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## Tritium—An Analysis of Key Environmental and Dosimetric Questions

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Technical Information Center  
P.O. Box 62, Oak Ridge, Tennessee 37830  
Printed Copy A07 ; Microfiche A01

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ORNL/TM-6990  
Dist. Categories UC-79b, -79c

Contract No. W-7405-eng-26

ORNL/TM--6990

DE88 003141

Health and Safety Research Division

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Date Published: May 1980

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## ACKNOWLEDGEMENTS

The authors wish to acknowledge the assistance of George Sherwood (DOE), and Keith Eckerman, Laura McDowell-Boyer, A. R. Olsen, D. E. Dunning, Jr., and F. O. Hoffman (ORNL), for their critical review and helpful suggestions regarding the content of this document.

We also thank A. G. Croff who arranged for ORIGEN computer runs to provide spent fuel compositions for calculation of source terms, and J. W. Elwood who assisted in the documentation of this report.

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## ABSTRACT

Tritium continues to be a major constituent of radioactivity released to the environment by nuclear reactors and proposed nuclear fuel reprocessing plants. The purpose of this study is to summarize new theoretical and experimental data that may affect the assessment of environmental releases of tritium, and to analyze the significance of this information in terms of the dose to man.

Calculated doses resulting from tritium releases to the environment are linearly dependent upon the quality factor chosen for tritium beta radiation. A value of 1.0 is currently being used as the quality factor for tritium betas and is based upon a recommendation by the International Commission on Radiological Protection (ICRP) in 1969. Based upon our study of data published since the ICRP recommendation, it is concluded that a reevaluation of the tritium quality factor by the ICRP is needed and that a value of 1.7 would seem to be more justifiable.

Several methodologies exist for evaluating exposures to man from tritium released to the environment. An analysis of four widely accepted methodologies indicates that they vary considerably in complexity yet the increased complexity does not necessarily mean a more accurate estimate of dose. A new model is proposed, based primarily upon the approach recommended by the National Council on Radiation Protection and Measurements, that maintains both simplicity and the ability to incorporate available site-specific data.

Several key parameters influence uncertainties in calculated dose from environmental tritium. Employing a "typical" LMFBR reprocessing facility source term, a "base case" dose commitment to total body (for a maximally exposed individual) was calculated to be  $4.0 \times 10^{-2}$  mSv, with  $3.2 \times 10^{-2}$  mSv of the dose due to intake of tritium. These results were used as the basis for examination of the following key variables: (1) use of U.S. regional (as opposed to U.S. average) absolute humidity values which vary the tritium dose to total body over a range of  $1.6 \times 10^{-2}$  to  $6.3 \times 10^{-2}$  mSv; (2) choice of a site-specific drinking water



dilution coefficient which for tritium may increase total-body dose by as much as 80%, under certain severe conditions; and (3) recent indications that rates of tritium production by ternary fission in fast reactor fuels may be an order of magnitude greater than previously estimated, which results in parallel increases in predicted dose. The influence of these variables, and the potential for significant tritium population doses due to world-wide implementation of nuclear fuel cycles, suggests the need for careful evaluation of tritium control technologies.

The study analyzes models which exist for evaluating the buildup of global releases of tritium from man-made sources. Our scenarios for the release of man-made tritium to the environment and prediction of collective dose commitment to future generations suggest that the dose from nuclear weapons testing will be less than that from nuclear energy even though the weapons source term is greater than that for any of our energy scenarios.

## INTRODUCTION

*John E. Till*

Tritium continues to be a radionuclide of major interest in the evaluation of the radiological impact associated with nuclear facilities. This interest has intensified recently for a number of reasons: (1) recognition of the significant contribution made by tritium to the total dose received by individuals living near certain nuclear plants; (2) the need for evaluation of the impact of potentially increased tritium releases from advanced fission energy systems; (3) the need for intercomparison of several complex methodologies for evaluating the dose from tritium released to the environment; (4) recent data suggesting that the biological effects from protracted low-level exposure to tritium have previously been underestimated; and (5) the potential for increases in the global release rate of tritium, with concurrent evaluation of potential health effects to present and future generations. The purpose of this study is to summarize new theoretical and experimental data that may affect the assessment of environmental releases of tritium, and to analyze the significance of this recent information in terms of the dose to man.

The report is divided into five chapters. Chapter 1 reviews the major sources of tritium entering the environment, then focuses more specifically on the potential for tritium releases from current and anticipated nuclear reactor fuel cycle sources. These data are utilized in later chapters as the basis for radiological dose calculations. Chapter 2 reviews four major methodologies available for the estimation of radiological dose from tritium in the environment and provides recommendations regarding choice of an appropriate model. Chapter 3 discusses the uncertainties associated with four parameters that have the potential for significantly modifying doses calculated for environmentally dispersed tritium. Chapter 4 quantitatively compares the impact of changes in the value of these parameters, using a set of radionuclide release rates typifying a modern design breeder reactor fuel reprocessing facility, but, for purposes of comparison, excluding the use of

tritium confinement technology. Chapter 5 develops in detail the tritium release rates associated with a global nuclear fuel cycle including a mix of power reactor types, then intercompares global tritium models and implements a specific model to estimate world population doses from global tritium releases.

The report summarizes current information regarding the impact of chronic exposure to tritium in the environment and provides both local and global perspectives on the need for containment of tritium at nuclear fuel reprocessing facilities.

## 1. PRODUCTION AND RELEASE OF TRITIUM TO THE ENVIRONMENT

*John E. Till, Elizabeth L. Etnier and E. S. Bomar*

### 1.1 Production of Tritium in Nature

Tritium is produced through natural processes involving interactions between cosmic rays and gases of the upper atmosphere. This rate may be enhanced through increased particle accretion which occurs during peak periods of solar flare activity. Literature concerning these sources is reviewed by Nir et al. (1966) and by Jacobs (1968). Nir et al. (1966) estimate the tritium production rate of  $0.19 \pm 0.09$  triton\*/ $\text{cm}^2\text{-sec}^\dagger$  due to cosmic rays, yet they calculate a material balance production rate of 0.5 triton/ $\text{cm}^2\text{-sec}$  overall. This discrepancy in natural tritium production is attributed to solar accretion. Flamm et al. (1962) estimate this latter phenomenon could account for an additional 0.4 triton/ $\text{cm}^2\text{-sec}$ .

The natural production rate of tritium found by Nir et al. (1966) falls within the range of reported values cited by Jacobs (1968) of 0.12 to 2.0 triton/ $\text{cm}^2\text{-sec}$  with a most probable value between 0.5 to 1.0 triton/ $\text{cm}^2\text{-sec}$ . These most probable values yield an annual production rate of 4 to 8 MCi (0.15 to 0.30 EBq) and a steady-state tritium inventory of 70 to 140 MCi (2.6 to 5.2 EBq).

The National Council on Radiation Protection and Measurements in NCRP 62 (1979a) considers the above estimates and recommends the use of 4 MCi/year (0.15 EBq/year) as an annual production rate and 70 MCi (2.6 EBq) as the natural level of the tritium world inventory. Assuming a mean surface area for the earth of  $5.096 \times 10^8 \text{ km}^2$ , a production rate of 0.52 triton/ $\text{cm}^2\text{-sec}$  gives an annual production of 4 MCi (0.15 EBq).

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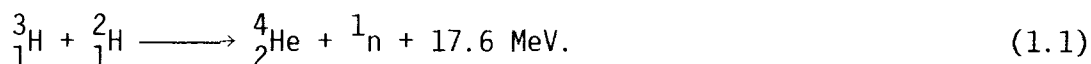
\* Triton = an atom of tritium.

† Per square centimeter of the earth's surface at sea level.

## 1.2 Production of Tritium by Man

### 1.2.1 Production and release of tritium from nuclear explosives

Tritium is produced in nuclear explosives by the interaction of fast neutrons with constituents of air. Relatively small amounts of tritium are created in fission weapons; however, greater quantities are produced in thermonuclear devices. The energetic interaction between deuterium and tritium, typical of the reaction taking place in a fission-fusion bomb, is represented by the following reaction



This reaction proceeds much more rapidly than other thermonuclear reactions and most of the tritium present initially is consumed in the explosion. The unreacted tritium, however, is released to the atmosphere following detonation (Glasstone and Dolan, 1977). Tritium is also produced by the interaction of high energy neutrons with nitrogen nuclei in the atmosphere and by  ${}^2\text{H}(\text{n},\gamma){}^3\text{H}$ , and  ${}^6\text{Li}(\text{n},\alpha){}^3\text{H}$  reactions with lithium deuteride in thermonuclear devices. Following formation of tritium in a nuclear weapon, it readily interchanges with  $\text{H}_2\text{O}$ , forming tritiated water,  ${}^3\text{HOH}$ , and enters the normal hydrologic cycle.

