/2} = 21.5 \pm 5.7$ s). The second component (4.74 ± 2 min) appears to be representative of haemoglobin clearance. The next slower component (19.8 ± 6.6 min) is most likely related to clearance of krypton from muscle. The two components with the slowest clearance rates can be related to body fat compartments. A half-time of about 2.4 h is attributed to a fat compartment not located in adipose tissue. The retention half-time of krypton in adipose tissue is the slowest component and is correlated significantly with the total body fat content. The relationship is $T_{1/2}(\text{h}) = 0.17$ (percentage fat) + 0.75 [C6].

E. DOSIMETRY

142. Krypton-85 released to the environment causes a radiation dose to man through external irradiation from amounts in air and through internal irradiation from amounts within the body. Tissues are irradiated both from the activity in the organ itself and from the activity present in the surrounding organs.

1. Dose per unit exposure

143. The equilibrium absorbed dose rates to body organs per unit concentration of krypton-85 in air are summarized in Table IV.4 [Ni]. For comparison, the recently published values of the ICRP are also listed [I3]. The ICRP values represent minor adjustments, except for the lungs, for which the beta dose due to ^{85}Kr in the airways of the lungs has been disregarded.

144. The dose equivalent rates in various organs are listed in Table IV.5. These are the ICRP values [I3]. The quality factor for ^{85}Kr radiation is one. Therefore the dose equivalent rates are numerically equal to the absorbed dose values. When combined with the tissue weighting factors suggested by the ICRP to account for varying incidence of health effects, the effective dose equivalent rate is obtained, which for ^{85}Kr is $8 \cdot 10^{-9}$ Sv/a per Bq/m³.

Table IV.4
Equilibrium absorbed dose rate to body organs per unit
air concentration from immersion in a semi-infinite cloud
of Krypton-85 (10^{-9} Gy/a per Bq/m³) [N1]

Source	Organ						
	Skin	Adipose tissue	Lungs	Red bone marrow	Skeleton	Ovaries	Testes
<u>Krypton-85 in air</u>							
Photons in air	4.1	3.2	3.0	3.8	4.1	1.3	3.5
Betas in air	490.0	-	-	-	1.1	0.30	0.68
Bremsstrahlung in air	0.6	0.57	0.51	0.97	1.1	0.0005	0.0027
Bremsstrahlung in skin	0.015	0.0018	0.0011	0.0010	0.0030	-	-
<u>Krypton-85 in the body</u>							
Photons in the body	0.0006	0.0006	0.0006	0.0007	0.0006	0.0008	0.0008
Betas in the body	0.10	0.30	0.16	0.21	0.10	0.10	0.10
Bremsstrahlung in the body	0.0001	0.0002	0.0002	0.0003	0.0003	0.0002	0.0003
Betas in airways of lung	-	-	4.9	-	-	-	-
Total	490	4.1	8.5	5.0	5.3	1.7	4.3
Total [13]	410		3.8	5.0	5.4		4.6

Table IV.5

Dose equivalent rates from submersion
in semi-infinite cloud of ^{85}Kr
 (10^{-9} Sv/a per Bq/m^3) [13]

	Dose equivalent rate	Weighting factor	Effective dose equivalent rate
Ovards	4.6	0.25	1.1
Breast	3.9	0.15	0.6
Red bone marrow	5.0	0.12	0.6
Lungs	3.8	0.12	0.46
Bone surface	5.4	0.03	0.16
Spleen	4.0	0.06	0.24
Small intestinal wall	3.8	0.06	0.23
Kidneys	3.5	0.06	0.21
Adrenal glands	3.5	0.06	0.21
Liver	3.3	0.06	0.20
Skin	410.0	0.01	4.1
Total			8.1

2. Dose per unit release

145. The collective effective dose equivalent commitment per unit release of ^{85}Kr into the atmosphere, S_1^C , can be determined, based on the procedures previously used by UNSCEAR [U7]. In the local region of the release, the following formula can be applied

$$S_1^C = \left(\frac{\chi}{Q}\right)_{1 \text{ km}} \delta_N \phi \int_1^{100 \text{ km}} \left(\frac{r}{1 \text{ km}}\right)^{-1.5} 2\pi r \text{ dr}$$

A dispersion factor, χ/Q , of $5 \cdot 10^{-7}$ s/m³ is assumed, which agrees with the data of Table IV.2. The population density, δ_N is assumed to be 100 man/km² in the local region extending to 100 km distance. The dose factor, ϕ is $8 \cdot 10^{-9}$ Sv/a per Bq/m^3 as given above. The concentration of ^{85}Kr in air is assumed to decrease as a function of the distance, r , from the release point. The integral of the distance dependence over the local region times the dispersion factor, the population density, and the dose factor gives the

collective effective dose equivalent commitment per unit ^{85}Kr activity released. The result is

$$S_1^C (\text{local}) = 1 \cdot 10^{-16} \text{ man Sv/Bq}$$

146. The collective dose equivalent commitment per unit release of ^{85}Kr to the global population can be determined from the following formula [U7]

$$S_1^C = \int_0^{\infty} \overline{D(t)} N(t) dt$$

where $\overline{D(t)}$ is the per caput dose rate per unit activity released and $N(t)$ is the population size.

147. After uniform mixing in the global atmosphere, the per caput dose rate from ^{85}Kr is equal to the product of the concentration in air, C_0 , which decreases due to radioactive decay of ^{85}Kr , and the dose factor, ϕ . The population is assumed to increase at a rate, ν , of 2% per year. The collective effective dose equivalent commitment is, thus,

$$S_1^C = \int_0^{\infty} C_0 e^{-\lambda t} \phi N_0 e^{\nu t} dt = \frac{C_0 \phi N_0}{\lambda - \nu}$$

148. The population-weighted surface air concentrations of Table IV.3 can be used to estimate the global collective dose. An initial world population of $4 \cdot 10^9$ is assumed. The following contributions are obtained in the first two years following the release of ^{85}Kr to the atmosphere.

$$\begin{aligned} S_1^C (\text{1st year}) &= 4 \cdot 10^9 \text{ man } 19.5 \cdot 10^{-20} \frac{\text{Bq a/m}^3}{\text{Bq}} 8 \cdot 10^{-9} \frac{\text{Sv/a}}{\text{Bq/m}^3} \\ &= 6 \cdot 10^{-18} \text{ man Sv/Bq} \end{aligned}$$

$$\begin{aligned} S_1^C (\text{2nd year}) &= 4.08 \cdot 10^9 \text{ man } 27.6 \cdot 10^{-20} \frac{\text{Bq a/m}^3}{\text{Bq}} 8 \cdot 10^{-9} \frac{\text{Sv/a}}{\text{Bq/m}^3} \\ &= 9 \cdot 10^{-18} \text{ man Sv/Bq} \end{aligned}$$

Thereafter, the formula in the preceding paragraph can be applied.

$$\begin{aligned} S_1^C (> 2 \text{ years}) &= \frac{4.16 \cdot 10^9 \text{ man } 22 \cdot 10^{-20} \frac{\text{Bq a/m}^3}{\text{Bq}} 8 \cdot 10^{-9} \frac{\text{Sv/a}}{\text{Bq/m}^3}}{\frac{\ln 2}{10.73 \text{ a}} - 0.02 \text{ 1/a}} \\ &= 1.6 \cdot 10^{-16} \text{ man Sv/Bq} \end{aligned}$$

The total global collective dose equivalent commitment per unit ^{85}Kr activity released is

$$S_1^C(\text{global}) = 1.8 \cdot 10^{-16} \text{ man Sv/Bq}$$

149. A more approximate estimation, namely assuming instant mixing of the ^{85}Kr in the global atmosphere of $4 \cdot 10^{18} \text{ m}^3$, gives the same result. The initial concentration is then $25 \cdot 10^{-20} \text{ Bq/m}^3$. Using the formula in paragraph 147, with an initial population of $4 \cdot 10^9$, gives the value of $1.8 \cdot 10^{-16} \text{ man Sv/Bq}$ for the global collective dose equivalent commitment per unit ^{85}Kr activity released.

150. It is seen by comparison that the local contribution to the collective dose equivalent commitment per unit release of krypton-85 is negligible. Therefore, the total dose estimate is independent of the location of the release.

E. REFERENCES

- A1 Apsimon, H.M. and A.J.H. Goddard. Modelling the atmospheric dispersal of radioactive pollutants beyond the first few hours of travel. p. 124-135 in The seventh International Technical Meeting on Air pollution Modelling and its Application. Proceedings of a symposium. Airlie, Va., U.S.A., 1976.
- B1 Bernhardt, D.C., A.A. Moghissi and J.W. Cochran. Atmospheric concentrations of fission product noble gases. p. 97-118 in The Noble Gases (A.A. Moghissi and J.W. Cochran, eds.). U.S. Government Printing Office, Washington, 1975.
- B3 Bonka, H., R. Schulten, K. Brüssermann et al. Zukünftige radioaktive Umweltbelastung in der BRD durch radionuklide aus kerntechnischen Anlagen im Normalbetrieb. Berichte der Kernforschungsanlage Jülich, Jül-1220 (1975).
- B4 Bieri, R.H., M. Koide and E.D. Goldberg. The noble gas contents of Pacific seawaters. J. Geophys. Res. 71: 5243-5247 (1966).
- C2 Crouch, E.A.C. Fission product yields from neutron induced fission, atomic data and nuclear data tables, Vol. 19, No. 5. Academic Press Inc., New York, 1977.
- C5 Commission of the European Communities. Methodology for evaluation the radiological consequences of radioactive effluents released in normal operations. V/3865/79 - EN, FR Directorate of Health Protection (1979).
- C6 Cohn, S.H., K.J. Ellis and H. Susskind. Evaluation of the health hazard from inhaled krypton-85. International Symposium on biological implications of radionuclides released from nuclear industries. IAEA-SM-237/46 (1979).
- D1 Diethorn, W.S. and W.L. Stockho. The dose to man from atmospheric krypton-85. Health Phys. 23: 653-662(1972).
- D3 Depres, A. and J. LeGrand. Une méthode d'évaluation des transferts atmosphériques à longue distance. p. 237-249 in La dispersion en milieu physique naturel. Proceedings of a seminar. Cadarache, France, 1978.
- E1 Erdman, C.A. and A.B. Reynolds. Radionuclide behaviour during normal operations of liquid metal cooled fast

- breeder reactors, part 1: Production. Nucl. Saf. 16: 43-52 (1975).
- H2 Heller, D., W. Roedel, K.O. Münnich et al. Decreasing release of krypton-85 into the atmosphere. Naturwissenschaften 64(7): 383 (1977).
- I1 International Atomic Energy Agency. Power reactors in member states IAEA, Vienna, 1980.
- I2 Islitzer, N.F. and D.H. Slade. Diffusion and transport experiments in meteorology and atomic energy. U.S. Atomic Energy Commission report USAEC-TID-24190 (1968).
- I3 International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. International Commission on Radiological Protection report 30, supplement to part I. Pergamon Press, Oxford, 1979.
- K1 Kirk, W.P. Krypton-85: A review of the literature and an analysis of radiation hazards, USEPA report EPA-NP-19251 (1972).
- K3 Kelly, G.N., J.A. Jones, P.M. Bryant et al. The predicted radiation exposure of the population of the European community resulting from discharge of krypton-85, tritium, carbon-14 and iodine-129 from the nuclear power industry to the year 2000. Commission of the European Communities, Directorate of Health Protection. V/2676/75 (1975).
- M1 Martin, A. and M. Apsimon. The forecasting of radioactive wastes arising from nuclear fuel reprocessing. p. 55-60 in Management of Radioactive Wastes from Fuel Reprocessing. Proceedings of a symposium jointly organized by OECD/NEA and International Atomic Energy Agency. Paris, 1972.
- M2 Machta, L., G.J. Ferber and J.L. Hefter. Regional and global scale dispersion of krypton-85 for population dose calculations. p. 411-424 in Physical Behaviour of Radioactive Contaminants in the Atmosphere. International Atomic Energy Agency publication STI/PUB/355. Vienna, 1974.
- N1 National Council on Radiation Protection and Measurements. Krypton-85 in the atmosphere - accumulation, biological significance and control technology. NCRP No. 44 (1975).

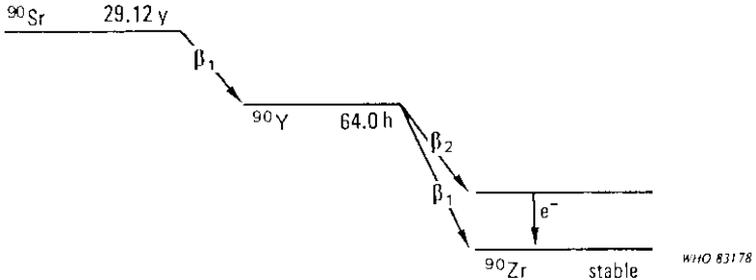
- R1 Rozanski, K. Krypton-85 in the atmosphere 1950-1977: A data review. *Envir. International* 2: 139-143 (1979).
- S1 Sutton, O.G. The theory of Eddy diffusion in the atmosphere. *Proc. R. Soc. (London), Ser. A* 135: 143-152 (1932).
- S2 Snyder, W.S., L.T. Dillman, M.R. Ford et al. Dosimetry for a man immersed in an infinite cloud of 85-K5, p. 119-122 in *The Noble Gases* (A.A. Moghissi and R.E. Stanley eds.). U.S. Government Printing Office, Washington, 1975.
- U1 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Thirteenth Session, Supplement No. 17 (A/3838). New York, 1958.
- U2 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Seventeenth Session, Supplement No. 16 (A/5216). New York, 1962.
- U3 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Nineteenth Session, Supplement No. 14 (A/5814). New York, 1964.
- U4 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-first Session, Supplement No. 14 (A/6314). New York, 1966.
- U5 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-fourth Session, Supplement No. 13 (A/7613). New York, 1969.
- U6 United Nations. Ionizing Radiation: Levels and Effects. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, with annexes. United Nations sales publication No. E.72.IX.17 and 18. New York, 1972.
- U7 United Nations. Sources and Effects of Ionizing Radiation. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General Assembly, with annexes. United Nations sales publication No. E.77.IX.1. New York, 1977.

V. STRONTIUM-90

A. INTRODUCTION

151. Strontium is element number 38 in the periodic table. It is an alkaline earth element and is therefore similar to calcium, barium and radium. It follows calcium through the food chains from environment to man, but some degree of discrimination exists against strontium. Both strontium and calcium are retained in the body largely in bone.

152. Since the early days of atmospheric nuclear testing the importance of ^{90}Sr as a contributor to the radiation exposure of man has been recognized. Strontium-90 is a radionuclide formed in the process of nuclear fission. It has a radioactive half-life of 29.1 years and decays by beta emission. Its daughter, ^{90}Y , is also radioactive with a half-life of 64.0 hours, and decays by beta emission to the stable isotope ^{90}Zr . The decay scheme for ^{90}Sr and ^{90}Y is given in Figure V.I. A summary of fission yields for ^{90}Sr is given in Table V.1.



Radiation type	Energy (keV)	Intensity (%)
^{90}Sr β_1	max. 546 avg. 196	100
^{90}Y β_1	max. 2284 avg. 935	99.984
β_2	max. 523	0.016

Figure V.I. Decay scheme informaton for ^{90}Sr and ^{90}Y . [N3]

Table V.1
Fission yields of strontium-90
[C3]

Nuclide	Fission yield (%)	
	Thermal	Fast
^{235}U	5.84	5.21
^{239}Pu	2.12	2.05
^{238}U		3.20
^{232}Th		7.66
Average for nuclear tests <u>a/</u>	3.50	

a/ From reference [H1].

153. Large amounts of ^{90}Sr were released in nuclear tests and dispersed throughout the world. Strontium-90 is also produced in the nuclear fuel cycle, but only small amounts are released to the environment. Strontium-90 in the environment is efficiently transferred to human diet. The absorption of ^{90}Sr by the body is relatively high and it has a long biological retention time.

154. Because of the correspondence in behaviour of strontium and calcium in the environment and in man, it has been the practice to express measurement results in diet and bone as quotients of ^{90}Sr to Ca concentrations. Discrimination is reflected as ratios of strontium to calcium quotients in samples to those in precursor samples in the transfer chain. Expressing results in terms of the strontium to calcium quotients has the practical advantage that for many environmental transfer processes, such as absorption into the body, secretion into milk and deposition in bone, the ratios remain relatively constant and predictable. However, since the average levels of calcium in diet and man are nearly constant, assessments of ^{90}Sr can also be made on the basis of amounts per unit mass or volume of material.

B. SOURCES

1. Nuclear explosions

155. Strontium-90 is produced in nuclear explosions in the amount of approximately $3.7 \cdot 10^{15}$ Bq per Mt of fission energy. Measurements of the fission debris from large nuclear tests gave a ^{90}Sr fission yield estimate of 3.5% [H1]. Assuming 1 kt of fission energy corresponds to $1.45 \cdot 10^{23}$

fissions [H1] and using the current best estimate of the ^{90}Sr half-life (29.12 ± 0.24 a) [N3], the ^{90}Sr production yield is estimated to be $3.8 \pm 0.1 \cdot 10^{15}$ Bq per Mt fission energy. Large deviations are possible for individual tests.

156. Approximate fission energy yields of nuclear weapons tests conducted in the atmosphere have been published. The total for tests through 1962 was 194 Mt of fission energy [F1, U3]. This would correspond to a production of about $74 \cdot 10^{16}$ Bq of ^{90}Sr . An additional $9 \cdot 10^{16}$ Bq have been produced in atmospheric tests to the end of 1980. A portion of the total amount of ^{90}Sr was local fallout, deposited in the immediate vicinity of testing regions. Local fallout is important especially for lower yield tests detonated on the land or water surface. The best estimates of the activity of globally distributed ^{90}Sr come from measurements of ^{90}Sr deposition. This amounts to $6 \cdot 10^{17}$ Bq for all tests conducted through 1980.

2. Nuclear fuel cycle

(a) Nuclear reactors

157. Strontium-90 is produced by fission in the fuel of nuclear reactors. The amounts produced vary depending on the fuel composition, reactor type, and degree of fuel burn-up achieved. The yields for various fission processes were given in Table V.1. In fairly high burn-up fuel ($33000 \text{ MW(t)d t}^{-1}$) of a PWR, the ^{90}Sr production is estimated to be $2.83 \cdot 10^{15}$ Bq per tonne of fuel, corresponding to $9.5 \cdot 10^{13}$ Bq per MW(e)a of electricity generated [01].

158. Small amounts of ^{90}Sr produced in the fuel in nuclear reactors may reach the coolant through defects in the fuel cladding. In coolant purification or following coolant leakage, ^{90}Sr may reach the gaseous and liquid effluent streams. In controlled amounts, some of the effluents are released to the environment.

159. The activity releases of ^{90}Sr were listed in the 1977 report of UNSCEAR [U6]. Average discharges were of the order of 0.01 to $4 \cdot 10^6$ Bq per MW(e)a in PWRs and BWRs, respectively, with most of the release in liquid effluents. Somewhat larger releases of ^{90}Sr in liquid effluents from GCRs arise primarily from spent fuel storage pools. There are considerable variations in the release amounts from individual reactors per unit electricity generated.

160. Assuming for each reactor type that the limited data of the ^{90}Sr activity released per unit of electrical energy

generated are representative, it is possible to obtain a very rough estimate of the total amount of ^{90}Sr released from reactors worldwide. Using the installed capacities of the various reactor types in 1980 [11] and assuming a reactor utilization of 60%, the estimated annual release from all reactors is about $2 \cdot 10^{12}$ Bq (Table V.2). In this calculation, it is assumed for the reactor types for which no data are available, that the releases are similar to those from BWRs.

Table V.2

Estimated global discharges of strontium-90
from nuclear power stations in 1980

Reactor type	Reactor number	Capacity [MW(e)a]	Production rate [Bq per MW(e)a]	Estimated strontium-90 discharge (Bq)
PWR	96	64239	$9 \cdot 10^4$	$0.3 \cdot 10^{10}$
BWR	62	35170	$4 \cdot 10^6$	$8 \cdot 10^{10}$
CCR	36	7086	$4 \cdot 10^6$	$170 \cdot 10^{10}$
Other	47	18490	$4 \cdot 10^6$	$4 \cdot 10^{10}$
Total	241	124985		$2 \cdot 10^{12}$

161. There have not been many reports on ^{90}Sr measurements in the environment surrounding nuclear reactors that can be attributed to reactor operation. In fact, ^{90}Sr would not be a likely radionuclide to be investigated, and furthermore results of measurements could hardly be made independent of fallout ^{90}Sr . The much more readily detectable ^{137}Cs , for example, is released in activity amounts up to several hundred times that of ^{90}Sr [U6]. Caesium-134 is released from reactors in amounts about 60% less than ^{137}Cs and, in addition, it would not be confused with weapons fallout in the environment. For these reasons, estimates of ^{90}Sr levels in the environment from reactor release must generally be based on measured releases and environmental dispersion calculations.

(b) Fuel reprocessing plants

162. In fuel reprocessing plants the fuel is dissolved to recover uranium and plutonium for reuse. All the ^{90}Sr and

other fission products as well go to waste streams. The radionuclide activities in airborne and liquid effluents from fuel reprocessing plants have been recorded by UNSCEAR [U6]. The data for ^{90}Sr are summarized in Table V.3.

Table V.3

Average normalized discharges of strontium-90
to the environment from fuel reprocessing plants
(Bq per MW(e)a) [U6]

Plant	Airborne effluents	Liquid effluents	Total
Windscale (United Kingdom)	$5 \cdot 10^6$	$2 \cdot 10^{11}$	$2 \cdot 10^{11}$
Nuclear Fuel Services (U.S.A.)	$2 \cdot 10^6$	$5 \cdot 10^6$	$5 \cdot 10^6$

163. Strontium-90 is released from fuel reprocessing plants primarily in liquid effluents. Relative to the total amounts of ^{90}Sr in spent fuel, the release amounts are not large. The fractional releases in liquid effluents are approximately $2 \cdot 10^{-3}$ from the Windscale plant in the United Kingdom and were about $5 \cdot 10^{-6}$ from the small Nuclear Fuel Services plant in the U.S.A., which is no longer in operation.

164. The fractional release of ^{90}Sr in airborne effluents is about $3 \cdot 10^{-6}$, compared to the amounts present in spent fuel. The experiences at Windscale and at Nuclear Fuel Services are similar in this regard.

165. The releases of radioactivity into the Irish Sea from the Windscale plant and the corresponding levels in the environment have been closely studied [N1]. Estimates of inventories of activity in the sea and sediments and of movements of the activity in its passage into the North Sea are being made [M1]. Some results of environmental surveys around the reprocessing plant at La Hague have also been published [S1]. Most of the attention, however, is focused on the greater quantities of other fission products released.

C. BEHAVIOUR IN THE ENVIRONMENT

1. Movement in soil

166. The downward penetration of ^{90}Sr in soil is slow, although it is more rapid than for ^{137}Cs or ^{239}Pu . Even after several years ^{90}Sr remains in the upper few centimetres in undisturbed soil. The rate of movement varies with soil type; a low content of clay and humus, a high content of electrolytes and a rapid movement of water increase penetration [U2]. The mechanism of movement is thought to involve both leaching and diffusion.

2. Transfer to plants

167. Plants acquire ^{90}Sr by direct deposition onto foliage and by root uptake of ^{90}Sr in the soil. Absorption into the leaves is relatively slow and superficial material is readily lost by weathering. The translocation of ^{90}Sr from plant leaf or grain surfaces to other parts of the plant is small.

168. Capture of ^{90}Sr on inflorescences of plants is of importance for entry to grain. Concentrations in husked grain will be higher than in the milled product during periods of deposition.

169. Uptake from soil is normally the primary mode of ^{90}Sr entry into plants. The quantity of absorbable calcium in soil is an important factor in determining the extent of ^{90}Sr absorption by plants. Uptake is greatest from soils of low calcium content. Uptake is thus reduced by the addition of lime, but usually not by a factor exceeding 3 [U2]. When soils contain adequate calcium for growth and the exchange capacity is largely saturated with calcium, the addition of lime has little or no effect.

170. Other factors which affect root uptake of ^{90}Sr include the clay and humus content of the soil, pH, the concentration of electrolytes other than calcium and the moisture content. The addition of organic matter and fertilizers to soil may have varying effects on plant uptake, which are, however, usually not large when these materials are applied at normal agricultural levels.

171. Plant-base absorption of ^{90}Sr has been noted to be quite efficient. The ^{90}Sr trapped in the surface root mat is relatively undiluted with the calcium in the soil and is in a particularly favourable position for absorption. This would help to explain the higher concentrations of ^{90}Sr in grain

in periods shortly after deposition and would be an important process in permanent pastures.

3. Transfer to milk

172. The total quantity of ingested ^{90}Sr secreted into the milk of cows is variable, depending on the milk yield. Values range from 0.5 to 2% of a single oral administration [U2]. With continuous ingestion under normal conditions of feeding, several investigations have shown that about 0.08% of the amount given daily is secreted per litre of milk. The transfer to goats milk may be more than ten times greater, corresponding to a higher proportion of dietary calcium secreted into the milk as well.

4. Transfer to diet

173. The transfer of fallout ^{90}Sr to diet has been extensively studied. Assessments by UNSCEAR have been based on application of generalized transfer models [U6]. The transfer from deposition to diet is quantitatively described by means of the transfer factor P_{23} defined as the time-integrated $^{90}\text{Sr}/\text{Ca}$ quotient in the diet divided by the ^{90}Sr integrated deposition density. The integrals may be replaced by summations if the relevant quantities are assessed over discrete intervals of time. In fact, annual values are the most generally available information. The transfer factor P_{23} is usually expressed in $\text{mBq a/gCa per Bq m}^{-2}$.

174. The transfer to diet from a specific deposition occurs over an extended period as long as ^{90}Sr remains in soil available for root uptake. The model used by UNSCEAR to describe the transfer of ^{90}Sr from deposition to diet is

$$C(n) = b_1 f(n) + b_2 f(n-1) + b_3 \sum_{m=1}^{\infty} f(n-m)e^{-\mu m}$$

where $C(n)$ is the $^{90}\text{Sr}/\text{Ca}$ quotient in total diet, in a food group, or in an individual food item, in the year n ; $f(n)$ is the annual deposition density in the year n , and b_1 , b_2 , b_3 , and μ are factors which can be derived from reported data by regression analysis [U5, U6]. The first term in the equation represents the contribution to dietary ^{90}Sr per unit deposition density in the current year, while the second term expresses separately the contribution from deposition in the previous year, reflecting also the use in the current year of stored food produced in the previous year. The third term expresses the contribution to dietary ^{90}Sr from the deposition density in all previous years, resulting from root uptake and taking into account decay and loss of availability due to downward movement in soil or to other physical or

chemical changes which may occur. The inverse of μ is the mean life of available ^{90}Sr in soil, which varies for individual foods and soil conditions.

175. The transfer factor P_{23} describes the cumulative transfer of ^{90}Sr to diet per unit deposition density. Using the model described above, the expression of the transfer factor is

$$P_{23} = b_1 + b_2 + b_3 \sum_{m=1}^{\infty} e^{-\mu m} = b_1 + b_2 + b_3 \frac{e^{-\mu}}{1-e^{-\mu}}$$

The transfer factor P_{23} can therefore be estimated from the parameters b_1 , b_2 , b_3 and μ obtained by regression analysis from reported data.

176. The values of the parameters obtained from the regression fits to ^{90}Sr deposition density and diet data from the fallout measurement programmes in New York and Denmark for dietary components and for the total diet have been reported by UNSCEAR [U6]. For total diet, the values of the parameters ($b_1 \approx 1.0$, $b_2 \approx 0.9$, $b_3 \approx 0.3$, $\mu \approx 0.1$) give estimates of the initial transfer ($b_1 + b_2$) of $1.9 \text{ mBq a (gCa)}^{-1}$ per Bq m^{-2} and of the long-term transfer (last term of equation above) of $2.9 \text{ mBq a (gCa)}^{-1}$ per Bq m^{-2} . The total transfer, the value of P_{23} , is $4.8 \text{ mBq a (gCa)}^{-1}$ per Bq m^{-2} .

177. Similar data from Argentina have also been evaluated, with reasonable agreement found for all three areas for individual foods and for total diet. Some differences are noted which are due to the different definition of the food groups or to different soil conditions and agricultural practices in the three countries.

178. The long-term transfer of ^{90}Sr to diet from a single input to the environment can be illustrated from the values of the transfer parameters obtained from component groups of diet. For example, it is determined that 90% of the cumulative transfer from a single release of ^{90}Sr is completed within 9 years for meat, fish and eggs, 12 years for grain products, 14 years for milk, 32 years for vegetables and 77 years for fruit. More rapid transfer indicates that direct deposition processes are more important. Slow transfer represents primarily uptake from the slowly decaying deposit of ^{90}Sr in soil. Since the exponential decreases of the transfer for the various groups differ, a direct fit to total diet data with a single exponential transfer function is expected to be less accurate than the summation of fits for the individual components.

Table V.4
Parameters of the transfer function between deposition density and $^{90}\text{Sr}/\text{Ca}$ in milk

Parameter	Northern hemisphere	San Francisco	New York	United Kingdom	Denmark	Argentina	Norway	Australia	Faroe Islands
b_1	0.84	0.61	0.69	0.89	0.99	1.39	0.70	2.07	2.70
b_2	0.54	0.61	0.23	0.47	0.46	1.24	0.66	1.27	1.38
b_3	0.22	0.19	0.19	0.15	0.73	0.12	1.02	0.30	0.81
μ	0.12	0.19	0.13	0.13	0.13	0.17	0.33	0.68	0.21
P_{23}	3.1	2.1	2.3	2.4	3.1	3.5	3.7	7.2	7.6

a/ The unit for parameters b_1, b_2, b_3 is $\text{mBq a}(\mu\text{Ca})^{-1}$ per Bq m^{-2}
 The unit for parameter μ is a^{-1}
 The unit for the transfer factor P_{23} is $\text{mBq a}(\mu\text{Ca})^{-1}$ per Bq m^{-2} .

179. The transfer of ^{90}Sr from deposition density to some individual foods, particularly to milk, has been studied for a number of different areas of the world. These results are listed in Table V.4. The transfer factors range from 2.1 to 7.6 mBq a/gCa per Bq m^{-2} .

180. Milk has been used as an indicator of the levels of ^{90}Sr in total diet in areas where foods other than milk were not analysed. This must be done, however, with some caution. It is usually the case that following a period of ^{90}Sr deposition, the $^{90}\text{Sr}/\text{Ca}$ quotient in milk declines somewhat more rapidly than the $^{90}\text{Sr}/\text{Ca}$ quotient in total diet. Where data are available, it is seen that the exponential factor is greater in milk than in total diet. This means that the diet-milk ratio of $^{90}\text{Sr}/\text{Ca}$ values changes with time during and after the period in which the ^{90}Sr deposition occurs.

181. The diet-milk ratios at particular times also show considerable variation from one country to another [U6]. In most countries where milk is an important component of the diet, the diet-milk ratio has averaged about 1.4 for most of the fallout years. A trend to increasing ratios in the future is expected if the deposition density rate remains at a very low level. In countries where milk is not an important component of diet, the diet-milk ratio will have a much higher value.

5. Aquatic behaviour

182. Strontium, like calcium, appears mainly in ionic form in water and is not strongly sorbed by suspended particulate materials. The fraction of strontium found in the particulate phase in several freshwater systems ranged from 1 to 10% [V1].

183. It has been of interest to determine the behaviour of ^{90}Sr in aquatic environments in the vicinity of nuclear installations. A number of determinations of the concentration factors (ratios of integrated concentrations or of equilibrium concentrations in organisms and in the water) have been performed in recent years for various marine biota by measurements of stable and radioactive strontium under laboratory and field conditions [C1, F2, N2, U4, U1]. Typical values are 100 for algae, 2 to 10 for crabs and lobster, about 1 for the flesh of ocean fish and 5 for fresh water fish.

184. The primary uptake of strontium and also calcium by fish occurs directly from the water. Therefore, accumulations in organisms are little dependent on trophic level [V1]. The concentration factor for fish depends inversely on the concentration in the water. Vanderploeg et al. [V1] have

suggested a quantitative relationship. The concentration factors for fish bone are about two orders of magnitude greater than for flesh.

185. The UNSCEAR aquatic model can be used to estimate the transfer of ^{90}Sr from the aquatic environment to diet for generalized discharge situations [U6]. Further discussion of this is presented in the section on dosimetry.

D. TRANSFER TO MAN

186. Strontium-90 is acquired by man primarily through ingestion of ^{90}Sr contaminated food. Terrestrial pathways are generally more important than aquatic pathways in transferring ^{90}Sr to man. From fallout experience it is noted that ^{90}Sr in drinking water always contributes less than 5% of the total ingestion intake and ^{90}Sr in fish is a minor contributor even in countries where consumption of fish is high. For example, it is estimated that only about 3% of the fallout ^{90}Sr intake by man in Japan between 1966 and 1971 came from fish [U1].

187. Correlations of fallout ^{90}Sr in diet with measurements of ^{90}Sr in bone have provided a relationship which is used to evaluate the transfer to man. The transfer function used by UNSCEAR [U5, U6] is:

$$C_b(n) = c C_d(n) + g \sum_{m=0}^{\infty} C_d(n-m) e^{-\mu m}$$

where $C_b(n)$ is the $^{90}\text{Sr}/\text{Ca}$ quotient in bone in the year n , $C_d(n)$ is the dietary $^{90}\text{Sr}/\text{Ca}$ quotient in the year n , and c , g , and μ are constants determined from regression analysis of the bone and diet data. The parameter c can be associated with that portion of ^{90}Sr intake which is retained for a short term on bone surfaces and is readily exchanged with plasma, and the parameter g with that portion more tightly retained in bone. The exponential term describes the effective removal rate of ^{90}Sr from bone due to radioactive decay and bone remodelling.

188. The transfer factor P_{34} linking diet and bone, is defined as the ratio of the time integrated $^{90}\text{Sr}/\text{Ca}$ quotient in bone to that in diet. It may also be thought of as the cumulative transfer and retention ^{90}Sr in bone per unit intake in diet. The expression for the transfer factor using

the above function is

$$P_{34} = c + g \sum_{m=0}^{\infty} e^{-\mu m} = c + \frac{g}{1 - e^{-\mu}}$$

Data are available for regression analysis applying this model only for ^{90}Sr in vertebrae. It is recognized that there is greater initial retention in cancellous bone such as vertebrae than in compact bone, but there is also more rapid turnover. Therefore, the time integrated results are expected to be representative of the skeleton as a whole.

189. The results of regression fits of fallout ^{90}Sr diet and adult vertebrae data for various localities have been given in UNSCEAR reports [U5, U6]. Representative values of the parameters are $0.02 \text{ Bq a (gCa)}^{-1}$ in bone per Bq a (gCa)^{-1} in diet for both c and g and about 0.2 a^{-1} for μ . The values determined for P_{34} range from 0.12 to $0.16 \text{ Bq a (gCa)}^{-1}$ in bone per Bq a (gCa)^{-1} in diet.

190. From the values of the exponential parameter μ it is possible to infer the mean residence time of ^{90}Sr in bone. This ranges from 3.4 to 6.7 years, corresponding to bone turnover rates of 12 to 23% per year. There is no reason to expect that the metabolic behaviour should differ in the various areas of the world, and as far as is known, dietary composition of usual foods does not affect ^{90}Sr availability. Therefore, the differences in the estimate are attributed to variations in the survey measurements.

191. To account for the bone $^{90}\text{Sr}/\text{Ca}$ quotients in children, Bennett [B2] has used age-dependent parameters in the above transfer function. For children under 9 years the parameter c was found to be zero, consistent with a single exponential transfer model as originally proposed by Rivera [R1]. The best fit was obtained with a turnover rate of ^{90}Sr varying from about 100% per year down to about 40% per year in the pre-teenage years and then falling with age to about 20% per year for adults. Beninson [B1] had reported a similar variation in turnover rate with age in Argentina. The fractional retention of strontium, the fraction of dietary intake incorporated into the skeleton, was also found by Bennett [B2] to vary with age being five to seven times higher for infants than for adults. Additional considerations of ^{90}Sr metabolism as a function of age have been presented by Papworth and Vennart [P1].

192. The initial ^{90}Sr concentration in the newborn must be determined from an empirical relationship with the mother's diet. The $^{90}\text{Sr}/\text{Ca}$ quotient in bone of newborn varies from

0.1 to 0.2 times the $^{90}\text{Sr}/\text{Ca}$ quotient in diet of the mother during the year prior to the birth. An average of about 0.15 is obtained from the survey data [B1, B2].

E. DOSIMETRY

1. Dose per unit intake

193. It is useful for radiological assessments to have expressions for the dose per unit ingestion intake and per unit inhaled amount. The absorbed doses in bone marrow and in bone-lining cells per unit integrated activity of ^{90}Sr in bone have been evaluated based on the work of Spiers [S2]. These are the transfer factors P_{45} relating activity in bone to the doses [U5, U6]. The values are P_{45} (bone marrow) = 0.38 mGy per Bq a (gCa) $^{-1}$; P_{45} (bone-lining cells) = 0.53 mGy per Bq a (gCa) $^{-1}$. If it may be assumed that the dietary calcium intake rate is 1 g daily (365 g/a), the transfer factor to bone per unit intake, P_{34} , is 0.14/365 Bq a (gCa) $^{-1}$ per Bq. This multiplied by the factor P_{45} gives the absorbed doses in bone marrow and bone-lining cells. The results are given in Table V.5.

Table V.5

Absorbed dose per unit intake of strontium-90 (Gy/Bq)

	Lung	Bone marrow	Bone-lining cells
Ingestion	-	$1.5 \cdot 10^{-7}$	$2.0 \cdot 10^{-7}$
Inhalation	$5.8 \cdot 10^{-9}$	$4.9 \cdot 10^{-7}$	$6.9 \cdot 10^{-7}$

194. The ICRP Task Group lung model gives guidance regarding the disposition of inhaled radioactivity [I3, I4] in the respiratory tract. The respiratory system is divided into the nasopharyngeal region (NP), the tracheobronchial region (TB), and the pulmonary region (P). For the present dosimetric assessments it will be assumed that the ^{90}Sr is associated with typical ambient aerosols of average diameter 0.5 μm . The ^{90}Sr compounds are grouped in Class D, retention in the lung being in the order of days, specifically 0.5 d for the portion deposited in the pulmonary region.

195. Fractional deposition of 0.5 μm particles in the NP, TB, and P regions are 0.14, 0.08, and 0.30 and subsequent fractional transfers to blood for the Class D compounds are 0.5, 0.95, and 1.0, respectively. Total transfer to blood is thus 0.446 of the inhaled amount. Fractional transfer from blood to bone is 0.3 [12].

196. The lung dose as a function of the inhaled activity can be calculated from the expression

$$D_{\text{lung}} = \frac{k A \bar{E} f 1.44 T_B}{M}$$

where k is a dosimetric constant, A is the activity inhaled, \bar{E} is the average energy per disintegration, f is the fraction of the activity retained in the lung, T_B is the retention half-time in the lung, and M is the mass of the lungs. The lung dose per unit activity of ^{90}Sr inhaled is therefore:

$$\begin{aligned} D_{\text{lung}} &= 13.8 \cdot 10^{-6} \frac{\text{Gy/d}}{\frac{\text{Bq MeV}}{\text{g dis}}} \cdot 1.13 \frac{\text{MeV}}{\text{dis}} \cdot 0.52 \frac{1.44 \cdot 0.5\text{d}}{1000 \text{ g}} \\ &= 5.8 \cdot 10^{-9} \text{ Gy/Bq} \end{aligned}$$

197. Assuming that the mean residence time of ^{90}Sr in bone is 10 years, applicable to the skeleton as a whole, the integrated concentration of ^{90}Sr in bone per unit inhaled amount is $0.446 \times 0.3 \times 10 \text{ years} \div 100 \text{ gCa} = 1.3 \cdot 10^{-3} \text{ Bq a/gCa}$ per Bq inhaled. The absorbed doses in bone marrow and bone lining cells are obtained by multiplying the values of the transfer factor P_{45} given above. The results are listed in Table V.5.

2. Dose per unit release

(a) Nuclear explosions

198. The dose commitment from ^{90}Sr released by nuclear explosions can be assessed using the environmental compartment model outlined in the introduction. The transfer factor of a sequence of steps in series is the product of the transfer factors of each step. The dose commitment, D^C , is related to the integrated deposition density of ^{90}Sr , F , by the following expression:

$$D^C = P_{23} P_{34} P_{45} F$$

where P_{23} , P_{34} and P_{45} are the transfer factors discussed previously. Average values for these transfer

factors, as assessed in the 1977 UNSCEAR report [U6], are $P_{23} = 5 \text{ mBq a/gCa per Bq m}^{-2}$, $P_{34} = 0.14$, and P_{45} as given previously.

199. The dose commitment from ^{90}Sr ingestion per unit of widespread deposition density of ^{90}Sr such as from nuclear explosions is thus $0.3 \text{ }\mu\text{Gy per Bq m}^{-2}$ for bone marrow and $0.4 \text{ }\mu\text{Gy per Bq m}^{-2}$ for bone lining cells.