The most important man-made sources of tritium in the atmosphere to date have been nuclear detonations. Production from this source was calculated from a recent compilation by Carter and Moghissi (1977) of estimated energy yields from nuclear devices detonated since 1945. These yields were summarized for  ${}^{14}\text{C}$  by Killough and Till (1978), who listed all individual detonations that have been announced by the six nations that have carried out nuclear tests (United States, United Kingdom, USSR, France, China, and India). The detonations for each country are listed in chronological order, and each detonation is categorized according to whether it occurred at high altitude (above 10 km), in the lower atmosphere (below 10 km), underground, or underwater. The yields are often given as ranges, upper limits, or lower limits (e.g., 20 to 200, <20, and >20 MT). To estimate tritium releases to the atmosphere

from these data, we used the midpoint of each range, or the single number shown in an inequality of either sense. Where no yield is shown for an event, a value of zero was assumed. Only detonations in the atmosphere were considered, thus we ignore the venting that has occurred in some underground tests.

As noted earlier, the production of tritium varies significantly between fission and fusion nuclear detonations. In fusion weapons, the expected yield is estimated to be approximately  $6.7 \times 10^6$  Ci (250 PBq) per megaton equivalent of TNT (Leipunsky, 1957). Fission devices yield about  $7.0 \times 10^2$  Ci (26 TBq) per megaton equivalent of TNT (Miskel, 1973).

Carter and Moghissi do not distinguish between fission and fusion explosions in their published data; however, the authors do note the year in which the first thermonuclear detonation occurred for each of the six countries. For our calculations, it was assumed that all detonations were of the thermonuclear type for a given country after the first fusion device was tested. The release rate of tritium produced by nuclear explosions is summarized in Table 1.1. Our data indicate that the cumulative release to the atmosphere by nuclear weapons between 1945 and 1975 is approximately 1900 MCi (70 EBq).

Figure 1.1 illustrates the buildup and removal of tritium in the biosphere as a result of nuclear weapons testing and natural decay. Equilibrium concentrations from naturally produced tritium are indicated. These data were calculated using a multicompartment model that is discussed in Section 5.0 of this report. Concentration levels for the atmosphere, ocean surface, deep ground water, and fresh-water streams and lakes are shown with highest values occurring in the mid-1950's to mid-1960's and decreasing thereafter.

### 1.2.2 Tritium in commercial products

Tritium has been used in the production of luminous devices for about 20 years. During most of this time it was applied as a tritiated paint onto the faces and hands of timepieces, compasses, dials, etc.

Table 1.1 Estimated annual tritium yields from nuclear weapons tests in the atmosphere<sup>a</sup>

Year	Tritium released (Ci) <sup>b</sup>	Year	Tritium released (Ci) <sup>b</sup>
1945	$4.0 \times 10^3$	1960	$7.1 \times 10^2$
1946	$1.4 \times 10^2$	1961	$4.9 \times 10^8$
1947	0	1962	$7.1 \times 10^8$
1948	$7.4 \times 10^2$	1963	0
1949	0	1964	$1.4 \times 10^2$
1950	0	1965	$1.4 \times 10^3$
1951	$1.1 \times 10^3$	1966	$4.7 \times 10^3$
1952	$7.1 \times 10^7$	1967	$2.0 \times 10^7$
1953	$1.8 \times 10^6$	1968	$3.9 \times 10^7$
1954	$1.0 \times 10^8$	1969	$2.0 \times 10^7$
1955	$7.8 \times 10^6$	1970	$3.6 \times 10^7$
1956	$9.3 \times 10^7$	1971	$5.2 \times 10^6$
1957	$6.4 \times 10^7$	1972	$8.7 \times 10^5$
1958	$2.1 \times 10^8$	1973	$1.7 \times 10^7$
1959	0	1974	$3.9 \times 10^6$

<sup>a</sup>Computed from detonation data compiled by Carter and Moghissi (1977) and summarized by Killough and Till (1978) for weapons testing by the United States, United Kingdom, USSR, France, China, and India. It is assumed that a one megaton fusion burst in the atmosphere releases 6.7 MCi (.25 EBq) of tritium and a one megaton fission burst in the atmosphere releases  $7.0 \times 10^2$  Ci (26 TBq) of tritium.

<sup>b</sup>1 Ci = 37 GBq.

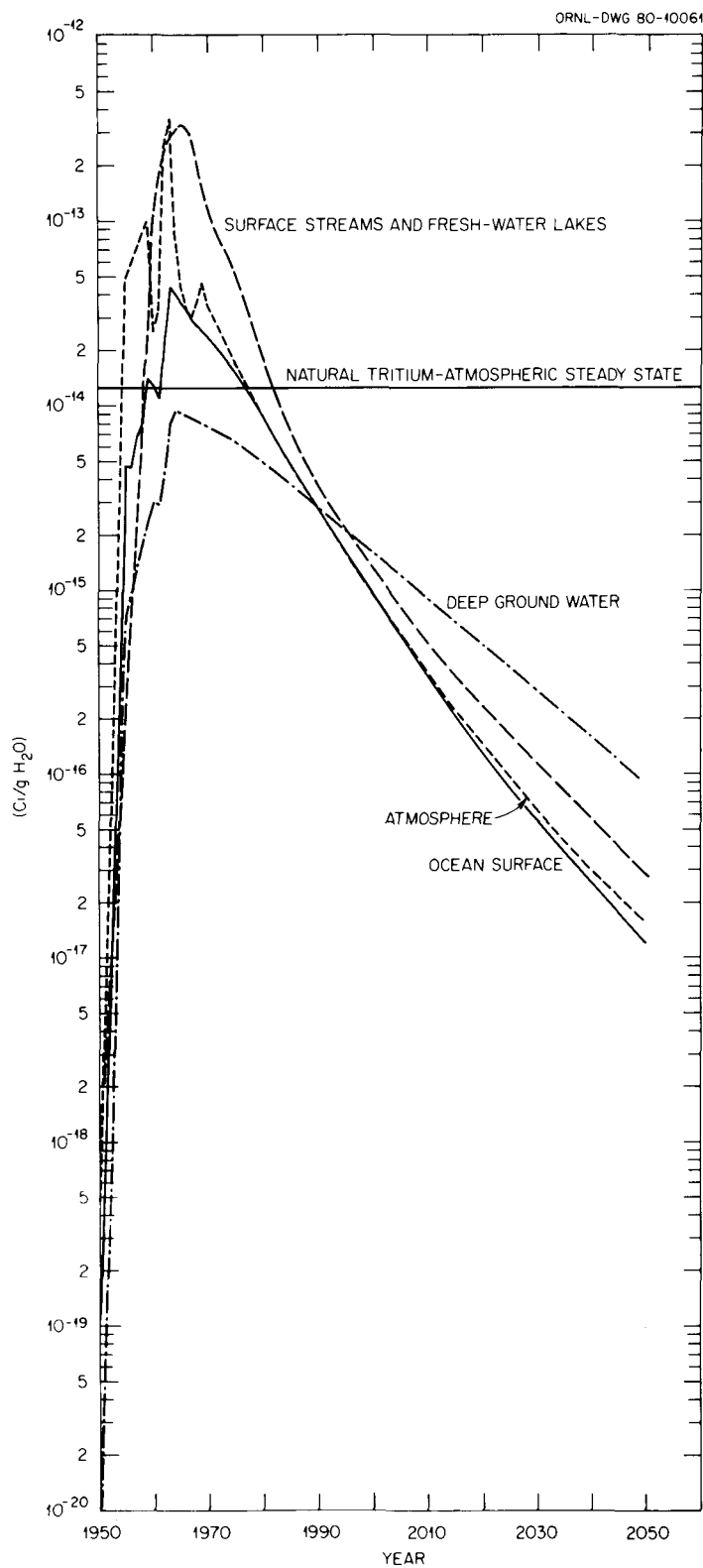


Fig. 1.1 Tritium in the biosphere as a result of nuclear weapons testing



More recently, elemental tritium has been used in the manufacture of products with self-luminous light sources. Liquid crystal display (LCD) watches may utilize self-luminous, background light sources containing tritium gas. This gas is enclosed in sealed borosilicate glass tubes coated internally with an inorganic phosphor. Beta particles emitted during radioactive decay of the enclosed tritium activate the phosphor, producing light. While watches containing tritium paint have an average activity of 2 mCi (74 MBq) (McDowell-Boyer and O'Donnell, 1978a), timepieces lighted with tritium in glass tubes can contain as much as 200 mCi (7.4 GBq) of tritium.

In the period 1969 to 1975, 0.13 MCi (4.8 PBq) of tritium was used in timepieces in the United States (McDowell-Boyer and O'Donnell, 1978a), whereas in 1978 alone, 0.4 MCi (15 PBq) were distributed in the United States for use in back-lit watches. For this reason, the distribution of tritium in the latter devices has made the major contribution to the global inventory from commercial products and will be the primary product of concern for estimates made later in this study.

#### 1.2.3. Tritium in the nuclear fission power industry

The production of tritium in various reactor systems was recently reviewed by the NCRP (1979a). Because of the high mobility of tritium in mechanical systems, and the potential contribution of tritium to dose, it is of interest to consider the mechanisms of escape of tritium from a fuel element. An understanding of these mechanisms is critical to the calculation of tritium release rates predicted for nuclear facilities.

Hydrogen in all of its isotopic forms is potentially very mobile, but its degree of mobility depends on the medium in which it is found and the temperature of the medium. These two factors, therefore, determine the rate at which tritium generated in fuel elements (from ternary fission or neutron activation of an impurity) moves through the fuel and its cladding into reactor coolant. Information has been obtained on the fraction of tritium retained in fuel elements, and its distribution in

different types of fuel elements, from dissolution experiments using irradiated fuels.

Goode and Vaughen (1970) examined the behavior of tritium in light water reactor (LWR), high temperature gas-cooled reactor (HTGR), and fast breeder reactor (FBR) fuel samples during head-end and fuel dissolution reprocessing operations. Intact rods or capsules were available for some of the fuel forms, but others were segments or small pieces of irradiated fuel. The tritium released during initial shearing of intact rods was generally somewhat less than 1% of the theoretical rod content.

The amount of tritium retained in a series of stainless-steel-clad fuel samples irradiated in water-cooled or liquid-metal-cooled reactors showed a strong dependence on fuel cladding temperature history. Most fuel samples contained oxide fuels, but two rods contained carbide fuel. A stainless-steel-clad pressurized water reactor (PWR) fuel rod irradiated at a linear heat rating of about 10 kw/m and a cladding temperature of approximately 105°C retained 99.8% of tritium calculated to have been generated during irradiation. There was, however, a pronounced drop in the fraction of tritium retained by FBR-type stainless-steel-clad fuel rods irradiated at linear heat ratings in the range of 30 to 92 kw/m under cooling conditions resulting in cladding temperatures of 500 to 1100°C. Under these conditions, the tritium retained in the fuel rods varied from 0.001 to 3.7% of that generated with one exception, where a linear heat rating of about 33 kw/m and a cladding temperature of 1000°C resulted in retention of approximately 50% of the calculated tritium content. Carbide fuel irradiated at a linear heat rating of 92 kw/m retained <1% of the calculated tritium generated.

Wozadlo et al. (1972) measured retention of <1% of the tritium produced in stainless-steel-clad mixed oxide (uranium and plutonium oxide) fuel irradiated in EBR-II (a fast-neutron experimental reactor) at average linear heat ratings of 36 to 46 kw/m, which produced cladding temperatures of 538 to 593°C. A somewhat larger retention was reported by Ebersole et al. (1971) for stainless-steel-clad driver fuel samples in the EBR-II. About 70 to 75% of the tritium was transferred to the primary sodium in these latter samples.

In liquid sodium cooled reactors, most of the tritium released to the primary coolant will be recovered from cold traps as sodium tritide. Smaller amounts will diffuse through the intermediate heat exchanger into the secondary coolant where cold trapping will also be employed. Some tritium will appear in the reactor cover gas and will have to be considered in handling that gas.

Goode and Cox (1970) found, on dissolving irradiated blanket rods from the Shippingport PWR, that essentially all of the tritium calculated to have been generated during irradiation was retained in the fuel element. Seven percent of the tritium was found in the Zircaloy-2 cladding and 93% in the  $\text{UO}_2$  fuel. The cladding temperature was calculated to be  $259^\circ\text{C}$  for a linear heat rating of 16 kw/m. At this temperature, movement of tritium through the cladding is effectively stopped by the zirconium oxide films inside and outside of the cladding and the formation of second phase platelets of zirconium hydride within the cladding.

Gainey (1976) has reviewed the behavior of tritium in HTGRs. The reference fuel for this reactor is in the form of microspheres of  $\text{UC}_2$ , about 350  $\mu\text{m}$  in diameter, coated with pyrolytic carbon, silicon carbide, and pyrolytic carbon in three layers. The principal sources of tritium in the HTGR are ternary fission and activation of  $^3\text{He}$  by the reaction  $^3\text{He}(n,p)^3\text{H}$ . Reactions with  $^6\text{Li}$ , which occurs as an impurity in the fuel and in core graphite, also form tritium by the reaction  $^6\text{Li}(n,\alpha)^3\text{H}$ . Boron-10, used in the control rods or as a "burnable poison," is converted to tritium by the reactions  $^{10}\text{B}(n,\alpha)^7\text{Li}$  followed by  $^7\text{Li}(n,n\alpha)^3\text{H}$ , and  $^{10}\text{B}(n,2\alpha)^3\text{H}$ .

Of the tritium generated in an HTGR, it is estimated that 62% will result from ternary fission and the balance from the several activation reactions mentioned above. Intact coatings on the fuel particles effectively retain tritium, but about 0.5% of that generated will be released to the coolant from the fuel due to coating failure. Graphite and  $\text{B}_4\text{C}$  are described as "extremely retentive toward tritium under reactor conditions" (Gainey, 1976); however, some portion of the tritium formed from  $^6\text{Li}$  impurity or  $^{10}\text{B}$  neutron absorber will be transferred to the

coolant, from which a fraction will eventually be released to the environment after diffusing through the heat exchanger tubes. Methods to reduce the permeation rate of tritium through the heat exchanger tubes, involving the formation of oxide "blocking" films, are being studied.

Heavy-water-moderated reactors (HWR) represent a special case in that the quantity of tritium produced from neutron activation of deuterium, by the reaction  $^2\text{H}(n,\gamma)^3\text{H}$ , is much greater than that resulting from fissioning of the fuel. Kouts and Long (1973) estimated that 40 times as much tritium would be produced in the HWR moderator as in the fuel. Special provisions for containment of the costly heavy water and its tritium content must be provided (Lewis and Foster, 1970).

Observations by various experimenters show that the amount of tritium retained in the fuel during irradiation in LWRs, FBRs, and HTGRs cannot be precisely predicted at present. Nevertheless, most of the generated tritium is released to the coolant in FBRs; a much smaller release to coolant in LWRs and HTGRs is indicated. The practical consequence of this variation is that the principal burden for control of tritium releases from LWR and HTGR fuels will be on the reprocessing plant, and for FBRs, on the reactor cooling system design. In the past, 100% retention of tritium in fuel has been assumed in studies of the environmental impact of reprocessing LWR fuels (USERDA, 1976a; Finney et al., 1977) and in the environmental survey of the uranium fuel cycle (USAEC, 1974a). A 10% retention factor in fuel was used in preparation of the environmental statement for the LMFBR program (USAEC, 1974b) and is currently used by the NCRP to forecast global tritium releases from LMFBR fuel reprocessing (NCRP, 1979a). We therefore also assume a 10% tritium retention value in the analyses of LMFBR reprocessing impacts presented in Chaps. 4 and 5 of this report.

In fuel reprocessing, the tritium content of any irradiated oxide fuel is transferred on dissolution in  $\text{HNO}_3$  to the aqueous stream by exchange with the hydrogen content of the acid solution. In the past, most of this tritium was assumed to be exhausted to the atmosphere as tritiated water after recovery of the  $\text{HNO}_3$  for reuse in the reprocessing plant. Since uncontrolled release of tritium to the atmosphere will

probably not be permitted by evolving environmental protection regulations, (e.g., USEPA, 1977) modified reprocessing flow sheets are being considered. A predissolution oxidation treatment of LWR, FBR, and HTGR fuels offers prospects for removing and recovering tritium for permanent storage (Goode and Vaughen, 1970; Finney et al., 1977). Other tritium confinement methods may become feasible in the future.