200. The dose commitment from ^{90}Sr via the inhalation pathway can be estimated from the time integrated concentration of ^{90}Sr in air. Multiplying the dose commitment per unit inhalation intake (Table V.5) by the inhalation intake rate of $22 \text{ m}^3 \text{ d}^{-1}$ gives the value of $1.3 \text{ } 10^{-7} \text{ Gy per Bq d m}^{-3}$ for the dose commitment to lungs per unit integrated concentration of ^{90}Sr in air.

201. The dose commitment to lungs can also be referred to measured values of the integrated deposition density. Dividing the integrated deposition density (Bq m^{-2}) by the average deposition velocity (m s^{-1}) gives the time integrated concentration in air. From long-term measurements of fallout ^{90}Sr , the average deposition velocity is 2.2 cm/s in New York, 2.0 cm/s in Argentina, and 1.5 cm/s in Denmark [A1, C2, E1]. The values determined from annual measurements at all three sites range from 1.2 to 2.9 cm/s . The value of 2 cm s^{-1} can be taken as representative. One Bq m^{-2} integrated deposition density thus corresponds to $5.8 \text{ } 10^{-4} \text{ Bq d m}^{-3}$ in air. The dose commitment to lung per unit integrated deposition density of ^{90}Sr is thus $7.5 \text{ } 10^{-11} \text{ Gy per Bq m}^{-2}$.

202. The total amount of ^{90}Sr released to the environment by nuclear tests, $6 \text{ } 10^{17} \text{ Bq}$, has given a population-weighted integrated deposition density of 1940 Bq m^{-2} in the world as a whole [U6]. The world population is $4 \text{ } 10^9$. With these values the collective dose commitments per unit activity of ^{90}Sr released may be estimated. The results, which are summarized later, apply to the geographic pattern of past nuclear tests.

(b) Nuclear installations

203. The activity of ^{90}Sr in airborne effluents is dispersed by turbulent air movement and is eventually deposited on the ground. It then enters the ingestion pathway. The average situation over several years of routine discharges will result in a nearly completed deposition within

a region with a radius R. The average integrated deposition density, for a discharged activity A is $F = A/\pi R^2$. The number of individuals exposed to that mean integrated deposition density, on the other hand, is $N = \delta_N \pi R^2$, provided the population density δ_N can be assumed to be constant over the relevant area.

204. It follows that the collective dose commitment per unit activity released, S_1^c , can be assessed by the expression

$$S_1^c = P_{23} P_{34} P_{45} \delta_N$$

Using the values for the transfer factors given previously, and assuming a population density of 25 man km⁻², the collective dose commitments per unit activity released are estimated to be about 7 10⁻¹² man Gy per Bq for bone marrow and about 9 10⁻¹² man Gy per Bq for bone-lining cells. These values apply provided that food is locally produced and that the production suffices for the population density under consideration. The estimates would probably be lower in actual circumstances.

205. The contribution of inhalation to the collective dose commitment, for effluent releases over many years, can be estimated by integration of the functions describing the atmospheric dispersion, assuming complete depletion by deposition within 100 km. The collective dose commitment contribution per unit activity released would be given by the expression

$$S_1^c = \left(\frac{X}{Q}\right)_{1 \text{ km}} I \delta_N \varphi \int_{1 \text{ km}}^{100 \text{ km}} \left(\frac{r}{1 \text{ km}}\right)^{-1.5} 2\pi r dr$$

where $\left(\frac{X}{Q}\right)_{1 \text{ km}}$ is the dispersion factor at 1 km from the release point, I is the individual intake rate of air, δ_N is the population density, φ is the dose per unit activity inhaled of ⁹⁰Sr and r is the distance from the release point [U6].

206. The collective dose commitment may also be estimated using the deposition velocity, thus avoiding the need to specify the deposition area. That is, the integrated deposition density from a discharged activity A is $A/\pi R^2$. Dividing by the deposition velocity gives the integrated concentration of ⁹⁰Sr in air. Multiplying by the population, $\delta_N \pi R^2$, removes the areal dependence.

Using a population density of 25 man km⁻², an air intake rate of 22 m³ d⁻¹, a deposition velocity of 0.5 cm s⁻¹ appropriate for particulates from near surface releases, and the dosimetric factors given in Table V.5, estimates of the collective r dose commitments per unit release are obtained. The results are listed in the summary Table V.6.

207. The collective dose commitment from the input of ⁹⁰Sr in water bodies, normalized per unit activity released, can be estimated [U5] using the expression

$$S_1^c = \frac{\sum_k N_k I_k f_k \varphi}{V(\lambda + 1/\tau)}$$

Here V is the volume of receiving waters, τ is the turnover time of receiving waters, λ is the decay constant of ⁹⁰Sr, N_k is the number of individuals exposed by pathway k, I_k is the individual consumption rate of pathway item k, f_k is the concentration factor for the consumed item in pathway k, and φ is the collective dose per unit activity ingested collectively by the exposed group.

208. The quantity $\frac{1}{V(\lambda + 1/\tau)}$ is the infinite-time integral of the water concentration per unit of activity released, while the quantity multiplied by f_k is the infinite-time integral of the concentration in the consumed item k. For radionuclide inputs into small volumes of water, the concentration in water and in fish will be high, but the population which can be served with drinking water or by fish consumption will be limited. For inputs into larger volume of water the concentrations will be smaller, but the population involved will be correspondingly larger. It seems reasonable to assume, as a first approximation, that the quantities N_k/V are relatively constant and independent of V.

209. A summary of the values used in the assessments presented by UNSCEAR [U6] is given in the following listing:

<u>Parameter</u>	<u>fresh water</u>	<u>sea water</u>
1. τ, turnover time of receiving water	10 a	1 a
2. Correction factor for sediment removal	0.78	1.0
3. $\frac{V}{N}$, water utilization factor	3 10 ⁷ l/man	3 10 ⁹ l/man

<u>Parameter</u>	<u>fresh water</u>	<u>sea water</u>
4. f_k , concentration factor for item k		
drinking water	0.5	
fish	5	1
shellfish		1
5. I_k , consumption rate for item k		
drinking water	440 l/a	
fish	1 kg/a	6 kg/a
shellfish		1 kg/a

Using these values and the dosimetric factors given in Table V.5, the collective dose commitments per unit activity released in liquid effluents are estimated to be about $9 \cdot 10^{-12}$ man Gy/Bq for bone marrow and $1 \cdot 10^{-11}$ man Gy/Bq for bone lining cells for discharges into fresh waters, and a rounded value of $4 \cdot 10^{-16}$ man Gy/Bq for both tissues for discharges into the sea. The values for sea discharges underestimate the collective dose commitment because they neglect the contribution from large-scale mixing with a longer residence time. However, due to the relatively short life of ^{90}Sr , this contribution can only have a small effect on the estimates.

3. Summary

210. Table V.6 summarizes the values obtained above for the collective dose commitments, normalized per unit activity released, for releases by atmospheric nuclear explosions and in effluents of the nuclear power industry. In both cases they are the result of generalized assessments and substantial variations should be expected in site-specific cases.

211. For ^{90}Sr and ^{90}Y radiations, the quality factor is one, and the weighting factors are 0.12 for lung and for bone marrow and 0.03 for bone lining cells. The effective dose equivalent commitment per unit intake of ^{90}Sr is thus $2.4 \cdot 10^{-8}$ Sv/Bq (ingestion) and $8.0 \cdot 10^{-6}$ Sv/Bq (inhalation). The collective effective dose equivalent commitments per unit release from nuclear installations and from nuclear tests are included in Table V.6.

Table V.6

Summary of collective dose commitments per unit
strontium-90 activity released
(10^{-14} man Gy per Bq)

	Lung	Bone marrow	Bone lining cells	Effective <u>a/</u>
<u>Nuclear explosions</u>				
Ingestion		400	500	60
Inhalation	0.1	8	10	1
<u>Nuclear installations</u>				
<u>Release to air b/</u>				
Ingestion		700	900	100
Inhalation	0.7	60	90	10
<u>Release to fresh water</u>				
Drinking water		900	1000	100
Fish		20	30	3
<u>Release to salt water</u>				
Fish		0.03	0.04	0.005
Shellfish		0.005	0.007	0.0008

a/ Collective effective dose equivalent commitment (10^{-14} man Sv/Bq).

b/ Assumes population density of 25 man/km² which is fully sustained by local food production.

F. REFERENCES

- A. Aarkrog, A. and J. Lippert. Environmental radioactivity in Denmark in 1971, 1972, 1973, 1974 and 1975. Danish Atomic Energy Commission. Risø reports 265 (1972), 291 (1973), 305 (1974), 323 (1975) and 363 (1976).
- B1 Beninson, D., A. Migliori de Beninson, C. Menossi et al. Radioestroncio en el hombre en función de la edad. Trabajo presentado en el Quinto Congreso Internacional de la "Société française de radioprotection". Grenoble, 1971.
- B2 Bennett, B.G. Strontium-90 in human bone - 1976 results for New York and San Francisco. p. I-69-I-84 in Health and Safety Laboratory environmental quarterly report HASL-328. New York, 1977
- C1 Cancio, D., J.A. Llauró, N.R. Ciallella et al. Incorporación de radioescio por organismos marinos. p. 347-356 in Radioactive Contamination of the Marine Environment. Proceedings of a symposium, Seattle, 1972. IAEA publication STI/PUB/313. Vienna, 1973.
- C2 Comisión Nacional de Energía Atómica. Argentina. Information submitted 1977.
- C3 Crouch, E.A.C. Fission product yields from neutron induced fission. Atomic Data and Nuclear Data Tables, Vol. 19, No. 5. Academic Press, New York, 1977.
- E1 Environmental Measurements Laboratory, U.S. Department of Energy. Environmental quarterly and appendix. Environmental Measurements Laboratory report EML-334. New York, 1978.
- F1 Federal Radiation Council. Estimates and evaluation of fallout in the United States from nuclear weapons testing conducted through 1962 - Report No. 4 (1963).
- F2 Foyn, L. Some marine radioecological problems at the nuclear power station establishment at Oslofjorden. Fisheries Directorate Sea Research Institute report series B, No. 10. Bergen, Norway, 1973.
- H1 Hallden, N.A., I.M. Fisenne, D.Y. Ong et al. Radioactive decay of weapons debris p. 194-199 in Health and Safety Laboratory fallout program quarterly summary report HASL-117. New York, 1961.

- I1 International Atomic Energy Agency. Power reactors in member states. IAEA, Vienna, 1980.12
- I2 International Radiological Protection. Report of Committee II on Permissible Dose for Internal Radiation. ICRP publication 2, Pergamon Press, 1959.
- I3 International Commission on Radiological Protection. Task group on lung dynamics. Deposition and retention models for internal dosimetry of the human respiratory tract. Health Phys. 12: 173-226 (1966).
- I4 International Commission on Radiological Protection. Task group of Committee 2. The metabolism of compounds of plutonium and other actinides. ICRP publication 19, Pergamon Press, 1972.
- M1 Mitchell, N.T. Radioactivity in surface and coastal waters of the British Isles, 1972-1973. U.K. Ministry of Agriculture Fisheries and Food. Fisheries Radiobiological Laboratory report FRL-7, FRL-8, FRL-9, FRL-10 (1971, 1973, 1975).
- N1 National Radiological Protection Board. The data submitted by the United Kingdom to the United Nations Scientific Committee on the Effects of Atomic Radiation for the 1977 Report to the General Assembly. NRPB report R47, Harwell (1976).
- N2 Norwegian Institute for Water Research. Release of radioactive materials from nuclear power stations. Report No. 2. Dispersal mechanisms, pathways and concentration factors for radionuclides in the cooling waters. Report 0-177/70 (1974).
- N3 National Council on Radiation Protection and Measurements. A handbook of radioactivity measurements procedures. NCRP report No. 58, Washington D.C. (1978).
- O1 Oak Ridge National Laboratory. Siting of fuel reprocessing plants and waste management facilities. Oak Ridge National Laboratory report ORNL-4451 (1970).
- P1 Papworth, D.G. and J. Vennart. Retention of 90-Sr in human bone at different ages and the resulting radiation doses. Phys. Med. Biol. 18: 169-186 (1973).
- R1 Rivera, J. and J.H. Harley. The HASL bone program 1961-1964. U.S. Atomic Energy Commission report HASL-163. New York, 1965.

- S1 Scheidhauer, J., R. Ausset, J. Planet et al. Programme de surveillance de l'environnement marin du centre de La Hague. p. 347-365 in Population Dose Evaluation and Standards for Man and His Environment. IAEA publication STI/PUB/375. Vienna, 1974.
- S2 Spiers, F.W., G.D. Zanelli, P.J. Darley et al. Beta-particle dose rates in human and animal bone. p. 130-148 in Biomedical Implications of Radiostrontium Exposure. U.S. Atomic Energy Commission Symposium Series 25 (1972).
- U1 Ueda, T., Y. Suzuki and R. Nakamura. Transfer of caesium-137 and strontium-90 from the environment to the Japanese population via the marine environment in Population Dose Evaluation and Standards for Man and His Environment. IAEA publication STI/PUB/375. Vienna, 1974.
- U2 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Seventeenth Session, Supplement No. 16 (A/5216). New York, 1962.
- U3 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Nineteenth Session, Supplement No. 14 (A/5814). New York, 1964.
- U4 United Nations. Report of the United Nations Scientific Committee on the effects of Atomic Radiation. Official Records of the General Assembly, Twenty-First Session, Supplement No. 14 (A/6314). New York, 1966.
- U5 United Nations. Ionizing Radiation: Levels and Effects. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, with annexes. United Nations sales publication, No. E.72.IX.17 and 18. New York, 1977.
- U6 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General Assembly, with annexes. United Nations sales publication No. E.77.IX.I. New York, 1977.
- V1 Vanderploeg, H.W., D.C. Porzyck, W.H. Wilcox et al. Bioaccumulation factors for radionuclides in freshwater biota. Oak Ridge National Laboratory report ORNL-5002 (1975).

VI. IODINE

A. INTRODUCTION

212. Iodine is a volatile element which is very mobile in the environment. It is non-uniformly distributed in nature, its abundance in the lithosphere being about 5 times higher than in the ocean waters. The iodine in the sea apparently originates from erosion of the land masses. Recycling to the terrestrial biosphere occurs through evaporation of sea water and decomposition of substances of marine origin.

213. There are at least 25 iodine isotopes with mass numbers ranging from 117 to 141. All except ^{127}I are radioactive. Omitting the very short-lived ^{140}I and ^{141}I , thirteen isotopes are produced by fission:

^{127}I (stable), ^{128}I (25 min), ^{129}I ($1.57 \cdot 10^7$ a), ^{130}I (12.4 h), ^{131}I (8.06 d), ^{132}I (2.3 h), ^{133}I (21 h), ^{134}I (52.8 min), ^{135}I (6.7 h), ^{136}I (83 s), ^{137}I (23 s), ^{138}I (5.9 s) and ^{139}I (2 s). From the point of view of environmental contamination and resulting doses to man, the most important isotopes of iodine are ^{131}I and ^{129}I . They are the only radioactive isotopes of iodine produced by fission with half-lives longer than one day. Iodine-131 is a beta-emitter with a half-life of 8.06 days and a maximum beta energy of 0.81 MeV emitting also gamma rays of 0.36 and 0.64 MeV and other energies. Iodine-129 has a very long half-life ($1.57 \cdot 10^7$ a); it is a beta-emitter (maximum energy: 0.15 MeV) with an accompanying gamma ray of 0.09 MeV in 8% of the disintegrations [D1]. The two isotopes are mainly found in the environment as a result of nuclear explosions and releases from nuclear reactors and fuel reprocessing plants. Only these two isotopes of iodine are considered in this report.

214. Iodine enters the metabolism of living organisms and is selectively taken up and concentrated in the thyroid gland; it plays a major role in the synthesis of the thyroid hormone and is secreted in milk. Owing to its decay properties, ^{131}I has been extensively used in the medical field for diagnosis and treatment of thyroid abnormalities.

B. SOURCES

1. Natural production

215. Like any other fission product, ^{129}I and ^{131}I are present in the environment as a result of spontaneous fission of natural uranium. In view of its very long half-life,

225. In liquid effluents from LWRs, ^{131}I is found in amounts comparable to the airborne releases from BWRs (10^8 Bq per MW(e)a) [U1]. It is not reported in the liquid effluents from other types of reactors.

226. In 1980, the installed nuclear capacity was $1.25 \cdot 10^5$ MW(e) on a worldwide scale [I2]. Assuming an average load factor of 0.6, the energy produced was $7.5 \cdot 10^4$ MW(e)a. The global production and release of ^{131}I at the reactor sites in 1980 are estimated to be about $7 \cdot 10^{21}$ Bq and $2 \cdot 10^{13}$ Bq, respectively, using the figures given previously for production and release, and assuming that the releases are similar to those from BWRs for the reactor types for which no data are available. The estimate of the global release is very crude, but nevertheless shows that only a tiny fraction of the order of 10^{-9} of the ^{131}I produced in the reactors is discharged into the environment. Table VI.1 provides a break-down of the releases from reactors according to reactor type.

227. Being a volatile element, iodine is readily released to the atmosphere in the case of a reactor accident. The two reported reactor accidents which have caused measurable irradiation of the public occurred at Windscale (U.K.) in October 1957 and at Three Mile Island in March 1979. The release of ^{131}I to the atmosphere was about $7 \cdot 10^{14}$ Bq in the Windscale accident [L1] and $6 \cdot 10^{11}$ Bq in the Three Mile Island accident [H3].

228. The activity of ^{129}I produced in a nuclear reactor is much lower than that of ^{131}I . The production of ^{129}I per unit energy generated is approximately $5 \cdot 10^7$ Bq per MW(e)a [U1] corresponding to an inventory of $1.5 \cdot 10^8$ Bq per MW(e) after three years of fuel irradiation. Iodine-129 has not been identified in routine discharges of nuclear reactors. A rough estimate of the activity of ^{129}I discharged per unit energy generated can be calculated assuming that the ratio of the release rate to the inventory in reactor fuel is the same for ^{129}I and ^{131}I . As the activity of ^{129}I in fuel is at least $2 \cdot 10^7$ times lower than that of ^{131}I , the release rate of ^{129}I per unit energy generated is at most 0.1 Bq per MW(e)a; activity of ^{129}I discharged from nuclear reactors in 1980 would thus be of the order of 10^4 Bq.

Table VI.1
 Estimated global discharges of ¹³⁷I from nuclear power stations in 1980

Reactor Type	Number	Capacity (MW(e)a)	Estimated release rates (Bq per MW(e)a)		Estimated discharges in 1980 (Bq)		
			Airborne	Liquid	Airborne	Liquid	Total
FRS	96	64239	10 ⁷	10 ⁸	5 10 ¹¹	6 10 ¹²	7 10 ¹²
BR	62	35170	10 ⁸	10 ⁸	4 10 ¹²	4 10 ¹²	8 10 ¹²
RR	14	5963	10 ⁷	10 ⁸	6 10 ¹⁰	5 10 ¹¹	7 10 ¹¹
GR	36	7086	10 ⁷	10 ⁸	7 10 ¹⁰	7 10 ¹¹	8 10 ¹¹
Other	33	12527	10 ⁸	10 ⁸	1 10 ¹²	1 10 ¹²	2 10 ¹²
Total	261	124985	-	-	6 10 ¹²	1 10 ¹³	2 10 ¹³

^{129}I has accumulated in the earth's crust and also in the ocean waters from where it is available to disperse in the whole biosphere.

216. In 1962, Edwards [E1] predicted the natural $^{129}\text{I}/^{127}\text{I}$ atom ratio in sea water to be about $3 \cdot 10^{-14}$ from spontaneous fission of uranium-238 and estimated that contributions of the same order of magnitude would arise from spontaneous fission of ^{235}U and from production in the upper atmosphere by interaction of energetic protons, neutrons and photons on isotopes of xenon. Experiments later confirmed the validity of Edwards' estimate, the $^{129}\text{I}/^{127}\text{I}$ atom ratios derived from measurements in iodine-rich minerals ranging from $2 \cdot 10^{-15}$ to 10^{-13} [M1, S1]. Taking the concentrations of ^{127}I in sea water to be $0.064 \mu\text{g g}^{-1}$ and the global volume of sea water to be $6 \cdot 10^{17} \text{ m}^3$, a natural activity of ^{129}I of $7 \cdot 10^9 \text{ Bq}$ in the oceans is obtained from an $^{129}\text{I}/^{127}\text{I}$ atom ratio of $3 \cdot 10^{-14}$.

2. Nuclear explosions

217. The activity of ^{131}I (or of ^{129}I) generated in nuclear explosions can be derived from the measurements of ^{90}Sr deposition [U1] and from the ratios of the fission yields of ^{131}I (or ^{129}I) and of ^{90}Sr for nuclear weapons tests [H2].

218. The total activity of ^{90}Sr produced in nuclear explosions through 1976, which has been globally distributed, is estimated to be $6 \cdot 10^{17} \text{ Bq}$ [U1]. This does not include the local fallout, which is deposited in the vicinity of the test area and which can be important for the lower yield tests detonated near or on the land surface.

219. On the basis of measured fission product yields of individual nuclides obtained by analysis of debris from megaton weapons, the yields for ^{90}Sr , ^{129}I and ^{131}I are estimated to be 0.035, 0.0126 and 0.029, respectively [H2].

220. The activities of ^{129}I and ^{131}I produced in nuclear tests through 1976, which gave rise to globally distributed fallout, are thus found to be $4 \cdot 10^{11} \text{ Bq}$ and $6 \cdot 10^{20} \text{ Bq}$. Owing to the half-lives of the two nuclides, practically all of the activity of ^{129}I is still present in the environment where it will remain for millions of years whereas practically all of the activity of ^{131}I has decayed.

3. Nuclear fuel cycle

(a) Nuclear power plants

221. Iodine-131 and iodine-129 are produced by fission in the fuel of nuclear reactors. The equilibrium activity of ^{131}I per unit of electrical power is established after a few weeks of irradiation in uranium fuel at about $3 \cdot 10^{15}$ Bq per MW(e) [U1] and increases slightly from the beginning of the fuel irradiation to the end, as the result of the larger fission yield of plutonium which contributes increasingly to power production as the burn-up proceeds. The corresponding activity of ^{131}I produced per unit energy generated is about $9 \cdot 10^{16}$ Bq per MW(e)a.

222. Small amounts of ^{131}I produced in the fuel may reach the coolant of the nuclear reactor through defects in the fuel cladding. In coolant purification or following coolant leakage, ^{131}I may reach the gaseous and liquid effluent streams. The reported ^{131}I releases in airborne effluents, as summarized by UNSCEAR [U1], show an extremely wide variability due mainly to different waste treatment systems. The overall activity discharged per unit energy generated is of the order of 10^7 Bq per MW(e)a from PWRs and 10^8 Bq per MW(e)a from BWRs [I1, U1] while the limited data from GCRs and HWRs indicate that the average airborne releases are comparable to those from PWRs [U1].

223. Airborne iodine can occur in various chemical forms in the airborne effluents. Elemental iodine (I and I_2), organic iodine with methyl iodide (CH_3I) as the simplest organic compound and hypoiodous acid (HOI) may be present in significant proportions, iodine is also to some extent bound to particulates.

224. Few data are available on the proportions of organic and inorganic forms of the iodine released to the atmosphere. Analyses at power stations in the Federal Republic of Germany show that only a very small fraction (usually less than 1%) of the iodine released in airborne effluents is in particulate form [W1]. Recent measurements in the U.S.A. indicate that, in airborne effluents from PWRs, on the average 31% of ^{131}I is organic, 40% HOI, 27% elemental and 2% in particulate form [P1], whereas in BWRs, the proportions were found to be somewhat different, namely 40% organic, 20% HOI, 28% elemental and 12% particulate [P2]. These proportions, however, are expected to vary significantly according to the type of waste treatment in use.

(b) Fuel reprocessing plants

229. At the fuel reprocessing stage of the nuclear fuel cycle (if it is undertaken), the elements uranium and plutonium in the irradiated nuclear fuel are recovered for reuse in fission reactors. Before reprocessing, the spent fuel elements are stored under water until ^{131}I has decayed to insignificant amounts. Storage times of six months and one year result in the reduction of the activities of ^{131}I originally present in the fuel by factors of $6.5 \cdot 10^6$ and $4.3 \cdot 10^{13}$, respectively.

230. In 1980, the only reprocessing plants operating commercially were at Windscale (U.K.) and La Hague and Marcoule (France): in addition there were several small experimental reprocessing facilities, such as the one at Karlsruhe (Federal Republic of Germany). The combined capacity of the reprocessing plants was much lower than the amount of fuel discharged from reactors worldwide.

231. The activity of ^{131}I released into the environment from reprocessing plants depends critically on the storage time: in practice with the growing backlog of fuel for reprocessing a storage time of one year is common but even a relatively small quantity of fuel with a short storage time will dominate the total ^{131}I releases. Luykx and Fraser [L2] have expressed the reported releases of ^{131}I from Windscale, La Hague, Marcoule and Karlsruhe during 1974-1978 in terms of activity discharged per unit of electricity generated. The results range from less than $1.5 \cdot 10^5$ to $7 \cdot 10^7$ Bq per MW(e)a and are presented in Table VI.2. The activities of ^{131}I discharged in liquid effluents have not been reported.

232. The discharges of ^{129}I depend upon the specific waste treatment at the reprocessing plant. With regard to airborne effluents, the reported activities released per unit of electricity generated were, on average during the 1975-1978 time period, $2.7 \cdot 10^6$ Bq per MW(e)a at Windscale and $4.8 \cdot 10^5$ Bq per MW(e)a at Karlsruhe, representing about 4% and 1%, respectively, of the fuel content [L2]. In a series of measurements from November 1975 to August 1977 the average values for the components of ^{129}I discharges from Karlsruhe were reported at 74% inorganic, 23% organic and 2% aerosol [B1].

Table VI.2

Average normalized activities of ^{129}I and ^{131}I
discharged into the environment by fuel reprocessing plants
 (Bq per MW(e)a)

Plant Location	Iodine-129			Iodine-131		
	Airborne effluents	Liquid effluents	Total	Airborne effluents	Liquid effluents	Total
Windscale	$2.7 \cdot 10^6$	$5.6 \cdot 10^7$	$5.9 \cdot 10^7$	$2.6 \cdot 10^6$	N.A. a/	$>2.6 \cdot 10^6$
La Hague	N.A.	N.A.	N.A.	$1.1 \cdot 10^7$	N.A.	$>1.1 \cdot 10^7$
Marcoule	N.A.	N.A.	N.A.	$7.4 \cdot 10^7$	N.A.	$>7.4 \cdot 10^7$
Karlsruhe	$4.8 \cdot 10^5$	N.A.	N.A.	$>1.5 \cdot 10^5$	N.A.	-

a/ N.A. = Data not available.

233. In recent years, the ^{129}I released in liquid effluents was only measured at Windscale. They average at $5.6 \cdot 10^7$ Bq per MW(e)a which corresponds fairly well with the theoretical fuel content [L2].

234. The information on ^{131}I and ^{129}I activities discharged per unit electricity generated is summarized in Table VI.2. If it is assumed for the four reprocessing plants, that all the ^{129}I contained in the fuel is discharged into the environment, the total ^{129}I released in 1978 was about $3 \cdot 10^{11}$ Bq, which is 7 orders of magnitude higher than the total activity estimated to be released from reactors. With regard to ^{131}I , it is much more difficult to assess the total activity released from fuel reprocessing plants, as the activities discharged into liquid effluents have not been reported. However, using the pessimistic assumptions that the activity contained in the airborne effluents represents 1% of the activity present in the fuel at the time of reprocessing and that the rest of the activity is discharged into liquid effluents, it is found that the total annual ^{131}I discharges from fuel reprocessing plants are about $5 \cdot 10^{11}$ Bq, which is much less than the global discharges from reactors.

C. BEHAVIOUR IN THE ENVIRONMENT

1. Nuclear explosions

235. The behaviour in the environment of ^{131}I produced in the nuclear explosions has been extensively studied, especially the air-vegetation-milk pathway which is generally the most significant route by which humans are exposed. Much of the literature has been referenced in UNSCEAR reports [U1-U5]. Environmental concentrations of ^{131}I following large nuclear explosions are significant; they are easily measured and this allows the transfer factors to be derived from observations. In contrast, the environmental concentrations (and the resulting dose rates) of ^{129}I are extremely low and have only been measured in a few studies. Although some of the aspects of environmental behaviour of ^{131}I following nuclear explosions apply also to ^{129}I , the discussion in this section will be limited to ^{131}I .

236. Radioactive fallout is observed to circle the earth in 20-30 days on average [R1] which is approximately the mean residence time of an aerosol in the troposphere and is longer than the mean life of ^{131}I . It is thus during its first pass around the earth that a given atom of ^{131}I formed in a nuclear explosion will either decay in the atmosphere or deposit on the earth's surface. It is unlikely that during such a short period the clouds of debris become well mixed. The ground-level air concentrations of ^{131}I at a particular station fluctuate according to meteorological conditions and are not necessarily representative of a larger region nor of a latitude band [P3].

237. Information on the physical and chemical nature of fallout ^{131}I is very limited. In the U.K., late 1961, an average 75% of the activity was in particulate form, the rest being in the gaseous state [E2] but in the U.S.A. in 1962 the particulate fraction was found to vary from 10 to 90% [P4]. These large variations may be partly explained by the physical and chemical transformations undergone by ^{131}I following its formation: Voillequé [V1] observed that the fraction of the total airborne ^{131}I associated with particulates is about 0.5 to 0.7 in the first few days following a nuclear explosion but that it later decreases to be approximately 0.3 after two months. In the gaseous fraction, the proportion of organic compounds was found to increase in the same two months time period from one fourth to about three fourths of the gaseous iodine [V1].

238. Iodine-131 deposition on the ground and on vegetation occurs by dry and wet deposition. The rate of dry deposition can be characterized by the deposition velocity on the vegetation which was derived to be about $2 \cdot 10^{-2} \text{ m s}^{-1}$ [C2, H4] from measurements performed during the series of tests of late 1961. When precipitation occurs, fallout ^{131}I is deposited at a much faster rate than in dry weather, essentially by rain-out, i.e., in-cloud mechanisms, rather than by wash-out, i.e., below the cloud processes [U5]. On the other hand, rain washes the surface of the leaves and thus removes some of the radio-iodine. Chamberlain and Chadwick in the U.K. [C2] and Hull in the U.S.A. [H4] calculated on the basis of their measurements that in late 1961 about 50% of the ^{131}I falling out in rain was retained on herbage.

239. Even though the observed ground-level air activity concentrations and deposited activities of ^{131}I vary widely from one area to another according to meteorological conditions, it is possible to obtain a rough estimate of the total activity density deposited, weighted over the world's population, from the average ratio of the $^{131}\text{I}/^{140}\text{Ba}$ deposited activity densities. Data from Argentina covering the years 1966 to 1973 [B2, B3, C3] reveal that the $^{131}\text{I}/^{140}\text{Ba}$ ratio of the annual deposited activity densities varied from 0.4 to 1.3 with a median value of 0.6. Relevant information is also provided by the air concentrations of ^{131}I and ^{140}Ba which are measured in the stations of the global network of the U.K. Atomic Energy Agency [C4]. The annual average of the integrated air activity concentration $^{131}\text{I}/^{140}\text{Ba}$ ratios of nine stations scattered over the world ranged from 0.19 to 3.1 with a median value of 0.46. However, the corresponding ratios of the deposited densities were higher as only the particulate fraction of ^{131}I was measured in the air. Assuming that the particulate fraction of ^{131}I represents half of the total activity of that nuclide in the air, the median value would be approximately 0.9, which is comparable to the Argentinian value of 0.6. An intermediate value of 0.7 will be adopted in this document.

240. As the average ratio of the $^{140}\text{Ba}/^{95}\text{Zr}$ deposited activity density is estimated to be 0.62 [U2] and the population-weighted global average deposition density of ^{95}Zr from all tests is approximately $2.4 \cdot 10^4 \text{ Bq m}^{-2}$ [U2], the population-weighted global average deposition density of ^{131}I is thus found to be on the order of 10^4 Bq m^{-2} .

241. Fresh milk is usually the main source of ^{131}I in food because of the concentration achieved by the grazing animal and the short storage period of milk. Besides, milk plays an important worldwide role in the diet of infants. The relationship between the integrated cow's milk concentration and the deposition density has been derived from measurements in Argentina [B2] to be $6.3 \cdot 10^{-4} \text{ Bq a l}^{-1}$ per Bq m^{-2} and to show little variation from year to year. The integrated milk concentrations observed throughout the world by the fallout network stations have been reported by UNSCEAR [U1-U5].

2. Industrial releases

242. The environmental behaviour of radio-iodines released from nuclear facilities differs in some aspects from that of fallout as the chemical forms are not in the same proportion and as the releases occur at discrete points on the surface of the earth both in the atmosphere and in the aquatic environment. As the authorities are concerned with the total impact resulting from the releases of radionuclides, all the important pathways leading to man have been investigated, mainly through laboratory and field experiments, and occasionally following unplanned releases. In comparison to ^{131}I and to fallout, much more ^{129}I is released from industrial operations and it will be considered in this section, together with ^{131}I in the discussion of the local and regional aspects, and on its own in the discussion of the global aspects.

(a) Local and regional aspects

(i) Atmospheric releases

243. The behaviour in the atmosphere of the radio-iodines released from nuclear facilities is complicated by the various forms that iodine may take (particulate, elemental, organic, or as hypoiodous acid). Elemental iodine readily deposits on forage and enters the cow-milk-man pathway. Organic iodine is retained much less efficiently by vegetation and its deposition velocity is 200 to 1000 times smaller than that of the elemental form [A1, H5]. Particulate associated iodine and hypoiodous acid will be deposited at rates intermediate between those for the elemental and organic forms [V2]. Physico-chemical transformations occurring during atmospheric transport may also affect the distribution of the various forms of iodine, since some of them are not stable in sunlight. On the basis of photochemical considerations, the atmospheric residence times for I_2 are estimated to be less

than a minute during the day, much shorter than the residence times for CH_3I and other organic iodides in plant effluents (about 60 hours in sunlight) [V1]. The elemental form would be expected to become rapidly associated with airborne aerosols, so that deposition at distances beyond the immediate vicinity of the release would be largely governed by the particulate behaviour. It must be pointed out, however, that the atmospheric residence times of CH_3I derived from environmental measurements is much longer than that obtained from photochemical considerations (about 100 days to be compared with 60 hours in sunlight) [V1].

244. Numerous field and laboratory experiments have been conducted to determine the deposition velocity on vegetation of various forms of iodine [B4, H6, V2]. Most of the experiments dealt with elemental iodine, for which the deposition velocity was found to vary with the temperature, the relative humidity of the air, the wind speed and the vegetation density. The best fit of the experimental data is obtained assuming that the deposition velocity on vegetation of elemental iodine is proportional to the wind speed and to the vegetation density and is an exponential function of the temperature and of the relative humidity of the air [A2]. Typical values of the deposition velocity on vegetation are $2 \cdot 10^{-2} \text{ m s}^{-1}$ for elemental iodine and $5 \cdot 10^{-3} \text{ m s}^{-1}$ for organic iodide. In the case of particulates, a value of 10^{-3} m s^{-1} was found representative for grass, about $2 \cdot 10^{-3} \text{ m s}^{-1}$ for clover and about $3 \cdot 10^{-4} \text{ m s}^{-1}$ for vegetation-free soil [H7]. Since the deposition velocity varies with the vegetation density, less variability is encountered by normalizing the values by the mass of dry vegetation per unit area. UNSCEAR [U1] adopted a normalized value of $5 \cdot 10^{-3} \text{ m}^3 \text{ kg}^{-1} \text{ s}^{-1}$ for the deposition velocity on grass of radio-iodine in effluents from nuclear installations.

245. Regarding atmospheric releases of radio-iodine, the main pathways to man are inhalation and consumption of fresh milk and fresh leafy vegetables; consumption of beef is also taken into consideration in the case of ^{129}I .

246. The assessment of the transfer of iodine to milk requires the knowledge of the value of the following parameters in addition to the deposition velocity; the residence half-time of iodine on vegetation, the average mass of grass consumed per cow and per day under average agricultural conditions during the grazing season, the fractional pasture grazing time and the fractional transfer of the daily ingested activity by the cow per unit volume of produced milk.

247. The residence half-time of ^{131}I on grass is 3-6 days, most estimates lying around 5 days [B4, B5]. This figure seems to be valid irrespective of the iodine production source (fallout, nuclear plants, experiments) and of the climatic characteristics of the region [B4]. The corresponding residence half-time of stable iodine (or of ^{129}I) on grass is about 14 days. The depletion mechanisms involved are: transfer to the roots; volatilization; leaching by atmospheric precipitation; mechanical removal by wind, rain or other agents; death or decomposition of the leaves or of their surface layer [B4, C2]. The opinions do not agree on the relative importance of those mechanisms [B4, C2].

248. The daily grass requirement of a lactating cow is estimated at 10 kg dry matter [B4, H6]. The pasture grazing time varies according to the climatic conditions and to the cattle management practices. An average grazing time of six months per year was assumed by UNSCEAR [U1]. During the winter months, the cows are held in the stable and consume dry fodder in which the activity concentration of ^{131}I will have decayed to insignificant levels and that of ^{129}I can be assumed to be the same as that in herbage.

249. The transfer of iodine is usually expressed as the fraction present in milk of the ingested activity under equilibrium conditions. This quotient was found to be around $5 \cdot 10^{-3} \text{ d l}^{-1}$ [B6, U1]. The value adopted by UNSCEAR in its 1977 report is an upper estimate of 10^{-2} d l^{-1} [U1]. In the case of a simple administration of ^{131}I to the cow, Lengemann and Comar [L3] observed that the maximum concentration is reached within one day and that it is followed by a rapid decrease (half-time of about 1.5 days) in the first 3-4 days and a slower decrease (half-time of about 3 days) afterwards. Among the different factors which can have an influence on the grass to milk transfer of iodine, the two most important may be the milk productivity by animals and the season, a higher iodine secretion in milk occurring with a productivity increase and in the warm season [B4, G1].

250. Taking account of all the parameters given above, an integrated air concentration of 1 Bq a m^{-3} of ^{131}I or ^{129}I would result in an integrated milk concentration for ^{131}I of 160 Bq a l^{-1} and for ^{129}I of 870 Bq a l^{-1} . Transfer via wet deposition is usually insignificant over the course of a year [U1].

251. The transfer of iodine from air to fresh leafy vegetables has been assessed by UNSCEAR [U1] on the basis of the values given above for the deposition velocity and the

residence time on the vegetation, and of a dry-to-wet vegetation weight ratio of 0.5. In addition, a fractional removal by washing of 0.4 [U6] and an average 7 day marketing delay (resulting in a decay factor of 0.55 for ^{131}I and 1.0 for ^{129}I) were taken into account. Time integrated conditions of 340 Bq a kg^{-1} (fresh weight) for ^{131}I and $1740 \text{ Bq a kg}^{-1}$ (fresh weight) for ^{129}I are calculated for the case of a time-integrated air concentration of 1 Bq a m^{-3} .

252. An estimate of the transfer of ^{129}I has also been carried out by UNSCEAR [U1]. Using the values given above for the deposition velocity on grass, residence half-time on grass and grass consumption rate, the resulting transfer factor is 260 Bq a kg^{-1} per Bq a m^{-3} of ^{129}I in air if the fractional transfer of daily ingested activity per unit mass of meat is taken to be $3 \cdot 10^{-3} \text{ d kg}^{-1}$ [P5].

253. In the assessment of the ^{129}I concentration in grass following deposition of that nuclide, the root uptake from soil has not been taken into account. It has been estimated [B7] that this pathway could contribute at equilibrium only about 20% of the concentration in grass arising from direct deposition.

(ii) Aquatic releases

254. Information on the behaviour of iodine in the aquatic environment is rather limited. The UNSCEAR aquatic model [U1] can be used to estimate the transfer of ^{131}I and ^{129}I from the aquatic environment to diet for generalized discharge situations. In that model, it is assumed that iodine is not removed to sediments but that during treatment of drinking water, 20% of the activity contained in raw water is removed. For the two isotopes of iodine considered, the concentration factors are taken to be 15 l kg^{-1} for fresh water fish, 20 l kg^{-1} for marine fish and 100 l kg^{-1} for shellfish. Further discussion is presented in the section on dosimetry.

(b) Global aspect

255. Because of its very long half-life ($1.57 \cdot 10^7 \text{ a}$) and of the mobility of iodine in the environment, ^{129}I may become widely distributed on the global scale. Whether released into the atmosphere or into the aquatic environment, ^{129}I will eventually reach the oceans in a time period very short in comparison with its half-life. Iodine-129 will then be recycled to the atmosphere and the terrestrial biosphere, mainly by evaporation of seawater. Atmospheric water is

exchanged within about 10 days and it follows therefore that there is a rapid exchange of iodine. The specific activity approach has been the usual method to assess the dose commitments and the long-term environmental concentrations from ^{129}I discharges. According to that approach, the specific activity of ^{129}I per unit mass of stable iodine of any environmental material of the terrestrial biosphere (including air and food-stuffs) will be in the long term equal to that of seawater. Assuming $6 \cdot 10^{23}$ g to be the mass of ocean waters with an iodine concentration of $0.064 \mu\text{g}$ per gram of water [T1], the specific activity of ^{129}I per unit mass of stable iodine obtained in sea water after a release of 1 Bq of that radionuclide into the environment is found to be $2.6 \cdot 10^{-17} \text{ Bq g}^{-1}$. If it is assumed that there is no environmental sink for iodine, ^{129}I will be recycled throughout its mean life of $2.3 \cdot 10^7 \text{ a}$.

D. TRANSFER TO MAN

256. Iodine is an element of fundamental importance for the human organism since it is an essential component of the thyroid hormone, which is necessary for the growth and metabolism of the body. The metabolic cycle of iodine in man, especially in the adult, is sufficiently well known in its fundamental behaviour, as a consequence of the large number of clinical studies carried out in the last years with radioactive isotopes of iodine [B4].

257. The absorption by the blood from the gastro-intestinal tract is complete and very rapid. It is absorbed at the rate of about 5% per minute and it can be considered to be complete after two hours [B4]. When inhaled in the form of inorganic iodide or as methyl iodide, a fraction of about 70% is absorbed [M2] whereas more than 90% of it is absorbed when inhaled in the form of elemental vapour [M3].

258. The uptake by the thyroid of the iodine contained in blood as well as the size of the thyroid gland are both very dependent upon the daily intake of stable iodine [D2]. The model adopted by ICRP [I3] for the metabolism of iodine applicable to adults is based on the three-compartment model of Riggs [R2]. ICRP [I3] assumes that 30% of the iodine entering the blood is translocated to the thyroid while the remainder goes directly to excretion. Iodine in the thyroid is assumed to be retained with a biological half-life of 120 days and to be lost from the gland in the form of organic iodine. Organic iodine is assumed to be uniformly distributed among all organs and tissues of the body other than the thyroid and to be retained there with a biological half-life

of 12 days. One-tenth of this organic iodine is assumed to go directly to faecal excretion and the rest is assumed to be returned to blood as inorganic iodine.

259. A quantitative assessment of the transfer of iodine to man must take into consideration the variation with age of the various parameters involved, which are essentially the mass of the thyroid gland, the fractional uptake by the thyroid, the effective residence half-time in the thyroid, the breathing rate and the consumption rate of foodstuffs. The values adopted for ^{129}I and ^{131}I by UNSCEAR in its 1977 report [U1] are presented in Table VI.3. It is to be noted that the metabolic parameters for adults are not in complete agreement with those adopted by ICRP [I3]; the resulting differences in the thyroid absorbed doses vary according to the pathway and the isotope considered but they are in all cases less than 50%.

Table VI.3

Variation with age of the parameters used in the assessment
of the transfer to man of ^{129}I and ^{131}I
 [U1]

	Age			
	6 months	4 years	14 years	Adult
Mass of the thyroid (g)	2	4	14	20
Effective half-time in the thyroid (d)				
^{131}I	6.0	6.3	6.9	7.6
^{129}I	23	28	48	136
<u>Inhalation pathway</u>				
Fractional uptake by the thyroid	0.30	0.26	0.26	0.26
Breathing rate ($\text{m}^3 \text{a}^{-1}$)	1150	3530	6440	8030
<u>Ingestion pathway</u>				
Fractional uptake by the thyroid	0.40	0.35	0.35	0.35
Consumption rate:				
Milk (l a^{-1})	330	180	150	90
Leafy vegetables (kg a^{-1})	0	13	20	30
Beef (kg a^{-1})	0	8	15	27
Drinking water (l a^{-1})	438	438	438	438
River fish (kg a^{-1})	1	1	1	1
Ocean fish (kg a^{-1})	6	6	6	6
Shellfish (kg a^{-1})	1	1	1	1

E. DOSIMETRY

1. Dose per unit intake

260. As iodine is selectively taken up in the thyroid gland, its concentration in that organ is considerably higher than in the other organs and tissues in the body. The stable iodine concentration in the thyroid tissue of an adult is of the order of $500 \mu\text{g g}^{-1}$ while in the rest of the body it is much less than $1 \mu\text{g g}^{-1}$ [S2]. Since the major contribution of the absorbed doses from ^{129}I and ^{131}I is due to the emission of the beta-particles, which have a short range in the human tissues, the absorbed doses in the thyroid are about 1000 times higher than those in the other organs and tissues, which will not be considered in this document.

261. The significant variation with age of the metabolic parameters is reflected in the thyroid absorbed doses per unit intake. Table VI.4 presents the values of the age-dependent thyroid absorbed doses per unit intake adopted by UNSCEAR in its 1977 report [U1]. These values are derived from the metabolic parameters presented in Table VI.3 and from the following figures for the energy absorbed in the thyroid (MeV) for disintegration: 0.18, 0.18, 0.19 and 0.19 MeV for ^{131}I and 0.060, 0.061, 0.063 and 0.064 MeV for ^{129}I for the ages of 0.5, 4, 14 years and adult, respectively.

Table VI.4

Age-dependent absorbed doses in the thyroid gland
per unit intake of ^{131}I and ^{129}I
(Gy Bq⁻¹)

Thyroid absorbed dose per unit intake	Age			
	6 months	4 years	14 years	Adult
Inhalation: ^{131}I	$3.2 \cdot 10^{-6}$	$1.5 \cdot 10^{-6}$	$4.9 \cdot 10^{-7}$	$3.8 \cdot 10^{-7}$
^{129}I	$4.1 \cdot 10^{-6}$	$2.2 \cdot 10^{-6}$	$1.1 \cdot 10^{-6}$	$2.3 \cdot 10^{-6}$
Ingestion: ^{131}I	$4.3 \cdot 10^{-6}$	$2.0 \cdot 10^{-6}$	$6.5 \cdot 10^{-7}$	$5.1 \cdot 10^{-7}$
^{129}I	$5.4 \cdot 10^{-6}$	$3.0 \cdot 10^{-6}$	$1.5 \cdot 10^{-6}$	$3.0 \cdot 10^{-6}$

2. Dose per unit release

(a) Nuclear explosions

262. The thyroid dose commitment via the ingestion (milk) pathway from ^{131}I released by nuclear explosions can be assessed from the sequential product of transfer factors

$$D^C = P_{23} P_{35} F$$

where F is the integrated deposition density, P_{23} is the deposition to milk transfer factor and P_{35} is the milk to thyroid dose transfer factor. The value of F weighted for the world's population and that of P_{23} for fallout deposition were given above as 10^4 Bq m^{-2} and $6.3 \cdot 10^{-4} \text{ Bq a l}^{-1}$ per Bq m^{-2} , respectively. The value of P_{35} can be derived from the age-dependent consumption rates of milk given in Table VI.3, the age-dependent thyroid doses per unit ingested activity presented in Table IV.4, and from the assumption that the three groups of children are representative of the age groups 0-1, 1-9 and 10-19 years, respectively, and that these groups contain respectively 2, 16, and 20% of the population [U]. The value of P_{35} (milk) is thus found to be $1.3 \cdot 10^{-4} \text{ Gy per Bq a l}^{-1}$. The thyroid dose commitment for the world's population arising from ^{131}I from global fallout of past nuclear explosions is therefore estimated to be about $8 \cdot 10^{-4} \text{ Gy}$. Most of the dose commitment was in fact delivered in the early 1960s. Taking the world's population at that time to be $3 \cdot 10^9$ persons, the collective dose commitment would be about $2 \cdot 10^6 \text{ man Gy}$. Since $6 \cdot 10^{20} \text{ Bq}$ of ^{131}I were estimated to give rise to global fallout, the individual and collective thyroid dose commitments per unit activity released are found to be $1.3 \cdot 10^{-24} \text{ Gy Bq}^{-1}$ and $3 \cdot 10^{-15} \text{ man Gy Bq}^{-1}$, respectively.

(b) Nuclear industry

(i) Local and regional contribution

263. Atmospheric releases. The contribution of the inhalation pathway to the collective dose commitments from effluent releases can be estimated from the integrated concentrations of ^{131}I and ^{129}I in ground-level air. Assuming that all the activity released in the atmosphere will eventually deposit on the ground, the integrated concentration in ground-level air is the total amount of ^{131}I or ^{129}I released per unit area of the deposition region divided by the deposition velocity, v_d . The population affected is the population density δ_N , times the area of the deposition region. The collective dose commitment per unit activity

released is given by the expression $S^c = \delta_N \varphi / v_d$ where φ is the age-weighted thyroid dose per unit integrated air concentration.

264. On the basis of a figure of $5 \cdot 10^{-3} \text{ m}^3 \text{ kg}^{-1} \text{ s}^{-1}$ for the deposition velocity on grass per unit area density of vegetation (see paragraph 244), using a value of 0.1 kg (dry) m^{-2} for the mass of grass per unit area, and assuming that the activity deposited on grass represents one fourth of the total activity deposited on ground, v_d would be $2 \cdot 10^{-3} \text{ m s}^{-1}$ or $6 \cdot 10^4 \text{ m a}^{-1}$. From the data contained in Tables VI.3 and VI.4 and the age distribution given above, the age-weighted thyroid dose per unit integrated air concentration would be $3.4 \cdot 10^{-3}$ and $1.4 \cdot 10^{-2}$ Gy per Bq a m^{-3} for ^{131}I and ^{129}I respectively. As the deposition area is expected to be very large, the population density is assumed to be about 25 persons km^{-2} , that is $2.5 \cdot 10^{-5}$ persons m^{-2} . The collective thyroid dose commitments are thus estimated to be $1.4 \cdot 10^{-12}$ and $5.8 \cdot 10^{-12}$ man Gy Bq $^{-1}$ for ^{131}I and ^{129}I , respectively.

265. The contribution of the ingestion pathway (consumption of milk, leafy vegetables and meat) to the collective dose commitments per unit activity released can be assessed by the expression $S^c = P_{13} P_{35} \delta_N$ where P_{13} is the air to dietary product to age-weighted thyroid dose transfer factor, and δ_N the population density. Using the values and assumptions given previously, the collective thyroid dose commitments are estimated to be, for ^{131}I , $8.7 \cdot 10^{-12}$ and $2.3 \cdot 10^{-12}$ man Gy Bq $^{-1}$ for consumption of milk and fresh leafy vegetables, respectively, while the values for ^{129}I would be $1.2 \cdot 10^{-10}$, $5.0 \cdot 10^{-11}$ and $6.4 \cdot 10^{-12}$ man Gy Bq $^{-1}$ for consumption of milk, fresh leafy vegetables and beef, respectively.

266. Aquatic releases. The collective thyroid dose commitment per unit activity of ^{131}I and ^{129}I discharged into the aquatic environment can be estimated, as in the 1977 UNSCEAR report [U1], using the expression

$$S^c = \frac{N I f \varphi}{V(\lambda + I/\tau)}$$

where V is the volume of the receiving waters, τ the turn-over time of receiving waters, λ the decay constant of the radionuclide considered, N the number of individuals exposed, I the individual consumption rate of the foodstuff considered, f the concentration factor of the radionuclide in that foodstuff, and φ the thyroid dose per unit activity ingested.

267. The quantity $\frac{1}{V(\lambda + 1/\tau)}$ is the infinite time integral of the water concentration per unit activity released, while that quantity multiplied by f is the infinite time integral of the concentration in the consumed item (fish, for example). For inputs into small volumes of water, the concentrations in water and in fish will be high, but the populations which can be served with drinking water or by fish consumption will be limited. For inputs into large volumes of water, the concentrations will be smaller, but the populations involved will be larger. It is reasonable, therefore, to assume as a first approximation that the quantities V/N are relatively constant; they are taken to be $3 \cdot 10^7$ and $3 \cdot 10^9$ litre per man, for fresh water and sea water, respectively. Using the values given previously in the text and in the tables, the collective thyroid dose commitments per unit activity of ^{131}I and ^{129}I can be estimated. The results are presented in Table VI.5.

Table VI.5
Collective thyroid dose commitments per unit activity
of ^{131}I and ^{129}I released in the aquatic environment
 (man Gy Bq⁻¹)

Type of release and pathway	^{131}I	^{129}I
<u>Release to fresh water</u>		
- Drinking water	$3.2 \cdot 10^{-13}$	$3.2 \cdot 10^{-10}$
- Fish	$1.4 \cdot 10^{-14}$	$1.4 \cdot 10^{-11}$
<u>Release to sea water</u>		
- Fish	$1.1 \cdot 10^{-15}$	$1.1 \cdot 10^{-13}$
- Shellfish	$8.8 \cdot 10^{-16}$	$1.1 \cdot 10^{-14}$

(ii) Global contribution

268. Using the specific activity approach described previously, the activity concentration of ^{129}I per unit mass of ^{127}I is the same in the sea water and in the human thyroid. Assuming that the concentration of stable iodine per unit mass of thyroid is 80, 180, 300 and 600 $\mu\text{g g}^{-1}$ at ages 6 months, 4 years and 14 years and for adults, respectively and using the age distribution given previously, a specific activity of 1 Bq per gram of stable iodine in the

thyroid would lead to an age-weighted annual thyroid dose of $1.5 \cdot 10^{-7}$ Gy. Since a release of 1 Bq ^{129}I results in a long-term concentration of $2.6 \cdot 10^{-17}$ Bq g^{-1} stable iodine per Bq (see paragraph 255), the collective thyroid dose commitment arising from discharges of ^{129}I would be about $9 \cdot 10^{-7}$ man Gy Bq^{-1} , assuming a world population of 10^{10} and no sink for iodine in the environment.

269. Most of the collective dose commitment to the thyroid is delivered in the far future, as the mean life of ^{129}I is $2.3 \cdot 10^7$ a. The average dose rate per unit activity released would be extremely low ($4 \cdot 10^{-24}$ Gy a^{-1} per Bq released). The estimate of the collective thyroid dose commitment could be significantly in error if there exists a mechanism that efficiently removed iodine from the biosphere at a rate significantly greater than the radioactive decay constant of ^{129}I of $4 \cdot 10^{-8}$ a^{-1} . Such a mechanism could be the retention by ocean sediments.

(c) Summary

270. Table VI.6 summarizes the results given above for the thyroid collective dose commitments per unit activity released, and provides also the collective effective dose equivalent commitments per unit activity released. The latter quantities are obtained by multiplying the former by 0.03.

Table VI.6

Summary of collective thyroid dose commitments per unit activity released and of collective effective dose equivalent commitments per unit activity released

	^{131}I	^{129}I ^a	^{131}I	^{129}I ^a
	(man Gy Bq ⁻¹)		(man Sv Bq ⁻¹)	
<u>Weapon tests</u>	$3 \cdot 10^{-15}$ ^b		$9 \cdot 10^{-17}$ ^b	
<u>Industrial releases</u>				
a) <u>Atmosphere</u>				
Inhalation	10^{-12}	$6 \cdot 10^{-12}$	$3 \cdot 10^{-14}$	$2 \cdot 10^{-13}$
Ingestion				
- Milk	$1 \cdot 10^{-11}$	$1 \cdot 10^{-10}$	$3 \cdot 10^{-13}$	$3 \cdot 10^{-12}$
- Leafy vegetables	$2 \cdot 10^{-11}$	$5 \cdot 10^{-11}$	$6 \cdot 10^{-14}$	$2 \cdot 10^{-12}$
- Beef	-	$6 \cdot 10^{-12}$	-	$2 \cdot 10^{-13}$
b) <u>Rivers</u>				
Ingestion				
- Water	$3 \cdot 10^{-13}$	$3 \cdot 10^{-10}$	$9 \cdot 10^{-15}$	$9 \cdot 10^{-12}$
- Fish	$1 \cdot 10^{-14}$	$1 \cdot 10^{-11}$	$3 \cdot 10^{-16}$	$3 \cdot 10^{-13}$
c) <u>Oceans</u>				
Ingestion				
- Fish	$1 \cdot 10^{-15}$	$1 \cdot 10^{-13}$	$3 \cdot 10^{-17}$	$3 \cdot 10^{-15}$
- Shellfish	$9 \cdot 10^{-16}$	$9 \cdot 10^{-14}$	$3 \cdot 10^{-17}$	$3 \cdot 10^{-15}$

^a First passage estimates. The long-term, global estimates for ^{129}I from all sources and by all pathways are $9 \cdot 10^{-7}$ man Gy Bq⁻¹ and $3 \cdot 10^{-8}$ man Sv Bq⁻¹.

^b Milk consumption.

F. REFERENCES

- A1 Atkins, D.H.F., R.C. Chadwick and A.C. Chamberlain. Deposition of radioactive methyl iodide vegetation. Health Phys. 13: 91 (1967).
- A2 Angeletti, A. and A. Sauve. Estimation de la vitesse de dépôt de l'iode vapeur sur les végétaux en fonction des éléments du climat. Congrès commun de radioprotection. SFRP-FS. Lausanne, 1981.
- B1 Berg, R. and H. Schuettelkopf. Die Messung der Verteilung in und der Abgabe von I-129 aus der Wiederaufarbeitungsanlage Karlsruhe. p.81 in Radioactive Effluents from Nuclear Fuel Reprocessing Plants. CEC document V/2266/78 (1978).
- B2 Beninson, D., A.M. Migliori de Beninson, and C. Menossi. Fallout radioactivo debido a las explosiones en el Pacífico sur en el período 1966-1970. Comisión Nacional de Energía Atómica. Informe RS 28/49 (1971).
- B3 Beninson, D., A.M. Migliori de Beninson and C. Menossi. Fallout radioactivo debido a las explosiones en el Pacífico sur en el período 1971-1972. Comisión Nacional de Energía Atómica. Informe RS 43/102 (1973).
- B4 Breuer, F. and M. de Bortoli. Behaviour of radioiodine in the environment and in man. Comitato Nazionale Energia Nucleare report RT/PROT(73)13 (1973).
- B5 Bergström, S.O.W. Transport of fallout 131-I into milk. p. 159-174 in Radiological Concentration Processes (Aberg, B. and F.P. Hungate, eds.). Pergamon Press, 1967.
- B6 Bustad, L.F., D.H. Wood, E.E. Elefson et al. 131-I in milk and thyroid of dairy cattle following a single contamination event and prolonged daily administration. Health Phys. 9: 1231-1234 (1963).
- B7 Bouville, A. Estimation des doses dues aux rejets d'iode 129 par les installations nucléaires. Echelles locale et régionale. p. 53-68 in I-129. Proceedings of an NEA Specialist Meeting. OECD, Paris, 1977.
- C2 Chamberlain, A.C. and R.C. Chadwick. Transport of iodine from atmosphere to ground. United Kingdom Atomic Energy

- Authority Research Group report AERE-R-4870. Harwell, Berkshire, 1965.
- C3 Comisión Nacional de Energía Atómica, Argentina. Fallout Radioactivo debido a las explosiones en el Pacífico sur en el período Enero-Octubre de 1973. Informe RS-47/118 (1973).
- C4 Cambray, R.S. et al. Radioactive fallout in air and rain. Reports covering the years 1955-1979. AERE-R-4094 (1962); AERE-R-4392 (1963); AERE-R-4687 (1964); AERE-R-4997 (1965); AERE-R-6556 (1970); AERE-R-7245 (1972); AERE-R-7832 (1974); AERE-R-8267 (1975); AERE-R-8671 (1976); AERE-R-9018 (1978); AERE-R-9441 (1979); AERE-R-9672 (1980).
- D1 Dillman, L.T. and F.C. Von der Lage. Radionuclide decay schemes and nuclear parameters for use in radiation-dose estimation. NM/MIRD Pamphlet No.10. Society of Nuclear Medicine, September 1975.
- D2 Dolphin, G.W. Dietary intakes of iodine and thyroid dosimetry. Health Phys. 21: 711-712 (1971).
- E1 Edwards, R.R. Iodine-129: Its occurrence in nature and its utility as a tracer. Science 137: 851-853 (1962).
- E2 Eggleton, A.E.J., D.H. Atkins and L.B. Cousins. Chemical and physical nature of fallout ^{131}I and carrier-free ^{131}I released in air. An abstract. Health Phys. 9: 1111 (1963).
- G1 Garner, R.J. and R. Scott Russell. Isotopes of iodine. Chapter 14 in Radioactivity and human diet (R. Scott Russell ed.). Pergmon Press, 1966.
- H2 Harley, N., I. Fisenne, L.D.Y. Ong et al. Fission yield product decay. p. 251-260 in Health and Safety Laboratory Fallout Program quarterly summary report HASL-164. New York, 1965.
- H3 Hull, A.P. Critical evaluation of radiological measurements and of the need for evacuation of the nearby public during the Three Mile Island incident. Paper presented at the IAEA International Conference on current nuclear power plant safety issues. Stockholm, October 1980.
- H4 Hull, A.P. Vegetation retention and vegetation-milk ratios of fallout ^{131}I . Health Phys. 9: 1173-1177 (1963).

- H5 Heinemann, K., M. Stoepler, K.J. Vogt et al. Untersuchungen zur Ablagerung und Desorption von Jod auf Vegetation. Kernforschungsanlage Jülich report Jül-1287 (1976).
- H6 Hoffman, F.O. A reassessment of the deposition velocity in the prediction of the environment transport of radioiodine from air to milk. Health Phys. 32: 437-441 (1977).
- H7 Horbert, J., K.J. Vogt and L. Angeletti. Untersuchung zur Ablagerung von Aerosolen auf Vegetation and anderen Grenzflächen. Kernforschungsanlage Jülich report Jül-1288 (1976).
- I1 International Atomic Energy Agency. Radioiodine removal in nuclear facilities. Methods and techniques for normal and emergency situations. Technical Reports Series No. 201. Vienna, 1980.
- I2 International Atomic Energy Agency. Power reactors in member states. 1980 edition. Vienna, 1980.
- I3 International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. ICRP Publication 30, Part 1. Annals of the ICRP, Vol. 2, No. 3/4 (1979).
- L1 Loutit, J.F., W.G. Marley and R.S. Russell. The nuclear reactor accident at Windscale, October 1957. Environmental aspects. Appendix H in The Hazards to Man of Nuclear and Allied Radiations. A second report to the Medical Research Council, London, 1960.
- L2 Luykx, F. and G. Fraser. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European Community. Discharge data 1974-1978. Radiological aspects. CEC document V/4116/80 (1980).
- L3 Lengemann, F.W.. Radioiodine in the milk of cows and goats after oral administration of radioiodate and radioiodide. Health Phys. 17: 565-569 (1969).
- M1 Manuel, O.K. Iodine-129. A study of its transport in the environment and distribution in biological systems. Report COO-2450-3 (1976).

- M2 Morgan, A., D.G. Morgan and G.M. Arkell. A study of retention and subsequent metabolism of inhaled methyl iodide in Inhaled Particles and Vapours II (C.N. Davies, ed.). Pergamon Press, 1967.
- M3 Morgan, A., D.J. Morgan and A. Black. A study of the deposition, translocation and excretion of radioiodine inhaled as iodine vapour. *Health Phys.* 15: 313-322 (1968).
- P1 Pelletier, C.A., J.E. Cline, E.D. Barefoot et al. Sources of radioiodine at pressurized water reactors. EPRI report NP-939 (1978).
- P2 Pelletier, C.A., J.E. Cline, E.D. Barefoot et al. Sources of radioiodine at boiling water reactors. EPRI report NP-495 (1978).
- P3 Peirson, D.H. and J.R. Keane. The characteristics of early fallout from the Russian nuclear explosions of 1961. *Nature* 196: 801-807 (1962).
- P4 Perkins, R.W. Physical and chemical form of ^{131}I in fallout. *Health Phys.* 9: 1113-1122 (1963).
- P5 Palms, J.M., V.R. Veluri and F.W. Boone. The environmental impact of iodine-129 released by a nuclear fuel reprocessing plant. *Nucl. Saf.* 16: 593-602 (1975).
- R1 République française. Retombées radioactives à la suite des tirs nucléaires en Polynésie. Mai-décembre 1970.
- R2 Riggs, D.S. Quantitative aspects of iodine metabolism in man. *Pharmacol. Rev.* 4: 284-370 (1952).
- S1 Srinivasan, B., E.C. Alexander Jr. and O.K. Manuel. Iodine-129 in terrestrial ores. *Science* 173: 327-328 (1971).
- S2 Spiers, F.W. Radioisotopes in the human body: physical and biological aspects. Academic Press, 1968.
- T1 Turekian, K.K. The oceans, streams and atmosphere in Handbook of Geochemistry (K.H. Wedepohl, ed.). Springer, Berlin, Heidelberg, New York, 1969.
- U1 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General

Assembly, with annexes. United Nations sales publication No. E.77.IX.I. New York, 1977.

- U2 United Nations. Ionizing Radiation: Levels and Effects. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, with annexes. United Nations sales publication No. E.72.IX.17 and 18. New York, 1972.
- U3 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-fourth Session, Supplement No. 13(A:7613). New York, 1969.
- U4 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-first Session, Supplement No. 14(A/6314). New York, 1966.
- U5 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Nineteenth Session, Supplement No. 14(A/5814). New York, 1964.
- U6 United States Environmental Protection Agency. Environmental analysis of the uranium fuel cycle. Part II. Nuclear power reactors. U.S. Environmental Protection Agency report EPA-520/9-73-003-C (1973).
- V1 Voillequé, P.G. Iodine species in reactor effluents and in the environment. EPRI report NP-1269 (1979).
- V2 Voillequé, P.G. and J.H. Keller. Air-to-vegetation transport of ¹³¹I as hypoiodous acid (HOI). Health Phys. 40: 91-94 (1981).
- W1 Winkleman, I. Bericht über die im Filterproben aus der Abluftüberwashingtonsanlage von Kernkraftwerken in der Bundesrepublik Deutschland in Jahre 1976 nachgewiesenen Einzelnuklide. STH-4/77 (1977).

VII. CAESIUM-137

A. INTRODUCTION

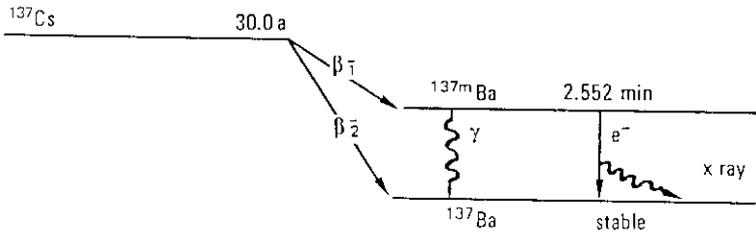
271. Caesium is element number 55 in the periodic table. It is an alkali metal like potassium, and it resembles potassium metabolically. Whereas potassium is an essential element for man, there is no evidence that caesium is also an essential trace element. In fact, stable caesium, ^{133}Cs , is fairly rare in the biosphere and in geological occurrence. Average occurrence in the earth's crust is $3 \mu\text{g g}^{-1}$. In specific rock types the estimated average concentration is $1 \mu\text{g g}^{-1}$ in basalts and $5 \mu\text{g g}^{-1}$ in granite. The K/Cs ratio in basalts is 7500 [T2]. Stable caesium occurrence in fresh water, lakes and rivers ranges between 0.01 and 1.2 ng g^{-1} and is 0.5 ng g^{-1} in the ocean [K2]. Stable potassium is more abundant, with usual concentrations of 0.2 to $10 \mu\text{g g}^{-1}$ in fresh waters and $380 \mu\text{g g}^{-1}$ in the ocean [VI].

272. The radioactive isotope ^{137}Cs is produced in nuclear fission and is one of the more significant fission products. The fission yield is relatively high, about 6 atoms per 100 fissions, independent of the type of fission in uranium or plutonium (Table VII.1). It has a radioactive half-life of 30.17 a and its beta decay is accompanied by a gamma ray of moderate energy. Figure VII.I shows the decay scheme and lists the primary transition energies.

Table VII.1

Fission yields of caesium-137
[C3]

Nuclide	Fission yield (%)	
	Thermal	Fast
^{235}U	6.21	6.12
^{239}Pu	6.64	6.50
^{238}U		5.93
^{232}Th		6.73



WHO 83179

Radiation type	Energy (keV)	Intensity (%)
<u>^{137}Cs</u>		
β_1^-	max. 511.6 avg. 173.5	94.6
β_2^-	max. 1173.2 avg. 415.4	5.4
Total β	avg. 186.6	100.0
<u>$^{137\text{m}}\text{Ba}$</u>		
γ_1	661.6	89.9

Figure VII.I Caesium-137 decay scheme information. [N2]

273. The chemical similarity of caesium and potassium and the opportunity to make simultaneous measurements by gamma spectrometry of ^{137}Cs and naturally-occurring ^{40}K has encouraged the expression of ^{137}Cs concentrations relative to the potassium concentration in a manner analogous to that used for strontium and calcium. However, caesium and potassium are not interdependent and do not behave in such a regular manner in biological systems as do strontium and calcium. As the levels of potassium in diet and man remain roughly constant (1.4 g per litre of milk and 2g per kilogram of body weight), the $^{137}\text{Cs}/\text{K}$ quotients can be converted easily to ^{137}Cs concentrations. Dietary intake of ^{137}Cs increases in proportion to the amount of food consumed, however the $^{137}\text{Cs}/\text{K}$ quotient in diet is relatively constant for adult and children diets for widespread contamination situations [G4]. An additional advantage of expressing

^{137}Cs levels in the body in terms of the $^{137}\text{Cs}/\text{K}$ quotient is that age and sex differences are minimized and the values correlate more closely to ^{137}Cs concentrations per unit of lean body mass, which seems to be a more important parameter for dosimetric purposes than the whole body mass. It is expected, however, that assessments in the future will be presented independently for ^{137}Cs , without so much reliance on the stable congener element.

274. A great deal of information has accumulated on ^{137}Cs in the environment, particularly the measurements of fallout ^{137}Cs in air, deposition, diet and man. Much of the literature has been referenced by UNSCEAR over the years. Recent reviews of ^{137}Cs data have been published by Moiseev and Ramzaev [M5] and the United States National Council on Radiation Protection and Measurements [N1]. This document is not extensive in terms of references cited. The representative references for most statements can be taken as the starting points for the more extensive literature available.

B. SOURCES

1. Nuclear explosions

275. Atmospheric testing of nuclear weapons has resulted in widespread distribution in the environment of radioactive fission and activation products. Extensive measurements of fallout radioactivity have been conducted. The data have been reported and discussed in each report of UNSCEAR [U2, U3, U4, U5, U6, U7, U8]. The stratospheric inventory of ^{90}Sr has been measured in a long-term programme [L1]. Global networks to measure fallout deposition have reported results for ^{90}Sr [F1] and ^{137}Cs [C1]. The activity ratio of ^{137}Cs to ^{90}Sr in long-term deposition has been found to be relatively constant at about 1.6 [U7], although variations for individual samplings are encountered [S1].

276. The total amount of ^{90}Sr produced in weapons testing through 1980, which has been globally dispersed, is estimated to be $6.0 \cdot 10^{17}$ Bq [F1]. Less than 1% of this amount remained in the stratosphere [L1]. The remainder has been deposited on the earth's surface. This corresponds to $9.6 \cdot 10^{17}$ Bq of ^{137}Cs produced in nuclear testing. Radioactive decay has reduced the cumulative deposit of ^{137}Cs to $6.9 \cdot 10^{17}$ Bq [C1], 76% of which is in the northern hemisphere and 24% in the southern hemisphere.

2. Nuclear fuel cycle

(a) Nuclear reactors

277. Caesium-137 is produced by fission in the fuel of nuclear reactors. The amounts produced depend on the degree of fuel burn-up achieved and to some extent on the type of fuel and the neutron spectrum in the reactor. In fairly high burn-up fuel (33000 MW[t]d t⁻¹) of a pressurized water reactor, the ¹³⁷Cs production is estimated to be 3.9 10¹⁵ Bq per tonne of fuel, corresponding to 1.3 10¹⁴ Bq per MW(e)a of electricity generated [01].

278. Small amounts of fission products produced in the fuel in nuclear reactors may reach the coolant through defects in the fuel cladding. In coolant purification or following coolant leakage, these fission products may reach gaseous and liquid effluent streams. In controlled amounts, some of the effluents are released to the environment.

279. Reported amounts of ¹³⁷Cs released to the environment from reactors have been summarized by UNSCEAR [U8]. The averaged release rates for some reactor types are included in Table VII.2.

Table VII.2

Estimated global discharges of ¹³⁷Cs
from nuclear power stations in 1980

Reactor type	Reactor number	Capacity [MW(e)a]	Release rate [Bq per MW(e)a]	Estimated discharge (Bq)
PWR	96	64239	6 10 ⁷	2 10 ¹²
BWR	62	35170	9 10 ⁸	19 10 ¹²
CCR	36	7086	2 10 ⁹	9 10 ¹²
Other	47	18490	9 10 ⁸	10 10 ¹²
Total	241	124985		4 10 ¹³

These are not necessarily the typical situation for a particular reactor. During a specific year it is normally the case that a somewhat larger release occurs from a single reactor with insignificant releases reported from all other sites. This means that there is a large range in release distribution, covering three or four orders of magnitude. For all reactor types the release of ^{137}Cs is primarily in liquid effluents; negligible amounts, in comparison, occur in airborne effluents.

280. Assuming the averaged normalized release rates to be representative for the reactor groups, it is possible to obtain a very rough estimate of the total amount of ^{137}Cs released from reactors worldwide. Using the installed capacities of the various reactor types as of 1980 [14] and assuming a reactor utilization of 60%, the estimated annual release from all reactors is about $4 \cdot 10^{13}$ Bq. In this calculation, it is assumed for the reactor types for which no data are available, that the releases are similar to those from BWRs.

(b) Fuel reprocessing plants

281. In fuel reprocessing plants the fuel is dissolved to recover uranium and plutonium for re-use. All of the ^{137}Cs and other fission products as well go to the waste streams. The radionuclide activities in airborne and liquid effluents from fuel reprocessing plants have been recorded by UNSCEAR [U8].

282. Caesium-137 is released from fuel reprocessing plants primarily in liquid effluents. Averaged release rates during the years 1971-1972 were 0.6, 90 and $520 \cdot 10^3$ Bq/MW(e)a from the Nuclear Fuel Services plant (U.S.A.) (no longer in operation), La Hague (France) and Windscale (U.K.), respectively [U8]. The release of caesium-137 in liquid effluents is small relative to the amount in spent fuel. The fractional liquid releases from the Windscale plant were approximately $4 \cdot 10^{-3}$. The only data for ^{137}Cs in airborne effluents is for the Nuclear Fuel Services plant, which corresponded to $4 \cdot 10^4$ Bq/MW(e)a [U8], several orders of magnitude less than in liquid effluents.

C. BEHAVIOUR IN THE ENVIRONMENT

1. Fixation in soil

283. Caesium is generally rather strongly fixed in soil. Downward migration and availability to plants is thereby

reduced. In mineral soils the movement of ^{137}Cs is appreciably less than that of ^{90}Sr . Three to four years after deposition on the soil surface, the median depth to which it has penetrated is usually less than 2 cm [F5]. Its mobility may be somewhat greater in organic soils. Much smaller amounts of ^{137}Cs than of ^{90}Sr are leached out of the soil to enter rivers and lakes.

284. There are exceptional areas, however, where caesium fixation in soil is much less, allowing enhanced transfer of caesium to plants. Marei et al. [M2] identified regions in the USSR where the soil is wet, peaty and podzolic, from which transfer of ^{137}Cs into the food chain is 10 times higher than for other areas. Other regions of the world where the soils give rise to high ^{137}Cs transfer into diet have also been identified, for example, in the Faroe Islands, New Zealand and Sweden [U8].

285. It has been reported that in clay minerals the important factor in fixation of caesium is the ability of certain layered silicates such as micas, vermiculites and illites to adsorb or fix trace quantities of caesium [T1]. Caesium ions are trapped in the interlayer regions of vermiculite or at the frayed edges of illites and micas. Caesium is thus more strongly retained in soils containing predominantly micaceous minerals. Soils which do not contain large quantities of micaceous minerals, such as tropical soils, peat soils, and podzolic soils, exhibit less retention and allow greater uptake of caesium by plants.

286. Fixation of caesium by sediments in aquatic environments occurs in a similar fashion to fixation in soil. The preferential adsorption of ^{137}Cs to the micaceous component of sediments has been demonstrated under environmental conditions.

2. Transfer to plants

287. Caesium-137 may be transferred to plants by direct deposition onto plant surfaces or by root uptake from accumulated deposits in soil. In general, direct foliar absorption is the predominant mode of plant contamination when the deposition rate is relatively high. Root uptake is low except in those cases mentioned above, when soil conditions allow low fixation of caesium.

288. Caesium depositing on plant surfaces is retained to the same extent as other particulate debris. A removal half-time

of 14 days due to weathering is generally assumed. Once absorbed by the plant, caesium is readily redistributed through the plant. Relationships between air concentrations and subsequent concentrations of ^{137}Cs in pasture plants have been discussed by Hawthorne et al. [H4] and Pelletier and Voilleque [P2].

289. Caesium may enter plants by plant base absorption before becoming fixed in soil. Thus, retention of ^{137}Cs in the root mat of pastures may allow ^{137}Cs to be relatively more available to plants for a period of a year or more [U3]. A high level of organic matter in soil can enhance the absorption of caesium by plants [B1]. Sorption of organic molecules on clay surfaces prevents the retention of caesium and also of potassium by these minerals. Thus, for permanent pastures in temperate regions, the frequent high organic matter content of the upper soil layer allows shallow rooted grass to absorb ^{137}Cs relatively more freely for a somewhat more extended period following deposition. Mushrooms have been shown to concentrate very effectively ^{137}Cs from soil [G2, M1], which may be associated with the highly organic areas of growing.

290. Uptake to plants of ^{137}Cs from soil low in available potassium may be somewhat increased. The addition of potassium may decrease absorption of ^{137}Cs in this case; however, this has no effect when the available potassium is high [N5, F4]. Root uptake of caesium is in general included in the range 0.01 to 1, which is the ratio of caesium concentrations in the dry plant material to that in dry soil [M3].

3. Transfer to milk

291. The fractional amount of ^{137}Cs transferred into milk is slightly greater than that of potassium. It has been shown that some 10% of orally ingested ^{137}Cs is secreted into the milk of dairy cows, corresponding to 1.3 to 1.5% of the amount ingested per litre of milk [G1, I1, L2]. The transfer of fallout ^{137}Cs in field conditions has been found to be somewhat less, ranging from 0.25 to 0.86% of intake per litre of milk [P2, M1, S4].

4. Transfer to meat

292. A correlation between ^{137}Cs concentration in beef and in milk has been noted, the ratio of concentrations in meat (Bq/kg) and in milk (Bq/l) averaging about 4 for production in

the same locality [L5, E1, J2]. This would correspond to a transfer of about 4% of the daily intake per kilogram of meat [F5]. Equilibrium conditions are reached in about 30 days in the cow [F5]. It cannot generally be expected that there will be a useful relationship between ^{137}Cs in meat and milk, since animals produced for the two purposes are frequently provided with different diets and are reared in different areas.

5. Transfer to diet

293. For general contamination situations, as for global fallout radioactivity, the main contributions to dietary intake of ^{137}Cs are generally from grain products, meat and milk. Fruit and vegetables contribute much smaller amounts of ^{137}Cs . This has been the pattern for western diets, such as Denmark [A1] and the United States [G4]. In Japan the main contribution has been from cereals [U1]. For all foods the transfer from a specific deposited amount seems to be rapid, being essentially completed within the first two years after deposition.

294. Marine food chains are of secondary importance in contributing to dietary intake of fallout ^{137}Cs , even in countries where fish is widely consumed. In Japan between 1966 and 1971, only about 8% of the ^{137}Cs in diet came from the consumption of fish products [U1]. As the contributions from other foods decrease, however, the relative contribution from fish can increase. During 1976 in Denmark, 15% of the ^{137}Cs intake was attributed to fish [A2]. It is possible that individuals consuming large amounts of freshwater fish may acquire ^{137}Cs burdens several times greater than individuals eating more diversified diets [G3].

295. The transfer of ^{137}Cs from deposition to diet has been studied quantitatively using the following transfer function between deposition and diet components in regression analyses of reported data [U8]:

$$C_j(n) = b_1^j f(n) + b_2^j f(n-1) + b_3^j \sum_{m=1}^{\infty} f(n-m) e^{-\mu m}$$

where $C_j(n)$ is the concentration of ^{137}Cs in the diet component j during the year (n) in mBq/gK ; $f(n)$ is the deposition density of ^{137}Cs in the current year (n) and in previous years $(n-1)$ in Bq/m^2 ; b_n^j are the proportionality factors, and μ is the decay constant accounting for radioactive decay and reduced availability of deposition in all previous years. The first term of the equations is the

rate term giving the contribution to dietary level from the current year's deposition amount. The second term is the long term contribution expressing separately the contribution from the previous year's deposition including storage of foods by market practices. The third term gives the contribution from the cumulative deposit of ^{137}Cs in soil.

296. The quotient of the time-integrated concentration of ^{137}Cs in diet and the integrated deposition density defines the transfer factor P_{23} . The integrals are replaced by summations if, as is usually the case, the relevant quantities are assessed over discrete intervals of time, such as annual averages

$$P_{23} = \frac{\sum_{n=1}^{\infty} C(n)}{\sum_{n=1}^{\infty} f(n)}$$

The transfer factor is usually expressed in mBq a/gK per Bq/m². It may be evaluated for total diet or for dietary components.

297. Using the model described above, the evaluation of the transfer factor reduces to

$$P_{23} = b_1 + b_2 + b_3 \sum_{m=1}^{\infty} e^{-\mu m} = b_1 + b_2 + b_3 \frac{e^{-\mu}}{1-e^{-\mu}}$$

The parameters of the transfer function, obtained by regression fits of the $^{137}\text{Cs}/\text{K}$ quotients in milk, dietary components and total diet from several countries have been reported by UNSCEAR [U8]. Some of these results are given in Table VII.3.