The LMFBR tritium source terms presented in this review are based on the ternary fission process alone. Any contribution to the tritium content of the spent fuel resulting from  $^6\text{Li}$  impurity has not been considered. Kabele (1974) has estimated that tritium derived from an unidentified level of lithium impurity in fuel irradiated in the fast flux test facility (FFTF) would equal about 50% of that from ternary fission. Kabele also estimated the tritium generated due to the  $\text{B}_4\text{C}$  content of the FFTF control rods would be about 7.4 times that resulting from ternary fission. Experimental observations show that between 20 and 80% of this tritium would be retained in the control rod. Tritium released to the sodium coolant from fuel elements and control rods would be recovered at the reactor by use of cold traps in which  $\text{Na}^3\text{H}$  would precipitate. The tritium content of the control rods would be permanently retained after removal from the reactor by the stainless steel cladding of these rods.

Chapter 5 of this report details nuclear fuel cycle tritium production estimates and impacts.

#### 1.2.4 Revised ternary fission yield data for fast reactor fuels

Data published by Buzzelli et al. (1976) and Buzzelli and Langer (1977) indicate, on a preliminary basis, that tritium in LMFBR fuels may be produced at levels higher than those previously anticipated. These tentative data and their implications are considered in detail in Chap. 4 of this report.

## 2. A REVIEW OF METHODOLOGIES FOR ESTIMATING THE DOSE FROM ENVIRONMENTALLY RELEASED TRITIUM

*John E. Till and Elizabeth L. Etnier*

The two primary chemical forms of tritium released to the environment from man-made sources are tritiated water vapor ( $^3\text{HOH}$ ) and tritium-hydrogen gas ( $^3\text{HH}$ ). Mason and Östlund (1979) point out that the two chemical forms may have different sources, distributions, and environmental sinks with the gas proceeding toward conversion to tritiated water vapor with an environmental half-time of 4.8 years. Small amounts of tritium also exist as tritium gas ( $^3\text{H}^3\text{H}$ ) and as tritiated methane ( $^3\text{HCH}_3$ ).

Radiological assessments generally assume that tritium is released from nuclear facilities as tritiated water vapor. Experimental data at the Savannah River Laboratory (Murphy and Pendergast, 1979) indicate, however, that although tritium released by reactors is primarily in the form of water vapor, releases from reprocessing nuclear fuel average approximately 40% tritium-hydrogen gas ( $^3\text{HH}$ ) or tritium gas ( $^3\text{H}^3\text{H}$ ), and 60% tritiated water vapor ( $^3\text{HOH}$ ). The effect of the chemical form of release upon the ultimate radiological dose to man is of interest due to the reduced biological impact of tritiated gases, and particularly because of possible releases of tritiated gases from fusion power reactors; additional research in the area is needed.

Numerous methodologies have been proposed to calculate the dose to man from tritium released to the environment. The following sections discuss four of these methodologies and include sample calculations of dose for each under chronic exposure conditions of  $1 \text{ pCi/m}^3$  ( $37 \text{ mBq/m}^3$ ) of tritium in the atmosphere. During review of Sects. 2.1-2.4, the reader should be aware that certain terms (e.g.,  $C_w$  and  $C_f$ ) are not defined consistently for all four methodologies. Ingestion rates as well as dose conversion factors also vary for the different methodologies. Definitions of terms are, of course, consistent within each of the four sections.

## 2.1 Specific Activity Methodology

Following analyses for tritium in deer, Evans (1969) suggested that long-term exposure to tritium results in significant incorporation of tritium in organic molecules in body tissues, in addition to mixing of  $^3\text{H}_2\text{O}$  (tritiated water) in body water. Based on his experimental data, Evans calculated an upper limit of the dose that man could receive from chronic exposure to tritium assuming body hydrogen is uniformly labeled. A reference man of 70 kg contains 7 kg of hydrogen, approximately 4.8 kg in body water and 2.2 kg in organic molecules (ICRP, 1975). If it is assumed that the tritium concentration in body water is  $1 \mu\text{Ci}/\ell$  ( $37 \text{ kBq}/\ell$ ), and that organic molecules are labeled to the same extent (i.e., exhibit the same  $^3\text{H}$  to  $^1\text{H}$  ratio as body water), this concentration results in a body burden of

$$\frac{1 \mu\text{Ci}}{\ell \text{ H}_2\text{O}} \times \frac{1 \ell \text{ H}_2\text{O}}{\text{kg H}_2\text{O}} \times \frac{18 \text{ kg H}_2\text{O}}{2 \text{ kg } ^1\text{H}} \times \frac{7 \text{ kg } ^1\text{H}}{\text{reference man}} = 63 \mu\text{Ci} \text{ (2.3 MBq)}. \quad (2.1)$$

Assuming a quality factor of 1.0 for beta particles of tritium, this body burden results in an annual dose rate of

$$\begin{aligned} & \frac{63 \mu\text{Ci}}{7 \times 10^4 \text{ g}} \times \frac{3.7 \times 10^4 \text{ dis}}{\text{sec} - \mu\text{Ci}} \times \frac{3.2 \times 10^7 \text{ sec}}{\text{year}} \times \frac{0.006 \text{ MeV}}{\text{dis}} \times \\ & \frac{1.6 \times 10^{-6} \text{ ergs}}{\text{MeV}} \times \frac{10^3 \text{ millirem} \cdot \text{g}}{100 \text{ ergs}} = 102 \text{ millirem/year (1.02 mSv/year)}. \end{aligned} \quad (2.2)$$

Evans reported further that his data indicated the labeling fraction in organic molecules to be between 0.62 and 1.0 in deer tissue, depending on the specific organ being considered, with a weighted average fraction of 0.85-1.0 extrapolated to the reference man. [A labeling fraction of 1.0 indicates that the  $^3\text{H}$  to  $^1\text{H}$  ratios are equal when comparing body water and organic (bound) components.] Assuming that tritium in body water is uniformly distributed, and assuming a labeling fraction of 0.85

for organically bound hydrogen in the body, one calculates a body burden of

$$\frac{[4.8 \text{ kg} + (0.85)(2.2 \text{ kg})]}{7.0 \text{ kg}} \times 63 \text{ } \mu\text{Ci} = 60 \text{ } \mu\text{Ci} \text{ (2.2 MBq)}, \quad (2.3)$$

which results in an annual dose of

$$\frac{60 \text{ } \mu\text{Ci}}{63 \text{ } \mu\text{Ci}} \times 102 \text{ millirem} = 97 \text{ millirem (0.97 mSv)}. \quad (2.4)$$

This annual dose of 97 millirem (0.97 mSv) resulting from chronic exposure to a concentration of 1  $\mu\text{Ci}/\ell$  (37 kBq/ $\ell$ ) in the body water can be used to evaluate the dose resulting from long-term exposures in the environment. Assuming an atmospheric concentration of tritium of 1 pCi/ $\text{m}^3$  (37 mBq/ $\text{m}^3$ ), a moisture content of 6 g  $\text{H}_2\text{O}$  (in air)/ $\text{m}^3$ , and that the concentration of tritium in man is in equilibrium with that in the atmosphere, the following annual dose is calculated using the data published by Evans:

$$\begin{aligned} & \frac{1 \text{ pCi}}{\text{m}^3} \times \frac{\text{m}^3}{6 \text{ g H}_2\text{O}} \times \frac{97 \text{ millirem}}{\mu\text{Ci}/\ell} \times \frac{10^3 \text{ g H}_2\text{O}/\ell}{10^6 \text{ pCi}/\mu\text{Ci}} \\ &= 1.6 \times 10^{-2} \text{ millirem (1.6} \times 10^{-7} \text{ Sv)}. \end{aligned} \quad (2.5)$$

The "specific activity" approach described here would be applicable to evaluating chronic exposures to tritium. This methodology represents an upper limit to the dose, since it assumes a maximum possible body burden of tritium. It is not recommended for evaluating exposures from acute releases to the environment (e.g., accidental releases), or in assessing doses near the point of release where the tritium concentrations in water, food, and air may vary considerably as a function of time.