298. The lowest values for the transfer factor for milk are obtained for the U.S.A., Denmark and the U.K. Most of the transfer of ^{137}Cs deposition to milk is from direct deposition. Less than 15% of the transfer is from uptake of ^{137}Cs from soil. Intermediate values of P_{23}^{milk} are obtained from the USSR and Argentina. Transfer from direct deposition is increased and uptake from soil is more significant. The highest values of P_{23}^{milk} are obtained for New Zealand, Australia, Norway and the Faroe Islands. A high value has also been reported for Finland, which is 15.8 mBq a (gK)⁻¹ per Bq m⁻² [C2]. These results can be explained by efficient transfer of direct deposition and soil conditions which allow only low fixation of ^{137}Cs .

Table VII.3
Parameters of the transfer functions between deposition density and ¹³⁷Cs/K in milk

Parameter $\frac{a}{a'}$	U.S.A. 1973	Denmark 1976	United Kingdom 1977	USSR 1972	Argentina 1976	New Zealand 1977	Australia 1973	Norway 1976	Faroe Islands 1976
b ₁	2.3	2.2	1.7	4.4	3.2	6.8	7.3	3.5	6.8
b ₂	1.3	1.2	1.6	0.2	0.0	0.9	4.8	2.4	5.5
b ₃	0.0	0.01	0.04	0.3	1.7	3.1	0.3	2.5	2.8
μ	0.05	0.03	0.07	0.2	0.3	0.5	0.2	0.2	0.2
P ₂₃	3.4	3.7	3.9	6.2	7.5	13.0	13.4	15.6	27.5

$\frac{a}{a'}$ The unit for parameters b₁, b₂, b₃ is mBq a(gK)⁻¹ per Bq m⁻².

The unit for parameter μ is a⁻¹.

The unit for the transfer factor P₂₃ is mBq a(gK)⁻¹ per Bq m⁻².

299. The range of values of transfer factors for milk, 3.4 to 27.5 mBq a (gK)⁻¹ per Bq m⁻², indicates that it is not easy to specify a typical transfer situation. To estimate the ¹³⁷Cs transfer to milk in a particular region requires some indication of soil and pasture conditions.

300. Using the comprehensive data from Denmark of ¹³⁷Cs in diet [A2], values of the parameters of the transfer function of various components of diet and for the total diet have been determined. For most foods, the major contribution to the value of the transfer factor comes from direct deposition. Only a small transfer is attributed to uptake from soil. The values of the parameters for the fit to total diet data are: $b_1 = 1.6$, $b_2 = 2.2$, $b_3 = 0.04$ mBq a(gK)⁻¹ per Bq m⁻², $u = 0.11$ a⁻¹ and $P_{23} = 4.1$ mBq a(gK)⁻¹ per Bq m⁻². Short-term transfer to diet is thus estimated to comprise 93% of the total transfer, $(b_1 + b_2)/P_{23}$.

301. The general pattern of ¹³⁷Cs transfer to diet can be expected to be similar to that for Denmark, although increased transfer can occur in specific areas, depending on soil conditions or particular consumption habits. UNSCEAR has used a rounded value of P_{23} of 4 mBq a(gK)⁻¹ per Bq m⁻² to assess the dose commitments from ¹³⁷Cs for the world population [U8].

6. The lichen-caribou-man foodchain

302. A terrestrial situation which allows much greater than usual transfer of caesium to man is the lichen-caribou-man foodchain. Caesium depositing on lichens is retained quite effectively. There is a very slow decrease in activity with time, approximately 5 to 10% of the ¹³⁷Cs being eliminated annually [U5]. Lichens provide the food base for grazing caribou and reindeer during the winter. A proportionality factor of ¹³⁷Cs in lichen to that in reindeer meat in northern Sweden during the winter averaged 4.9 ± 0.4 [L4]. In summer also grass and herbaceous plants are consumed by the animals, and therefore the ¹³⁷Cs levels in meat show marked seasonal variation.

303. High concentrations of ¹³⁷Cs arise in Lapp and Eskimo populations who eat the caribou and reindeer meat. Levels of 740-1300 Bq per kg of body weight were observed in individuals during 1964 from fallout [H1, M4], about a factor of 100 greater than burdens of individuals from temperate northern hemisphere regions. Other fallout radionuclides, such as ⁵⁴Mn and ⁵⁵Fe, and the natural isotopes ²¹⁰Pb-²¹⁰Po are also concentrated along the lichen-caribou-man foodchain.

7. Aquatic behaviour

304. Caesium in the aquatic environment is strongly adsorbed by suspended particulate materials, especially clays. Therefore, the amount of caesium in the soluble phase decreases with increasing suspended solid concentrations. Potassium is also sorbed, but to a much less degree.

305. Caesium levels in fish are inversely related to the potassium content of the water [K1]. Because of the high concentration of potassium in the ocean, the transfer of ^{137}Cs to fish is of primary concern in the freshwater environment. The activity of fresh water fish may be 100 times that of ocean fish, given the same caesium concentrations in water.

306. The low mineral content of fresh water also enhances the absorption of ^{137}Cs by aquatic plants. Aquatic plants from fresh water areas, which are sometimes important in cattle feed, may have increased levels of ^{137}Cs compared to ^{137}Cs which may have deposited on nearby pasture ground.

307. Caesium in aquatic animals is accumulated primarily from the food chain. Absorption efficiency of potassium and caesium from food is high. In animals the excretion rate of potassium is about 3 times larger than that of caesium. As a result, the caesium concentration per unit amount of potassium in tissues increases by a factor of about 3 with each trophic level [P1].

308. The food web also accumulates caesium from suspended and bottom sediments. Filter feeders may accumulate caesium adsorbed to particulate matter. Benthic invertebrates obtain caesium absorbed to ingested bottom sediments. Fish ingest those invertebrates and also some sediment particles along with the prey. Absorption efficiency in the fish depends on the caesium fixation ability of the sediment minerals.

309. The concentration factors (ratio of concentration in organism to that in the water) for caesium must be related to the potassium concentration in the water and to the turbidity. From a literature review of values for fresh water systems [V1], suggested values of the concentration factors are 1000 for algae and plants, molluscs and invertebrates in all waters and $5000/K_w$ and $1500/K_w$ for non-piscivorous and piscivorous fish, respectively, in clear waters, where K_w is the stable potassium concentration of water in $\mu\text{g/g}$. The factors for fish are a factor of 5 less in turbid waters

(> 50 µg/g suspended solids). The concentration factors for caesium in the ocean are 10 for algae and molluscs, 30 for fish and 50 for molluscs [F6].

D. TRANSFER TO MAN

1. Absorption and distribution in tissues

310. As a general rule, caesium compounds are soluble in body fluids. Intestinal absorption is complete (100%) under experimental conditions [R2, S3], but from normal diets is probably less efficient, ranging from 50 to 80% [F5]. In man caesium is secreted into the gastrointestinal tract, between the stomach and small intestine, and is readily reabsorbed [I2]. One basis for therapeutic treatment in internal contamination cases is to administer solutions of Prussian blue, which binds with caesium in the gastrointestinal tract preventing reabsorption [I2, D1].

311. Caesium migrates rapidly into cells of the body following intake and becomes relatively uniformly distributed in soft tissues [R2, L3, R4]. The metabolism in mothers and infants has also been studied. There seems to be no placental discrimination, as the newborn has ^{137}Cs concentrations about equal to that of the mother [B3].

312. Concentrations of caesium and potassium are low in fat tissues. Therefore, for equal ^{137}Cs concentrations in intake, the concentrations of ^{137}Cs (Bq per kg body weight) in males are higher than in females, due to the higher average proportion of fat tissue in the female body. However, a difference is also expected due to longer retention time of caesium in males. Expressed in Bq ^{137}Cs per gK, the difference between males and females is somewhat reduced.

313. It has been inconclusive for some time whether ^{137}Cs concentrates in bone. One study reported that the concentration of ^{137}Cs in rib bones, which were free of muscle but not of marrow, was comparable to the concentration in soft tissues [Y2]. The results were variable, and subsequent studies both did and did not confirm these results [A3, H3, N4]. From the results of a recent study it appears that caesium associated with bone is present in the marrow portion with only slight uptake by the hard tissue [H2].

314. A slower turnover of caesium in bone could allow concentrations in bone to lag behind those in tissue, causing higher relative concentrations in bone during periods of

decreasing intake. A longer retention half-time would eventually be noted in whole-body measurements. However, such a component has not been identified in over 1000 days of measurement following an acute intake case [R4].

2. Retention half-time

315. A great many investigations of the biological half-time in man have been conducted. Many of the references are collected in the discussion by Lloyd [L6]. The half-time in man varies considerably, depending on age and other factors. The half-time is less in women than in men, and the half-time in children and infants is less than in adults. Pregnant women have shorter caesium half-times than in their non-pregnant conditions. Table VII.4 shows the summary of ^{137}Cs retention half-time reported recently by the NCRP [N1], using the data of Lloyd et al. [L7] and Zundel et al. [Z1].

Table VII.4

Retention half-time of ^{137}Cs in the human body
{N1}

Subjects	Number	Age	Half-time (d)
Men	26	23-55 a	105 ± 25
Women	15	20-51 a	84 ± 20
Pregnant women	24	16-39 a	49 ± 16
Children	7	5-17 a	57 ± 20
Infants	5	17-143 d	19 ± 8

316. The biological half-time for caesium in man can be considered a function of age for juveniles and of sex for adults, but it is not determined by body mass [L6]. Half-time and body mass may, however, be dependent on some other common factors. The rate of caesium turnover may be under hormonal influence or control or may reflect the general metabolic rate [L6].

317. Shorter half-time components of ^{137}Cs in man have been reported, including one of only 2 to 3 hours [N3]. In general, two components of the ^{137}Cs half-time in man have been established: a small fraction (10 to 15%) excreted with

a short half-time (1 to 1.5 days) and the remainder excreted more slowly (50 to 150 days) [R1, R3]. The ICRP suggests representative values of retention of 10% with a half-time of 2 days and 90% with a half-time of 110 days. Integral retention is 143 Bq d per Bq intake, contributed almost entirely by the long-term component.

3. Transfer factor

318. The value of the transfer factor P_{34} relating concentrations of ^{137}Cs in diet and man can be derived from the integral retention by dividing by the potassium content of the body (140 g) and multiplying by the daily potassium intake (3.3 g d^{-1}) [I5]. The result is $3.4 \text{ Bq a (gK)}^{-1}$ in man per Bq a (gK)^{-1} in diet.

319. The relatively short biological half-time of caesium in the body makes it possible to assess the transfer factor P_{34} from the measured $^{137}\text{Cs}/\text{K}$ quotients in diet and man integrated over a few years. Using this procedure, an average value of 3 Bq a (gK)^{-1} per Bq a (gK)^{-1} diet is derived [U6, U7, U8].

E. DOSIMETRY

1. Dose per unit intake

320. The dose from ^{137}Cs in tissue is due to the beta particles from ^{137}Cs decay and to the photon, x rays, and conversion and Auger electrons from decay of the daughter, $^{137\text{m}}\text{Ba}$. A portion of the photon energy will escape from the body, depending on the body size. Calculations have been performed for uniform distributions of ^{137}Cs in the body for a range of proportions and masses corresponding to infants, children and adults [N1, F1].

321. The average dose rate within the body for a uniform ^{137}Cs concentration of one Bq per kg body weight is about 3.5 nGy/d from beta particles in both adults and infants plus 3.2 nGy/d from photons in the adult and about 1.6 nGy/d in infants. The totals are $6.7 \text{ nGy/d per Bq/kg}$ in the adult and $5.1 \text{ nGy/d per Bq/kg}$ in the infant.

322. For 140 gK in the 70 kg adult body, the dose rate corresponding to 1 Bq ^{137}Cs per gK would be

$$0.0067 \frac{\mu\text{Gy d}^{-1}}{\text{Bq}(\text{kg})^{-1}} \cdot \frac{140 \text{ gK}}{70 \text{ kg}} \cdot \frac{365 \text{ d}}{\text{a}} = 4.9 \frac{\mu\text{Gy/a}}{\text{Bq/gK}}$$

Spiers [S2] also obtains this result and estimates the corresponding dose rate in a child weighing 8 kg of

$$4.1 \quad \frac{\mu\text{Gy/a}}{\text{Bq/gK}}$$

323. UNSCEAR [U6, U7, U8] assessed the transfer factor between tissue and dose, P_{45} , as $4.9 \mu\text{Gy per Bq a (gK)}^{-1}$. The value is nearly independent of age, being only slightly less for the child. For a single uptake of ^{137}Cs , the integral retention per unit intake is $143 \text{ Bq d per Bq intake}$ (paragraph 317). The average absorbed dose in the body per unit intake is thus

$$0.0067 \frac{\mu\text{Gy/d}}{\text{Bq/kg}} \quad 143 \frac{\text{Bq d}}{\text{Bq}} \quad \frac{1}{70 \text{ kg}} = 1.4 \cdot 10^{-8} \text{ Gy per Bq intake.}$$

324. This result applies to intake by ingestion. For inhalation the value is less by a factor of 0.63 (for particles of $1 \mu\text{m}$ size) due to fractional deposition in the lungs of inhaled amounts. Following inhalation there is only a short retention in the lung (half-time = 0.5 d) for the soluble caesium compounds and a small dose primarily from the beta particles before the ^{137}Cs becomes distributed throughout the body.

2. Dose per unit release

(a) Nuclear explosions

325. The dose commitment via the ingestion pathway from ^{137}Cs released by nuclear explosions can be assessed from the sequential product of transfer factors

$$D^c = P_{23} P_{34} P_{45} F$$

where F is the integrated deposition density. The values of the transfer factors as derived above are: $P_{23} = 4 \cdot 10^{-3} \text{ Bq a (gK)}^{-1}$ per Bq m^{-2} , $P_{34} = 3 \text{ Bq a (gK)}^{-1}$ per Bq a (gK)^{-1} , and $P_{45} = 4.9 \cdot 10^{-6} \text{ Gy per Bq a (gK)}^{-1}$. The dose commitment from ^{137}Cs ingestion per unit widespread deposition density, such as from nuclear explosions, is thus $6 \cdot 10^{-8} \text{ Gy per Bq m}^{-2}$.

326. The total amount of ^{137}Cs released to the environment by nuclear tests, $9.6 \cdot 10^{17} \text{ Bq}$, has given a population-weighted integrated deposition density of 3100 Bq m^{-2} in the world as a whole [U8]. The world population is $4 \cdot 10^9$. With

these values, the collective dose commitment per unit activity of ^{137}Cs released is estimated to be $8 \cdot 10^{-13}$ man Gy per Bq (ingestion).

327. The dose commitment via the inhalation pathway is determined from the integrated concentration of ^{137}Cs in air, which is estimated from the integrated deposition density (Bq m^{-2}) divided by the deposition velocity (m s^{-1}). As for ^{90}Sr , the average deposition velocity can be taken to be 2 cm s^{-1} . Thus, 1 Bq m^{-2} integrated deposition density corresponds to $5.8 \cdot 10^{-4} \text{ Bq d m}^{-3}$ in air. With the above estimates of integrated deposition density, total amount of ^{137}Cs released by nuclear tests, world population, breathing rate ($22 \text{ m}^3 \text{ d}^{-1}$) and dose per unit intake ($8.8 \cdot 10^{-9} \text{ Gy Bq}^{-1}$), the collective dose commitment per unit activity of ^{137}Cs released is estimated to be $1 \cdot 10^{-15}$ man Gy per Bq (inhalation).

328. For the external exposure pathway it is assumed that the deposited ^{137}Cs becomes exponentially distributed in soil with a mean depth of 3 cm. This gives a dose rate in air of $8.9 \cdot 10^9 \text{ Gy per Bq m}^{-2}$ [B2]. The mean life of ^{137}Cs in soil is 43.5 years, determined by its radioactive decay. The dose to air from the integrated deposition density is, thus, $3.9 \cdot 10^{-7} \text{ Gy per Bq m}^{-2}$. The dose to tissue is determined by a factor of 0.8 to account for the change of material (air to tissue) and back-scatter and shielding afforded by other tissues of the body and a factor of 0.4 to account for building shielding and time spent indoors [U8]. The combined factor is $0.8 \times 0.4 = 0.32$. The transfer factor P_{25} relating integrated deposition of ^{137}Cs to tissue dose is thus $3.9 \cdot 10^{-7} \times 0.32 = 1.2 \cdot 10^{-7} \text{ Gy per Bq m}^{-2}$. Using the above values of integrated deposition density, total amount of ^{137}Cs released, and world population, the collective dose commitment per unit activity of ^{137}Cs released is estimated to be $1.6 \cdot 10^{-12} \text{ Gy per Bq}$ (external exposure).

(b) Nuclear installations

329. The contribution of the inhalation pathway to the collective dose commitment for effluent releases can be estimated from the integrated concentration of ^{137}Cs in air. This can be derived from a dispersion formula or from an estimate of the deposition velocity. In the latter case, the integrated concentration in air is the total amount of ^{137}Cs released per unit area of the deposition region divided by the deposition velocity, v_d . The population affected is the population density δ_N times the area of the deposition

region. The areal dependence is removed by the product of these quantities. The collective dose commitment per unit activity released is given by the expression

$$S_1^c = I \delta_N \phi / v_d$$

where I is the individual intake rate of air and ϕ is the dose per unit activity of ^{137}Cs inhaled.

330. Using a deposition velocity of 0.5 cm s^{-1} for near surface releases, a population density of 25 man km^{-2} , an air intake rate of $22 \text{ m}^3/\text{d}$ and the dosimetric factor of $8.8 \cdot 10^{-9} \text{ Gy per Bq intake}$, the collective dose commitment for the inhalation pathway per unit activity released is estimated to be $1 \cdot 10^{-14} \text{ man Gy/Bq}$.

331. The contribution of the ingestion pathway from airborne effluents to the collective dose commitment per unit activity released, S_1^c , can be assessed by the expression

$$S_1^c = P_{23} P_{34} P_{45} \delta_N$$

Using the values for the transfer factors given previously, and assuming a constant population density of 25 man km^{-2} in the region of deposition, the collective dose commitment for the ingestion pathway per unit activity released is estimated to be $2 \cdot 10^{-12} \text{ man Gy/Bq}$. This value assumes that food is locally produced and that the production suffices for the population density under consideration. It also applies for the case of ^{137}Cs becoming relatively rapidly fixed in soil ($P_{23} = 4 \text{ mBq a [gK]}^{-1} \text{ per Bq m}^{-2}$). For other types of soil conditions or special consumption patterns and also for other population densities, the estimate should be adjusted accordingly.

332. For the external exposure pathway the transfer factor P_{25} relating integrated deposition of ^{137}Cs in soil to the tissue dose has been assessed above with regard to nuclear explosions to be $1.2 \cdot 10^{-7} \text{ Gy per Bq m}^{-2}$. This value is of general applicability. Similarly to the ingestion pathway, the collective dose commitment per unit activity released, S_1^c , can be assessed by the expression

$$S_1^c = P_{25} \delta_N$$

Assuming a population density of 25 man km^{-2} , the collective dose commitment from external exposure per unit activity released is estimated to be $3 \cdot 10^{-12} \text{ man Gy/Bq}$.

333. For aquatic ingestion pathways from the input of ^{137}Cs to water bodies the collective dose commitment, normalized per unit activity released, can be estimated [U8], using the expression

$$S_1^C = \frac{\sum_k N_k I_k f_k \varphi}{V(\lambda + 1/\tau)}$$

where V is the volume of the receiving waters, τ is the turnover time of receiving waters, λ is the decay constant of ^{137}Cs , N_k is the number of individuals exposed by pathway k , I_k is the individual consumption rate of pathway item k , f_k is the concentration factor for the consumed item in pathway k , and φ is the collective dose per unit activity ingested collectively by the exposed group.

334. The quantity $\frac{1}{V(\lambda+1/\tau)}$ is the infinite time integral of the water concentration per unit of activity released, while the quantity multiplied by f_k is the infinite time integral of the concentration in the consumed item k . For radionuclide inputs into small volumes of water, the concentrations in water and in fish will be high, but the population which can be served with drinking water or by fish consumption will be limited. For inputs into larger volumes of water, the concentrations will be smaller, but the populations involved will be correspondingly larger. It is reasonable, therefore, to assume as a first approximation that the quantities N_k/V are relatively constant, independent of V . The values for these quantities as well as values for the other parameters of the above expression have been extensively discussed by UNSCEAR [U8].

335. A listing of the values used in the assessments presented by UNSCEAR is given below [U8]:

<u>Parameter</u>	<u>fresh water</u>	<u>sea water</u>
1. τ , turnover time of receiving water	10 a	1 a
2. Correction factor for sediment removal	0.3	1.0
3. $\frac{V}{N}$, water utilization factor	$3 \cdot 10^7$ l/man	$3 \cdot 10^9$ l/man
4. f_k , concentration factor for item k		
drinking water	0.2	
fish	400	30
shellfish		30

<u>Parameter</u>	<u>fresh water</u>	<u>sea water</u>
5. Ik, consumption rate for item k		
drinking water	440 l/a	
fish	1 kg/a	6 kg/a
shellfish		1 kg/a

(c) Summary

336. Table VII.5 summarizes the values obtained above for the collective dose commitments per unit of ^{137}Cs activity released in airborne and liquid effluents. These are also the values of the collective effective dose equivalent commitments with Sv replacing Gy, since the quality factor is one and the dose is assumed to be uniform in all tissues. The largest collective dose commitments result from airborne discharges due to the external exposure and ingestion pathways. These estimates are for a generalized release situation, and substantial variations could be expected in site-specific cases.

Table VII.5

Summary of collective dose commitments per unit ^{137}Cs activity released
[10^{-1} man Gy per Bq]

<u>All tissues</u>	
<u>Nuclear explosions</u>	
External exposure	160
Ingestion	80
Inhalation	0.1
<u>Nuclear installations</u>	
Release to air <u>a/</u>	
External exposure	300
Ingestion	200
Inhalation	1
Release to fresh water	
Fish	50
Drinking water	10
Release to salt water	
Fish	0.08
Shellfish	0.02

a/ Assumes population density of 25 man km⁻².

F. REFERENCES

- A1 Aarkrog, A. Prediction models for strontium-90 and caesium-137 levels in the human food chain. *Health Phys.* 20: 297 (1971).
- A2 Aarkrog, A. and J. Lippert. Environmental radioactivity in Denmark in 1976. Risø National Laboratory report No. 361 (1977).
- A3 Anderson, R.W. and P.F. Gustafson. Concentration of caesium-137 in human rib bone. *Science* 137: 668 (1962).
- B1 Barber, D.A. Influence of soil organic matter on the entry of caesium-137 into plants. *Nature* 204: 1326-1327 (1964).
- B2 Beck, H.L. and G. de Planque. The radiation field in air due to distributed gamma-ray sources in the ground. U.S. Atomic Energy Commission report HASL-195. New York, 1968.
- B3 Bengtsson, L.G., Y. Naversten and K.G. Svensson. Maternal and infantile metabolism of caesium. p. 21-32 in *Assessment of Radioactivity in Man. Proceedings of a symposium, Heidelberg, May 1964.* International Atomic Energy Agency publication STI/PUB/Vol. 2. Vienna, 1964.
- C1 Cambray, R.S., E.M.R. Fisher, K. Playford et al. Radioactive fallout in air and rain: results to the end of 1977. United Kingdom Atomic Energy Authority report AERE-R9016 (1978).
- C2 Castrén, O. UNSCEAR transfer coefficients P-23 (milk) and P-234 for 90-Sr and 137-Cs in Finland. p. 73-76 in *Studies on Environmental Radioactivity in Finland 1971-1975.* Institute of Radiation Protection report STL-A21. Helsinki, 1977.
- C3 Crouch, E.A.C. Fission product yields from neutron induced fission. *Atomic Data and Nuclear Data Tables*, Vol. 19, No. 5 Academic Press, New York, 1977.
- D1 Ducouso, R., A. Causse and C. Pasquier. Comparative effects of acetazolamide and Prussian blue on 137-Cs retention in the rat. *Health Phys.* 28: 75-78 (1975).
- E1 Ellis, F.B. and B.T. Barnes. Relationship between the concentration of caesium-137 in beef and milk. p. 77-78 in *Agricultural Research Council Radiobiological Laboratory annual report ARCRL-14* (1965).

- F1 Feely, H.W. World-wide deposition of strontium-90 through 1977. p. 1.19-1.41 in Environmental Measurements Laboratory environmental quarterly report EML-344. New York, 1978.
- F4 Fredriksson, L., B. Eriksson and B. Rasmunson. Plant uptake of 90-Sr and 137-Cs from soils, Vol. 18. p. 449-470 in Proceedings of the Second International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958.
- F5 Fredriksson, L., R.J. Garner and R.S. Russell. Caesium-137. p. 317-352 in Radioactivity and Human Diet (R.S. Russell, ed.). Pergamon Press, 1966.
- F6 Freke, A.M. A model for the approximate calculation of safe rates of discharge of radioactive wastes into marine environments. Health Phys. 13: 743-758 (1967).
- G1 Gragle, R.G. Uptake and excretion of caesium-134 and potassium-42 in lactating dairy cows. J. Dairy Sci. 44: 352-357 (1961).
- G2 Grueter, H. Radioactive fission product 137-Cs in mushrooms in West Germany during 1963-1970. Health Phys. 20: 655-656 (1971).
- G3 Gustafson, P.F. Comments on radionuclides in aquatic ecosystems. p. 853-858 in Radioecological Concentration Processes (B. Aberg and F. Hungate, eds.). Pergamon Press, 1967.
- G4 Gustafson, P.F. and J.E. Miller. The significance of 137-Cs in man and his diet. Health Phys. 16: 167-183 (1969).
- H1 Hanson, W.C. Radioecological concentration processes characterizing arctic ecosystem. p. 183-191 in Radioecological Concentration Processes (B. Aberg and F. Hungate, eds.). Pergamon Press, 1967.
- H2 Hardy, E.P. On 137-Cs and 90-Sr in bone. p. I.64-I.69 in Health and Safety Laboratory Fallout Program Quarterly Summary report HASL-278. New York, 1974.
- H3 Harrison, G.E., A. Sutton, K.H. Edwards et al. Concentrations of radioactive and stable caesium in bone and soft tissues. Br. J. Radiol. 36: 745-748 (1963).

- H4 Hawthorne, H.A., S.D. Zellner, L.L. Eberhardt et al. 137-Caesium cycling in a Utah dairy farm. Health Phys. 30: 447-464 (1976).
- I1 Ilin, D.I. and Y.I. Koskalev. On the metabolism of caesium, strontium and a mixture of beta-emitters in cows. J. Nucl. Energ. 5: 413-420 (1957).
- I2 International Atomic Energy Agency. Manual on early medical treatment of possible radiation injury. International Atomic Energy Agency publication Safety Series No. 47 and IAEA/STI/PUB/506. Vienna, 1978.
- I4 International Atomic Energy Agency. Power reactors in member states. IAEA, Vienna, 1980.
- I5 International Commission on Radiological Protection. Report of the task group on reference man. ICRP publication 23. Pergamon Press, 1975.
- J2 Johnson, J., G.M. Ward and A.H. Dal. Comparisons of 137-Cs levels in feed and meat of cattle fed on pasture and in the dry lot. Health Phys. 10: 612 (1964) (Abstract).
- K1 Koehmainen, S., E. Häsänen and J.K. Miettinen. 137-Cs in fish, plankton and plants in Finnish lakes during 1964-1965. p. 913-919 in Radioecological Concentration Processes (B. Aberg and F.P. Hungate, eds.). Pergamon Press, 1967.
- K2 Krumholz, L., E. Goldberg and H. Boroughs. Ecological factors involved in the uptake, accumulation and loss of radionuclides by aquatic organisms in The Effects of Atomic Radiation on Oceanography and Fisheries. National Academy of Sciences, National Research Council Publication 551. Washington, 1957.
- L1 Leifer, R. and L. Toonkel. Updating stratospheric inventories to April 1977. p. I.3-I.14 in Environmental Measurements Laboratory environmental quarterly report EML-334. New York, 1978.
- L2 Lengemann, F.W. and R.A. Wentworth. The transfer coefficient of 137-Cs into cows milk as related to the level of milk production. Health Phys. 34: 720-722 (1978).

- L3 Lidén, K. The metabolism of caesium. p. 33 in Assessment of Radioactivity in Man. Proceedings of a symposium, Heidelberg, May 1964. International Atomic Energy Agency publication STI/PUB/84/Vol. 2. Vienna, 1964.
- L4 Lidén, K. and M. Gustafsson. Relationship and seasonal variation of ^{137}Cs in lichen, reindeer and man in northern Sweden, 1961 to 1965. University of Helsinki, Department of Radiochemistry annual report 1965. Helsinki, 1966.
- L5 Lindell, B. and A. Magi. The occurrence of ^{137}Cs in Swedish food, especially dairy milk and in the human body after the nuclear test explosions in 1961 and 1962. Ark. Fys. 29: 69-96 (1965).
- L6 Lloyd, R.D. Caesium-137 half-times in humans. Health Phys. 25: 605 (1973).
- L7 Lloyd, R.D., W.S. Zundel, C.W. Mays et al. Short caesium half-times in patients with muscular dystrophy. Nature 220: 1029-1031 (1968).
- M1 Marei, A.N., R.M. Barkhudarov, N.J. Novikova et al. Effect of natural factors on caesium-137 accumulation in the bodies of residents in some geographical regions. Health Phys. 22: 9-15 (1972).
- M2 Marei, A.N., R.M. Barkhudarov and N.J. Novikova. Radiation safety aspects of the geochemical areas with increased transfer of caesium-137. Manuscript No. 0020-R-4.
- M3 Menzel, R.G. Soil-plant relationships of radioactive elements. Health Phys. 11: 1325-1332 (1965).
- M4 Miettinen, J.K. and E. Hasanen. ^{137}Cs in Finnish Lapps and other Finns in 1962-1966. p. 221-231 in Radioecological Concentration Processes (B. Aberg and F. Hungate, eds.). Pergamon Press, 1967.
- M5 Moiseev, A.A. and P.U. Ramzaev. Caesium-137 in the biosphere. Atomizdat, Moscow, 1975 (in Russian).
- N1 National Council on Radiation Protection and Measurements. Caesium-137 from the environment to man: metabolism and dose. National Council on Radiation Protection and Measurements report No. 52. Washington D.C., 1977.

- N2 National Council on Radiation Protection and Measurements. A handbook on Radioactivity measurements procedures. National Council on Radiation Protection report No. 58. Washington, D.C., 1978.
- N3 Naversten, Y. and K. Lidén. Half-life studies of radiocaesium in humans. p. 79-87 in Assessment of the Radioactive Body Burdens in Man. Proceedings of a symposium, Heidelberg, May 1964. International Atomic Energy Agency publication STI/PUB/84/Vol. 2. Vienna, 1964.
- N4 Nay, U., W. Stahlhofen and A. Kaul. Distribution of caesium-137 in samples consisting of soft tissues, bone and bone marrow. p. 49-60 in Assessment of Radioactivity in Man. Proceedings of a symposium, Heidelberg, May 1964. International Atomic Energy Agency publication STI/PUB/84/Vol.2 Vienna, 1964.
- N5 Nishita, H., E.M. Romney and K.H. Larson. Uptake of radioactive fission products by crop plants. J. Agric. Food Chem. 9: 101-106 (1961).
- O1 Oak Ridge National Laboratory. Siting of fuel reprocessing plants and waste management facilities. Oak Ridge National Laboratory report ORNL-4451 (1970).
- P1 Pendleton, R.C., C.W. Mays, R.D. Lloyd et al. A trophic level effect on 137-Cs concentration. Health Phys. 11: 1503 (1965).
- P2 Pelletier, C.A. and P.G. Voilleque. The behaviour of 137-caesium and other fallout radionuclides on a Michigan dairy farm. Health Phys. 21: 777-792 (1971).
- R1 Richmond, C.R., J.E. Furchner and W.H. Langham. Long-term retention of radio-caesium by man. Health Phys. 8: 201-205 (1962).
- R2 Rosoff, B., S.H. Cohn and H. Spencer. Caesium-137 metabolism in man. Radiat. Res. 19: 643-654 (1963).
- R3 Rundo, J., J.I. Mason, D. Newton et al. Biological half-life of caesium in man in acute and chronic exposure. Nature 200: 188-189 (1963).
- R4 Rundo, J. The metabolism of biologically important radionuclides. VI. A survey of the metabolism of caesium in man. Br. J. Radiol. 37: 108 (1964).

- S1 Sherill, R.D., N.G. Sumerlin, J.N. Beck et al. Variation of the ratio of caesium-137 to strontium-90 in the atmosphere. *Health Phys.* 28: 335-340 (1975).
- S2 Spiers, F.W. *Radioisotopes in the human body: physical and biological aspects.* Academic Press, New York, 1968.
- S3 Stara, J.F. Tissue distribution and excretion of caesium-137 in the guinea pig after administration by three different routes. *Health Phys.* 11: 1195-1202 (1965).
- S4 Stewart, H.F., G.W. Ward and J.E. Johnson. Availability of fallout 137-Cs to dairy cattle from different types of feed. *J. Dairy Sci.* 48: 709 (1965).
- T1 Tamura, T. and D.G. Jacobs. Structural implications in caesium sorption. *Health Phys.* 2: 391-398 (1960).
- T2 Taylor, S.R. Abundance of chemical elements in the continental crust: a new table. *Geochim. Cosmochim. Acta* 28: 1273-1285 (1964).
- U1 Ueda, T., Y. Suzuki and R. Nakamura. Transfer of 137-Cs and 90-Sr from the environment to the Japanese population via the marine environment. p. 501-511 in *Dose Evaluation and Standards for Man and His Environment.* International Atomic Energy Agency publication STI/PUB/375. Vienna, 1974.
- U2 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Thirteenth Session, Supplement No. 17(A/3838). New York, 1958.
- U3 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Seventeenth Session, Supplement No. 16(A/5216). New York, 1962.
- U4 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Nineteenth Session, Supplement No. 14(A/5814). New York, 1964.
- U5 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-first Session, Supplement No. 14(A/6314). New York, 1966.

- U6 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. Official Records of the General Assembly, Twenty-fourth Session, Supplement No. 13(A/7613). New York, 1969.
- U7 United Nations. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, with annexes Volume I: Levels, Volume II: Effects. United Nations sales publication No.E.72.IX.17 and 18. New York, 1972.
- U8 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General Assembly, with annexes. United Nations sales publication No.E.77.IX.I. New York 1977.
- V1 Vanderploeg, H.A., D.C. Parzyck, W.H. Wilcox et al. Bioaccumulation factors for radionuclides in freshwater biota. Oak Ridge National Laboratory report ORNL-5002 (1975).
- Y2 Yamagata, N. and T. Yamagata. The concentration of 137-Cs in human tissues and organs. Bull. Inst. Publ. Health 9: 72-78 (1960).
- Z1 Zundel, W.S., F.H. Tyler, C.W. Mays et al. Short half-times of caesium-137 in pregnant women. Nature 221:89 (1969).

VIII. RADON

A. INTRODUCTION

337. Radon is element number 86, but the term generally refers to the isotope ^{222}Rn , the decay product of ^{226}Ra . Other isotopes of radon, ^{220}Rn and ^{219}Rn , are generally referred to by their historical names, thoron and actinon, respectively. Radon has a half-life of 3.82 days and decays by alpha-particle emission to a polonium isotope (^{218}Po), which by further decay through isotopes of lead, bismuth, polonium and thallium ends the uranium (^{238}U) decay chain with stable lead (^{206}Pb).

338. Radon is a chemically inert gas. It arises from decay of radium which occurs in soil and other common materials. Part of the radon produced diffuses into the surrounding environment.

339. The short-lived decay products of radon, called radon daughters, are ^{218}Po (3.05 min), ^{214}Pb (26.8 min), ^{214}Bi (19.7 min) and ^{214}Po ($1.6 \cdot 10^{-4}\text{s}$). They become largely attached to aerosols in air and if inhaled they are partly deposited in the human respiratory tract. The radiation doses caused by inhalation of radon daughters in air constitute the main part of the natural radiation dose to man. The radiation dose caused by radon itself is minor by comparison with that of radon daughters.

340. The levels of radon and radon daughters in air depend on the source and on the dilution in the air. The levels are normally higher indoors than outdoors. Reduced ventilation may cause radon released from building materials to build up in enclosed spaces. The radon levels may be very high in underground mines, particularly in uranium mines.

B. SOURCES

1. Outdoors

(a) Natural radon

341. The main source of radon outdoors is radium in the earth's crust. The concentration of uranium and radium in the ground varies with the types of rocks and minerals. The concentration of radium in rocks and soil is often (but not always) the same as that of uranium. Fractionated dissolution and transport of uranium-234 and/or radium can cause breaks in the uranium chain [S2].

342. The total amount of radium in the outer 10 km of the earth's crust is of the order of 10^{24} Bq. Most of the radon produced by decay of radium is physically attached to the radium-bearing material and only a small part diffuses out into the air. Other relatively less important sources of radon in air outdoors are plants, ground water, oceans, etc. The sources contributing to the total amount of radon are given in Table VIII.1. It is assumed that the radon exhalation rate from the land (soil) is $0.02 \text{ Bq m}^{-2} \text{ s}^{-1}$ and from the oceans is $70 \text{ } \mu\text{Bq m}^{-2} \text{ s}^{-1}$. The total production rate of radon is of the order of $10^{20} \text{ Bq a}^{-1}$; the equilibrium inventory in the atmosphere, determined from the total production rate divided by the decay constant (66.2 a^{-1}) is estimated to be $1.5 \cdot 10^{18} \text{ Bq}$.

Table VIII.1

Sources of radon in the global atmosphere

[H5]

Source	Radon production per year (Bq)
Soil	$9 \cdot 10^{19}$
Plants and ground water	$< 2 \cdot 10^{19}$
Oceans	$9 \cdot 10^{17}$
Houses ^{a/}	$3 \cdot 10^{16}$
Natural gas	$3 \cdot 10^{14}$
Coal	$2 \cdot 10^{13}$

^{a/} The value for houses is estimated in this document assuming 10^9 reference houses (see Table VIII.3). The true value may be between $5 \cdot 10^{15}$ - $10^{17} \text{ Bq a}^{-1}$.

(b) Mines and mine tailings

343. Sources of radon of local interest include the tailings from uranium and phosphate mining and milling and from geothermal power stations. The radon exhalation rate from tailings depends on the radium content of the tailings, on the emanation factor (fractional release of radon) and on the land reclamation (overburden). The radon exhalation rate from uncovered uranium tailings varies from $0.5 \text{ Bq m}^{-2} \text{ s}^{-1}$ or less to $10 \text{ Bq m}^{-2} \text{ s}^{-1}$ or more [S9, U4]. The thickness and area of the tailings per unit mass of uranium in the ore can also vary depending on the tailings engineering and therefore

the radon exhalation rate varies in the range of about 10-100 Bq s⁻¹ per MW(e)a [S9, U4]. By covering the tailings with a few meters of soil the radon exhalation rate is reduced by one to several orders of magnitude. The radon exhalation from tailings of phosphate mining and milling also depends on land reclamation and the type of ore. A range of exhalation rates of about 0.01-1 Bq m⁻² s⁻¹ has been reported [R1]. Geothermal power stations may cause radon releases into the air by releases of radon from the water which is depressurized at the surface. A radon release of the order of 10¹¹ Bq per MW(e)a has been reported [M3]. The radon release from coal-fired plants is 3 to 4 orders of magnitude less.

344. The sources of radon in underground spaces like mines are radium in the rock and minerals of the mine and radon in water. The total release of radon into the mine depends on many factors: the uranium-radium concentration of the ore; the number and size of cracks in the ore; the isolation of abandoned spaces; the radon concentration and amount of water; the isolation of water; and the ventilation system principles. By using a normal range of total ventilation rates (10-1000 m³ s⁻¹) and a normal radon concentration (0.1-10 kBq m⁻³) the range of radon release into the mine may be estimated at 1-10⁴ kBq s⁻¹. Since there is normally an inverse proportionality between radon concentration and ventilation rate, lower ventilation rates are more often related to higher radon concentrations and vice versa. There is also a radon release from the mine to outside air and it can be compared with the radon release from uncovered tailings. Mining of 100 tonnes uranium per year results in 5000-15000 m² of tailings per year depending on the percentage of uranium in the ore and the tailings engineering. The radon release will be in the range of 1-10 kBq s⁻¹. In non-uranium mines radon-rich water is often a significant source of radon to the mine [S5]. The radon concentration of the water may be in the range of 100-1000 kBq m⁻³.

2. Indoors

(a) Building materials

345. Radon in houses comes from building materials, the soil under the house, the water and the domestic gas. Radium concentrations in building materials have been investigated. The data indicate that some materials such as aerated concrete with alum shale and phospho-gypsum from sedimentary ores have

significantly higher radium concentrations than others and cause enhanced radon concentrations indoors. If these materials are excluded, the average concentrations of radium in building material is about 100 Bq kg^{-1} . Materials with low activity are wood, natural gypsum, sand and gravel. The radon exhalation rate from walls, floors and ceilings is dependent on the radium concentration, the emanation power, the diffusion coefficient in the material and the qualities and thickness of any applied sealant on the surfaces.