## 2.2 Methodology of the National Council on Radiation Protection and Measurements

The National Council on Radiation Protection and Measurements (NCRP, 1979a) proposes a methodology for calculating the dose from tritium when the concentration of tritium is known in the water, food products, and air to which the individual is exposed. This technique for calculating the dose applies to an equilibrium situation only and is not recommended to evaluate exposures resulting from pulse releases of tritium. The NCRP methodology assumes that the dose from tritium via the various pathways of exposure depends upon the relative contributions to total water intake as listed in Table 2.1. The annual dose per unit concentration for 3.0 l/day water intake is described by the following expression:

$$D = [1.22 C_w + 1.27 C_{f1} + 0.29 C_{f2} + 0.22 C_a] \frac{1}{3.0} \times DRF, \quad (2.6)$$

where\*

$D$  = annual dose (millirem),

$C_w$  = concentration of tritium in drinking water (pCi/l),

$C_{f1}$  = concentration of tritium in water in food (pCi/l),

$C_{f2}$  = concentration of tritium oxidized to water upon metabolism of food (pCi/l),

$C_a$  = concentration of tritium in atmospheric water (pCi/l), and

$DRF$  = dose rate factor ( $\frac{\text{millirem/year}}{\text{pCi/l}}$ ).

The dose rate factor (DRF) used by the NCRP is  $95 \times 10^{-6}$  millirem·l (pCi·year) $^{-1}$ , [ $2.6 \times 10^{-8}$  Sv·l (Bq·year) $^{-1}$ ] assuming a quality factor of 1.0. This value is based upon a three compartment model of hydrogen in the body, published by Bennett (1973). The model assumes a water balance of 3.0 l/day and retention half-time components of 9, 30, and 450 days for tritium in the body. The value for DRF reported here is defined as the committed dose per integrated intake or the equilibrium

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\*1 rem = 10 mSV; 1 pCi = 37 mBq.

Table 2.1 Contributions to total water intake of reference man<sup>a</sup>

Source	Intake (ℓ/d)	Fraction
Drinking water	1.22	0.41
Food products <sup>b</sup>	1.27	0.42
Oxidation of food <sup>c,d</sup>	0.29	0.10
Inhalation <sup>e</sup>	0.13	0.04
Skin absorption <sup>a</sup>	<u>0.09</u>	<u>0.03</u>
Total	3.0	1.00

<sup>a</sup>NCRP (1979a).

<sup>b</sup>In food 0.72 ℓ/day.  
In milk 0.53 ℓ/day.  
In juice 0.02 ℓ/day

<sup>c</sup>Oxidation of food 0.25 ℓ/day.  
Oxidation of milk 0.04 ℓ/day.  
Oxidation of juice 0.002 ℓ/day.

<sup>d</sup>Tritium entering body as organically bound hydrogen which is oxidized to <sup>3</sup>HOH during metabolism.

<sup>e</sup>Assuming an absolute humidity of 6 g H<sub>2</sub>O/m<sup>3</sup> in air.

dose rate in millirem per year ( $\mu\text{Sv}/\text{year}$ ) per constant intake concentration ( $\text{pCi}/\ell$ ,  $\text{mBq}/\ell$ ). According to Bennett (1973), 84% of the dose is due to tritium in body water and 16% is due to organically bound tritium.

In order to calculate the dose from a chronic exposure to  $1 \text{ pCi}/\text{m}^3$  ( $37 \text{ mBq}/\text{m}^3$ ) of tritium in the atmosphere using the methodology proposed by the NCRP, several assumptions are needed. First, it is assumed that the absolute humidity is  $6 \text{ g H}_2\text{O}/\text{m}^3$  (the same as that used in NCRP Report No. 62). The second assumption is that the concentrations of tritium in drinking water, food, and air are equal, and as given by

$$\begin{aligned} \frac{1 \text{ pCi}}{\text{m}^3} \times \frac{\text{m}^3}{6 \text{ g H}_2\text{O}} &= 1.7 \times 10^{-1} \text{ pCi/g H}_2\text{O}. \\ &= 1.7 \times 10^{-1} \text{ pCi/ml H}_2\text{O} \text{ (6.3 mBq/ml H}_2\text{O)}. \end{aligned} \quad (2.7)$$

Then, from Eq. (2.6), the dose is given by

$$\begin{aligned} &[(1.22 + 1.27 + 0.29 + 0.22) (1.7 \times 10^{-1} \text{ pCi/ml H}_2\text{O})] \times \frac{1}{3.0} \\ &\times 10^3 \frac{\text{ml}}{\ell} \times 95 \times 10^{-6} \frac{\text{millirem/year}}{\text{pCi}/\ell} = 1.6 \times 10^{-2} \text{ millirem/year} \\ &\quad (1.6 \times 10^{-7} \text{ Sv/year}). \end{aligned} \quad (2.8)$$

This result is identical to the value calculated using the specific activity approach proposed by Evans. A primary reason that the two doses are identical is that in this example we have assumed that the activity concentration of tritium in the water content of air is equal to that in drinking water and foodstuffs. This assumption is not always valid for chronic exposure conditions. One example occurs when the source of drinking water is relatively uncontaminated, and thus the concentration of tritium in water,  $C_w$ , is significantly less than that in air,  $C_a$  (Sect. 4.5). Another example is the case in which food products are grown away from the point of release of the tritium but are consumed at a site of higher tritium air concentration. As an example, we use the NCRP methodology and assume  $1 \text{ pCi}/\text{m}^3$  ( $37 \text{ mBq}/\text{m}^3$ ) of tritium in air and  $6 \text{ g H}_2\text{O}/\text{m}^3$  absolute humidity, but we adjust the concentration in

drinking water to 1% of that in air, and the concentration in all food products to 50% of that in air.

Then, from Eq. (2.6), and adjusting the concentration factors [Eq. (2.7)] as above:

$$\begin{aligned}
 & [1.22(1.7 \times 10^{-3}) + 1.27(8.5 \times 10^{-2}) + 0.29(8.5 \times 10^{-2}) + \\
 & 0.22(1.7 \times 10^{-1})] \times 10^3 \times \frac{1}{3.0} \times 95 \times 10^{-6} \frac{\text{millirem/year}}{\text{pCi/l}} \\
 & = 5.5 \times 10^{-3} \text{ millirem/year } (5.5 \times 10^{-8} \text{ Sv/year}). \quad (2.9)
 \end{aligned}$$

The effect of incorporating simulated site-specific data is to reduce the dose to approximately one-third in this example. The NCRP model, therefore, would be applicable to chronic exposure conditions in which differences exist in the concentration of tritium in water, food, and air, but where time-averaged concentrations remain constant.

### 2.3 Methodology of AIRDOS-EPA

Moore et al. (1979) proposed a comprehensive computerized methodology (AIRDOS-EPA) that is capable of estimating the dose from environmentally released tritium and other radionuclides. Their code includes an atmospheric dispersion model that predicts concentrations of tritium at distances up to 50 miles (80 km) from the point of release. Tritium is assumed to move with water through the environment. Doses from ingestion of food and drinking water at a particular location are assumed proportional to the concentration of tritium in air at that location. The AIRDOS-EPA model is a further refinement over the NCRP method previously described, since it explicitly allows one to account for food products grown both at the site and elsewhere, where tritium concentrations may be different.

The dose from ingestion of tritium is given by:

$$D_{\text{ing}} = C_f X + C_w X \quad (2.10)$$

where\*

$D_{\text{ing}}$  = total dose from ingestion of tritium (rem/year)

$C_f$  = dose rate factor for food ( $\frac{\text{rem/year}}{\text{pCi/cm}^3}$ )

$C_w$  = dose rate factor for water ( $\frac{\text{rem/year}}{\text{pCi/cm}^3}$ )

$\chi$  = ground level concentration of tritium in air at an environmental location (pCi/cm<sup>3</sup>).

The ingestion dose from food ( $C_f\chi$ ) is artificially broken down into ingestion doses from vegetables ( $D_v$ ), meat ( $D_b$ ) and milk ( $D_c$ ), weighted according to the fraction of water entering the body from each of these food products. The equations used in the code to calculate  $C_f\chi$  are:

$$C_f\chi = D_v + D_b + D_c, \quad (2.11)$$

where\*

$$D_v = 0.505 C_f(f_{v1}\chi + f_{v2}\chi_v),$$

$$D_b = 0.185 C_f(f_{b1}\chi + f_{b2}\chi_b),$$

$$D_c = 0.310 C_f(f_{c1}\chi + f_{c2}\chi_c),$$

and

$D_v$ ,  $D_b$ , and  $D_c$  = tritium food ingestion dose from vegetables, meat, and milk, respectively (rem/year),

$\chi_v$  = average ground-level concentration of tritium in air over the assessment area weighted by quantities of vegetables produced as a function of location (pCi/cm<sup>3</sup>)

$\chi_b$  = same as above, except applied to meat,

$\chi_c$  = same as above, except applied to milk,

$f_{v1}$  = fraction of vegetable intake which is produced at the individual's location,

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\*1 rem = 10 mSv; 1 pCi = 37 mBq.