346. The radon exhalation rate from uncovered building materials varies by several orders of magnitude from about $10^{-6} \text{ Bq m}^{-2} \text{ s}^{-1}$ (gypsumboard, fiberboard, chipboard, bricks) to $(0.1-10) 10^{-3} \text{ Bq m}^{-2} \text{ s}^{-1}$ for concrete of different origins and qualities [J5, P3, S17, M8, W3]. The radon exhalation rate per Bq Ra/kg varies less, e.g., $(1.6 \pm 0.88) 10^{-5} \text{ Bq m}^{-2} \text{ s}^{-1}$ per Bq Ra/kg for some building materials in the Federal Republic of Germany [W3]. If the same values are normalized to an emanating power of 1%, the radon exhalation rate is $(4.4 \pm 1.9) 10^{-6} \text{ Bq m}^{-2} \text{ s}^{-1}$ per Bq Ra/kg per percentage emanating power. In the first case the standard deviation is 54% and in the last case it is 43%. By painting, plastering or application of wall-paper on the wall the radon exhalation may be reduced by less than a factor of 5 [W3, M8].

(b) Soil

347. The contribution of radon from the soil into a building depends on the thickness and tightness of the base structure. The exhalation from the soil is of the order of $10^{-2} \text{ Bq m}^{-2} \text{ s}^{-1}$ and a concrete floor in cellars should normally reduce the radon exhalation from soil into the building by a factor of 10 or more. Even so, radon from soil may contribute significantly to the radon concentrations in a house, particularly in the cellar and in wooden houses. In some areas houses are built on natural uranium deposits (Canada) [L2, K2], on phosphate-related land (Florida, U.S.A.) [U3, U13], on waste products from uranium industry (Colorado, U.S.A.) [C6], and on waste products from alum production from radium-rich alum shales (Sweden) [S22]. In these cases the radon exhalation rate may be several orders of magnitude higher than from normal soil.

(c) Water

348. Another source of radon in a house may be radon-rich water. The relative radon release depends on the use of water. Boiling and splashing of the water increase releases

and consequently the highest radon releases occur in washrooms, at shower-baths and in the kitchen during cooking. The resultant radon concentration in a house depends on the amount of water used, the volume of the house and the ventilation. Several studies have been made to estimate the relative significance of radon from water [P1, C2, N1], and a typical value of the air-to-water radon concentration quotient is about 10^{-4} . Measurements of radon in water are most often made in areas with suspected high concentrations because of uranium deposits and estimates of the weighted average radon concentration in water for a country are rare.

349. Population-weighted average concentrations of radon in drinking water have been estimated for a few countries and are found to be 40 kBq m^{-3} in Finland [A5, C3], 7 kBq m^{-3} in Sweden [S8] and $0.4\text{-}4 \text{ kBq m}^{-3}$ in the Federal Republic of Germany [M9]. The corresponding radon release into a house can be estimated by assuming a daily use of 500 l per person and 10-100% relative release from the water.

(d) Natural gas

350. Natural gas containing radon may also be a source of radon in houses. Gas is transported as purified gas in long transmission lines and distributed to the homes or bottled under pressure as propane for sale as liquified petroleum gas (LPG). The radon concentrations in natural gas at the production wells are found to vary from undetectable values up to about 40 kBq m^{-3} [U12, H6]. During supply, transit, storage and delivery the radon concentration decreases to an approximate average of the order of 1 kBq m^{-3} for both natural gas and LPG (in U.S.A.) [U12, B1, G1].

(e) Summary

351. The relative contribution of different radon sources to the total radon input in a house is estimated in Table VIII.2 with some typical values of radon concentration and releases. The radon in outside air is brought into the house by ventilation. The volume of the house is assumed to be 200 m^3 and the inner surface area 350 m^2 .

Table VIII.2
The relative significance of different radon sources
in a reference house

Source	Radon flux (Bq d ⁻¹)	Comments
Building material	70 10 ³	Emanation rate 2 10 ⁻³ Bq m ⁻² s ⁻¹
Water	4 10 ³	1000 l d ⁻¹ and 4 kBq m ⁻³ , 100% release
Outside air	9 10 ³	Radon concentration outdoors 0.004 kBq m ⁻³ ; ventilation rate 0.5 per hour
Natural gas	3 10 ³	
Liquified petroleum gas	0.2 10 ³	

C. BEHAVIOUR IN THE ENVIRONMENT

1. Release from soil

(a) Emanation

352. The mechanism of radon release from rock, soil and other materials is not very well understood and is probably not always the same. The main physical phenomena are recoil and diffusion of the radon atom through imperfections of the crystalline structures of the radium-bearing particle followed by a secondary diffusion, which depends on the porosity of the material [A1]. High porosity increases the diffusion rate. The release rate from a material depends also on its moisture content: if the moisture content is very low the radon release is decreased by the effect of re-adsorption of radon atoms on surfaces in the pores. If the moisture content increases slightly, the radon release increases up to a certain moisture content, above which the release of radon decreases again owing to a decreasing diffusion rate in water-filled pores [M7].

(b) Diffusion

353. Once radon has entered the air or water surrounding the emanating radium-bearing particle, it is transported by

diffusion, earth-mechanical and convective flow, percolation of rain water and flow of ground water. The diffusion mechanism can be expressed by the equation

$$C_x = C_0 \exp \{-x/\sqrt{(D/\lambda)}\}$$

where C_x is the radon concentration at distance x in air or water from the emanating surface; C_0 is the radon concentration at the surface; D is the diffusion coefficient (gas kinetic) and λ is the decay constant [G2]. The diffusion constant D is about $10^{-2} \text{ cm}^2 \text{ s}^{-1}$ in air and $10^{-3} \text{ cm}^2 \text{ s}^{-1}$ in water. This means that it takes on the average about 13 days for a radon atom to diffuse 5 m in air or 5 cm in water. In that time the radon would decay by almost a factor of 10. Accordingly, long distance transport of radon in air and water mainly depends on the other mechanisms mentioned above, which are the transport of air and water itself.

(c) Exhalation

354. The radon concentration C_z in soil air at depth z below the surface depends on the diffusion coefficient D , the emanating factor a ($0 < a < 1$), the fractional pore space of the soil f , the radium activity concentration C_r (per unit volume of soil) and the decay constant of radon λ according to the equation [J1]

$$C_z = \frac{a \cdot C_r}{f} [1 - \exp(-\sqrt{(\lambda/D)} z)]$$

The exhalation rate is expressed by the equation [J1]

$$R = D \left[-\frac{d(C_z)}{dz} \right]_{z=0}$$

The combination of the above two equations gives

$$R = \frac{\lambda a C_r}{f} \sqrt{D/\lambda}$$

If $a = 0.1$; $C_r = 0.07 \text{ Bq cm}^{-3}$; $f = 0.3$ and $D = 0.01 \text{ cm}^2 \text{ s}^{-1}$; $\lambda = 2 \cdot 10^{-6} \text{ s}^{-1}$, the exhalation rate R is $3 \cdot 10^{-2} \text{ Bq m}^{-2} \text{ s}^{-1}$.

355. The diffusion rate and thereby the exhalation rate is influenced by meteorological factors such as rainfall, snowfall, freezing and variations in atmospheric pressure. An increase in these parameters will decrease the exhalation rate. Measured values of radon exhalation rate from soil vary

between about 0.0002 and 0.07 Bq m⁻² s⁻¹ [G2, W1]. The radon exhalation from sea water per unit area and time is about two orders of magnitude less.

2. Dispersion in air

356. The dispersion of radon in air is influenced by the vertical temperature gradient, the direction and strength of the wind and the air turbulence. The dispersion of radon daughters is also influenced by precipitation. The vertical distribution of radon and its daughters in air can be calculated from the following system of differential equations [J2]:

$$\frac{d}{dz} \left(K \frac{C_1}{dz} \right) - \lambda_1 C_1 = 0$$

$$\frac{d}{dz} \left(K \frac{C_i}{dz} \right) + \lambda_{i-1} C_{i-1} - (\lambda_i + A) C_i = 0$$

where C_1 is the concentration of radon atoms in air at the height z ; n_i is the concentration of radon daughter i in air at the height z ; λ_1 is the decay constant of radon; λ_i is the decay constant of radon daughter i ; A is the removal rate of radon daughters caused by washout and rainout. Boundary conditions to the above equations are

$$C_i(z=0) = 0 \text{ for } i > 1 \text{ and } C_i(z \rightarrow \infty) = 0 \text{ for } i = 1, 2, 3, \dots$$

By assuming a constant radon exhalation from an infinite plane (ground surface) which equals the radioactive decay of the total radon content in the atmosphere it is possible to solve the first two equations in this paragraph, which in combination with different values of the turbulent diffusion coefficient K [J1, J2] give the vertical distribution of radon and radon daughters for different atmospheric stabilities.

357. Measured values of the relative distribution of radon in air are shown in Figure VIII.I. Although measured values in this case are found to follow the predicted vertical distribution fairly well, the models described above should only be taken to serve as rough guidance for the prediction of radon daughter levels. The varying radon exhalation rate on land and on sea and varying meteorological conditions may cause distribution patterns different from those predicted by the model.

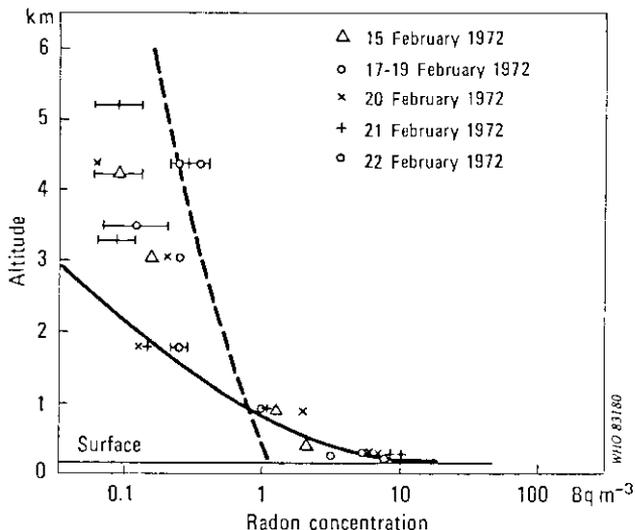


Figure VIII.I Radon concentration at different altitudes over the Yukon Valley in Alaska. The figure represents the average of 12 measurements over the Gulf and Alaska. The solid curve represents the theoretical profile for an exhalation rate of $0.33 \text{ atom cm}^{-2} \text{ s}^{-1}$ and a strong inversion. The broken curve is representative of theoretical profiles with normal conditions of turbulence. [L1]

358. At ground level the time-variation of radon concentrations depends on the variation of the radon exhalation rate and of the vertical dispersion of radon. The effect of increased vertical dispersion of radon by turbulence during spring, as compared with autumn, outweighs the greater exhalation rate of radon during late spring and summer, as compared with autumn and winter. The overall effect is a seasonal variation of the radon concentration at ground level with a minimum in the spring and summer and a maximum in the autumn and winter observed in several measurements [M1, B2, R2, Me]. Diurnal variations of the radon concentration in air at ground level also occur because of different varying turbulent mixing; the concentrations are maximum in the early morning and minimum in the afternoon. The variations are generally less than one order of magnitude [R2, J3].

359. For estimation of the dispersion of radon released from a point (for instance, a geothermal plant or a mine ventilation outlet) the most commonly used statistical model

is the Gaussian plume equation [Si8, P2, G4]. The estimated concentrations at different distances are therefore dependent on local meteorological conditions, terrain roughness, etc. However, in the case of a continuous release, the daily variations are smoothed out and an annual average is obtained, which differs from place to place only according to persistent and substantial local differences.

360. The dispersion of radon released from extended sources like mill tailings can be estimated from dispersion formulas, assuming the extended source to consist of a number of small point sources. The relative concentration of radon released from a point source is approximately inversely proportional to the p -power of the distance d from the source. If the concentration C_d at distance d is expressed relative to the concentration C_1 at the reference distance d_1 the expression is

$$C_d = \left(\frac{d}{d_1}\right)^{-p} C_1$$

The formula approximately gives the relative concentration at distances more than 1 km if the reference distance $d_1 = 1$ km and $p = 1.2-1.5$.

361. The dispersion and relative vertical distribution of the radon daughters in air mainly follow the behaviour of radon. Owing to deviating atmospheric parameters for radon daughters as compared with radon (e.g., precipitation by rainout and washout) there is seldom equilibrium between radon daughters and radon and between the different radon daughters. The long-lived decay products of radon (^{210}Pb , ^{210}Bi and ^{210}Po) behave in the troposphere as aerosols with residence times of the order of ten days and more. Because of their long physical half-life there is no simple correlation between these nuclides and radon.

3. Indoor behaviour

362. For closed spaces (e.g., a mine or a house) a theoretical correlation may be established between radon concentration in air and radon input (exhalation and transport by inlet air) and ventilation rate. The change of the radon concentration in the enclosure is given by the following equation

$$\frac{d C(t)}{dt} = R \left(\frac{S}{V}\right) + \frac{A_k}{V} + C_0 \lambda_v - C(t) (\lambda + \lambda_v)$$

where $C(t)$ is the radon concentration in the air at the time t ; R is the radon exhalation rate from unit surface in the room; S is the emanating surface area; V is the volume of the space, A_k is the radon release from the source (water, gas); C_0 is the radon concentration in the inlet air; λ_v is the ventilation rate (h^{-1}); and λ is the decay constant of radon.

363. At equilibrium the radon concentration in the enclosure is

$$C = \frac{R\left(\frac{S}{V}\right) + \frac{A_k}{V} + C_0 \lambda_v}{\lambda + \lambda_v}$$

In homes $0.1 < \lambda_v < 3 \text{ h}^{-1}$, and since $\lambda = 7.6 \cdot 10^{-3} \text{ h}^{-1}$ and $\lambda_v \gg \lambda$, the above equation takes the form

$$C = \frac{R\left(\frac{S}{V}\right) + \frac{A_k}{V}}{\lambda_v} + C_0$$

As long as $\lambda_v \gg \lambda$ and C_0 is negligible, the radon concentration indoors increases in direct proportion to the decrease in ventilation rate. As the ventilation rate increases from 0 to 0.1 and to 1 h^{-1} , the radon concentration decreases by factors of 13 and 10, respectively.

364. In view of the strong influence of the ventilation rate, there are great variations of the radon levels as the effective ventilation of a room is changed. This is caused by meteorological conditions (wind, pressure, temperature) and by human activities like opening doors and windows. There may be variations of the radon concentration in air caused by changes of the radon exhalation rate from surfaces, which in turn can be caused by changes of atmospheric pressure [J6]. Diurnal variation in houses have been studied in several long-term measurements [S11, D1, S21, H4, J7, S12, M6] and variations of the order of ten and more may occur. Maxima during the night and early morning and minima at noon are usually found, but for several reasons that is not always the case. Only a few studies have been reported on the seasonal variations of radon concentration indoors. The variations of the monthly averages are found to be less than a factor 3 [F1, S13]. Examples of measured variations of radon concentration in houses are shown in Figure VIII.II [M10].

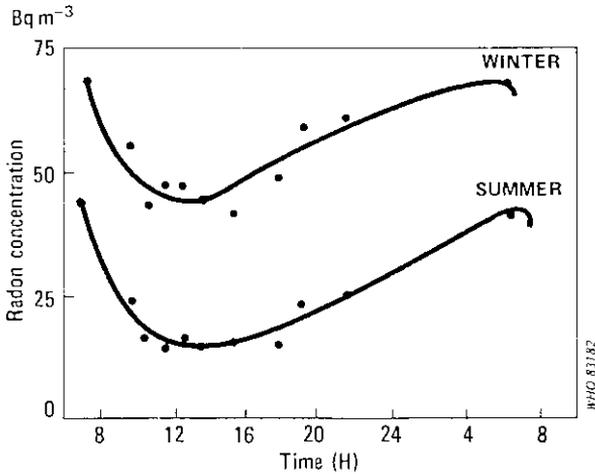


Figure VIII.II Diurnal variation of radon concentration in 16 dwellings in winter and summer. [M10]

365. In mines and other underground spaces there are also diurnal and seasonal variations of the radon concentration. The diurnal variations are most often minor if the ventilation is unchanged by the seasonal variations may be large with maxima during the summer and minima during the winter. This is caused by the change from winter to summer in the temperature gradient from outside to inside the mine (Figure VIII.III).

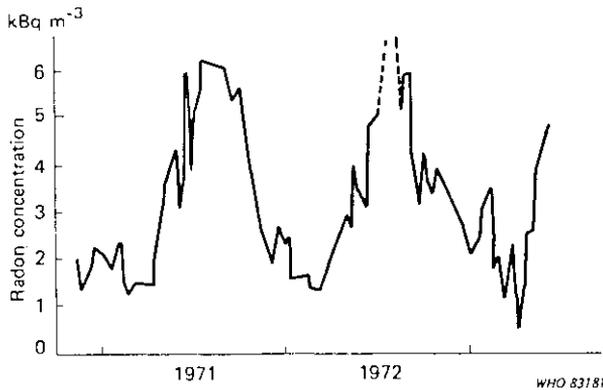


Figure VIII.III Radon concentration in the return air of Persberg Mine in Sweden measured once a week. [S1]

4. Radon daughter concentrations

(a) Concentration expression

366. The concentration of radon daughters can be expressed in terms of their activity or of their potential alpha energy, the latter being the total alpha energy emitted during the decay of the atoms present down to ^{210}Pb . For any mixture of radon daughters in air the potential alpha energy is the sum of the potential alpha energy of all daughter atoms in the air. A unit of exposure which is used in mines is the working level (WL). It is defined as any combination of short-lived radon daughters per litre of air that will result in the emission of $1.3 \cdot 10^5$ MeV of alpha energy in their decay to ^{210}Pb .

367. Another quantity of interest in connection with radon daughters is the equilibrium factor F defined as the ratio of the total potential alpha energy for the given daughter concentration to the total potential alpha energy of the daughters if they are in equilibrium with radon. If the unit WL is used, the equilibrium factor F can be calculated as

$$F = \frac{a C_{\alpha}}{C}$$

where C_{α} is the potential alpha-energy concentration in WL of radon daughters; C is the radon activity concentration in Bq l^{-1} ; and a is a constant ($a = 3.7 \text{ Bq l}^{-1}/\text{WL}$).

368. For a room having a known ventilation rate λ_v (air changes per hour) it is possible to calculate the equilibrium factor F. The relationship between F and λ_v is shown in Figure VIII.IV.

369. The product $C \times F$, where C is the radon concentration and F is the equilibrium factor is called the equilibrium equivalent concentration of radon (EEC); it corresponds to a concentration of radon for which the radon daughters in equilibrium with radon have the same potential alpha energy as the actual daughter concentration of interest.

(b) Attachment

370. The radon daughters in air may be unattached (free atoms or ions) or attached to aerosols. The first daughter, ^{218}Po , is at the time of formation an unattached ion or neutral atom. But within a few seconds most of the ^{218}Po becomes attached to an aerosol and the subsequent decay

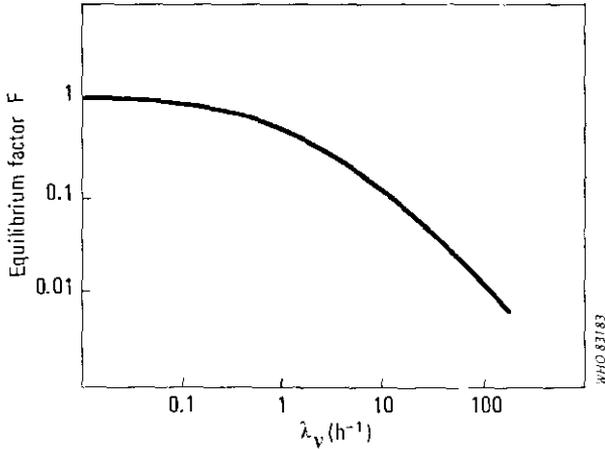


Figure VIII.IV Equilibrium factor F as a function of ventilation rate λ_v (h^{-1}).

products ^{214}Po and ^{214}Bi are therefore to a great extent attached to aerosols at their formation.

371. The attachment rate of a free radon daughter depends therefore on the number and size distribution of the aerosols in the air. These parameters vary in different rooms and will thus affect the attachment rate. In a house with normal aerosol concentration ($\sim 10^4 \text{ cm}^{-3}$) and size distribution, the attachment rate will be about 10^{-2} s^{-1} , i.e., the mean life of the free radon daughter will be about 100 s. In a mine with higher aerosol concentration the corresponding values may be about 0.3 s^{-1} and 4 s, respectively.

372. Radon daughters in room air will also attach to the surfaces in the room. The deposition rate for radon daughters attached to aerosols is dependent on the diffusion rate of the aerosols and the proportion between the surface area and the volume of the room. If that proportion is 2 m^{-1} , the mean life of the attached radon daughters (as far as deposition is concerned) is of the order of one hour. Unattached radon daughters have much higher diffusion rate than aerosols and therefore the deposition rate is also higher. The corresponding mean life is of the order of one minute.

373. The fraction of unattached radon daughters in room air is also dependent on radioactive decay and ventilation rate.

With given values of the attachment rate of free atoms and deposition rate of unattached atoms, the fraction of unattached daughters in air increases with increasing decay constant and ventilation rate. This means that the fraction of unattached ^{218}Po atoms ($\lambda = 13.6 \text{ h}^{-1}$) is normally higher than, e.g., that of ^{214}Pb ($\lambda = 1.6 \text{ h}^{-1}$). Measured values are in the range of 1-30%.

(c) Equilibrium variations

374. Ventilation and deposition to surfaces both prevent the radon daughters reaching equilibrium with radon in air. Only the dependence of the equilibrium factor on the ventilation rate was considered in Figure VIII.IV. However, because of deposition, measured values of the factor F are often lower than the predicted value from Figure VIII.IV. The deviation is larger in air with low aerosol concentrations. An approximate expected value of F is obtained by multiplying the value in Figure VIII.IV by 0.5.

375. Measured values of F in houses show great variation mainly due to differing ventilation conditions. In the UNSCEAR 1977 report [U12] an average value of F for houses of 0.5 was adopted. In outdoor air the equilibrium factor is also dependent on meteorological factors. Measured values indicate an average value of 0.6, which was used by UNSCEAR in its 1977 report. For uranium mines with good ventilation a factor of 0.3 may be appropriate.

D. TRANSFER TO MAN

376. The transfer to man of radon and radon daughters occurs from the inhalation of air. A negligible amount arises from decay of radium in ingested food and water. From a dose standpoint, it is most important to know the intake amount of radon daughters in air.

377. The amount of radon daughters inhaled depends upon the concentration in air and on the breathing rate. The breathing rate varies with different levels of physical activity and age. For the adult, the average breathing rates are 20 l min^{-1} during light activity, 7.5 l min^{-1} resting and 12.5 l min^{-1} for intermediate activity [I1].

378. To compute the average air intake rate, it will be assumed that the time spent indoors per day (19 h) consists of 5.5 h light activity, 8 h resting and 5.5 h intermediate activity. The time spent outdoors (5 h) is assumed to consist

of 2 h light activity and 3 h intermediate activity. This gives estimated intake rates of approximately $15 \text{ m}^3 \text{ d}^{-1}$ indoors and $5 \text{ m}^3 \text{ d}^{-1}$ outdoors.

379. The deposition of radon daughters in the respiratory system depends on the size distribution of the aerosols to which they are attached and on the fraction of unattached radon daughters. The deposition is also influenced by the manner of breathing. The attached radon daughters are deposited in the pulmonary region. However, the deposition is not 100 per cent. Some is exhaled and some is transported by mucus before decay. An approximate value of the fractional retention may be about 50% [U12] but great variations have been reported.

380. The unattached radon daughters are mainly deposited in the upper respiratory tract. The efficient deposition of unattached daughters has been experimentally verified in a model lung by Chamberlain and Dyson [C4]. However, a major part of the unattached daughters is removed by nasal deposition [U12].

E. DOSIMETRY

1. Dose per unit exposure

381. The doses of radon gas in air are negligible in comparison with those of the daughters. On rare occasions, when there is a great disequilibrium between radon and radon daughters, the relative contribution from radon to effective dose equivalent could be significant.

382. Inhalation of radon daughters leads to inhomogeneous irradiation of the respiratory tract. The maximum dose is received by the basal cells in the epithelium of the upper bronchi in the tracheobronchial region of the lungs due to deposition of unattached radon daughters. Normally, the fraction of unattached radon daughters in air is small (a few per cent). Attached radon daughters are mostly deposited in the pulmonary region.

383. Jacobi [J3] has calculated the doses from radon daughters to the tracheobronchial region of the lung (assumed mass 45 g) and to the pulmonary region (955 g) as a function of the unattached fraction, f , of the potential alpha energy. The absorbed energy fractions are $0.03 (1 + 6f)$ joule in the tracheobronchial region per joule inhaled and $0.38 (1 - f)$ joule in the pulmonary region per joule inhaled. Total energy

absorbed in the lung, reflecting deposition and clearance for an average aerosol size and breathing rate is of the order of 40%.

384. The value of the unattached fraction, f , of radon daughters in air is usually in the range 0.02 to 0.1 [U12]. For a mean value of 0.06, the absorbed doses in the lungs are 0.9 Gy per joule inhaled in the tracheobronchial region and 0.4 Gy per joule inhaled in the pulmonary region.

385. The dose equivalent is obtained by multiplying by a quality factor, which for alpha radiation is 20. The effective dose equivalent is obtained by multiplying by the tissue weighting factor, which for the lungs is 0.12. It might be appropriate to apply a weighting factor of 0.06 to the dose equivalent to the basal cell layer of the tracheobronchial region and 0.06 for the pulmonary region. The contributions to the effective dose equivalent to the lungs per unit energy inhaled are thus 1.1 Sv J^{-1} (tracheobronchial region) and 0.5 Sv J^{-1} (pulmonary region).

386. The dosimetry of radon daughters in the lungs is, at the moment, under review. The comparisons of various dosimetric models may lead to recommendations for adjustments of some of these results. For this report a value of 1.6 Sv per joule inhaled will be used for the total effective dose equivalent to the lungs, applicable to exposures indoors and outdoors and also to occupational exposures in mines.

387. The potential alpha energy of an atom in the decay chain of radon is the total alpha energy emitted during decay of the atom up to ^{210}Pb . Dividing by the decay constant of the radionuclide gives the potential alpha energy per unit of activity. The values for radon and its short-lived daughters are given in Table VIII.3.

388. The measured concentration of radon in air must be multiplied by the equilibrium concentration (EEC). For an EEC of 1 Bq m^{-3} and a breathing rate of $20 \text{ m}^3 \text{ d}^{-1}$ ($7300 \text{ m}^3 \text{ a}^{-1}$), the effective dose equivalent rate is

$$1 \frac{\text{Bq}}{\text{m}^3} 7300 \frac{\text{m}^3}{\text{a}} 34500 \text{ MeV} 1.6 \text{ J}^{-1} \frac{\text{J}}{\text{MeV}} 1.6 \frac{\text{Sv}}{\text{J}} = 6 \cdot 10^{-5} \text{ Sv a}^{-1}$$

The effective dose equivalent per unit integrated concentration of radon in air is $6 \cdot 10^{-5} \text{ Sv per Bq a m}^{-3}$.

Table VIII.3

Potential alpha energy of radon and short-lived decay products

Radionuclide	Energy per atom (MeV)	Energy per unit activity (MeV Bq ⁻¹)
²²² Rn	19.2	9.15 10 ⁶
²¹⁸ Po	13.7	3620
²¹⁴ Pb	7.69	17800
²¹⁴ Bi	7.69	13100
²¹⁴ Po	7.69	0.002
Total <u>a/</u>		34500

a/ The total is the sum of the potential energies of the daughters only.

389. In terms of WLM for occupational exposures, the potential alpha energy inhaled is calculated as follows:

$$1.3 \cdot 10^5 \frac{\text{MeV l}^{-1}}{\text{WL}} \cdot 1200 \frac{\text{l}}{\text{h}} \cdot 170 \frac{\text{h}}{\text{M}} \cdot 1.6 \cdot 10^{-13} \frac{\text{d}}{\text{MeV}}$$

$$= 4.2 \cdot 10^{-3} \text{ J (WLM)}^{-1}$$

where the breathing rate is 1200 l h⁻¹ (20 l min⁻¹) during an 8 h working day. The effective dose equivalent per unit exposure is

$$4.2 \cdot 10^{-3} \frac{\text{d}}{\text{WLM}} \cdot 1.6 \frac{\text{Sv}}{\text{J}} = 7 \cdot 10^{-3} \text{ Sv (WLM)}^{-1}$$

The results are summarized in Table VIII.4.

Table VIII.4

Effective dose equivalent per unit exposure from short-lived radon decay products

Public (indoors and outdoors)	5.10 ⁻⁵ Sv (Bq a m ⁻³) ⁻¹
Workers	7 10 ⁻³ Sv (WLM) ⁻¹

390. The effective dose equivalent caused by inhalation of radon without daughters at a concentration of 1 Bq m^{-3} is about $7 \cdot 10^{-7} \text{ Sv a}^{-1}$ [H8]. This is only about one per cent of the effective dose equivalent caused by inhalation of radon daughters in equilibrium with radon.

391. Radon in water may cause a radiation dose to man by ingestion of water and by inhalation of the radon daughters produced by decay of the radon released to air. Consumption of 0.5 l/day of radon-rich water with a radon concentration of 1 kBq l^{-1} will lead to an effective dose equivalent of 0.5 mSv a^{-1} (by ingestion) [S10].

392. The levels of radon have been measured repeatedly in different places of the world. The levels found in outdoor air vary between about 0.1 to 10 Bq m^{-3} , lower values having been found over oceans and islands, higher values over continents. For the estimation of an average dose equivalent, a radon concentration in air of 3.7 Bq m^{-3} might be used, corresponding to an equilibrium equivalent concentration of 2.2 Bq m^{-3} (equilibrium factor = 0.6). If it is assumed that people are outdoors 20% of the time as an annual average (i.e., the occupancy factor is 0.2) the corresponding average effective dose equivalent would be $1.5 \cdot 10^{-2} \text{ mSv per year}$ from radon daughters.

393. In houses the radon levels may be high because of radium-rich building materials and/or poor ventilation. However, normal values are of the order of 10 Bq m^{-3} . An inhabitant-weighted average value for countries in which comprehensive measurements have been carried out is an equilibrium equivalent concentration of radon of 14 Bq m^{-3} . Using the effective dose equivalent per unit exposure given in Table VIII.4 for the public, $0.06 \text{ mSv (Bq a m}^{-3})^{-1}$, and an occupancy factor of 0.8, an equilibrium equivalent concentration of 14 Bq m^{-3} will correspond to an effective dose equivalent of 0.7 mSv per year . A summary of the dose estimates for indoor and outdoor exposures is given in Table VIII.5.

2. Dose per unit release

394. Estimates of the dose per unit release of radon are quite variable, depending on the location of the release, the population density, and the conditions of the dispersion. The most generalized estimates of the collective effective dose equivalents to the world population ($4 \cdot 10^9$ person) may be made by combining dose values of Table VIII.5 with the total

Table VIII.5
Average effective dose equivalent from inhalation
of radon daughters in air

	Equilibrium equivalent concentration (Bq m ⁻³)	Occupancy factor	Dose factor (mSv per Bq a m ⁻³)	Effective dose equivalent (mSv a ⁻¹)
Outdoors	2.2	0.2	0.06	0.03
Indoors	14.0	0.8	0.06	0.7

amounts of radon released of Table VIII.1. For a release of radon outdoors (10^{20} Bq a⁻¹ worldwide) the estimate is $1 \cdot 10^{-15}$ man Sv per Bq, and for a release indoors ($3 \cdot 10^{16}$ Bq a⁻¹ world-wide) the result is $9 \cdot 10^{-11}$ man Sv per Bq.

395. Releases of radon from nuclear installations result from mining and milling operations and from the residual tailings. These generally occur in areas of low population density - from 3 man km⁻² in mining areas to about 25 man km⁻² around the mills [U12]. The local collective dose per unit release can be estimated by an integration over the distance from 1 to 100 km, assuming an atmospheric dispersion factor of $5 \cdot 10^{-7}$ s m⁻³ at 1 km from the release and a reduction in concentration inversely proportional to the 1.5 power of the distance expressed in kilometres [U12]. Using the dose factor of 0.06 mSv per Bq a m⁻³ and population densities of 3 and 25 man km⁻², the range of estimates of the collective effective dose equivalent commitment is $3 \cdot 10^{-16}$ to $3 \cdot 10^{-15}$ man Sv per Bq. This does not yet account for limited outdoor occupancy. Thus, an average estimate of the collective dose per unit release is roughly the same as the generalized estimate obtained above for an outdoor release, namely $1 \cdot 10^{-15}$ man Sv per Bq.

F. REFERENCES

- A1 Andrews, J.N. and D.F. Wood. Mechanism of radon release in rock matrices and entry into groundwaters. Trans. Inst. Min. Metall, (IMM) Sect. B: Applied Earth Science 81: 198-209 (1972).
- A5 Asikainen, M. and H. Kahlos. Natural radioactivity of ground- and surface-water in Finland. Institute of Radiation Protection report STL-A24. Helsinki, 1977 (in Finnish).
- B1 Barton, C.J., R.E. Moore and P.S. Rohwer. Contribution of radon in natural gas to the natural radioactivity dose in homes. Oak Ridge National Laboratory report ORNL-TM-4154 (1973).
- B2 Blifford, J.H. Jr., H. Friedman, L.B. Lockhart, Jr. et al. Geographical and time distribution of radioactivity in the air. J. Atmos. Terr. Phys. 9: 1-17 (1956).
- C2 Castrén, O., M. Asikainen, M. Annanmäki et al. High natural radioactivity of bored wells as a radiation hygienic problem in Finland, in Proceedings of the Fourth International Congress of the International Radiological Protection Association (IRPA). Paris, April 1977.
- C3 Castrén, P. The contribution of bored wells respiratory radon daughter exposure in Finland. p. 1364-1370 in Natural Radiation Environment III. CONF-780422 (1980).
- C4 Chamberlain, A.C. and E.D. Dyson. The dose to the trachea and bronchi from the decay products of radon and thoron. Br. J. Radiol. 29: 317-329 (1956).
- C6 Culot, M.V., H.G. Olson and K.J. Schiager. Radon progeny control in buildings. Final report of the U.S. Atomic Energy Commission COO-22731. Fort Collins, Colorado State University, 1973.
- D1 Davies, B.L. and J. Forward: Measurement of atmospheric radon in and out of doors. Health Phys. 19: 136 (1970).
- F1 Fisenne, I.M. and N.H. Harley. Lung dose estimates from natural radioactivity measured in urban air (1973).
- G1 Gesell, T.F., R.H. Johnson, Jr. and D.E. Bernhardt. Assessment of potential radiological population health

- effects from radon in liquified petroleum gas. U.S. Environmental Protection Agency report EPA-520/1-75-002 (1977).
- G2 George, A.C. Environmental radon and radon daughters, U.S. Department of Energy Environmental Measurements Laboratory Report EML-383 New York (1980).
- G4 Gyllander, C. and U. Widemo. Concentration statistics based on experimental data of atmospheric diffusion. AB Atomenergi Studsvik report S-447. Sweden, 1972.
- H5 Harley, J.H. Environmental radon. p. 109-114 in Noble Gases (R.E. Stanley and A.A. Moghissi, eds.). U.S. Energy Research and Development Administration report CONF-730915 (1973).
- H6 Heijde, R.B. van der, H. Beens and A.R. de Monchy. The occurrence of radioactive elements in natural gas. Ecotoxicology and Environmental Safety 1: 49-87 (1977).
- H8 Hofmann, W., F. Steinhäusler and E. Pohl. Age-, sex- and weight-dependent dose patterns due to inhaled natural radionuclides, p. 1116-1144 in Natural Radiation Environment III. CONF-780422 (1980).
- H9 Holleman, D., D. Martz and K. Schiager, Total respiratory deposition of radon daughters from inhalation of uranium mine atmospheres. Health Phys. 17: 187-192 (1969).
- I1 International Commission on Radiological Protection. Report of the task group on reference man. ICRP Publication 23. Pergamon Press, Oxford, 1974.
- J1 Jacobi, W. Die natürliche Radioaktivität der Atmosphäre und ihre Bedeutung für die Strahlenbelastung des Menschen. Hahn-Meier-Institut für Kernforschung Berlin report HMJ-B 21 (1962).
- J2 Jacobi, W. and K. André. The vertical distribution of radon-222, radon-220 and their decay products in the atmosphere. J. Geophys. Res. 68: 3799-3814 (1963).
- J3 Jacobi, W. Relations between the inhaled potential alpha-energy of 222-Rn and 222-Rn and 220-Rn-daughter and the absorbed alpha-energy in the bronchial and pulmonary region. Health Phys. 23: 3-11 (1972).

- J5 Jonassen, N. and J.P. McLaughlin. Exhalation of radon-222 from building materials and walls, p. 1211-1236 in Natural Radiation Environment III. CONF-780422 (1980).
- J6 Jonassen, N. On the effect of atmospheric pressure variations on the radon-222 concentration in unventilated rooms. Health Phys. 29: 216-220 (1975).
- K2 Knight, G.B. and C.E. Makepeace. Modification of the natural radionuclide distribution by some human activities in Canada. p. 1494-1560 in Natural Radiation Environment III. CONF-780422 (1980).
- L1 Larson, R.E. Radon profiles over Kilauea, the Hawaiian Islands and Yukon snow cover. Pageoph 112: 203-208 (1974).
- L2 Letourneau, E.G., R.G. McGregor and H. Taniquchi. Background levels of radon and radon-daughters in Canadian homes. Paper presented at the Nuclear Energy Agency (NEA) Meeting on Personal Dosimetry and Area Monitoring Suitable for Radon and Daughter Products. Paris, November 1978.
- M1 Malakhov, S.G. and P.G. Chernysheva. On the seasonal variations in the concentration of radon and thoron in the surface layer of the atmosphere. p. 60-68 in Radioactive Isotopes in the Atmosphere and Their Use in Meteorology (J.L. Karol et al., eds.). U.S. Atomic Energy Commission report AEC-tr-6711 (1964).
- M3 Mastinu, G.G. The radiological impact of geothermal energy. Paper presented at the EEC/SCPRI seminar on the radiological burden of man from natural radioactivity in the European community. Le Vesinet, France, December 1979.
- M4 Mattsson, R. Seasonal variation of short-lived radon progeny, 210-Pb and 210-Po, in ground level air in Finland. J. Geophys. Res. 75: 1741-1744 (1970).
- M7 Megromi, K. and R. Mamuro. Emanation and exhalation of radon and thoron gases from soil particles. J. Geophys. Res. 79: 3357-3360 (1974).
- M8 Mustonan, R. Measurements of the radon inhalation rates from building materials. Presented at the Nordic Society for Radiation Protection meeting "Radiation in our Environment". Geilo, Norway, January 1980.

- M9 Muth, H. Natürliche Radioaktivität in Trinkwasser, Nahrungsmitteln und in Menschen in Deutschland. Paper presented at EEC/SCPRI seminar on the radiological burden of man from natural radioactivity in the European community. Le Vesinet, Paris, December 1979.
- M10 Mäkeläinen, I. Preliminary survey on radon in Finnish dwellings. Presented at the Nordic Society for Radiation Protection meeting "Radiation in our Environment". Geilo, Norway, January 1980.
- N1 Nuclear Energy Agency (OECD). Radiological implication of the natural radioactivity in building materials - physical aspects. Report NEA (78) 12. Paris, 1978.
- P1 Partridge, J.E., T.R. Horton and E.L. Sensintaffar. A study of radon-222 released from water during typical household activities. U.S. Environmental Protection Agency. ORP/EERF-79-1 (1979).
- P2 Pasquill, E. The estimation of the dispersion of windborne material. Meteorol. Magazine 90: 33 (1961).
- P3 Porstendorfer, J.A., A. Schraub and A. Wiche. Bestimmung der Radon-Exhalation aus Baumaterialien. Paper presented at the Annual Meeting of the Deutsche Gesellschaft für Biophysik e.V. Freiburg. Federal Republic of Germany, 1974.
- R1 Raghavayya, M. and J.H. Jones. A wire screen-filter paper combination for the measurement of fractions of unattached radon daughters in uranium mines. Health Phys. 36: 417-429 (1974).
- R2 Rangarajan, C., S.S. Gopalakrishnan and C.D. Eapen. The diurnal and seasonal changes in short-lived radon-thoron daughters' concentrations in the coastal and inland regions of India and their possible relation to regional climatology. Pageoph 112: 941-953 (1974).
- S2 Sandström, O. Study on radioactive minerals occurring in Swedish mines. Gruvforskningen Serie B, Nr. 242 (1978) (in Swedish).
- S5 Snihs, J.O. The significance of radon and its progeny as natural radiation sources in Sweden. p. 115-130 in Nobel Gases (R.E. Stanley and A.A. Moghissi, eds.). U.S. Energy Research and Development Administration report CONF-730915 (1973).

- S7 Snihs, J.O. The approach to radon problems in non-uranium mines in Sweden. p. 900-912 in Proceedings of the Third International Congress of the International Radiological Protection Association (IRPA). Washington, 1973.
- S8 Snihs, J.O. Personal communication (1979).
- S9 Snihs, J.O. and P.O. Agnedal. The radiological impacts of uranium mill tailings - a review with special emphasis on the tailing at Ranstad in Sweden. p. 105-113 in Proceedings of the NEA Seminar on Management, Stabilization and Environmental Impact of Uranium Tailings. Albuquerque, 1978.
- S10 Soumela, M. and H. Kahlos. Studies on the elimination rate and the radiation exposure following ingestion of 222-Rn rich water. Health Phys. 23: 641-652 (1972).
- S11 Spitz, H.B. and M.E. Wrenn. The diurnal variation of the radon-222 concentration in residential structures in Grand Junction, Colorado. Report prepared for the Workshop on Environmental Radiation, February 1974. U.S. Atomic Energy Commission report HASL-287. New York, 1974.
- S13 Steinhäusler, F. Long-term measurements of 222-Rn, 220-Rn, 214-Pb and 212-Pb concentrations in the air of private and public buildings and their dependence on meteorological parameters. Health Phys. 29: 705-713 (1975).
- S17 Strandén, E. and L. Berteig. Radon in dwellings and influencing factors. Health Phys. 39: 275-284 (1980).
- S18 Sutton, O.G. Micrometeorology. McGraw Hill, London, 1953.
- S21 Swedjemark, G. Radon in dwellings. Some preliminary results of long-term measurements. National Institute of Radiation Protection report SSI-1974-020. Stockholm, 1974 (in Swedish).
- S22 Swedjemark, G.A. and B. Hakansson. Radon concentration and gamma-radiation in one-family houses built with unusually large fraction of aerated concrete based on alum shale. National Institute of Radiation Protection report SSI: 1978-022. Stockholm, 1978.
- U3 United States Environmental Protection Agency. Preliminary findings of radon-daughter levels in

structures constructed on reclaimed Florida phosphate land. U.S. Environmental Protection Agency Technical note ORP/CSD-75-4. Washington D.C., 1975.

- U4 United States Environmental Protection Agency. Environmental analysis of the uranium fuel cycle. Part I. Fuel Supply. U.S. Environmental Protection Agency report EPA-520/9-73-003-B (1973).
- U12 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 1977 report to the General Assembly, with annexes. United Nations sales publication No.E.77.IX.I. New York, 1977.
- U13 University of Florida. Radioactivity of lands and associated structures. Fourth Semi-annual Technical Report submitted to Florida Phosphate Council. Cumulative Summary report March 1976 to February 1978.
- W1 Wilkening, M.H., W.E. Clemento and D. Stanley. Radon-222 flux measurements in widely separated regions, p. 717-730 in Natural Radiation Environment II (J.A.S. Adams and W.L. Lowden, eds.). The University of Chicago Press, Chicago (1975).
- W3 Wicke, A., J. Postendörfler and A. Schraub. Der Einfluss unterschiedlicher Baumaterialien auf die Radonkonzentration in Wohnräumen. Tagungsbericht Radioaktivität und Umwelt. 12. Jahrestagung. Norderney, Oct. 1978.

B. SOURCES

1. Nuclear explosions

402. The major source which has introduced plutonium into the environment has been atmospheric nuclear testing. Of nearly $1.5 \cdot 10^{16}$ Bq of ^{239}Pu , ^{240}Pu activity released, about $1.2 \cdot 10^{16}$ Bq has been dispersed and deposited world-wide [H2]. The remainder has been deposited locally at the sites of the tests. The amount deposited in the northern hemisphere, where most of the tests were conducted, was three times that deposited in the southern hemisphere.

403. The amounts of the globally dispersed plutonium isotopes produced in all nuclear tests are listed in Table IX.2. Of the total mass of plutonium released world-wide ($4 \cdot 10^6$ g), 96% is comprised of ^{239}Pu and ^{240}Pu . These two isotopes are not separately distinguished in alpha spectrometry and the combined amounts are usually reported. The production data and also analysis of environmental samples indicate that of ^{239}Pu , ^{240}Pu total amounts, 85% by mass or 60% by activity is due to ^{239}Pu .

Table IX.2
Production of globally dispersed plutonium isotopes
in atmospheric nuclear testing

Isotope	Half-life (a)	Specific activity (Bq/g)	Amount produced	
			Activity (Bq)	Mass (t)
^{239}Pu	87.74	$6.3 \cdot 10^{11}$	$3.3 \cdot 10^{14}$	0.00051
^{239}Pu	24065	$2.3 \cdot 10^9$	$7.4 \cdot 10^{15}$	3.26
^{240}Pu	6537	$8.4 \cdot 10^9$	$5.2 \cdot 10^{15}$	0.59
^{241}Pu	14.4	$3.8 \cdot 10^{12}$	$1.7 \cdot 10^{17}$	0.041
^{242}Pu	376000	$1.5 \cdot 10^8$	$1.6 \cdot 10^{13}$	0.11

404. Most of the alpha activity of plutonium produced in nuclear explosions is due to ^{239}Pu , ^{240}Pu . ^{241}Pu is a beta-emitter which decays to ^{241}Am . Little ^{241}Am was produced directly in the tests, but the activity amounts are accumulating in the environment as ^{241}Pu decays. The

activity of fallout ^{241}Am in soil is currently about 25% of that of $^{239},^{240}\text{Pu}$. Decay of ^{241}Pu this far and subsequent decay will result in a total production of ^{241}Am from nuclear tests of $5.5 \cdot 10^{15}$ Bq [B4].

2. Nuclear fuel cycle

405. There are about 240 nuclear reactors used to generate electric power throughout the world. Upon fuel discharge, for each MW(e)a of electricity produced, it can be calculated that $1.2 \cdot 10^{12}$ Bq of $^{239},^{240}\text{Pu}$ are produced, $4.0 \cdot 10^{12}$ Bq of ^{238}Pu , $2.1 \cdot 10^{14}$ Bq of ^{241}Pu and $2.8 \cdot 10^{11}$ Bq of ^{241}Am [H4, K1]. Generally more than half of the ^{239}Pu produced during reactor operation undergoes fission, thus contributing to the energy produced by the reactor. Routine operation of the reactors probably has not resulted in the release to the environment, world-wide, of more than trace amounts of plutonium and americium.

406. Discharge of plutonium to rivers and oceans from fuel reprocessing plants can be much more significant. It has been estimated that 0.1-1% of the plutonium throughput is released in liquid effluent from the Windscale plant in the U.K. [M1]. Little plutonium is released to the air from reprocessing plants. For example, it has been estimated that future reprocessing activities may result in the airborne release of about 40 to 4000 Bq of plutonium and 4 to 40 Bq of americium per MW(e)a [K1].

3. Other sources

407. Processes involved in the production of nuclear weapons have resulted in the release of plutonium to the surrounding environment. Releases occur from fabrication and particularly from reprocessing plants. Leakage from an oil storage area at Rocky Flats plant in Colorado resulted in release of about $2 \cdot 10^{11}$ Bq of ^{239}Pu , half of it offsite but in the near vicinity of the plant. At Mound Laboratory in Miamisburg, Ohio, $4 \cdot 10^{11}$ Bq of ^{239}Pu were washed into an abandoned canal and another $2 \cdot 10^{10}$ Bq was estimated to have been released to the air. Plutonium has been released to onsite disposal areas at several laboratories.

408. A few accidents involving nuclear weapons have been reported that introduced plutonium into the environment. The crash of an airplane at Thule, Greenland resulted in about $9 \cdot 10^{11}$ Bq of plutonium being deposited on the shore and in the bottom sediments. A much smaller quantity is believed to have been carried by winds away from the accident site [F1].

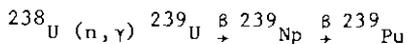
IX. PLUTONIUM

A. INTRODUCTION

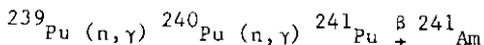
396. Plutonium, the element of atomic number 94, is a member of the actinide series of elements, those of atomic number 89 (actinium) through 103. The actinide elements have similar chemical properties and are also similar to the lanthanide or rare earth elements of atomic number 57 (lanthanum) through 71. The actinides are considered a second rare earth series. Elements beyond uranium (atomic number 92) in the periodic table are called transuranium elements. Some environmental information on another transuranium element, americium (atomic number 95), is also available and will be considered here.

397. Plutonium occurs naturally in very small quantities. It is formed continuously in uranium ores by neutron capture, the neutrons being produced by spontaneous fissioning of uranium. The uppermost layers of the earth's crust contain a few kilograms of ^{239}Pu and about the same amount of primordial ^{244}Pu . Plutonium naturally occurring can only be detected in the richest uranium ore.

398. The most important of the 15 plutonium isotopes, ^{239}Pu , has a half-life of 24065 years and is produced from uranium in nuclear reactors:



One of the plutonium isotopes, ^{241}Pu , decays by beta-particle emission with a 14.4 year half-life to americium-241 which has a half-life of 432.2 years. The decay properties of ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{241}Am are listed in Table IX.1. Americium is produced by successive neutron capture reactions by plutonium isotopes in a reactor:



399. Plutonium can exist in four valence states in aqueous solutions: III, IV, V and VI. The IV state is the most common under physiological conditions where it will exist in solution only as a strongly complexed ion. Weak complexes of Pu(IV) in neutral solutions will form polymeric hydroxides. Plutonium oxidizes rapidly and, thus, the very insoluble PuO_2 is the most common form in the environment, although Pu(VI) has been reported in oceans [N1] and drinking water [L1]. Some plutonium will be complexed with biological ligands and incorporated in micro-organisms or in plant or animal tissues.

400. Americium can exist in three valence states in aqueous solutions: III, IV and VI. The trivalent state is the stable form under physiological conditions. The most common oxide is AmO₂, which is more soluble than PuO₂. Americium appears to be more readily incorporated into biological materials when dispersed to the environment than is plutonium.

401. Plutonium isotopes, ²³⁸Pu, ²³⁹Pu and ²⁴⁰Pu and the americium isotope ²⁴¹Am, emit alpha radiation. Since the x rays accompanying the alpha emissions are low energy, concentrations of these isotopes that might occur in the environment would not cause biological effects unless they are incorporated in biological material. Deposition in the lungs and absorption from the gastro-intestinal tract following ingestion are the most important routes of entry into the bodies of animals and human beings.

Table IX.1
Decay information for plutonium and americium isotopes

Isotope	Half-life (years)	Decay mode	Energy <u>a</u> / (MeV)	Intensity (%)
²³⁸ Pu	87.74	α	5.59	0.716
			5.55	0.283
			5.45	0.001
²³⁹ Pu	24065	α	5.24	0.739
			5.23	0.152
			5.19	0.107
²⁴⁰ Pu	6537	α	5.26	0.734
			5.21	0.265
			5.11	0.0009
²⁴¹ Pu	14.4	β	5.24 · 10 ⁻³ (average)	1.0
²⁴¹ Am	432.2	α	5.58	0.852
			5.54	0.128
			5.48	0.014
			5.64	0.003
			5.61	0.002

a/ Principal transitions.

The collision of two military planes resulted in plutonium from two weapons being dispersed at Palomares, Spain. Much soil was removed in an attempt to clean up the plutonium. Three aberrant missiles were deliberately destroyed in flight and another burned on the launch pad at Johnson Island in the Pacific [F1]. Although the launch pad was decontaminated, undoubtedly several kilograms of plutonium were dispersed to the ocean from the three accidents. It is estimated that from $4 \cdot 10^{11}$ to $40 \cdot 10^{11}$ Bq of plutonium remain available to be incorporated into biological systems from these accidents [F1].

409. The use of plutonium in thermoelectric generation systems of spacecraft has resulted in a relatively small amount of ^{238}Pu and ^{239}Pu being introduced into the environment. Of nearly $350 \cdot 10^{14}$ Bq of ^{238}Pu with an accompanying $260 \cdot 10^{11}$ Bq of ^{239}Pu carried into space by 19 U.S. spacecraft, $6.3 \cdot 10^{14}$ Bq of ^{238}Pu and $4.8 \cdot 10^{11}$ Bq of ^{239}Pu were dispersed into the environment when one spacecraft re-entered the atmosphere and burned over the Indian Ocean. Nearly 80% of this was dispersed in the stratosphere of the southern hemisphere and about 20% in the stratosphere of the northern hemisphere [H1, P5]. Another $16.5 \cdot 10^{14}$ Bq of ^{238}Pu and $12 \cdot 10^{11}$ Bq of ^{239}Pu entered, with containers intact into the Pacific Ocean as a result of an aborted flight. In another aborted flight, the plutonium source was recovered intact from the ocean floor [D4].

410. Large usage is now being made of smoke detectors containing ^{241}Am as an ionization source, with an average of $1 \cdot 10^5$ Bq of ^{241}Am in each detector. About $28 \cdot 10^{11}$ Bq of ^{241}Am has been distributed throughout the U.S.A. in this form [U2]. Since the useful life of these detectors is estimated to be 10 years, it can be assumed that some of these detectors have already been disposed of in sanitary landfills, by incineration, and by other means.

411. Other uses of transuranium elements are made in consumer or medical devices, notably ^{238}Pu in heart pacemakers. These are sealed sources, which when handled properly in normal circumstances, should not allow the contents to be released to the environment.

C. BEHAVIOUR IN THE ENVIRONMENT

1. Movement in soil

412. When plutonium enters soil as fallout, or is added in solutions containing hydrolyzable Pu(IV), it is usually highly insoluble, regardless of soil type [W4]. Diffusion

coefficients for surface soils [G1] are universally low (approximately 10^{-7} cm² s⁻¹). Therefore, the major inventory normally remains in the top cm of undisturbed soils, even with considerable water percolation [E6]. The relative immobility of plutonium in soils under these circumstances may be attributed to the initial low solubility of fallout particles and interaction of Pu(IV) hydrolysis products with soil, mineral and organic surfaces [W4].

413. A small fraction (< 0.1%) of plutonium in soils is soluble, accounting for limited plant uptake from soil and chemical mobility under certain conditions in subsoils. This may be due to the presence of complexing agents or valence states less subject than Pu(IV) to hydrolysis and insolubilization. When Pu(IV) is added to soil as a synthetic or natural organic complex, solubility is initially greatly increased (several orders of magnitude) because of reduced hydrolysis; subsequent mobility is a function of complex stability, competition with other ligands, and resistance of the ligand to chemical and microbial degradation [W6]. Empirical evidence suggests the presence of Pu(III) under reduced conditions in ground waters, and Pu(V)-Pu(VI) have been identified in certain natural waters [W4, B6].

414. Physical processes also account for some vertical movement in soil. Cultivation results in redistribution within the plow layer (to 30 cm) and longterm field studies have traced plutonium migration to 30 cm in undisturbed arid soil [N4]. In the latter case, the increased mobility over that predicted by diffusion alone has been attributed to biological transport and particle movement.

415. Since most plutonium is strongly absorbed on surface soils, wind and water erosion become primary environmental transport mechanisms [W4]. Transport distance will generally be a function of the size of the particle with which plutonium is associated. Particles in the fine silt-clay size range are the most likely to contain the highest concentrations of plutonium, to be transported the greatest distance by wind and water, and to remain attached to biological surfaces.

416. Detailed investigations of the behaviour of americium in soils are lacking. In contrast to plutonium, disproportionation does not occur readily, and Am(III) would be the expected stable species [W4]. Hydrolysis reactions also influence the behaviour of Am(III) in soil, but the products of Am(III) hydrolysis are more soluble than those of Pu(IV) [R3].

2. Transfer to plants

417. Principal mechanisms of plutonium and americium transport to vegetation are foliar interception and root uptake. Foliar uptake is dependent upon chemical form and size of the particle intercepted, residence time and weathering reactions of the leaf. Translocation to the seeds and roots after deposition on the leaf of soyabeans is approximately 10^{-5} of intercepted amounts [W4]. The primary mode of entry into plants is root uptake, with reported soil-to-plant concentration ratios for plutonium ranging from 10^{-3} to 10^{-8} [E5]. Increasing evidence suggests that the solubility in soil, rather than discrimination at the plant root level, is the limiting factor in plutonium uptake by plants [W6].

418. Evidence suggests that plutonium is transported across the root as Pu(IV) [W6, D3]. Complexed Pu(IV) is probably the major translocated species in plants, and several anionic and cationic complexes of Pu(IV) have been determined in the xylem of plants supplied with Pu(IV) [B3]. Plutonium is not uniformly distributed in the plant. The plutonium concentration decreases up the stem of soyabeans, and lowest plutonium concentrations occur in the seeds of barley and soyabeans [W6]. Systematic investigations of americium translocation and deposition in the plant after root uptake have not been conducted.

3. Transfer to animals

419. The primary sources of plutonium and americium to domestic animals are inhalation and consumption of plant tissues containing plutonium in surface-absorbed particles or in tissues. In grazing herbivores, plutonium is primarily associated with the gastro-intestinal tract and pelt, and to a lesser degree, with the lungs [W4]. Gastro-intestinal absorption requires the presence of soluble plutonium and hydrolysis/complexation reactions are likely to govern solubility. The reducing potential of the gut appears sufficient to maintain principally the Pu(IV) state, which is subject to insolubilization by hydrolysis. The effect is more pronounced in the presence of additional reducing substances, such as food residue [S8]. The fraction of ingested amount absorbed and deposited in the bone and liver is approximately 10^{-4} [W6]. However, administration of Pu(VI) in solution to starved animals, or Pu(IV) in complexed forms (synthetic chelates or plant tissues) may increase gut absorption [W6, W5, B2].

420. The gastro-intestinal absorption of americium from gavigated solutions is slightly greater than that of plutonium perhaps reflecting the reduced tendency of americium for hydrolysis. Information on absorption of americium incorporated in plant tissues is not yet available.

4. Transfer to diet

421. The transfer of plutonium and americium to diet from fallout has not been as extensively studied as the transfer of ^{90}Sr and ^{137}Cs . A complete diet sampling, conducted annually in New York, was analysed for $^{239,240}\text{Pu}$ in 1972 [B5] and 1974 [B4]. The 1974 samples were also analysed for ^{241}Am . A few samples of selected foods from 1963 and 1964 were also analysed for $^{239,240}\text{Pu}$.

422. The highest concentrations of $^{239,240}\text{Pu}$ and ^{241}Am were found in shellfish, followed by grain products and fresh fruits and vegetables. The lowest concentrations were in meat, milk, eggs, fresh fish and in processed foods. The values indicate that external contamination is a factor in the occurrence of plutonium in foods. For the shellfish sample, comprising clams and shrimp, most of the plutonium and americium were found in the clams. The muscle in the fresh fish sample, comprising halibut, snapper and flounder, had a $^{239,240}\text{Pu}$ concentration 10 times less and a ^{241}Am concentration 50 times less than the shellfish sample.

423. Based on the New York sampling, the intake by ingestion during 1974 was estimated to be 60 mBq a^{-1} for $^{239,240}\text{Pu}$ and 16 mBq a^{-1} for ^{241}Am . The ratio of americium to plutonium was 0.27 in the total diet indicating little increase of americium relative to plutonium compared to the americium and plutonium in the soil.

424. A $^{239,240}\text{Pu}$ dietary intake record has been calculated based upon the annual fallout deposition rate and the cumulative deposit in soil and compared with the New York food sample surveys conducted in 1963, 1964, 1972 and 1974 [B4]. Assuming no further atmospheric injections, the $^{239,240}\text{Pu}$ dietary intake will remain relatively constant at 0.03 Bq a^{-1} owing to uptake from the 81 Bq m^{-2} cumulative deposit in soil. For ^{241}Am the calculation indicates that the uptake of ^{241}Am from the cumulative deposit in soil is a factor of two greater than plutonium. The estimated ^{241}Am dietary intake continues to increase as the cumulative deposit in soil increases owing to ingrowth from ^{241}Pu decay. When the cumulative deposit reaches its projected maximum of

29 Bq m⁻², the dietary intake will also be at a maximum of 0.03 Bq a⁻¹.

425. The cumulative transfer of plutonium and americium to diet depends very much on the assumed residence times in soil. These times are no doubt shorter than the radioactive mean lives due to leaching and fixation in soil. Extremes of transfer estimates are obtained by taking the mean residence times to be 50 years in one case and the radioactive mean lives in the other case. The geometric mean of these extremes then gives a tentative estimate of the transfer factor P₂₃ from deposition density to diet. The results are 0.6, 0.3 and 0.2 Bq per Bq m⁻² for ²³⁹Pu, ²⁴⁰Pu and ²⁴¹Am, respectively. For ²³⁸Pu and ²⁴¹Pu the estimates of P₂₃ are based on the radioactive mean lives. Most of the transfer is attributed to direct deposition [B4]. The values are 0.08 and 0.04 Bq per Bq m⁻² for ²³⁸Pu and ²⁴¹Pu, respectively.

5. Aquatic behaviour

426. Plutonium is mobilized off watersheds to rivers and coastal waters [H1, M5, H3, S3, S2]. Estimates available for plutonium indicate input ranges from 0.05% per year for heavily cultivated watersheds [M5, S2] to 0.005% per year for a heavily forested watershed, indicating a residence time of 10³ to 2 10⁴ years [W4].

427. Environmental studies have shown that in a variety of comparatively shallow bodies of water, both fresh water and marine, more than 96% of the total plutonium released to or deposited on these environments is rapidly transferred to sediment [E1, L2, H5, H6, P6, S1, N2, H3, E4, P1, W3, S2]. However, in the deep oceans there is only slow transfer of the total plutonium in the ocean water column to deposited sediments. It is estimated that this may represent about 30% by 1980 of deposited fallout plutonium [B8].

(a) Freshwater systems

428. The behaviour of plutonium and americium has been studied in a wide range of fresh water systems [S2, E3, D1, R1, B3, W3, B7]. The concentrations of plutonium in the water of these systems varied by more than four orders of magnitude [W4]. Higher concentrations of plutonium have been observed in lakes with low pH, lakes with high sulfate concentrations and other acidic lakes [W1]. Chemical analyses indicate that while the plutonium in Lake Michigan in the U.S.A. was predominantly in the Pu(V) and (VI) states, in all other lakes

studied Pu(III) and (IV) predominated [N1, W2]. The results strongly suggest that the solubility of plutonium is governed by different complexing agents. In waters of high pH, the concentration of CO_3^{-2} and HCO_3^- is relatively high, and carbonate complexes can form. In waters of low pH, such complexes cannot exist, and the solubility must be due to complexing with other ligands, such as natural organic compounds.

429. A relationship has been shown between the concentration of plutonium in water and the concentration in sediments or particulate matter [N1]. Values for the distribution constant, K_D , vary between 10^4 and $5 \cdot 10^5$, with most values not varying more than fivefold. Considering the wide variety of the systems, including sediment types, size, source terms, etc., this small range in values suggests a commonality in the behaviour of plutonium in these systems. There is some evidence that the plutonium absorbed by sediment particles is predominantly in the (III) and (IV) states, yet on re-equilibration of sediment with water, it has been shown that there is a conversion of Pu(III) and Pu(IV) back to Pu(V) or (VI). This strongly supports the hypothesis that the concentration of plutonium in many fresh water lakes and rivers is controlled by an equilibrium between water and sediment [B7].

(b) Marine systems

430. The behaviour and fate of transuranic elements in the marine environment were given very little attention before the mid-1960s. By far the greatest effort for the next decade was applied to determine the residence time of plutonium in the oceans [B8, M4]. Comparison with ^{90}Sr and ^{137}Cs indicates that the residence time of plutonium in the water column is less than that of both ^{90}Sr and ^{137}Cs . The observed distribution has been explained in terms of a distribution of particles settling at various velocities [N2].

431. In the Irish Sea, the concentration of dissolved $^{239,240}\text{Pu}$ is only 6% as large as that of ^{137}Cs , normalized to a unit discharge rate; the value for ^{241}Am is even lower at 3%. These values suggest that plutonium and americium leave the water phase very rapidly. Measurements in the Irish Sea indicate that plutonium is in solution predominantly as Pu(VI) and on particles as Pu(III) and Pu(IV) [N1], a situation similar to that which exists in the Great Lakes in the U.S.A.

432. Distribution coefficients, K_D , between water and suspended sediments for plutonium in the oceans are similar to those in the Great Lakes and do not appear to be source related. Similar K_D values (10^4 to 10^5) for sediments from the Irish Sea and Enewetak Lagoon suggest that similar chemical reactions are occurring. In the Irish Sea, the overall order of K_D values for transuranic nuclides is $^{241}\text{Am} > ^{242}\text{Cm}$ and $^{244}\text{Cm} > ^{239,240}\text{Pu}$ [P2].

(c) Bioaccumulation

433. Trophic-level studies in freshwater and marine environments indicate that plutonium concentration factors for organisms relative to water generally decrease at higher trophic levels [H5, H6, B7, D2, E2, P4]. Typical values for plutonium in the edible portions used for assessment purposes are 10 for fish, 100 for crustacea and 1000 for molluscs and algae [I1, N3]. Whole organisms values may be 10 to 50 times higher depending upon the degree of contamination by sediment. Limited field data indicate increased concentration factors for ^{241}Am over plutonium in lower trophic levels and in fish [P3, W1, P4].

434. Laboratory studies indicate that marine teleost fish can absorb Pu(VI) by direct uptake from sea-water with limited absorption across the gut from labelled food or sediment. Marine elasmobranchs on the other hand appear to absorb plutonium across the gut relatively easily [P3]. Crustacea, such as crabs, have high assimilation efficiency for plutonium when fed labelled food [F3, G2], and some biomagnification in a simple laboratory invertebrate food chain has been observed [F2].

D. TRANSFER TO MAN

435. Information on the transfer of plutonium to man is available from autopsy measurements on persons exposed to plutonium from weapons test fallout, from occupational sources, and from intentionally administered plutonium in terminally ill patients. The data from fallout plutonium are most pertinent to general environmental considerations. In addition, the results from many animal studies provide supporting data on the probable magnitude of the biokinetic parameters that determine this transfer.

436. Extensive data on the fallout plutonium content of persons in the general population have been published, including data from several areas in the U.S.A. [M2, W7], from Finland [M3] and Japan [O1]. Although great variability was

noted from sample to sample, particularly where only small quantities of tissue were available, the average results agreed reasonably well with computed tissue plutonium burdens [B5], based on estimated plutonium intake by inhalation (ingestion was shown to be insignificant relative to inhalation), assuming metabolic parameters as employed by ICRP.

437. Considering all of the human data now available, there would seem to be no reason to alter the ICRP assumption of an equal distribution of systemic plutonium between skeleton and liver (45% in each) [I4]. The deposition and retention of fallout plutonium in the lung seems also to be well described by the ICRP lung model, assuming behaviour as a Class Y compound [I5]. The ICRP lung model appears to overestimate substantially the transfer of fallout plutonium to tracheobronchial lymph nodes. From extensive studies in experimental animals it is known that the extent of translocation from lung to lymph nodes varies widely with the chemical and physical form of the particles inhaled [B1]. The human data also suggest that gonadal deposition may be slightly higher than the fraction of 10^{-5} g^{-1} assumed by ICRP [I2]. More precise analytical data are required, however, to support such a conclusion. Studies of plutonium deposition in the gonads of several species of experimental animals support the ICRP assumption [R2].

438. Based on comparative studies in experimental animals, ICRP has assumed that inhaled americium will behave in humans in a manner identical with plutonium [I4], except that all americium compounds are assumed to follow Class W lung model kinetics [I2]; i.e., americium is more rapidly cleared from the lung and more efficiently translocated to bone and liver than is plutonium oxide. Applying these assumptions to the estimated intake by inhalation of fallout americium and ^{241}Pu (which will decay to ^{241}Am), estimates have been made of human americium burdens [B4]. These indicate a $^{241}\text{Am}/^{239,240}\text{Pu}$ ratio of 0.24 in 1978, which will increase to 0.38 by the year 2000 because of further decay of deposited ^{241}Pu . Pooled samples from 18 autopsies done from 1970 to 1974 showed a measured $^{241}\text{Am}/^{239,240}\text{Pu}$ ratio, in vertebrae, of 0.22 [B4]. The close agreement between measured and calculated ratios lends support to the ICRP assumptions.

439. Data on the gastro-intestinal absorption of plutonium and americium are available only from studies in experimental animals. Such studies were summarized by an ICRP task group in 1972, which led to assumed values for the fraction absorbed of 10^{-6} for plutonium oxide and 3×10^{-5} for other commonly

occurring compounds of plutonium [I4]. It was recognized that a much higher absorption might be expected for complexed forms of plutonium. More recent data obtained in a variety of animal species [S4, S6] resulted in a modification of ICRP estimates to 10^{-5} for oxides and hydroxides of plutonium, 10^{-4} for other commonly occurring plutonium compounds, and 5×10^{-4} for all compounds of americium [I2]. Qualitative support for the plutonium numbers is provided by autopsy data from a group of reindeer-herding northern Finns who ingest large quantities of plutonium-rich reindeer liver [M3]. Their plutonium burdens seem to be no higher than those of southern Finns, who do not ingest these relatively large quantities of plutonium. A difference should have been apparent if the fraction absorbed from the gastro-intestinal tract had been much greater than 10^{-4} .

440. There is evidence from animal studies that plutonium incorporated into alfalfa [S7] or liver [S4] may be absorbed to a greater extent than inorganic plutonium; the effect is not large, however, and is reversed in the case of americium [S4]. Concern has been expressed that the gastro-intestinal absorption of plutonium in the hexavalent state, such as may be produced by chlorination of water supplies, may be markedly increased as compared to tetravalent plutonium [L1]. It has since been shown, however, that under normal conditions in the gastro-intestinal tract of experimental animals, no such increase of gastro-intestinal absorption is observed [S8].

441. A marked increase in the gastro-intestinal absorption of plutonium and other actinides has been reported in neonatal animals of several species [I4, S5]. This increase may be as much as several hundredfold in the case of rats and several thousandfold in the case of miniature swine. In addition to increased absorption, there is a prolonged retention of the actinide within the mucosa of a small intestine [S5]. It must be assumed that the human infant will also show an increased absorption, although the magnitude of this increase and its duration is unknown.

442. In addition to the inhalation and ingestion routes, actinides may under unusual circumstances of occupational exposure, enter the body by absorption through the intact or punctured skin [I4, B1]. Normally, however, intact skin is an effective barrier to plutonium entry, and this route of entry should not be of concern for general environmental exposure.

443. Once deposited systemically, plutonium is tenaciously retained. This fact is qualitatively evident from extensive data on the excretion of plutonium by occupationally exposed

humans [V1]. It has also been quantitatively evaluated in a few human cases and in a variety of experimental animals [I4, D5]. Based on these data, ICRP has employed a biological half-time of 100 years for plutonium in the skeleton, and a half-time of 40 years for plutonium in liver; plutonium in gonads is assumed to be retained without loss [I4, I2]. The same parameters are assumed to apply in the case of americium.

E. DOSIMETRY

1. Dose per unit intake

444. The doses to the various tissues following inhalation or ingestion of plutonium and americium are determined using the parameters and models suggested by ICRP. A variety of estimates are possible, depending on particle size of inhaled particles and the solubility class of both inhaled and ingested material. The values given below are for the representative 1 μ m aerosol size and for the insoluble oxide or hydroxide forms of plutonium.

445. The ICRP lung model divides the respiratory system into three compartments. Deposition fractions in each region for the 1 μ m particle size are 29% in the nasopharyngeal region, 8% in the tracheo-bronchial region and 23% in the pulmonary region. Smaller particles have a progressively greater deposition fraction in the pulmonary region and less retention in the nasopharyngeal region.

446. Inhaled, insoluble plutonium particles are assigned Class Y parameters, retention in the lungs of the order of years (500 d half-time for 60% of the pulmonary deposition). Because of greater mobility of americium, all of its compounds are assigned Class W parameters, with retention in the lungs of the order of weeks (50 d half-time for 60% of the pulmonary deposition). In both cases, 40% of the pulmonary deposition is cleared with a half-time of 1 d by mucociliary action through the tracheo-bronchial region. Translocation from the naso-pharyngeal and tracheo-bronchial region is rapid, within one day, with most of the material swallowed and small amounts absorbed to blood.

447. For both inhaled and ingested material reaching blood, fractional transfer to blood and liver is assumed to be 0.45 each and $3.5 \cdot 10^{-4}$ to gonads (testes) for both plutonium and americium. Uptake to blood following ingestion of insoluble plutonium is 10^{-5} and $5 \cdot 10^{-4}$ for all compounds of americium.

Table IX.3

Absorbed dose equivalent commitments per unit
intake of plutonium and americium
(Sv per Bq)

Isotope	Lung	Liver	Gonads	Red bone marrow	bone lining cells	Effective <u>a/</u>
<u>Inhalation</u>						
^{238}Pu	$3.2 \cdot 10^{-4}$	$1.8 \cdot 10^{-4}$	<u>b/</u>	$6.6 \cdot 10^{-5}$	$8.3 \cdot 10^{-4}$	$8.2 \cdot 10^{-5}$
^{239}Pu	$3.2 \cdot 10^{-4}$	$2.1 \cdot 10^{-4}$	<u>b/</u>	$7.6 \cdot 10^{-5}$	$9.5 \cdot 10^{-4}$	$8.9 \cdot 10^{-5}$
^{240}Pu	$3.2 \cdot 10^{-4}$	$2.1 \cdot 10^{-4}$	<u>b/</u>	$7.6 \cdot 10^{-5}$	$9.5 \cdot 10^{-4}$	$8.9 \cdot 10^{-5}$
^{241}Pu	$3.2 \cdot 10^{-6}$	$4.4 \cdot 10^{-6}$	$2.8 \cdot 10^{-7}$	$1.7 \cdot 10^{-6}$	$2.1 \cdot 10^{-5}$	$1.6 \cdot 10^{-6}$
^{242}Pu	<u>b/</u>	$5.5 \cdot 10^{-4}$	$3.2 \cdot 10^{-5}$	$2.0 \cdot 10^{-4}$	$2.5 \cdot 10^{-3}$	$1.4 \cdot 10^{-4}$
<u>Ingestion</u>						
^{238}Pu	<u>b/</u>	$4.0 \cdot 10^{-8}$	$2.3 \cdot 10^{-9}$	$1.5 \cdot 10^{-8}$	$1.8 \cdot 10^{-7}$	$1.5 \cdot 10^{-8}$
^{239}Pu	<u>b/</u>	$4.4 \cdot 10^{-8}$	$2.6 \cdot 10^{-9}$	$1.6 \cdot 10^{-8}$	$2.1 \cdot 10^{-7}$	$1.6 \cdot 10^{-8}$
^{240}Pu	<u>b/</u>	$4.4 \cdot 10^{-8}$	$2.6 \cdot 10^{-9}$	$1.6 \cdot 10^{-8}$	$2.1 \cdot 10^{-7}$	$1.6 \cdot 10^{-8}$
^{241}Pu	<u>b/</u>	$8.6 \cdot 10^{-10}$	$5.7 \cdot 10^{-11}$	$3.4 \cdot 10^{-10}$	$4.2 \cdot 10^{-9}$	$2.5 \cdot 10^{-10}$
^{242}Pu	<u>b/</u>	$2.3 \cdot 10^{-6}$	$1.4 \cdot 10^{-7}$	$8.4 \cdot 10^{-7}$	$1.1 \cdot 10^{-5}$	$5.9 \cdot 10^{-7}$

a/ Effective dose equivalent commitment per unit intake (Sv per Bq).

b/ Doses which contribute < 10% to the effective dose equivalent commitment.
Note: for plutonium - Class Y and $f_1 = 10^{-5}$
for americium - Class W and $f_1 = 3 \cdot 10^{-4}$.
Reference [13].

448. The estimates of the absorbed dose equivalents per unit intake are given in Table IX.3. These are computed over a 50 year period following intake. They correspond to the transfer factors P_{25} and P_{35} relating intake in air and diet, respectively, to tissue dose. For these dose equivalent values, the quality factor of 20 has been used for the alpha-emitters and 1 for the beta-emitters (^{241}Pu).

2. Dose per unit release

(a) Nuclear explosions

449. The dose equivalent commitments for plutonium and americium released in nuclear explosions can be assessed using the expressions

$$D^c = P_{25} I_a \quad (\text{inhalation})$$

$$D^c = P_{23} P_{35} F \quad (\text{ingestion})$$

where I_a is the cumulative intake from air (the integrated concentration in air (Bq a m^{-3}) times the breathing rate ($22 \text{ m}^3 \text{ d}^{-1}$) and F is the integrated deposition density. The values of the transfer factors were discussed above.

450. For the past pattern of nuclear tests the population-weighted global integrated concentration in air and deposition density can be determined from comparisons with ^{90}Sr [U1], from $\text{Pu}-^{90}\text{Sr}$ production ratios, and from ^{241}Pu decay considerations [B4]. The results are for air: 1.5, 37, 25, 830 and $1.7 \mu\text{Bq a m}^{-3}$ and for deposition density: 0.85, 21, 14, 470 and 16 Bq m^{-2} for ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{241}Am , respectively. With this information, the dose equivalent commitments can be determined by using the above expressions, and the values per unit activity released can be obtained by dividing the results by the estimated input amounts from nuclear testing (Table IX.2).

451. Finally, the collective dose equivalent commitments are determined by multiplying by the appropriate global population. For inhalation, the activity in air has by now been nearly depleted, so the present global population applies ($4 \cdot 10^9$ persons). For ingestion of the short half-life ^{241}Pu , the present population applies and for the other isotopes, the long-term transfer can be assumed to apply to the projected equilibrium value of 10^{10} persons. The results for the collective effective dose equivalent commitment per unit release are included in the summary Table IX.4.

(b) Nuclear installation

452. The contribution of the inhalation pathway to the collective dose commitments for plutonium and americium in airborne effluents from nuclear installations can be estimated

from the integrated concentrations in air in the dispersion region. It has previously been shown that the appropriate formula is

$$S_1^c = \frac{I \delta_N P_{25}}{v_d}$$

where I is the air breathing rate ($22 \text{ m}^3 \text{ d}^{-1}$), δ_N is the population density in the region (25 man km^{-2}), P_{25} is the dose per unit intake factors (Table IX.3) and v_d is the deposition velocity (0.5 cm s^{-1}). The integrated concentration in air is determined by the amount released (unit activity) per unit area of the deposition region divided by the deposition velocity. The areal dependence is removed by multiplying by the population of the region (times the area). The results from evaluating this expression are summarized in Table IX.4.

Table IX.4
Summary of collective effective dose equivalent commitments
per unit activity released of plutonium and americium
 ($10^{-14} \text{ man Sv per Bq}$)

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am
<u>Nuclear explosions</u>					
Inhalation	1000	1000	1000	30	5 <u>a/</u>
Ingestion	3	30	10	0.01	300
<u>Nuclear installations</u>					
<u>Release to air <u>b/</u></u>					
Inhalation	10000	10000	10000	200	20000
Ingestion	3	20	10	0.03	300
<u>Release to fresh water</u>					
Drinking water	20	20	20	0.2	900
Fish	5	5	5	0.06	200
<u>Release to salt water</u>					
Fish	0.01	0.01	0.01	0.0001	0.4
Shellfish	0.2	0.2	0.2	0.002	6

a/ Per Bq of ^{241}Pu released.

b/ Assumes population density of 25 man km^{-2} .

453. The contribution of the ingestion pathway from airborne effluents to the collective dose commitment per unit activity released, S_1^c , can be determined by using the expression

$$S_1^c = P_{23} P_{35} S_N$$

Using the values of the transfer factors given previously and a population density, δ_N , of 25 man km⁻² in the region of deposition, the values summarized in Table IX.4 are obtained.

454. For the aquatic ingestion pathways the generalized UNSCEAR model is utilized [U1]

$$S_1^c = \frac{N_k I_k f_k P_{35}}{V(\lambda + 1/\tau)}$$

to be evaluated for each pathway k. The quotient of water receiving volume, V, and the population involved, N_k, is the water utilization factor [U1], assumed to be a constant for each pathway. A summary of the values used in the assessments by UNSCEAR is given in the following listing:

<u>Parameter</u>	<u>fresh water</u>	<u>sea water</u>
1. τ , turnover time of receiving water	10 a	1 a
2. Correction factor for sediment removal	1.0	1.0
3. $\frac{V}{N}$, water utilization factor	3 10 ⁷ l/man	3 10 ⁹ l/man
4. f_k , concentration factor for item k		
drinking water	0.1	
fish	10	3
shellfish		300
5. I_k , consumption rate for item k		
drinking water	440 l/a	
fish	1 kg/a	6 kg/a
shellfish		1 kg/a

455. Due to insufficient data, the values of the above listing will be assumed to apply to all isotopes of plutonium and americium. The values of the dose factor, P₃₅, are given in Table IX.3. A summary of the evaluated results is given in Table IX.4.

456. Assessments using the above model only account for that portion of the dose given during the mean residence time of the water in the receiving area. These are essentially complete collective dose commitments for the release to fresh water and for the shorter-lived isotopes (^{241}Pu). The removal of the longer-lived isotopes to the sediments of the deep ocean will largely preclude any further contributions to the dose estimates.