$f_{v2}$  = fraction of vegetable intake whose source represents an average produced over the assessment area,

$f_{b1}$ ,  $f_{b2}$ ,  $f_{c1}$ ,  $f_{c2}$  = same as above, except for meat and milk, respectively; and

0.505, 0.185, 0.310 = fraction of water entering the body in vegetable, meat, and milk, respectively.

The value of  $C_f$  is derived from: (1) the tritium total-body dose conversion factor for ingestion,  $8.3 \times 10^{-5}$  rem/ $\mu$ Ci ( $2.2 \times 10^{-11}$  Sv/Bq), published by Killough et al. (1978); (2) an average absolute humidity of 8 g  $H_2O/m^3$ ; and (3) a consumption rate of water in food products of 1638 g/day.

The dose conversion factor for ingestion is calculated assuming a 350-g daily intake of hydrogen and assuming that under conditions of chronic exposure the specific activity in the body is equal to that in the daily intake of food and water. A quality factor of 1.0 is assumed for the tritium beta particles. The resulting value of  $C_f$  is

$$6.18 \text{ (rem/year)/(pCi/cm}^3\text{) or } 1.67 \text{ (Sv/year)/(Bq/cm}^3\text{)}.$$

In order for the absolute humidity to be consistent throughout our analysis, the  $C_f$  was recalculated assuming an absolute humidity of 6 g  $H_2O/m^3$ , resulting in a value of

$$8.24 \text{ (rem/year)/(pCi/cm}^3\text{) or } 2.23 \text{ (Sv/year)/(Bq/cm}^3\text{)}.$$

The value of  $C_w$  as reported in AIRDOS-EPA for an assumed daily drinking water intake of 1512 g is

$$5.70 \text{ (rem/year)/(pCi/cm}^3\text{) or } 1.54 \text{ (Sv/year)/(Bq/cm}^3\text{)}.$$

This value is increased to 7.60 (rem/year)/(pCi/cm<sup>3</sup>) or 2.05 (Sv/year)/(Bq/cm<sup>3</sup>) for an absolute humidity of 6 gH<sub>2</sub>O/m<sup>3</sup>.

The AIRDOS-EPA code accounts for tritium doses via inhalation and skin absorption by using a separate dose conversion factor, also published by Killough et al. (1978). Complete absorption is assumed for inhaled tritium, and skin absorption is accounted for by adding 50% to the inhalation dose conversion factor. The dose conversion factor for inhalation is  $1.3 \times 10^{-4}$  rem/ $\mu$ Ci ( $3.4 \times 10^{-11}$  Sv/Bq) inhaled.

The dose from exposure to 1 pCi/m<sup>3</sup> ( $10^{-6}$  pCi/cm<sup>3</sup>, 37 nBq/cm<sup>3</sup>) of tritium in the atmosphere was calculated with the AIRDOS-EPA methodology to allow comparison with the methods of Evans (1969) and the NCRP (1979a) described earlier. The dose was estimated assuming an absolute humidity of 6 g H<sub>2</sub>O/m<sup>3</sup> and assuming that all food products are grown at the point of interest. The total dose from ingestion is

$$\begin{aligned}
 D_{\text{ing}} &= C_f \chi + C_w \chi \\
 &= \frac{(8.24 + 7.60) \text{ rem/year}}{\text{pCi/cm}^3} \times 10^{-6} \text{ pCi/cm}^3 \\
 &= 1.6 \times 10^{-5} \text{ rem/year, or} \\
 &= 1.6 \times 10^{-2} \text{ millirem/year } (1.6 \times 10^{-7} \text{ Sv/year}).
 \end{aligned} \tag{2.12}$$

The dose from inhalation and skin absorption,  $D_{\text{inh}}$ , is given by

$$\begin{aligned}
 D_{\text{inh}} &= \frac{1.3 \times 10^{-10} \text{ rem}}{\text{pCi}} \times \frac{10^{-6} \text{ pCi}}{\text{cm}^3} \times \frac{9.6 \times 10^5 \text{ cm}^3}{\text{h}} \times \\
 &\quad \frac{8.76 \times 10^3 \text{ h}}{\text{year}}, \\
 &= 1.1 \times 10^{-6} \text{ rem/year} \\
 &= 1.1 \times 10^{-3} \text{ millirem/year } (1.1 \times 10^{-8} \text{ Sv/year}).
 \end{aligned} \tag{2.13}$$

Therefore, the annual dose when chronically exposed to 1 pCi/m<sup>3</sup> (37 mBq/m<sup>3</sup>) in the atmosphere is

$$\begin{aligned}
 D &= 1.6 \times 10^{-2} \text{ millirem} + 1.1 \times 10^{-3} \text{ millirem} \\
 &= 1.7 \times 10^{-2} \text{ millirem } (1.7 \times 10^{-7} \text{ Sv}).
 \end{aligned} \tag{2.14}$$

To provide a comparison between AIRDOS-EPA and the NCRP methodology using the previous example where the concentration in air is 1 pCi/m<sup>3</sup> (10<sup>-6</sup> pCi/cm<sup>3</sup>), the concentration in food is 50% of that in air, and the concentration in drinking water is 1% of that in air, the following parameter values are assumed:

$$f_{v1}, f_{b1}, \text{ and } f_{c1} = 0,$$

$$f_{v2}, f_{b2}, \text{ and } f_{c2} = 1, \text{ and}$$

$$\chi_v, \chi_b, \text{ and } \chi_c = 0.5 \text{ pCi/m}^3 = 5 \times 10^{-7} \text{ pCi/cm}^3 \text{ (190 } \mu\text{Bq/cm}^3\text{)}.$$

The dose from ingestion is given by

$$\begin{aligned} D_{\text{ing}} &= C_f(5 \times 10^{-7} \text{ pCi/cm}^3) + C_w(1 \times 10^{-8} \text{ pCi/cm}^3) \\ &= 4.2 \times 10^{-3} \text{ millirem/year (4.2} \times 10^{-8} \text{ Sv/year)}. \end{aligned} \quad (2.15)$$

Since the dose from inhalation and skin absorption is not changed, the total dose rate is

$$\begin{aligned} D &= 4.2 \times 10^{-3} \text{ millirem/year} + 1.1 \times 10^{-3} \text{ millirem/year} \\ &= 5.3 \times 10^{-3} \text{ millirem/year (5.3} \times 10^{-8} \text{ Sv/year)}. \end{aligned} \quad (2.16)$$

The effect of including simulated site-specific data (see discussion on p. 21) is a reduction in the dose by a factor of ~3.

## 2.4 Methodology of the U.S. Nuclear Regulatory Commission

Methodology proposed by the U.S. Nuclear Regulatory Commission (1977) for calculating the concentration of tritium in food products is based upon a model published by Anspaugh et al. (1972) which assumes that the concentration in vegetation is one-half that in surrounding air. The concentration in vegetation is given by the following equation



$$C_T^V(r, \theta) = 3.17 \times 10^7 Q_T [\chi/Q(r, \theta) (0.75)(0.5/H)], \quad (2.17)$$

where

$C_T^V(r, \theta)$  = concentration of tritium in vegetation grown at location  $(r, \theta)$ , (pCi/kg),

$3.17 \times 10^7$  = unit conversion factor ( $\frac{\text{pCi}}{\text{Ci}} \cdot \frac{\text{g}}{\text{kg}} \cdot \frac{\text{year}}{\text{sec}}$ ),

$[\chi/Q(r, \theta)]$  = atmospheric dispersion factor at distance  $r$  and direction  $\theta$ , ( $\frac{\text{sec}}{\text{m}^3}$ ),

$Q_T$  = annual release rate of tritium (Ci/year),

$H$  = absolute humidity of the atmosphere at location  $(r, \theta)$ , ( $\text{g}/\text{m}^3$ ),

$0.5$  = ratio of tritium concentration in plant water to tritium concentration in atmospheric water (dimensionless);

$0.75$  = fraction of total plant mass that is water (dimensionless).