F. REFERENCES

- B1 Bair, W.J. Recent animal studies on the deposition, retention and translocation of plutonium and other transuranic compounds. p. 51-83 in Diagnosis and Treatment of Incorporated Radionuclides. Proceedings of a seminar. Vienna, 1976.
- B2 Ballou, J.E., K.R. Price, R.A. Gies et al. The influence of DTPA on the biological availability of transuranics. Health Phys. 34: 445 (1978).
- B3 Bartelt, G.E., C.W. Wayman, S.E. Groves et al. 238-Pu and 239,240-Pu distribution in fish and invertebrates from the Great Miami River, Ohio. p. 517-530 in Transuranics in Natural Environments (M.G. White and P.B. Dunaway, eds.). Proceedings of a symposium, Gatlinburg, Tennessee, 1976. ERDA report NVO-178, Nevada Operations Office, NTIS, 1977.
- B4 Bennett, B.G. Environmental aspects of americium. Environmental Measurements Laboratory report EML-348, New York, 1978.
- B5 Bennett, B.G. Transuranic element pathways to man. p. 367-383 in Transuranium Nuclides in the Environment. IAEA publication STI/PUB/410, Vienna, 1976.
- B6 Bondietti, E. and F.H. Sweeton. Transuranic speciation in the environment. p. 449-476 in Transuranics in Natural Environments (M.G. White and P.B. Dunaway, eds.). Proceedings of a symposium, Gatlinburg, Tennessee, 1976. ERDA report NVO-178, Nevada Operations Office, NTIS, 1977.
- B7 Bowen, V.T. Plutonium and americium concentration along fresh water chains of the Great Lakes, U.S.A. USAEC report COO-3568-13, Woods Hole Oceanographic Institution, NTIS, 1976.
- B8 Bowen, V.T., K.M. Wong and V.E. Noshkin. 239-Pu in and over the Atlantic Ocean. J. Mar. Res. 29: 1 (1971).
- D1 Dahlman, R.C. Transuranium elements in aquatic and terrestrial environments. p. 131-147 in Environmental Sciences Division annual progress report. USAEC report ORNL-5257, Oak Ridge National Laboratory, NTIS, 1976.
- D2 Dahlman, R.C., E.A. Bondietti and L.D. Eyman. Biological pathways and chemical behaviour of plutonium and other actinides in the environment. p. 47-80 in Actinides in

the Environment. ACS symposium series 35, American Chemical Society, 1976.

- D3 Delaney, M.S. and C.W. Francis. The relative uptake of ^{237}Pu (IV) and Pu (VI) oxidation states from water by bushbeans. *Health Phys.* 34: 492 (1978).
- D4 Dobry, Jr., T.J. Transuranic elements in space nuclear power systems. p. 83-85 in *Transuranic Elements in the Environment* (W.C. Hanson, ed.). DOE publication series TID-22800, Washington D.C., 1980.
- D5 Durbin, P.W. Plutonium in man: a new look at the old data. p. 469-537 in *Radiobiology of Plutonium* (B.J. Stover and W.S.S. Jee, eds.). J.W. Press, Salt Lake City, 1972
- E1 Edgington, D.N. and J.A. Robbins. The behaviour of plutonium and other long-lived radionuclides in Lake Michigan. II. Patterns of deposition in the sediments. p. 245-260 in *Impacts of Nuclear Releases into the Aquatic Environment. Proceedings of a symposium.* Otaaniemi, Finland 1975. IAEA publication STI/PUB/406, Vienna, 1975.
- E2 Edgington, D.N., M.A. Wahlgren and J.S. Marshall. The behaviour of plutonium in aquatic ecosystems: a summary of studies on the Great Lakes. p.45-80 in *Environmental Toxicity of Aquatic Radionuclides: Model and Mechanisms* (M.W. Miller and J.N. Stannard, eds.). Ann Arbor Science Publishers, Ann Arbor, Michigan, 1976.
- E3 Emery, R.M. and D.C. Kopfer. The distribution of transuranic elements in a freshwater pond ecosystem. p. 269-286 in *Environmental Toxicity of Aquatic Radionuclides: Models and Mechanisms* (M.W. Miller and J.N. Stannard, eds.). Ann Arbor Science Publishers, Ann Arbor, Michigan, 1976.
- E4 Emery, R.M., D.C. Kopfer and M.C. McShane. The migration of plutonium from a freshwater ecosystem at Hanford in *Transuranic Elements in the Environment* (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C. 1980.
- E5 Energy Research and Development Administration. Environmental research for transuranic elements in *Workshop on Environmental Research for Transuranic Elements. Proceedings of the workshop*, Battelle Seattle Research Centre, Seattle, Washington, November 1975. ERDA report ERDA-76/134, NTIS (1976).

- E6 Essington, E.H., E.B. Fowler, R.O. Gilbert et al. Plutonium, americium and uranium concentration in Nevada Test Site soil profiles. p. 157-173 in Transuranium Nuclides in the Environment. Proceedings of a symposium, San Francisco, 1975. IAEA publication STI/PUB/410, Vienna, 1976.
- F1 Facer, G. Quantities of transuranic elements in the environment from operations relating to nuclear weapons. p. 86-91 in Transuranic Elements in the Environment (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.
- F2 Fowler, S.W. and T.M. Beasley. Plutonium and americium in fish. *Nature* 265: 384 (1977).
- F3 Fowler, S.W. and J.C. Guary. High Absorption efficiency for ingested plutonium in crabs. *Nature* 266: 827-828 (1977).
- G1 Garland, T.R. and R.E. Wildung. Physiochemical characterization of mobile plutonium species in soils, p. 254-263 in Biological Implications of Metals in the Environment (H. Drucker and R.E. Wildung, eds.). DOE symposium series, Richland, Washington, 1975. CONF-750929 NTIS (1977).
- G2 Guary, J.C. and A. Fraizier. Influence of trophic level and calcification on the uptake of plutonium observed, in situ, in marine organisms. *Health Phys.* 32: 21-28 (1977).
- H1 Harley, J.H. Plutonium in the environment - a review. Prepared for the 1979 annual meeting of the Radiation Research Society of Japan, Osaka City, Japan, 1979.
- H2 Hardy, E.P. World-wide distribution of plutonium. p. 115-128 in Plutonium and Other Transuranium Elements: Sources, Environmental Distribution and Biomedical Effects (B.W. Wachholz, ed.). Presented at the Environmental Protection Agency Plutonium Standards Hearings, Washington, D.C., 1974. U.S. Atomic Energy Commission, Springfield, Virginia, 1974.
- H3 Hayes, D.W. and H.J. Horton. Plutonium and americium behaviour in the Savannah River marine environment in Transuranic Elements in the Environment (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.

- H4 Heeb, C.M. and E.T. Merrill. Significant actinide activities in the LWR and LMFBR nuclear fuel cycle. EPA-520/2-75-006 (1974).
- H5 Hetherington, J.A., D.F. Jefferies and M.B. Lovett. Some investigations into the behaviour of plutonium in the marine environment. p. 193-212 in Impacts of Nuclear Releases into the Aquatic Environment. Proceedings of a symposium, Ontaniemi, Finland, 1975. IAEA publication STI/PUB/406, Vienna, 1975.
- H6 Hetherington, J.A., D.F. Jefferies, N.T. Michell et al. Environmental and public health consequences of the controlled disposal of transuranic elements to the marine environment. p. 139-156 in Transuranium Nuclides in the Environment. Proceedings of a symposium, San Francisco, 1975. IAEA publication STI/PUB/410, Vienna, 1976.
- I1 International Atomic Energy Agency. The radiological basis of the IAEA revised definition and recommendations concerning high level radioactive waste unsuitable for dumping at sea. IAEA-211 (1978).
- I2 International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. ICRP publication 30, Pergamon Press, 1979.
- I3 International Commission on Radiological Protection. Limits for intakes of radionuclides by workers, supplement to Part I. ICRP publication 30, Pergamon Press, 1979.
- I4 International Commission on Radiological Protection. The metabolism of compounds of plutonium and other actinides. ICRP publication 19, Pergamon Press, 1972.
- I5 International Commission on Radiological Protection. Task group on lung dynamics. Deposition and retention models for internal dosimetry of the human respiratory tract. Health Phys. 12: 173-226 (1966).
- K1 Kreiter, M.R., J.E. Mendel and R.W. McKee. Transuranic wastes from the commercial light-water-reactor cycle. p. 92-106 in Transuranic Elements in the Environment (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.
- L1 Larsen, R.P. and R.D. Oldham. Plutonium in drinking water: effects of chlorination on its maximum permissible concentration. Science 201: 1008-1009 (1978).

- L2 Livingstone, H.L. and V.T. Bowen. Americium in the marine environment - relationships to plutonium. p. 107-130 in Environmental Toxicity of Aquatic Radionuclides: Models and Mechanisms (M.W. Miller and J.N. Stannard, eds.). Ann Arbor Science Publishers, Ann Arbor, Michigan, 1976.
- M1 Morley, F. and G.N. Kelly. Radiological protection and transuranic wastes from the nuclear fuel cycle. Proceedings of a NEA/CEA Seminar on the management of plutonium contaminated solid wastes. Marcoule, October, 1974.
- M2 McInroy, J.F., E.E. Campbell, W.D. Moss et al. Plutonium in autopsy tissue: a revision and updating of data reported in LA-4875. Health Phys. 37: 1-136 (1979).
- M3 Miettinen, J.K., H. Mussalo, M. Hakanen et al. Distribution of plutonium and americium in human and animal tissues after chronic exposures. p. 265-268 (Vol.III) in Book of Papers. Fifth International Congress of the International Radiation Protection Association, Jerusalem, 1980.
- M4 Miyake, Y. and Y. Sugimura. The plutonium content of Pacific Ocean waters. p. 91-104 in Transuranium Nuclides in the Environment. Proceedings of a symposium, San Francisco, 1975. IAEA publication STI/PUB/410, Vienna, 1976.
- M5 Muller, R.N., D.G. Sprugel and B. Kohn. Erosional transport and deposition of plutonium and caesium in two small midwestern watersheds. J. Environ. Qual. 7(2): 171-174 (1978).
- N1 Nelson, D.M. and M.B. Lovett. The oxidation state of plutonium in the Irish Sea. Nature 276: 599-601 (1978).
- N2 Noshkin, V.E. and V.T. Bowen. Concentrations and distributions of long-lived fallout radionuclides in open ocean sediments. p. 671-684 in Radioactive Contamination of the Marine Environment. IAEA, Vienna, 1973.
- N3 National Radiological Protection Board/CEA. Methodology for evaluating the radiological consequences of radioactive effluents released in normal operations. Joint report of NRPB/CEA, CEC (1979).

- N4 Nyhans, J.W., F.R. Miera, Jr. and R.E. Neher. Distribution of plutonium in trinity soils after 28 years. *J. Environ. Qual.* 5(4): 431-437 (1976).
- 01 Okabayashi, H., H. Watanabe and Y. Takizawa. Measurement of plutonium in Japanese human organs. *J. Radiat. Res.* 19: 62-69 (1978).
- P1 Paine, D. Plutonium in Rocky Flats freshwater systems in Transuranic Elements in the Environment (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.
- P2 Pentreath, R.J., D.F. Jefferies, M.B. Lovett et al. The behaviour of transuranium and other long-lived radionuclides in the Irish Sea and its relevance to the deep sea disposal of radioactive wastes. Proceedings of the Third NEA Seminar on Marine Radioecology, OECD, Paris, 1980.
- P3 Pentreath, R.J. and M.B. Lovett. Transuranic nuclides in plaice, *pleuronectes platessa* from the northeastern Irish Sea. *Marine Biol.* 48: 19-26 (1978).
- P4 Pentreath, R.J., D.S. Woodhead, B.R. Harvey et al. A preliminary assessment of some naturally-occurring radionuclides in marine organisms (including deep sea fish) and the absorbed dose resulting from them. Proceedings of the Third NEA Seminar on Marine Radioecology, OECD, Paris, 1980.
- P5 Perkins, R.W. and C.W. Thomas. World-wide fallout. p. 53-82 in Transuranic Elements in the Environment (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.
- P6 Pillai, K.C. and E. Mathew. Plutonium in the aquatic environment, its behaviour, distribution and significance. p. 25-45 in Transuranium Nuclides in the Environment. Proceedings of a symposium, San Francisco, 1975. IAEA publication STI/PUB/410, Vienna, 1976.
- R1 Rees, T.F., J.M. Cleveland and W.C. Gottschall. Dispersion of plutonium from contaminated pond sediments. *Environ. Sci. Technol.* 12(9): 1085-1087 (1978).
- R2 Richmond, C.R. and R.L. Thomas. Plutonium and other actinide elements in gonadal tissue of man and animals. *Health Phys.* 29: 241-250 (1975).

- R3 Routson, R.C., G. Jansen and A.V. Robinson. 99-Tc, 237-Np and 241-Am sorption on two United States subsoils from differing weathering intensity areas. Health Phys. 33: 311-317 (1977).
- S1 Schell, W.R., F.G. Lowman and R.P. Marshall. Geochemistry of transuranic elements at Bikini Atoll in Transuranic Elements in the Environments (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington D.C., 1980.
- S2 Simpson, H.J., R.M. Trier and C.R. Olsen. Transport of plutonium by rivers in Transuranic Elements in the Environment (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.
- S3 Sprugel, D.H. and G.E. Bartelt. Erosional removal of fallout plutonium from a large midwestern watershed. J. Environ. Qual. 7(2): 175-177 (1978).
- S4 Stather, J.W., J.D. Harrison, P. Rodwell et al. The gastrointestinal absorption of plutonium and americium in the hamster. Phys. Med. Biol. 24: 396-407 (1979).
- S5 Sullivan, M.F. Absorption of actinide elements from the gastrointestinal tract of neonatal animals. Health Phys. 38: 173-185 (1980).
- S6 Sullivan, M.F. Absorption of actinide elements from the gastrointestinal tract of rats, guinea pigs and dogs. Health Phys. 38: 159-171 (1980).
- S7 Sullivan, M.F., T.R. Garland, D.A. Cataldo et al. Absorption of plutonium from the gastrointestinal tracts of rats and guinea pigs after ingestion of alfalfa containing 238-Pu. Health Phys. 38: 215-221 (1980).
- S8 Sullivan, M.F., J.L. Ryan, L.S. Gorham et al. The influence of oxidation state on the absorption of plutonium from the gastrointestinal tract. Radiat. Res. 80: 116-121 (1979).
- T1 Till, J.E., S.V. Kaye and J.R. Trabalka. Toxicity of uranium and plutonium to the developing embryos of fish. ERDA report ORNL-5160, Oak Ridge National Laboratory, NTIS, 1976.
- U1 United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, 1977 report to the General

Assembly, with annexes. United Nations sales publication No. E.77.IX.I. New York, 1977.

- U2 U.S. Nuclear Regulatory Commission. Environmental Assessment of Ionization Chamber Smoke Detectors Containing Am-241 (R. Belanger, D.W. Buckley and J.B. Swenson, eds.). Prepared for the U.S. Nuclear Regulatory Commission by Science Applications, Inc., La Jolla, California, 1979.
- V1 Voelz, G.L., L.H. Hempleman, J.N.P. Lawrence et al. A 32-year medical follow-up of Manhattan project plutonium workers. *Health Phys.* 37: 445-485 (1979).
- W1 Wahlgren, M.A., J.J. Alberts, K.A. Orlandini et al. A comparison of the concentrations of fallout-derived plutonium in a series of freshwater lakes. p. 92-94 in Radiological and Environmental Research Division annual report, (1977). ERDA report ANL-77-65 (Part 3), Argonne National Laboratory, NTIS (1977).
- W2 Wahlgren, M.A., J.J. Alberts, D.M. Nelson et al. Study of the occurrence of multiple oxidation states of plutonium in natural water systems. p. 95-98 in Radiological and Environmental Research Division annual report (1977). ERDA report ANL-77-65 (Part 3), Argonne National Laboratory, NTIS (1977).
- W3 Wahlgren, M.A., J.A. Robbins and D.N. Edgington. Plutonium in the Great Lakes in *Transuranic Elements in the Environment* (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.
- W4 Watters, R.L., D.N. Edgington, T.E. Hakonson et al. Synthesis of the research literature in *Transuranic Elements in the Environment* (W.C. Hanson, ed.). DOE publication series, TID-22800, Washington, D.C., 1980.
- W5 Weeks, M.H., J. Katz, W.O. Oakley et al. Further studies on the gastrointestinal absorption of plutonium. *Radiat. Res.* 4: 339 (1956).
- W6 Wildung, R.E., T.R. Garland and D.A. Cataldo. Environmental processes leading to the presence of organically-bound plutonium in plant tissues consumed by animals in *Biological Implications of Radionuclides Released from Nuclear Industries*. Proceedings of a symposium, Vienna, 1978. IAEA, Vienna, 1979.

- W7 Wrenn, M.E. and N. Cohen. Determination of Pu-239/240 tissue concentrations in non-occupationally exposed residents of New York City. Annual report to the Department of Energy. New York University Medical Centre report COO-2968-2. New York, 1978.

X. RADIATION EFFECTS

457. The interaction of radiation with matter results in the liberation of energy carried by the alpha-particle, beta-particle, gamma or x-rays and the ionization or excitation of the irradiated material. In biological material, damage may be caused directly to cell components by the radiation interactions or indirectly by the actions of free radicals, the charged fragments of ionization events. The damage may result in cell death or in cell transformations, which at some later time may cause harmful effects in the irradiated individual or in his offspring. The amount of damage depends on the amount of radiation, which may be from external irradiation or from radionuclides within the body, on the type of radiation and on the sensitivity of the tissue.

458. Radiation effects in man are usually classified as somatic and genetic or hereditary, according to whether they affect somatic or germinal cells. Somatic damage is expressed therefore by definition within the lifetime of the irradiated individual, while genetic damage is expressed at some stage in his progeny. Somatic effects are - somewhat loosely - further distinguished as immediate or late depending on the time of their appearance.

A. SOMATIC EFFECTS

1. Early somatic effects

459. The immediate or early somatic effects are expressed in man within a few days or a few weeks after exposure as a result of damage to one or more of the self-renewing tissues. These effects are also called functional because they are due to the inactivation of a great number of functional cells of a given differentiative line. Selective irradiation of a given tissue (as in the case of exposure to some internal emitters) usually leads to effects localized in that tissue; whole-body irradiation above a given dose results, on the contrary, in the appearance of generalized effects, usually under the form of a specific syndrome which depends on dose. The clinical severity of the immediate effects changes considerably with the dose, dose rate, type and energy of the absorbed radiation and part of the body irradiated.

460. The nature of immediate effects is non-stochastic or deterministic, in the sense that these effects are expected to occur in an exposed individual absorbing doses in excess of a minimum amount of radiation referred to as the threshold

dose. The threshold dose is extremely variable depending on the effect and the tissue considered. It is of the order of a few tenths of a Gy for most functional effects of importance for radiation protection. The existence of a threshold of dose is an important characteristic of these effects; it makes it virtually impossible for them to appear for doses below the threshold, thus allowing their complete avoidance.

461. Another important aspect of early effects to be emphasized in relation to irradiation from internal emitters is the variability of the dose threshold as a function of dose rate. Low dose rate irradiation is normally a condition leading to an increase of the threshold. Since irradiation from radionuclides (particularly at the levels usually present in the environment) takes place at very low doses and dose rates it is virtually impossible, except under exceptional conditions of emergency, that immediate effects might be observed. They do not deserve therefore any more extended treatment.

2. Late somatic effects

462. Late somatic effects are those appearing in irradiated individuals after a latency period and are expressed mainly in the form of leukaemias or solid tumours. These effects are stochastic or statistical in nature, in so far as it is impossible to identify a causal relationship for them in any given case. The correlation between radiation dose and induction of these conditions may only be shown on large populations of irradiated individuals as an increase of the above diseases over their apparently spontaneous background incidence.

463. Since it is impossible to establish with any certainty the shape of the dose-induction relationship for late tumorous effects, particularly at low doses and dose rates, it is usually assumed that the frequency of their occurrence is linear with dose and without threshold. The clinical severity of these conditions is variable, but it is commonly assumed for purposes of radiation protection, that they might be of a uniform and maximum severity, namely the death of an individual.

B. GENETIC EFFECTS

464. Radiation-induced hereditary effects may appear in the progeny of irradiated individuals within the first generation following irradiation - in which case the damage is called dominant - or within subsequent generations when the genes

that carry the same mutation in the male and in the female genetic complement happen to match in the genome of the zygote. In this latter case the genetic damage is called recessive. Clinically, radiation-induced hereditary conditions have a large spectrum of severity from the relatively trivial to the very harmful.

465. As in the case of the somatic late effects, radiation protection usually makes reference to the most severe hereditary diseases which are either incompatible with life or very disabling for the individual. Radiation protection also assumes that for this class of effects a linear non-threshold induction relationship with dose may apply.

C. DOSE-RESPONSE RELATIONSHIPS

466. In experimental animals and in man late somatic and hereditary effects may exhibit different shapes of the dose-effect relationships, according to a large number of physical and biological variables operating in each particular system. Linear, linear-quadratic, quadratic or complex relationships have been described in various circumstances. No generalization may be gained by the consideration of all existing experience, except perhaps that each specific system responds according to different kinetics of action and that biologically complex effects usually correspond to more complex types of relationships. It would be impossible to set up a rationale for a system of radiation protection by considering each case separately. To overcome this difficulty the assumption is made that late somatic and hereditary effects of irradiation follow a non-threshold linear function of dose. This assumption is simple and there is evidence that it is also a conservative assumption in most cases.

467. It is important to stress the meaning of the assumption of non-threshold linearity. It postulates, on the one hand, that there is no dose, however small, that may in principle be considered safe and no dose increment, however small, which could not produce a corresponding increase of effect and therefore of risk. The summation of doses taken as a measure of total risk and calculations of collective doses as expression of the total detriment in the exposed population have little meaning outside the assumption of non-threshold linearity.

468. On the other hand, adoption of the assumption of linearity involves implicitly the adoption of other important principles. Actually, if there is a linear relationship between the dose and the induction of stochastic effects, it

becomes possible to use the average dose received by a given organ or tissue as a significant reference quantity. Under these conditions it becomes unnecessary to consider the dose variability within the given organ or tissue because the response of the component cells (taken to be of uniform sensitivity) will in any case be linear with the absorbed dose. The reactions of the component cells will in any case sum-up to produce an overall effect corresponding to that expected from the mean dose in that organ or tissue.

469. In case of internal irradiation there may be problems related to the presence of point sources. The following considerations apply in these cases. Firstly, with regard to non-stochastic effects, cell death resulting from high doses within microscopic volumes of the tissue are expected to produce less harm than for the same dose uniformly distributed within large volumes: this is because killing of transformed cells by high doses would be expected to lead to inactivation of potentially transformed cells. Moreover, loss of cells around the zones of highest dose absorption would not be expected to result in significant decrease in tissue or organ function, unless the functional reserve of the organ or tissue is impaired for other reasons.

D. RISK ESTIMATES

470. UNSCEAR has extensively reviewed in its 1977 report most information on the subject of tumour induction in man by ionizing radiation. The Committee concluded that the risk for all fatal malignancies after whole-body irradiation at low doses and dose rates of low-LET radiation - as an average of both sexes and all ages - is in the region of 10^{-2} Sv⁻¹. The risk of inducing non-fatal malignancies under the same conditions would probably fall in the same range.

471. It should be emphasized that for doses of the order of those received annually from natural sources no direct information is available. The above estimate is derived predominantly from observations conducted on people exposed to absorbed doses of over 1 Gy. While the rate per unit dose or doses in the region of the natural background would unlikely be higher, it could, however, be substantially lower. There is no evidence that irradiation from internal sources would produce rates of tumour induction differing from those from external irradiation, if account is taken of the mean absorbed doses in tissues.

472. Concerning hereditary effects, the Committee estimated that when a population is continuously exposed to low doses of

low-LET radiation at rates of the order of 10^{-2} Gy per generation (a generation corresponds to about 30 years) about 50 genetic diseases might be expected to occur per one million first generation progeny. At equilibrium, the total genetic damage expressed over all generations (or the value in each generation reached after prolonged continuous exposure) would be of the order of 150 cases per million progeny.

473. It is often convenient to be able to estimate the total detriment to the individual from irradiation of specific organs or tissues, taking into account the various types of effects from various irradiations. This could be a difficult procedure, however, in cases when different irradiation modalities lead to different effects. Such a situation applies for irradiation by internally deposited radionuclides, when the nuclides produce different doses in tissues of varying sensitivity and the effects must be added to the effects of whole-body irradiation.

474. In order to overcome this difficulty, ICRP, in its publication 26, has designed a system allowing combined estimates of risk in various organs and tissues, based on their susceptibility to various effects. These risks of effects of irradiations of specific tissues are weighted relative to the total effect from whole-body irradiation. The risks apply to one individual or the whole exposed population, making use of the hypothesis of linearity. It is realized, of course, that the applicability of risk estimates may vary according to the characteristics of the individual (genetic make-up, sex, age, etc.) or to the structure of the exposed population. But it is also held that an acceptable level of precision may be reached by assuming an average risk value to be applied to all members of the population irrespective of the above mentioned variability.

475. The mortality risks and weighting factors recommended by ICRP are shown in Table X.1. These factors have been derived for the protection of workers, but may also be applied to large population groups, provided allowance is made for the hereditary effects which would be expected to appear in all generations subsequent to the second.

476. It must be emphasized that the risk estimates for induction of somatic and genetic effects should be regarded as the best possible numerical conclusions to be drawn from a very heterogeneous data base affected by various types of dosimetric and epidemiological uncertainties. Although it is felt that such estimates are reasonably precise for the

purpose of radiation protection they are to be interpreted and used in a statistical sense simply as illustrations of the order of magnitude of potential risks. The actual validity of these estimates could not possibly be tested empirically under normal circumstances owing to the small levels of the risks compared with the far higher "spontaneous" background of similar conditions in the general population.

Table X.1
Mortality risk and weighting factors for different organs
(from ICRP publication 26)

Tissue	Mortality risk (Sv ⁻¹)	Weighting factor
Breast	0.25 10 ⁻²	0.15
Red bone marrow	0.2 10 ⁻²	0.12
Lungs	0.2 10 ⁻²	0.12
Thyroid	0.05 10 ⁻²	0.03
Bone surface	0.05 10 ⁻²	0.03
Remainder	0.5 10 ⁻²	0.30
Gonads (hereditary effects in the first two generations)	0.4 10 ⁻²	0.25
Total	1.62 10 ⁻²	1.00

XI. CONCLUSIONS

A. RADIONUCLIDES AND THE ENVIRONMENT

477. Radiation is a natural feature of man's environment - from the high energy charged particles which make up cosmic radiation to the radioactive decay of radionuclides in the earth's crust and in the biosphere. Several of the radionuclides considered in this document have important natural sources, including tritium, carbon-14, krypton-85 and radon. The first three are produced mainly by cosmic ray interactions in the atmosphere. Radon arises from the decay of radium present in the earth's crust. All of these radionuclides are widely dispersed in air. Tritium and ^{14}C enter more general bio-geochemical cycles, the hydrological cycle and the carbon cycle, respectively.

478. Several activities of man result in the production of radionuclides and contribute to the radiation background. An important source has been the testing of nuclear weapons in the atmosphere. Large scale atmospheric testing was completed prior to the Test Ban Treaty of 1963, but additional tests by some countries have continued. The radionuclides produced in atmospheric nuclear explosions become widely dispersed in the atmosphere, primarily of the hemisphere in which the test was conducted, but with some interhemispheric mixing contribute to the global exposures to fallout radioactivity. Important fission radionuclides in terms of the doses delivered include ^{131}I , ^{90}Sr and ^{137}Cs . In the long-term, the radionuclides ^{14}C and isotopes of plutonium become important contributors to the radiation dose.

479. The generation of electricity using nuclear power reactors also results in the production of radionuclides and in some radiation exposure of man. The radioactive materials are largely contained within the fuel elements in the reactor or in waste treatment systems at the fuel reprocessing plant. Releases of controlled amounts occur in liquid and airborne effluents from the nuclear installations. Accidents could result in potentially greater releases of radioactive materials.

480. The natural and man-made sources of radionuclides in the environment have been discussed for the various radionuclides considered in this document. The amounts of radioactive materials released from the various sources have been reviewed and the relevant values have been shown to depend very much on specific past practices. Even the normalized release amounts,

for example the amounts per Mt in nuclear explosions or per MW(e)a of electricity generated, have limited validity. The amounts released in atmospheric nuclear testing depend on the types of devices tested and on the geographic pattern of past testing. Releases from nuclear power installations depend on the efficiency and integrity of present designs and on the specific waste management practices currently utilized.

481. The behaviour of radionuclides in the environment has been studied rather extensively, so that by now, the dispersion of these pollutants in the environment is fairly well understood. Values have been derived for the various transfer factors which describe the transport of radionuclides in the environment and their transfer to man. For the purpose of dose assessment it is necessary to consider only the more specific aspects of particular release situations and to adjust the more generally valid parameters to the local conditions.

B. DOSE ASSESSMENTS

482. Exposure of man to radionuclides in the environment occurs by inhalation of amounts in air, ingestion of amounts incorporated into diet, or from external exposure to radionuclides in air or deposited on the ground. The dose assessments for the radionuclides considered in this document have been directed toward obtaining estimates of the collective effective dose equivalent commitments per unit amount of activity of the radionuclide released. This expression of dose gives the absorbed dose, weighted for radiation type and sensitivity of irradiated tissues to the entire population and for as long as the exposures from a specific release continue. This quantity is expected to be most directly related to the total health detriment which may result. The results of the dose assessments are summarized in Table XI.1.

483. The degree of transfer of the radionuclides to man and thus the dose estimates vary depending on the source of release of the radionuclides. Natural production of tritium, ^{14}C and ^{85}Kr occurs primarily in the upper atmosphere, following which there is widespread dispersion, and for ^3H and ^{14}C generalized cycling throughout the environment. Nuclear explosions conducted in the atmosphere, particularly large scale tests, result in injection of debris into the stratosphere, from where the radionuclides are globally dispersed. Releases of radionuclides from nuclear installations are near surface emissions in airborne effluents

or in liquid effluents to rivers and lakes or to the marine environment. Exposures are primarily to the local and regional populations, although the longer-lived radionuclides may also become more widely dispersed.

Table XI.1

Collective effective dose equivalent commitments
per unit activity released
 (10⁻¹² man Sv per Bq)

Radio-nuclide	Source				
	Natural	Nuclear explosions	Nuclear installations		
			Airborne release	Release to fresh water	Marine release
³ H	0.0005	0.0008	0.0009	0.003	0.0008
¹⁴ C	120	120	120	120	120
⁸⁵ Kr	0.0002	0.0002	0.0002	0.0002	0.0002
⁹⁰ Sr	-	0.6	1	1	0.00006
¹²⁹ I	-	30000	30000	30000	30000
¹³¹ I	-	0.00009	0.4	0.009	0.00006
¹³⁷ Cs	-	2	5	0.6	0.001
²²² Rn	0.001	-	0.001	-	-
²³⁸ Pu	-	10	100	0.3	0.002
²³⁹ Pu	-	10	100	0.3	0.002
²⁴⁰ Pu	-	10	100	0.3	0.002
²⁴¹ Pu	-	0.3	2	0.pp3	0.00002
²⁴¹ Am	-	3	200	10	0.06

These values are only approximations of a very generalized nature. The text contains a full discussion of limitations and of the specific assumptions utilized.

484. Release of unit activity of a radionuclide to the environment generally results in the lowest collective effective dose equivalent commitment when the release is to the marine environment. In this case, for the generalized situation considered here, transfer of radionuclides to man occurs only through ingestion of fish and shellfish. There may, however, be important specialized pathways of transfer in more specific release circumstances.

485. Somewhat greater collective doses result from releases to freshwater systems, particularly if the water is subsequently used for drinking. Transfer of radionuclides to fish may also be somewhat more in fresh water than in the ocean.

486. The largest doses generally result from airborne releases. The radionuclides in air may be inhaled, although the contributions to the dose from this pathway are usually small. More importantly, the deposited radionuclides are available for incorporation into the general diet. Long-term transfer of the radionuclides to man may then occur.

487. The greatest contributors to the collective dose commitments per unit activity released are the longest-lived radionuclides. Specifically, the very low dose rates from ^{129}I result in substantial collective doses over the millions of years mean radioactive lifetime of this radionuclide. The same is true of ^{14}C and some isotopes of plutonium, for which the mean radioactive lifetimes are of the order of thousands of years. Of course, the validity of assessments which involve such long-term projections must be questioned. These results can not be related to equilibrium dose rates to the world's population. For this purpose, limiting the integration periods to the duration of the practices, (for example a few hundred years at the most for nuclear power production) obtaining the incomplete collective dose commitments, has some merit.

C. EFFECTS EVALUATION

488. The final step in evaluating the consequences of releasing radionuclides into the environment, once exposures have been determined, is to estimate the health effects. A great deal of study has been made of radiation exposure-response relationships and risk estimates have been formulated for the various effects. There is uncertainty, however, in knowing whether the risk estimates, which are generally only obtainable at higher doses and dose rates, will

Other publications in the IAEA Safety Series and those of the other sponsoring organizations should be consulted for practical detailed guidance in implementing these Basic Safety Standards.

For IAEA operations or operations undertaken with the assistance of IAEA, WHO and ILO, this publication should be applied in the light of relevant national rules and regulations.

Section I

OBJECTIVE AND SCOPE OF THE BASIC SAFETY STANDARDS

- 101 The objective of these standards is to provide guidance for the protection of man from undue risks of the harmful effects of ionizing radiation, while still allowing beneficial practices involving exposure to radiation.
- 102 These standards shall apply to the introduction of sources or practices and to existing sources or practices involving exposure to ionizing radiation subject to control by the competent authority.

Section IV

SYSTEM OF DOSE LIMITATION

- 401 Doses resulting from sources and practices involving exposure to ionizing radiation or to radioactive substances shall be restricted by a system of dose limitation which shall include justification of the practice, optimization of radiation protection and annual dose equivalent limits.

Justification of a practice

- 402 In order to prevent unnecessary exposure, no practice involving exposure to ionizing radiation shall be authorized by the relevant competent authorities unless the introduction of the practice produces a positive net benefit.

Optimization of radiation protection

- 403 The design, plan and subsequent use and operation of sources and practices shall be performed in a manner to ensure that exposures are as low as reasonably achievable, economic and social factors being taken into account.

Dose limitation

A. GENERAL

Limits and reference levels

404 Limits and reference levels are two different concepts in radiation protection. A limit is the value of a quantity which must not be exceeded. A reference level is the value of a quantity which is used to determine a particular course of action; it is not a limit.

405 The limits used in radiation protection are as follows:

(1) *Primary dose equivalent limits.* The primary limits relate to the dose equivalent (para.912.2), effective dose equivalent (para.912.3), committed dose equivalent or committed effective dose equivalent (para.912.5), depending on the exposure circumstances (see also Annex IV, paras 116–119). These limits apply to an individual or, in the case of exposure of the public, to the critical group (see para.908).

(2) *Secondary limits* are needed when the primary dose limits cannot be applied directly. In the case of external exposure, secondary limits may be expressed in terms of dose equivalent index, $H_{1,d}$ or $H_{1,s}$ (see para.912.7). In the case of internal exposure, secondary limits may be expressed in terms of annual limits on intake (see para.903).

(3) *Derived limits* are related to the primary limits by a defined model such that if the derived limits are observed, it is likely that the primary limits would also be observed.

(4) *Authorized limits*

(a) Limits for any quantity may be specified by the competent authority or by the management of an installation and are called authorized limits. These should generally be lower than the primary or derived limits. When authorized limits are specified by the management, they are designated as operational limits.

(b) Authorized primary and secondary limits shall be specified by the competent authority to apply to individual persons, irrespective of the radiation's source. In addition, for particular installations or practices, the competent authority may specify authorized limits for various quantities, relating to, for example, the sources of radiation, or the environment or the exposed individuals. Such authorized limits take precedence over derived limits. Operational limits shall not exceed authorized

be valid for the low level chronic exposures of environmental situations.

489. It is not the intention of this document to provide detailed guidance regarding effects evaluations. This requires more detailed specifications of the radionuclides present in the sources and the amounts released and closer consideration of the environmental conditions and the consequent exposures to the population involved. Only a brief summary has been presented (Chapter X) of the main aspects of radiation effects and of the considerations involved in assigning risk estimates.

490. In the generally accepted philosophy of radiation protection, all exposures are considered to increase the risk of harmful effects. Increased risks are only justified when balanced by net positive benefits from the radiation operations. The conceptual basis for measuring benefits and accounting for acceptability of risks are topics under review by national and international bodies.

491. The first part of the procedure of assessing the consequences of releasing radionuclides into the environment - the specification of sources, of environmental behaviour of radionuclides and of the consequent exposures - appears, in general, to be well founded, as shown by the reviews of radionuclide behaviour and dosimetry of this document. This contributes in a general sense to the establishment of health criteria for these radionuclides. The usual precautions must be expressed in applying general criteria to specific situations. Considerable judgement is always required to make meaningful evaluations to serve as proper guides for future actions. It should also be appreciated that the results of radiological assessments are but one aspect of the considerations on which decisions must be made. The better knowledge that has developed on these aspects should not unduly condition other factors, such as socio-economic considerations, which are also of importance in the rational selection of various possible options.

ANNEX

Excerpts from "Basic Safety Standards for Radiation Protection 1982 Edition", Safety Series No. 9, IAEA

INTRODUCTION

The International Atomic Energy Agency, the World Health Organization, the International Labour Organisation and the Nuclear Energy Agency of the OECD have undertaken to provide jointly a world-wide basis for harmonized and up-to-date radiation protection standards. The new Basic Safety Standards for Radiation Protection are based upon the latest recommendations by the International Commission on Radiological Protection (ICRP) which are essentially contained in its Publication No. 26. These new Basic Safety Standards have been elaborated by an Advisory Group of Experts which met in Vienna from 10-14 October 1977, from 23-27 October 1978 and from 1-12 December 1980 under the joint auspices of the IAEA, ILO, WHO and the Nuclear Energy Agency of the OECD. Comments on the draft Basic Safety Standards received from Member States and relevant organizations were taken into account by the Advisory Group in the process of preparation of the revised Basic Safety Standards for Radiation Protection, which are published by the IAEA on behalf of the four sponsoring organizations.

One of the main features of this revision is an increased emphasis on the recommendation to keep all exposures to ionizing radiation as low as reasonably achievable, economic and social factors being taken into account; consequently, radiation protection should not only apply the basic dose limits but also comply with this recommendation. Detailed guidance is given to assist those who have to decide on the implementation of this recommendation in particular cases. Another important feature is the recommendation of a more coherent method for achieving consistency in limiting risks to health, irrespective of whether the risk is of uniform or non-uniform exposure of the body.

This publication is intended for use by the appropriate competent authorities in the Member States of the sponsoring organizations and also to provide general guidance for organizations representing employers or workers. The main text, based on ICRP recommendations, has been written in the form of regulations. For some competent authorities it may serve as a regulatory basis for the radiation protection of workers and members of the public, while other competent authorities may wish to refer to this text and make regulations more specifically adapted to their particular needs and conditions.

Annex IV to the publication has not been written in the form of regulations and is intended to provide supplementary information and guidance only.

The sponsoring organizations recognize that the ICRP's recommendations cannot be converted into regulations without loss of flexibility in their application to individual situations. However, this loss of flexibility should be minimized, both in the formulation of the regulations and in their administration, to the greatest extent compatible with the nature of the problem and with good regulatory practice.

limits. When a limit is expressed as an average value over some period of time, it is understood that the actual value of the limited quantity over shorter periods of time may show considerable fluctuations.

406 The reference levels used in radiation protection are as follows:

(1) *Recording level* is a level defined by the competent authorities for dose equivalent or effective dose equivalent or intake above which the information is of sufficient interest from a radiation protection point of view to be worth recording and keeping.

(2) *Investigation levels* are defined as values of dose equivalent or effective dose equivalent or intake above which the results are considered sufficiently important to justify further investigations.

(3) *Intervention levels* are usually specified for use in abnormal situations (para.201.2). Such a level is specified in advance by a competent authority or management of an installation, so that if the value of a quantity does not exceed or is not predicted to exceed the intervention level, then it is highly improbable that intervention will be warranted.

(4) A reference level may be established for any quantity used in radiation protection whether or not there is a limit for the quantity.

B. ANNUAL DOSE EQUIVALENT LIMITS

407 No individual shall be exposed, as a result of controlled sources and practices, in excess of the limits presented in paras 411- 413 and 417-419.

408 The dose equivalent limits do not apply to doses incurred from medical exposures of patients or from natural background radiation:

(1) Except as required in para. 602 where dose equivalent limits as defined by the competent authority shall apply to medical exposure of patients in research applications of ionizing radiation or radioactive substances, and

(2) Except as required in para. 706 for cases of technologically enhanced exposure to natural sources of radiation for which the competent national authority gives special requirements.

409 There are separate limits for workers (paras 411-417), and for members of the public (paras 418, 419).

410 The limits for the control of stochastic effects (Annex IV, paras 101, 104--107) are expressed in terms of effective dose equivalents (para.912.3) and the limits for the prevention of non-stochastic effects (Annex IV, paras 102, 103) are expressed in terms of dose equivalent (912.2). Limits for the annual effective dose equivalent, and, in the case of particular organs and tissues, annual dose equivalents, are specified in paras 411 and 412 for workers and in paras 418 and 419 for members of the public respectively. Where limits are expressed in terms of effective dose equivalent they shall apply to the sum of the effective dose equivalents resulting from external exposures during one year and the lifetime¹ effective dose equivalents resulting from the intake of radionuclides during that year.

Workers

411 The limit for the annual effective dose equivalent for workers is 50 mSv (5 rem). In addition, the annual dose-equivalent limit for the individual organs and tissues of workers is 500 mSv (50 rem) except in the case of the lens of the eye; for the lens the limit is 150 mSv (15 rem).