The concentration of tritium in milk and meat depends upon the tritium concentration in vegetation ingested by cattle, and is given by the following expression

$$C_T^{m,B}(r, \theta) = F_{m,B} C_T^V(r, \theta) Q_F, \quad (2.18)$$

where

$C_T^{m,B}(r, \theta)$  = concentration of tritium in milk (Ci/l), or in meat (Ci/kg),

$C_T^V(r, \theta)$  = concentration of tritium in vegetation (Ci/kg),

$F_{m,B}$  = average fraction of the animal's daily intake of tritium that appears in each liter of milk ( $1.0 \times 10^{-2}$  day/l), or in each kg of meat ( $1.2 \times 10^{-2}$  day/kg); and

$Q_F$  = amount of feed consumed by a cow (kg/day).

The ingestion dose is then given by

$$D_{\text{ing}} = [C_T^V f_g(U_V) + C_T^{m,B}(U_{m,B})] \times \text{DCF} \quad (2.19)$$

where

DCF = dose conversion factor (millirem/pCi ingested);

$U_{V,m,B}$  = ingestion rate of vegetables, milk and beef,  
respectively;

$f_g$  = fraction of ingested produce grown locally, here  
assumed = 1.

It is important to understand that the methodology proposed here assumes that the maximum concentration of tritium in plants is one-half of that in the atmosphere. This assumption is made because a significant portion of the water in plants is taken up from the soil water, in which the tritium activity concentration is assumed to be lower than in air.

The Regulatory Guide 1.109 methodology does not provide guidance for assumptions regarding the tritium concentration in drinking water when only atmospheric concentrations are known. (The lack of guidance regarding this pathway may be assumed to indicate that it is anticipated that contamination of drinking water with atmospheric tritium contributes less than 10% of the total tritium dose at LWR sites.) The dose to total body from tritium in drinking water may be calculated from a measured or assumed concentration in water using a dose conversion factor for ingestion of tritium, and a consumption rate of 370 l/year for adults.

Dose conversion factors in the Regulatory Guide were derived by Hoenes and Soldat (1977) and are listed in units of millirem per picrocurie ingested or inhaled. A breathing rate of 8000 m<sup>3</sup>/year is recommended. The inhalation dose conversion factor accounts for absorption through the skin by increasing the factor calculated from inhalation alone by 50%. This is identical to the approach taken by Killough et al. (1978), discussed earlier. The dose conversion factors published by

Hoenes and Soldat assumed a quality factor for tritium beta particles of 1.7. In order to provide a consistent comparison of the NRC methodology with the methodologies previously discussed, the dose conversion factors have been recalculated for a quality factor of 1.0 resulting in values of  $6.2 \times 10^{-8}$  millirem/pCi ( $1.7 \times 10^{-11}$  Sv/Bq) and  $9.4 \times 10^{-8}$  millirem/pCi ( $2.5 \times 10^{-11}$  Sv/Bq) for ingestion and inhalation, respectively.

Assuming an atmospheric concentration of tritium of 1 pCi/m<sup>3</sup> (37 mBq/m<sup>3</sup>), an absolute humidity of 6 g H<sub>2</sub>O/m<sup>3</sup>, and that the activity concentration of tritium in drinking water is the same as that in air, an annual dose of  $5.9 \times 10^{-3}$  millirem ( $5.9 \times 10^{-8}$  Sv) may be calculated using NRC methodology for a one-year chronic tritium exposure to the average individual. This total dose is broken down as follows:  $1.2 \times 10^{-3}$  millirem ( $1.2 \times 10^{-8}$  Sv) from ingestion of contaminated foods;  $3.9 \times 10^{-3}$  millirem ( $3.9 \times 10^{-8}$  Sv) from ingestion of contaminated water; and  $7.6 \times 10^{-4}$  millirem ( $7.6 \times 10^{-9}$  Sv) from skin absorption and inhalation.

If it is assumed that all of the ingested food products are grown at a different site where the atmospheric concentration is 50% of that at the point of interest, and the concentration in drinking water is 1% of the atmospheric concentration, the NRC methodology yields a dose of  $1.4 \times 10^{-3}$  millirem ( $1.4 \times 10^{-8}$  Sv). Incorporating these assumptions thus reduces the dose by a factor of ~4.

## 2.5 Recommendations for Calculating the Dose from Chronic Exposures to Tritium in the Environment

Table 2.2 summarizes the dose from 1 pCi/m<sup>3</sup> (37 mBq/m<sup>3</sup>) of tritium calculated using each of the preceding methodologies, with and without simulated site-specific parameters. Each of the methodologies reviewed is applicable to chronic exposure conditions only and is not recommended for estimating the dose following an acute release of tritium to the environment. The primary advantage of the methodologies of Evans (1969) and the NCRP (1979a) is simplicity. The NCRP method, however, allows one to account for variations in the concentration of tritium in water

Table 2.2 Summary of the annual dose from chronic exposure to 1 pCi/m<sup>3</sup> (37 mBq/m<sup>3</sup>) of tritium in air, calculated using four methodologies<sup>a</sup>

Methodology	No site-specific data <sup>b</sup> (millirem) <sup>d</sup>	Including certain site-specific data <sup>c</sup> (millirem)
Evans, 1969	$1.6 \times 10^{-2}$	<sup>e</sup>
NCRP, 1979	$1.6 \times 10^{-2}$	$4.8 \times 10^{-3}$
AIRDOS-EPA, 1979	$1.7 \times 10^{-2}$	$5.3 \times 10^{-3}$
USNRC, 1977	$5.9 \times 10^{-3}$	$1.4 \times 10^{-3}$

<sup>a</sup>Calculations are made for 6 g H<sub>2</sub>O/m<sup>3</sup> and a quality factor of 1.0 for tritium betas.

<sup>b</sup>Assuming that all food products are grown at point of interest and that the specific activity of tritium in drinking water equals that in the atmosphere.

<sup>c</sup>Assuming that all food products are grown at another location where the air concentration of tritium is 50% of that at the point where the dose is calculated, and the concentration in drinking water is 1% of that where the dose is calculated.

<sup>d</sup>1 millirem = 10 μSv.

<sup>e</sup>The methodology proposed by Evans does not provide for the inclusion of site-specific data.

taken into the body while the Evans procedure does not. The methodologies of AIRDOS-EPA (Moore et al., 1979) and the USNRC (1977) permit incorporation of site-specific data such as the fraction of food products harvested from a location other than the point of interest. It is obvious from our calculations that consideration of data describing reduced tritium concentrations in drinking water and food products may significantly reduce the calculated dose. Therefore, these features available in the AIRDOS-EPA and NRC methods are important. The primary disadvantage to these two methodologies is complexity.

Our analysis of the four methodologies leads to the conclusion that the model recommended by the NCRP provides the best combination of simplicity and relative accuracy for calculating the dose from chronic exposures to tritium. Acute environmental exposures should be evaluated on a case by case basis using analytical measurements of tritium in the atmosphere, water, and food products.

Two minor modifications to the NCRP methodology would maintain its simplicity and would allow for incorporation of the dose from food products grown elsewhere. First, because current data concerning oxidized vs non-oxidized tritium components in food products are preliminary (Sect. 3.2), we combine these components into a single value of 1.56 (Table 2.1). Second, the concentration of tritium in food products is broken into two parts: (1) that fraction grown at the point where the dose is being calculated, and (2) that fraction grown at another location where the air concentration is different than at the point of interest. The model is described by the following equation, a modification of Eq. (2.6):

$$D = \left[ 1.22 C_w + 1.56 \left( \sum_{n=1, 2, 3, \dots} C_{fn} \delta_n \right) + 0.22 C_a \right] \frac{1}{3.0} \times DRF, \quad (2.20)$$

which simplifies to

$$D = \left[ 0.41 C_w + 0.52 \left( \sum_{n=1, 2, 3, \dots} C_{fn} \delta_n \right) + 0.07 C_a \right] DRF,$$

where\*

$D$  = annual dose (millirem),

$C_w$  = concentration of tritium in drinking water (pCi/l),

$C_{fn}$  = concentration of tritium in water of food products grown at location  $n$  (pCi/l),

$\delta_n$  = fraction of food products grown at location  $n$  (dimensionless),

$C_a$  = concentration of tritium in air (pCi/l),

$DRF$  = dose rate factor  $\left( \frac{95 \times 10^{-6} \text{ millirem/year}}{\text{pCi/l}} \right)$ ,  
 $(2.6 \times 10^{-8} \frac{\text{Sv/year}}{\text{Bq/l}})$ .