Women of reproductive capacity

417 No separate provisions are required for women of reproductive capacity except that any necessary exposure should be as uniformly distributed with time as is practicable. The purpose of this provision is the protection of the embryo before a pregnancy is known. When a woman is known to be pregnant she should work only in Working Condition B (para. 507.2).

Members of the public

418 The limit for the annual effective dose equivalent for members of the public is 5 mSv (0.5 rem). The annual dose equivalent limit for the individual organs and tissues of members of the public is 50 mSv (5 rem). These dose limits are to be applied to the critical group of the population.

419 When the same individual members of the public could otherwise be exposed at or near the annual effective dose equivalent limit for prolonged periods (many years) it would be prudent to take measures to restrict their lifetime effective dose equivalent to a value corresponding to an annual average of 1 mSv (0.1 rem).

¹ For workers this period shall be taken to be 50 years.

C. SECONDARY LIMITS FOR OCCUPATIONAL EXPOSURE

420 In order to demonstrate compliance with the primary limits for occupational exposure given in paras 411 and 412, limits of dose equivalent indices and annual limits on intake (ALI) may be used. Values of ALI are given in Annex III.

421 When both external and internal exposures are received in a given year, the annual dose limits will not be exceeded if both the following conditions are met:

$$\frac{H_{I,s}}{500 \text{ (mSv)}} \leq 1$$
$$\frac{H_{I,d}}{50 \text{ (mSv)}} + \sum_j \frac{I_j}{I_{j,L}} \leq 1,$$

where $H_{I,s}$ is the shallow dose-equivalent index, $H_{I,d}$ is the deep dose-equivalent index, I_j is the annual intake of radionuclide j , and $I_{j,L}$ is the annual limit on intake for radionuclide j .

D. SECONDARY LIMITS AND QUANTITIES FOR MEMBERS OF THE PUBLIC

422 For members of the public, the effective dose equivalent can be estimated by use of the equation in para. 912.3 (using the same weighting factors $\{w_T\}$ as used for workers). In computing the dose equivalent incurred from intake of radionuclides account shall be taken of the biological, metabolic parameters and other factors such as food habits, demographic distribution and land use that are characteristic of the critical group.

423 In cases involving exposure of a critical group of adults only, one-tenth or one fiftieth² of the relevant ALI value given in Annex III may be used where appropriate as the secondary quantity which corresponds to or is less than the dose limit given in para. 418 and the value given in para. 419. In other cases, when the critical group involves infants or children, one hundredth³ of the relevant ALI value given in Annex III may be used where appropriate as the secondary quantity which corresponds to or is less than the dose limit given in para. 418 and the average annual value given in para. 419, or special assessments shall be made (see Annex IV, para. 500).

² The value of the fraction is a provisional one pending the publication of the ICRP values.

³ The factor of $\frac{1}{100}$ could be used if for some reason it is not possible to calculate an individual ALI but it should be recognized that this might be overcautious in some circumstances. For purposes of optimization, such an oversimplified ALI value should not be used, as a realistic value is required.

- 424 In the case of external exposure the dose equivalent indices may be used, where appropriate, as relevant quantities for the secondary limits, applying the annual effective dose equivalent limit to the deep dose equivalent index $H_{I,d}$ and the annual dose equivalent limit for skin to the shallow dose equivalent $H_{I,s}$.

Section VII

EXPOSURES OF THE PUBLIC, EXCLUDING MEDICAL EXPOSURES

- 701 Practices involving radiation exposures of members of the public shall only be carried out within the requirements of the system of dose limitation. The dose limits for members of the public shall apply to the dose in "critical groups" (para. 908), the selection of which shall require the approval of the competent authority.
- 702 In the case of continuing practices, the dose commitment from each year of operation shall be limited in a manner such that the future annual doses to critical groups (para. 908), at the time when they reach their maximum value⁵, will not exceed the appropriate dose limits. In applying this limitation to the dose commitment from one year of operation, sufficient margin shall be provided to take account of other practices exposing the given critical groups.
- 703 The management responsible for practices which lead to exposure of members of the public shall ensure that these comply with all relevant requirements of the competent authority aimed at providing protection to the public.

Radioactive releases to the environment

- 704 The release of radioactive substances to the environment at levels above the exemption limits set by the competent authority shall require prior authorization by that authority. Pre-operational studies shall be carried out to identify the critical group and critical pathways in a manner which satisfies the competent authority. The management of any installation releasing radioactive substances to the environment shall be responsible for establishing control of the releases in order to respect

⁵ See: Principles for Establishing Limits for the Release of Radioactive Materials into the Environment, Safety Series No.45, IAEA, Vienna (1978) with Annex 1982.

the authorized limits of release. In order to show this compliance it is responsible for carrying out adequate monitoring and accounting of the radioactive substances released. When the competent authority so requires, effluent monitoring shall be complemented by an appropriate environmental monitoring programme.

Exposure from consumer products

- 705 The competent authority shall regulate the introduction of consumer products involving exposure to radiation, and shall make provisions for the assessment of such products even when their commercialization and use is exempted from these standards. When authorizing the introduction of a new product, its future disposal shall be taken into account.

Technologically enhanced exposures to natural radiation sources

- 706 The principles of justification and optimization generally apply to all procedures that may influence the exposure from natural sources of radiation. Although the dose equivalent limits do not as a rule apply to doses from natural background radiation, the competent national authority may specify under which circumstances administratively controllable, technologically enhanced exposures to natural sources of radiation shall be subject to the dose equivalent limits mentioned in paras 405, 411, 412, 418 and 419, or to other, special dose limitation. Existing high levels of natural background should be assessed on the basis of the expected net benefit of actions to reduce the exposure. The appropriate intervention levels should be decided by the competent authority in each case, recognizing that the dose equivalent limits have little relevance in this case.

Section IX

DEFINITIONS AND EXPLANATION OF TERMS USED

- 902 **activity:** The activity, A , of an amount of radioactive nuclide in a particular energy state at a given time is the quotient of dN by dt , where dN is the expectation value of the number of spontaneous nuclear transformations from that energy state in the time interval dt [ICRU Report 33]:

$$A = \frac{dN}{dt}$$

The special name for the SI unit of activity is becquerel (Bq):
 $1 \text{ Bq} = 1 \text{ s}^{-1}$.

The special unit of activity, curie (Ci), may be used temporarily:
 $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$ (exactly).

903 **annual limit on intake (ALI):** The ALI is a secondary limit for occupational internal exposure (see para. 405.2) and is the smaller value of intake of a given radionuclide in a year by Reference Man [ICRP Publication 23] which would result in either a committed effective dose equivalent of 50 mSv or a committed dose equivalent in the lens of the eye of 150 mSv or in any other organ or tissue of 500mSv (paras 405 and 411). ALI values for radionuclides are given in Annex III.

908 **critical group:** For a given source or group of sources, the group of members of the public whose exposure is reasonably homogeneous and is typical of individuals receiving the highest dose.

909 **derived air concentration (DAC):** The DAC for a given radionuclide is a derived limit (para. 405.3) and is the activity concentration of that radionuclide in air (Bq/m³) which, if breathed by Reference Man [ICRP Publication 23] for a working year of 2000 hours under conditions of light physical activity (breathing rate 1.2 m³/h), would result in an inhalation of one ALI, or the concentration which for 2000 hours of air immersion would lead to the irradiation of any organ or tissue to the appropriate limit.

910 **derived limit:** See **limits**, para. 405.

912 **dose:** A term denoting the quantity of radiation energy absorbed by a medium. Although the terms 'dose' or 'radiation dose' are often used in a general sense, they should usually be qualified, for example as absorbed dose (D), dose equivalent (H), effective dose equivalent (H_E), effective dose-equivalent commitment (H_{E,C}), committed effective dose equivalent (H_{E,50}), or collective effective dose equivalent (S_E), etc. The named terms are defined below.

912.1 **absorbed dose:** The absorbed dose, D, is the quotient of $d\epsilon$ by dm , where $d\epsilon$ is the mean energy imparted by ionizing radiation to matter of mass dm [ICRU Report 33]:

$$D = \frac{d\epsilon}{dm}$$

The special name for the SI unit of absorbed dose is gray (Gy):

$$1 \text{ Gy} = 1 \text{ J} \cdot \text{kg}^{-1}.$$

The special unit of absorbed dose, rad, may be used temporarily:

$$1 \text{ rad} = 10^{-2} \text{ Gy}.$$

- 912.2 **dose equivalent:** The dose equivalent, H , is the product of D , Q and N at the point of interest in tissue, where D is the absorbed dose, Q is the quality factor (see para. 927 and Annex II), and N is the product of all other modifying factors [ICRU Reports 25, 33]:

$$H = DQN$$

The SI unit H is the same as that for D (joule per kilogram).

The special name for the SI unit of dose equivalent is sievert (Sv):

$$1 \text{ Sv} = 1 \text{ J} \cdot \text{kg}^{-1}.$$

The special unit of dose equivalent, rem, may be used temporarily:

$$1 \text{ rem} = 10^{-2} \text{ Sv}.$$

Currently ICRP assigns a value of unity to N .

- 912.3 **effective dose equivalent:** The effective dose equivalent, H_E (see also Annex IV, paras 117 to 120), is defined as:

$$H_E = \sum_T w_T H_T$$

where H_T is the mean dose equivalent in tissue T and w_T is a weighting factor representing the proportion of the detriment from stochastic effects resulting from tissue T to the total detriment from stochastic effects when the body is irradiated uniformly.

The values of w_T are specified by ICRP [ICRP Publication 26] and are:

Tissue	w_T	Tissue	w_T
Gonads	0.25	Thyroid	0.03
Breast	0.15	Bone surfaces	0.03
Red bone marrow	0.12	Remainder	0.30
Lung	0.12		

A value of w_T of 0.06 is applicable to each of the five organs or tissues of the remainder receiving the highest dose equivalents, and the exposure of all other remaining tissues may be neglected. (The following parts of the GI tract - stomach, small intestine, upper large intestine and lower large intestine - are to be treated as four different organs.)

The dose equivalents in hands and forearms, feet and ankles, the skin, and the lens of the eye are not considered in computing the effective dose equivalent. However, to assess the detriment from exposure of population groups due to a small risk of fatal cancer resulting from exposure of the skin, a value of w_T of 0.01 is assigned.

- 912.4 **effective dose-equivalent commitment:** The effective dose-equivalent commitment, $H_{E,C}$, from a given decision or practice is the infinite time integral of the per caput effective dose-equivalent rate, $\bar{H}_E(t)$, for a specified population:

$$H_{E,C} = \int_0^{\infty} \bar{H}_E(t) dt$$

The exposed population is not necessarily constant in size. It is also possible to define a collective effective dose-equivalent commitment. This may be obtained by integration of the collective effective dose-equivalent rate.

- 912.5. **committed effective dose equivalent:** The committed effective dose equivalent, $H_{E,50}$, resulting from an intake of radioactive material into the body is the effective dose equivalent that will be accumulated during the 50 years following the intake:

$$H_{E,50} = \int_{t_0}^{t_0 + 50 \text{ years}} \dot{H}_E(t) dt$$

where $\dot{H}_E(t)$ is the relevant effective dose-equivalent rate from the intake and t_0 is the time of intake. Sometimes it may be necessary to extend the integration time beyond 50 years in order to assess the "lifetime dose".

- 912.6 **collective effective dose equivalent:** The collective effective dose equivalent, S_E , gives a measure of the total health detriment from a given radiation source, and is defined as:

$$S_E = \int_0^{\infty} H_E P(H_E) dH_E$$

where $P(H_E) dH_E$ is the number of individuals receiving an effective dose equivalent between H_E and $H_E + dH_E$ from the given source. The units of S_E are man-sieverts (man·Sv).

- 912.7 **dose-equivalent indices:** For the purpose of these standards, two dose-equivalent indices are defined:
(a) **deep dose-equivalent index, $H_{I,d}$,** at a point is the maximum dose equivalent within the 28 cm diameter core of a 30 cm diameter sphere centred at this point and consisting of material equivalent to soft tissue

with a density of $1 \text{ g}\cdot\text{cm}^{-3}$.

(b) **shallow dose-equivalent index**, $H_{1,s}$, at a point is the maximum dose equivalent within the spherical shell extending from a depth of 0.07 mm to a depth of 1 cm from the surface of a 30 cm diameter sphere centred at this point and consisting of material equivalent to soft tissue with a density of $1 \text{ g}\cdot\text{cm}^{-3}$.

(For a definition of dose equivalent index, see ICRU Reports 25, 33.)

- 915 **exposure**: As used in these Standards, any exposure of persons to ionizing radiation. A distinction is made between:
- (a) **external exposure**, being exposure to sources outside the body;
 - (b) **internal exposure**, being exposure to sources inside the body;
 - (c) **total exposure**, being the sum of the external and internal exposures.
- 916 **ionizing radiation**: Electromagnetic radiation (e.g. X-ray or gamma-ray photons) or corpuscular radiation capable of producing ionization in its passage through matter.
- 919 **natural radiation exposure**: Exposure of persons resulting from natural radioactive substances inside the body and from sources of external radiation including cosmic rays and sources of terrestrial origin, i.e. radionuclides naturally present in the crust of the earth and in air.
- 920 **technologically enhanced natural radiation exposure**: Exposure resulting from natural sources of radiation whose original state has been changed by human activity. Such exposures may result from the utilization of mineral raw materials or industrial discharge products. Examples are utilization of fossil fuel, utilization of fertilizers containing phosphates, operation of certain mines, building materials, air travel.
- 921 **nuclide**: An atomic species characterized by the number of protons and the number of neutrons in its nucleus, and by the energy state of the nucleus.
- 922 **occupational exposure**: Exposure of a worker during a period of work.
- 930 **radiation source**: Substance or apparatus producing or capable of producing ionizing radiation.
- 931 **radioactivity**: For the quantity relating to this phenomenon *see activity*, para. 902.
- 932 **sievert:(Sv)**: SI unit of dose equivalent (see para. 912.2).

Annex III

ANNUAL LIMITS ON INTAKES (ALIs) AND DERIVED AIR
CONCENTRATIONS (DACs) OF RADIONUCLIDES
FOR OCCUPATIONAL EXPOSURE

Introduction

- A.III.1. For each radionuclide a table is given indicating the class of the chemical compound which is to be used for selecting the appropriate ALI value (para. 405.2).
- A.III.2. The ALIs and DACs for inhalation are given for the three ICRP classes¹ (D, W, Y) of radioactive materials which refer to their biological retention in the pulmonary region². This classification applies to a range of biological half-lives for D of less than 10 days, for W from 10-100 days and for Y greater than 100 days.
- A.III.3. The values of ALIs and DACs are those given in the ICRP report entitled Limits for Intakes of Radionuclides by Workers, ICRP Publication 30, Parts 1, 2 and 3.
- A.III.4. The DAC values are derived limits (para. 405(3)) intended to control exposure over prolonged periods of up to one year. The relationship between DAC and ALI is given by:

$$\begin{aligned} \text{DAC} &= \text{ALI}/(2000 \times 60 \times 0.02) \\ &= \text{ALI}/2.4 \times 10^3 \text{ Bq} \cdot \text{m}^{-3} \end{aligned}$$

where 0.02 m^3 is the volume of air breathed at work by 'Reference Man' per minute under working conditions of "light activity" (see ICRP Publication 30).

¹ See also ICRP Publication 30, Parts 1, 2 and 3, where these are described in full.

² See: Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract, a report prepared by the ICRP task group on lung dynamics; also Health Physics 12 (1966) 173-207, reproduced in Inhalation Risks from Radioactive Contaminants, Technical Report Series No. 142, IAEA (1973). Some modifications have however been introduced in ICRP Publication 30, Parts 1, 2 and 3, and these are reflected here.

A.III.5. ALI and DAC values relate to intake by the specified route of entry into the body of the single radionuclide named and include an appropriate allowance for any daughter radionuclides produced in the body during the decay of the parent nuclide. However, intakes which include both parent and daughter radionuclides should be treated by the general method appropriate to mixtures (see main text para. 421).

A.III.6. The values of ALI or DAC do not apply directly when the worker both ingests and inhales a radionuclide, when he is exposed to a mixture of radionuclides or when he is exposed to both internal and external irradiation (see definition of ALI in para. 903). In such a case, the method and summation formula specified in para.421 of the main text shall apply.

A.III.7. In the special case of inhalation of the short-lived daughters of ^{222}Rn and ^{220}Rn , $I_{j,L}$ may be expressed in terms of inhaled potential alpha energy, the annual limit for which is:

0.02 J for the daughters of ^{222}Rn , and
0.06 J for the daughters of ^{220}Rn .

Expressed as time-integrated exposure, in the widely used practical units of working level months (WLM), the limits are:

5 WLM for the daughters of ^{222}Rn , and
15 WLM for the daughters of ^{220}Rn .

A.III.8. The following tables give for various nuclides of each of the elements listed below the annual limits on intake, ALI (Bq), and derived air concentrations, DAC ($\text{Bq}\cdot\text{m}^{-3}$), the figures being based on a forty-hour week. To obtain the ALI expressed in Ci the relevant value should be divided by 3.7×10^{10} . To obtain DAC expressed in $\text{Ci}\cdot\text{m}^{-3}$ the relevant value should be divided by 3.7×10^{10} .

In the following tables, the following abbreviations have been used:

LLI wall	lower large intestine wall
St. wall	stomach wall.

HYDROGEN

Radionuclide		Oral	Inhalation
^3H (Tritiated water)	ALI	3×10^9	3×10^9
	DAC	-	8×10^5
^3H (Elemental tritium)	ALI	-	-
	DAC	-	2×10^{10}

Absorption of tritium through the skin is included in the calculations for DAC.

CARBON

Radionuclide		Oral	Inhalation
		a	b
^{11}C	ALI	2×10^{10}	2×10^{10}
	DAC	-	6×10^6
^{14}C	ALI	9×10^7	9×10^7
	DAC	-	4×10^4

a, b All labelled organic compounds of carbon except carbon monoxide and carbon dioxide.

CARBON MONOXIDE

Radionuclide		Inhalation
^{11}C	ALI	4×10^{10}
	DAC	2×10^7
^{14}C	ALI	6×10^{10}
	DAC	3×10^7

CARBON DIOXIDE

Radionuclide		Inhalation
^{11}C	ALI	2×10^{10}
	DAC	1×10^7
^{14}C	ALI	8×10^9
	DAC	3×10^6

KRYPTON

Radionuclide	Semi-infinite cloud	1000 m ³ room	500 m ³ room	100 m ³ room
⁷⁴ Kr	1 × 10 ⁵	1 × 10 ⁶ (3 × 10 ⁶) Skin	1 × 10 ⁶ (3 × 10 ⁶) Skin	1 × 10 ⁶ (6 × 10 ⁶) Skin
⁷⁶ Kr	3 × 10 ⁵	7 × 10 ⁶	9 × 10 ⁶	2 × 10 ⁷
⁷⁷ Kr	1 × 10 ⁵	2 × 10 ⁶ (3 × 10 ⁶) Skin	2 × 10 ⁶ (4 × 10 ⁶) Skin	2 × 10 ⁶ (7 × 10 ⁶) Skin
⁷⁹ Kr	6 × 10 ⁵	1 × 10 ⁷	2 × 10 ⁷	3 × 10 ⁷
⁸¹ Kr	2 × 10 ⁷	1 × 10 ⁸ (5 × 10 ⁸) Lens	1 × 10 ⁸ (6 × 10 ⁸) Lens	1 × 10 ⁸ (9 × 10 ⁸) Lens
⁸³ Kr ^m	4 × 10 ⁸ (7 × 10 ⁹) Lens	4 × 10 ⁸ (7 × 10 ⁹) Lens	4 × 10 ⁸ (7 × 10 ⁹) Lens	4 × 10 ⁸ (8 × 10 ⁹) Lens
⁸⁵ Kr ^m	8 × 10 ⁵	5 × 10 ⁶ (2 × 10 ⁷) Skin	5 × 10 ⁶ (3 × 10 ⁷) Skin	5 × 10 ⁶ (4 × 10 ⁷) Skin
⁸⁵ Kr	5 × 10 ⁶ (5 × 10 ⁷) Skin	5 × 10 ⁶ (1 × 10 ⁹) Skin	5 × 10 ⁶ (1 × 10 ⁹) Skin	5 × 10 ⁶ (2 × 10 ⁹) Skin
⁸⁷ Kr	2 × 10 ⁶	8 × 10 ⁵ (5 × 10 ⁶) Skin	8 × 10 ⁵ (6 × 10 ⁶) Skin	8 × 10 ⁵ (1 × 10 ⁷) Skin
⁸⁸ Kr	7 × 10 ⁸	2 × 10 ⁶	2 × 10 ⁶	3 × 10 ⁶ (4 × 10 ⁶) Skin

Exposure in a cloud of radioactive noble gas is mainly determined by external irradiation, since dose-equivalent rate from gas absorption in tissue or contained in the lungs will be negligible in comparison with dose-equivalent rate to organs and tissues from external irradiation.

STRONTIUM

Radionuclide		Oral		Inhalation	
		a	b	c	d
⁸⁰ Sr	ALI	2×10^8	2×10^8	4×10^8	5×10^8
	DAC		..	2×10^5	2×10^5
⁸¹ Sr	ALI	9×10^8	9×10^8	3×10^9	3×10^9
	DAC			1×10^6	1×10^6
⁸³ Sr	ALI	1×10^8	8×10^7	3×10^8	1×10^8
	DAC		..	1×10^5	5×10^4
⁸⁵ Sr ^m	ALI	8×10^9	8×10^9	2×10^{10}	3×10^{10}
	DAC		..	9×10^6	1×10^7
⁸⁵ Sr	ALI	9×10^7	1×10^8	1×10^8	6×10^7
	DAC			4×10^4	2×10^4
⁸⁷ Sr ^m	ALI	2×10^9	1×10^9	5×10^9	6×10^9
	DAC			2×10^6	2×10^6
⁸⁹ Sr	ALI	2×10^7 (2×10^7) LLI Wall	2×10^7	3×10^7	5×10^6
	DAC	..		1×10^4	2×10^3
⁹⁰ Sr	ALI	1×10^6 (1×10^6) Bone surface	2×10^7	7×10^5 (8×10^5) Bone surface	1×10^5
	DAC	3×10^2	6×10^1
⁹¹ Sr	ALI	8×10^7	6×10^7	2×10^8	1×10^8
	DAC			9×10^4	5×10^4
⁹² Sr	ALI	1×10^8	1×10^8	3×10^8	2×10^8
	DAC		..	1×10^5	1×10^5

a Soluble salts of strontium.

b SrTiO₃.

c All soluble components except SrTiO₃.

d All insoluble components and SrTiO₃.

IODINE

Radionuclide		Oral	Inhalation
		a	b
¹²⁰ I	ALI	1 × 10 ⁸ (3 × 10 ⁸) Thyroid	3 × 10 ⁸ (5 × 10 ⁸) Thyroid
	DAC	-	1 × 10 ⁵
^{120m} I	ALI	4 × 10 ⁸ (5 × 10 ⁸) Thyroid	8 × 10 ⁸
	DAC	-	3 × 10 ⁵
¹²¹ I	ALI	4 × 10 ⁸ (1 × 10 ⁹) Thyroid	7 × 10 ⁸ (2 × 10 ⁹) Thyroid
	DAC	-	3 × 10 ⁵
¹²³ I	ALI	1 × 10 ⁸ (4 × 10 ⁸) Thyroid	2 × 10 ⁸ (7 × 10 ⁸) Thyroid
	DAC	-	9 × 10 ⁴
¹²⁴ I	ALI	2 × 10 ⁶ (6 × 10 ⁶) Thyroid	3 × 10 ⁶ (1 × 10 ⁷) Thyroid
	DAC	-	1 × 10 ³
¹²⁵ I	ALI	1 × 10 ⁶ (5 × 10 ⁶) Thyroid	2 × 10 ⁶ (8 × 10 ⁶) Thyroid
	DAC	-	1 × 10 ³
¹²⁶ I	ALI	8 × 10 ⁵ (3 × 10 ⁶) Thyroid	1 × 10 ⁶ (4 × 10 ⁶) Thyroid
	DAC	-	5 × 10 ²
¹²⁸ I	ALI	2 × 10 ⁹ (2 × 10 ⁹) St. wall	4 × 10 ⁹
	DAC	-	2 × 10 ⁶

IODINE (cont.)

Radionuclide		Oral	Inhalation
		a	b
¹²⁹ I	ALI	2 × 10 ⁵ (7 × 10 ⁵) Thyroid	3 × 10 ⁵ (1 × 10 ⁶) Thyroid
	DAC	-	1 × 10 ²
¹³⁰ I	ALI	1 × 10 ⁷ (4 × 10 ⁷) Thyroid	3 × 10 ⁷ (7 × 10 ⁷) Thyroid
	DAC	-	1 × 10 ⁴
¹³¹ I	ALI	1 × 10 ⁶ (4 × 10 ⁶) Thyroid	2 × 10 ⁶ (6 × 10 ⁶) Thyroid
	DAC	-	7 × 10 ²
¹³² I	ALI	1 × 10 ⁸ (3 × 10 ⁸) Thyroid	3 × 10 ⁸ (6 × 10 ⁸) Thyroid
	DAC	-	1 × 10 ⁵
^{132m} I	ALI	1 × 10 ⁸ (4 × 10 ⁸) Thyroid	3 × 10 ⁸ (7 × 10 ⁸) Thyroid
	DAC	-	1 × 10 ⁵
¹³³ I	ALI	5 × 10 ⁶ (2 × 10 ⁷) Thyroid	1 × 10 ⁷ (3 × 10 ⁷) Thyroid
	DAC	-	4 × 10 ³
¹³⁴ I	ALI	8 × 10 ⁸ (1 × 10 ⁹) Thyroid	2 × 10 ⁹
	DAC	-	7 × 10 ⁵
¹³⁵ I	ALI	3 × 10 ⁷ (9 × 10 ⁷) Thyroid	6 × 10 ⁷ (2 × 10 ⁸) Thyroid
	DAC	-	2 × 10 ⁴

a All commonly occurring compounds of iodine.

b All compounds of iodine.

CAESIUM

Radionuclide		Oral	Inhalation
		a	b
¹²⁵ Cs	ALI	2 × 10 ⁹ (3 × 10 ⁹) St. wall	5 × 10 ⁹
	DAC	-	2 × 10 ⁶
¹²⁷ Cs	ALI	2 × 10 ⁹	4 × 10 ⁹
	DAC	-	1 × 10 ⁶
¹²⁹ Cs	ALI	9 × 10 ⁸	1 × 10 ⁹
	DAC	-	5 × 10 ⁵
¹³⁰ Cs	ALI	2 × 10 ⁹ (4 × 10 ⁹) St. wall	7 × 10 ⁹
	DAC	-	3 × 10 ⁶
¹³¹ Cs	ALI	8 × 10 ⁸	1 × 10 ⁹
	DAC	-	5 × 10 ⁵
¹³² Cs	ALI	1 × 10 ⁸	1 × 10 ⁸
	DAC	-	6 × 10 ⁴
¹³⁴ Cs	ALI	3 × 10 ⁶	4 × 10 ⁶
	DAC	-	2 × 10 ³
¹³⁴ Cs ^m	ALI	4 × 10 ⁹ (4 × 10 ⁹) St. wall	5 × 10 ⁹
	DAC	-	2 × 10 ⁶
¹³⁵ Cs	ALI	3 × 10 ⁷	4 × 10 ⁷
	DAC	-	2 × 10 ⁴
¹³⁵ Cs ^m	ALI	4 × 10 ⁹	7 × 10 ⁹
	DAC	-	3 × 10 ⁶
¹³⁶ Cs	ALI	2 × 10 ⁷	2 × 10 ⁷
	DAC	-	1 × 10 ⁴
¹³⁷ Cs	ALI	4 × 10 ⁶	6 × 10 ⁶
	DAC	-	2 × 10 ³
¹³⁸ Cs	ALI	7 × 10 ⁸ (1 × 10 ⁹) St. wall	2 × 10 ⁹
	DAC	-	9 × 10 ⁵

a,b All compounds of caesium

PLUTONIUM

Radionuclide		Oral		Inhalation	
		a	b	c	d
²³⁴ Pu	ALI	3×10^8	3×10^8	8×10^6	7×10^6
	DAC	-	-	3×10^3	3×10^3
²³⁵ Pu	ALI	3×10^{10}	3×10^{10}	1×10^{11}	9×10^{10}
	DAC	-	-	5×10^7	4×10^7
²³⁶ Pu	ALI	8×10^5 (1×10^6) Bone surface	6×10^6	7×10^2 (1×10^3) Bone surface	1×10^3
	DAC	-	-	3×10^{-1}	6×10^{-1}
²³⁷ Pu	ALI	5×10^8	5×10^8	1×10^8	1×10^8
	DAC	-	-	5×10^4	5×10^4
²³⁸ Pu	ALI	3×10^4 (5×10^5) Bone surface	3×10^6 (3×10^6) Bone surface	2×10^3 (4×10^2) Bone surface	6×10^2 (6×10^2) Bone surface
	DAC	-	-	9×10^{-2}	3×10^{-1}
²³⁹ Pu	ALI	2×10^5 (4×10^5) Bone surface	2×10^6 (3×10^6) Bone surface	2×10^2 (4×10^2) Bone surface	5×10^2 (6×10^2) Bone surface
	DAC	-	-	8×10^{-2}	2×10^{-1}
²⁴⁰ Pu	ALI	2×10^5 (4×10^5) Bone surface	2×10^6 (3×10^6) Bone surface	2×10^2 (4×10^2) Bone surface	5×10^2 (6×10^2) Bone surface
	DAC	-	-	8×10^{-2}	2×10^{-1}
²⁴¹ Pu	ALI	1×10^7 (2×10^7) Bone surface	1×10^8 (2×10^8) Bone surface	1×10^4 (2×10^4) Bone surface	2×10^4 (3×10^4) Bone surface
	DAC	-	-	4	1×10^1
²⁴² Pu	ALI	3×10^5 (5×10^5) Bone surface	3×10^6 (3×10^6) Bone surface	2×10^2 (4×10^2) Bone surface	6×10^2 (6×10^2) Bone surface
	DAC	-	-	9×10^{-2}	2×10^{-1}
²⁴³ Pu	ALI	6×10^8	6×10^8	1×10^9	1×10^9
	DAC	-	-	5×10^5	6×10^5

PLUTONIUM (cont.)

Radionuclide		Oral		Inhalation	
		a	b	c	d
²⁴⁴ Pu	ALI	3 × 10 ⁶ (5 × 10 ⁵)	3 × 10 ⁶ (3 × 10 ⁶)	2 × 10 ² (4 × 10 ²)	6 × 10 ² (6 × 10 ²)
	DAC	Bone surface	Bone surface	Bone surface 9 × 10 ⁻²	Bone surface 2 × 10 ⁻¹
²⁴⁵ Pu	ALI	8 × 10 ⁷	8 × 10 ⁷	2 × 10 ⁸	2 × 10 ⁸
	DAC	-	-	7 × 10 ⁴	6 × 10 ⁴

- a For all commonly occurring compounds of plutonium.
- b For oxides and hydroxides of plutonium.
- c All commonly occurring compounds of plutonium except those in d.
- d PuO₂.

It should be noted that data are reported which indicate a much higher gastro-intestinal absorption for certain compounds of plutonium that are unlikely to be encountered in occupational exposure, e.g. hexavalent plutonium compounds, citrates, and other organic complexes; absorption is also increased in the very young.

AMERICIUM

Radionuclide		Oral	Inhalation
		a	b
²³⁷ Am	ALI	3×10^9	1×10^{10}
	DAC	-	4×10^6
²³⁸ Am	ALI	1×10^9	1×10^8 (2×10^8) Bone surface
	DAC	-	4×10^4
²³⁹ Am	ALI	2×10^8	5×10^8
	DAC	-	2×10^5
²⁴⁰ Am	ALI	8×10^7	1×10^8
	DAC	-	4×10^4
²⁴¹ Am	ALI	5×10^4 (9×10^4) Bone surface	2×10^2 (4×10^2) Bone surface
	DAC	-	8×10^{-2}
²⁴² Am ^m	ALI	5×10^4 (9×10^4) Bone surface	2×10^2 (4×10^2) Bone surface
	DAC	-	8×10^{-2}
²⁴² Am	ALI	2×10^8	3×10^6 (3×10^6) Bone surface
	DAC	-	1×10^3
²⁴³ Am	ALI	5×10^4 (9×10^4) Bone surface	2×10^2 (4×10^2) Bone surface
	DAC	-	8×10^{-2}
²⁴⁴ Am ^m	ALI	2×10^9 (3×10^9) Bone surface	1×10^8 (2×10^8) Bone surface
	DAC	-	6×10^4

AMERICIUM (cont.)

Radionuclide		Oral	Inhalation
		a	b
²⁴⁴ Am	ALI	1×10^8	6×10^6 (1×10^7)
	DAC	-	Bone surface 3×10^3
²⁴⁵ Am	ALI	1×10^9	3×10^9
	DAC	.	1×10^6
²⁴⁶ Am ^m	ALI	2×10^9 (2×10^9) St. wall	6×10^9
	DAC	-	3×10^6
²⁴⁶ Am	ALI	1×10^9	4×10^9
	DAC	.	2×10^6

a, b All compounds of americium.

It should be noted that greater gastro-intestinal absorption might be expected for complexed forms of the element and that enhanced absorption has been reported in very young rats.

EXPOSURES OF THE PUBLIC, EXCLUDING MEDICAL EXPOSURES

(see also Section VII of main text)

General

A.IV.500. *Individual members of the general public are not normally subject to supervision. Therefore, exposure of the members of the public is mainly limited through control at the source. This differs from control of exposure of the workers, who are subject to supervision. In assessing and controlling exposure of the public it is important to recognize that there will often be differences between the public and worker population with respect to age distribution, metabolic parameters, critical pathways, intake mechanisms, and so forth. Thus for the special cases where there is internal exposure of infants or children, due to the more restrictive biological and metabolic parameters used in these cases, the intake values should be chosen as follows:*

One-hundredth⁸ of the relevant ALI value in Annex III may be used where appropriate as the relevant quantity for the secondary limit (see para. 423 of main text) in cases where the exposure is not likely to be repeated over long periods of time. This fraction is made up of two components. One component is one-tenth and is to be applied to ALI values to take account of the lower dose limit for members of the public stated in para. 418 of the main text. The other component is an additional one-tenth to take account of the smaller mass of organs of the critical infant or child.

One hundredth⁹ of the relevant ALI value in Annex III may be used as the relevant quantity for the average intake value (see para. 423 of main text) in cases where the exposure of individuals of the critical group is extended for long periods of time (up to the entire lifetime of the individual). This fraction is made up of two components. One component is one-fiftieth and is to be applied to the ALI values to take account of the lower annual average value for members of the public. The second component is one half and is to take account of the mass of organs averaged over a lifetime, as the organ masses increase as individuals become adult.

^{8,9} The value of the fraction is a provisional one pending the publication of the ICRP values.

The factor of one-hundredth could be used if for some reason it is not possible to calculate an individual ALI but it should be recognized that this might be overcautious in some circumstances. For purposes of optimization, such an oversimplified ALI value should not be used, as a realistic value is required.

However, the annual effective dose equivalent limit and annual average values are those given in paras 418 and 419 in the main text, i.e. 5 mSv (0.5 rem) when exposure is limited to a short period of the individual's lifetime or an average of 1 mSv (0.1 rem) over the entire lifetime of the individual.

A.IV.501. The basic principles of justification, optimization and individual dose limitation are also applicable in the limitation of exposure of the members of the public. Therefore, the limitation of exposure of the public would take into account:

- (1) Individual-related radiation protection criteria. The dose limits for the members of the public should be respected.
- (2) Source-related radiation protection criteria. Each source or practice should be justified and subject to the optimization principle.

A.IV.502. For source-related protection assessments it would be useful to develop long-term forecasts of the trends of various contributions to the total collective and individual dose equivalent from various sources of exposure.

A. Individual-related radiation protection criteria

A.IV.503. The individual-related dose assessment aims at the estimation of the total radiation exposure received by given individuals from all sources and practices involving radiation for which the individual dose limits apply.

A.IV.504. Dose equivalent limits for workers and for members of the public are not intended for design or planning purposes, but are the lower boundary of the range of values that is generally considered to be unacceptable. Values above the limits are to be specifically avoided, but values below the limits are not automatically permitted. In this sense, the limits are constraints for the optimization process.

A.IV.505. When several practices are likely to contribute significantly to the exposure of the same exposed population, either simultaneously or successively, the determination of the critical groups must take account of these separate contributions.

- A.IV.506. It must be recognized that the group which is critical for a multitude of sources is not necessarily identical to any of the groups which are critical for the individual sources. In general, the critical group for a given source or group of sources is the group of individuals whose exposure is relatively homogeneous and typical of individuals receiving the highest dose. It is independent of political boundaries.
- A.IV.507. Because the dose limits apply to the combined exposure from many practices, they cannot be used as operational limits. In fact exposures at the limit from one single practice would leave no scope for other exposures of the same critical group. Furthermore, each year of a continuing practice can, in some cases, cause exposures which will be delivered in the future and which add to the contributions of other years.
- A.IV.508. It is possible to control the future average effective dose equivalent from all practices by applying practical limits to the effective dose-equivalent commitment per unit practice for each of the practices. This concept has been used in some countries to set authorized limits to the collective effective dose-equivalent commitment per MW · year of electrical energy produced by nuclear means.
- A.IV.509. The use of environmental models allows the establishment of relationships between releases, environmental levels and resulting doses, making it possible to relate releases of radioactive materials into the environment to the resulting effective dose equivalents in the critical groups. The sequence of events leading from the primary release of radioactive substances to irradiation of human tissues can be schematically represented by compartment models, in which the rates of transfer of radioactivity between compartments are specified by constants or by time functions. The use of compartment models, even if they are complex, implies considerable simplification of the real transfer process of the network of pathways linking the release or radioactive material to human exposure¹⁰.

¹⁰ International Atomic Energy Agency, *Generic Models and Parameters for Assessing the Environmental Transfer of Radionuclides from Routine Releases: Exposures of Critical Groups*, Safety Series No. 57, IAEA, Vienna (1982).

A.IV.510. For individual-related assessments, regional and world-wide contributions from other sources and practices have to be taken into account in so far as the exposure affects the same critical group. In addition, allowance has to be made for exposures of other types, taking into account present and future conditions, to the same critical group.

B. Source-related radiation protection criteria

A.IV.511. Source-related assessments ideally aim at the estimation of the total radiation detriment from a given source or practice. This detriment, when it involves a small individual dose lower than the dose limits, is composed only of stochastic effects; under the proportionality assumption the health component of the total detriment is proportional to the collective effective dose-equivalent commitment.

A.IV.512. The general methodology for assessing the collective dose-equivalent commitment is given in the IAEA Safety Series¹¹.

C. Control of exposures

A.IV.513. Control of exposure that might result from a source or practice should involve the examination and testing of protective arrangements, including:

- (1) Examination and approval of proposals involving radiation risk. These should include pre-operational studies if required.
- (2) Acceptance into service of new installations and modifications of existing installations with regard to protection against any radiation exposure or contamination liable to extend beyond the perimeter of the installation, taking into account demographic, meteorological, geological, hydrological and ecological conditions.
- (3) Checking the effectiveness of protective devices, establishment of authorized limits for discharges and, wherever necessary, the establishment of emergency plans.

A.IV.514. All releases of radioactive substances to the environment including the disposal of solid wastes should be subject to

¹¹ See: Principles for Establishing Limits for the Release of Radioactive Materials into the Environment, Safety Series No.45, IAEA, Vienna (1978).

authorization and control by the competent national authority unless specifically exempted. In setting such limits, the authorities should apply, with an appropriate degree of detail, the process of optimization and should take account of present and foreseeable future sources of exposure. In any event the authorized limits of release as fixed by the national authorities should not exceed limits derived using the methodology outlined in para. A.IV.509 above.

A.IV.515. Direct external exposure from sources used for industrial, medical or scientific purposes should be restricted. It may be necessary to define an area of restricted access or use in the vicinity of the source.

D. Monitoring and surveillance programmes for the individual and collective dose-equivalent commitments

A.IV.516. Surveillance should be established:

- (1) To ensure compliance with authorized limits
- (2) To assess dose equivalents of members of the public from the sources under consideration, and
- (3) To evaluate trends of exposure levels in the environment.

Monitoring of exposure of the population can be achieved by:

- (a) Monitoring of the source, which would include monitoring of discharges and estimating the dose rate in the case of direct exposure of members of the public. The monitoring of the source constitutes the basic control requirement in contrast to environmental monitoring of pathways.
- (b) Monitoring of environmental pathways, to ensure that the critical groups of the population are adequately protected. Surveillance should be carried out in all places where critical groups may be present. Environmental monitoring should also be done in other areas where people could be affected by a given source. It should be emphasized, however, that the levels in the environment are usually very low and thus primary reliance is based on monitoring of the source.

A.IV.517. The results of radiation surveillance should be examined with a view to verifying or modifying the assumptions and environmental models used in deriving release limits and estimating dose commitments.

A.IV.518. Records relating to the measurement of external exposure and radioactive contamination, and to the estimates of doses received by the population, should be properly filed. All values of releases of radioactive material to the environment should be recorded in conformity with the requirements of the competent national authorities.

A.IV.519. Before the establishment of a practice involving exposure of members of the public, it may be necessary to carry out appropriate pre-operational studies to provide the information needed for the relevant exposure pathway parameters, and for planning the monitoring required during operations.