If the concentrations of tritium in food products and in drinking water are not known, it is recommended that the concentration in food be assumed to be 50% of that in air<sup>†</sup> at location "n," and that the concentration in drinking water be 1% of that in air at location "n." This "modified" NCRP model incorporates the simplicity of the specific activity and NCRP approaches and allows use of site-specific information on tritium concentrations in food products grown both locally and remotely. Use of such a model is recommended for estimating dose from chronic exposure to <sup>3</sup>H<sub>2</sub>O in the environment.

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\* 1 millirem = 10  $\mu$ Sv, 1 pCi = 37 mBq.

<sup>†</sup> Although this value has been widely accepted in the assessment of tritium released to the atmosphere, it is emphasized that the 50% value is based upon the model published by Anspaugh et al. (1972). More recent contradictory information published by Murphy and Pendergast (1979) indicates that the concentration of tritium in vegetation may be nearly equal to that in air under chronic exposure conditions. Therefore, because of the importance of the value assumed in the calculation of dose, additional research to determine the concentration of tritium in food products relative to that in air is badly needed.



### 3. ANALYSIS OF KEY PARAMETERS INCLUDED IN THE CALCULATION OF DOSE FROM TRITIUM

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This chapter considers four key parameters used in the calculation of the dose from tritium: (1) the quality factor for tritium betas, (2) the biological half-life and the effect of organic binding of tritium, (3) the absolute humidity, and (4) the concentration of tritium in drinking water. The value assumed for each of these parameters could significantly modify the calculated value of dose resulting from environmental exposures.

#### 3.1 The Tritium Quality Factor

The concept of dose equivalent (DE) was introduced by the International Commission on Radiological Protection (ICRP, 1966) and the International Commission on Radiation Units and Measurements (ICRU, 1962) in order to define a uniform scale of damage from exposure to different types and energies of radiation. The unit of the dose equivalent is the rem\*, which is calculated using the following expression:

$$DE = D \times Q \times (RDF)_1 \times (RDF)_2 \dots \quad (3.1)$$

where

DE = dose equivalent in rem,

D = absorbed dose in rads,

Q = quality factor (dimensionless),

$(RDF)_1, (RDF)_2 \dots$  = radionuclide distribution factors.

The term quality factor (Q) has been accepted for use in the calculation of dose and has been related to linear energy transfer (LET) on a common scale for all ionizing radiation. A distribution factor (RDF) may be

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\*1 rem = 10 mSv.



used to express the modification of biological effectiveness due to non-uniform distribution of internally deposited isotopes. Since tritium is essentially uniformly deposited in body tissue, its distribution factor is assumed to be 1. Some disagreement still exists, however, as to the best value of Q for tritium betas.

In ICRP Publication 9 (1966), the Commission recommended a value of 1.7 be used as the Q for  $\beta^-$ ,  $\beta^+$  and  $e^-$  radiation with maximum energies  $\leq 0.03$  MeV. An amendment to ICRP Publication 9, in April 1969 (ICRP, 1969), reduced Q to 1.0 for all  $\beta^-$ ,  $\beta^+$ ,  $e^-$ ,  $\gamma$ , and x rays. It was concluded that a value of unity is appropriate within the degree of precision required for the purposes of radiological protection. The decision to reduce the quality factor from 1.7 to 1.0 was also based upon the lack of scientific evidence to support the higher value as well as the variability in radiological end points and reference radiation reported in the literature.

The ICRP decision in 1969 followed a review of the experimental literature by Vennart (1968). He concluded that in view of experimental evidence on the quality factor of  $\beta$  particles from tritiated water a value different from unity could hardly be justified. Vennart also based his conclusion on the fact that since the ICRP in Publication 8 (1966) recommended expressing risk per unit dose only in terms of orders of magnitude, those factors included in the calculation of dose should be rounded to whole numbers. A later review of the literature by Rohwer (1976) stated that most of the information on tritium exposure supported the value of 1.0, although he pointed out that this was an area needing further study and evaluation.

Variability still exists in experimental end points and reference radiation, reported in the literature, which are used to determine the quality factor for  $\beta^-$  radiation. Cumming et al. (1979) demonstrated that total radiation dose from a single injection of tritiated water can be greatly influenced by minor changes in experimental conditions. Improvements continue to be made, however, in evaluating exposures to tritium at very low dose rates and in choosing physiological and biochemical end points leading to a significant increase in the sensitivity of the tests.

The potential genetic consequences of chronic, low-level exposure to tritium may be of particular importance for environmental releases from the nuclear industry. Carsten and Commerford (1976) studied mice exposed to 3  $\mu\text{Ci/ml}$  drinking water. Second generation females were sacrificed in late pregnancy to determine mutation frequency. Analysis of their results showed a significant reduction in the number of viable embryos resulting from mating between animals exposed to tritium but showed no effect on breeding efficiency. The authors noted that no direct parallelism exists between man and the data deduced from mice; however, the significant effect seen in their study suggests that further investigation at lower tritium concentrations is necessary.

Another study by Dobson et al. (1977) also suggests that genetic effects produced from internally deposited tritium may warrant renewed consideration. In their experiment, female germ cells in both mouse and monkey were shown to be extremely sensitive to destruction by low-level, chronic tritium exposure. Their results appear to be inconsistent with previous reports which conclude that oocytes in both monkey and man are relatively radioresistant, with X-ray doses lethal to 50% of the population ( $\text{LD}_{50}$ 's) of up to 5000 rads (50 Gy) (administered in single, acute doses at various times during development). Dobson and his colleagues suggest that the greater sensitivity they observed resulted from chronic exposure to tritium in body water that acted on cells passing through highly vulnerable periods of early development. The exposure conditions established in their experiments are likely to be more representative of low-level environmental exposures from tritium (such as releases by nuclear facilities) than those described by most investigators; therefore, the results of their study must be given serious consideration in the design of further bioeffects research involving tritium, and in current radiological assessment methodologies.

Table 3.1 summarizes selected experiments reported since the review published by Vennart (1968). The table includes only animal and mammalian studies in which values for relative biological effectiveness (RBE)\* are calculated. Additional experiments have been reported using

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\*See footnote a, Table 3.1.

Table 3.1 RBE<sup>a</sup> values for tritium from selected animal and cell studies<sup>b</sup>

Description of experiment	Reference radiation	Reported RBE	Reference
Irradiated mouse testes using internally distributed tritium from injected tritiated thymidine and tritiated water. Used inability of spermatogonia to divide twice as damage criteria.	200 kVp X rays	1.3 - 2.4	Lambert (1969)
Irradiated aqueous solutions of ribonuclease using tritiated water. Measured residual enzymatic activity and observed survival.	<sup>60</sup> Co γ rays	0.94 ± 0.11	Richold et al. (1971)
Exposed rats internally to tritiated water. Damage criteria were change in weight of spleen, thymus, adrenals, and liver, as well as biochemical indices.	<sup>137</sup> Cs γ rays	1.4 - 2.2	Moskalev et al. (1972)
Exposed rats internally to tritiated water. Damage criteria were survival rates, peripheral blood response and thymic and splenic weight changes.	<sup>137</sup> Cs γ rays	1.4 - 1.9	Moskalev et al. (1973)
Exposed two mammalian cell lines to tritiated water and tritiated thymidine. Measured cell survival.	<sup>60</sup> Co γ rays	1.7 - 1.9	Bedford et al. (1975)
Exposed weanling mice to tritiated water and observed survival of primary oocytes.	<sup>60</sup> Co γ rays	1.1 - 1.7	Dobson et al. (1975)
Exposed developing mice from conception to 14 days after birth. Observed primary oocyte survival.	<sup>60</sup> Co γ rays	1.6 - 3.0 (with varied rate)	Dobson and Kwan (1976)
Exposed mice to tritiated water, observed gene mutations transmitted.	<sup>60</sup> Co γ rays	2.2	Russell et al. (1979)

<sup>a</sup>Relative biological effectiveness (RBE) is a factor expressing the relative effectiveness of radiations that have different linear energy transfer (LET) values, in producing a given biological effect. The unit is limited to use in radiobiology but is similar to the value of quality factor used in the calculation of dose.

<sup>b</sup>Studies published since the literature review by Vennart (1968).

plants or in which radiosensitivity of organisms is studied but where no RBE is calculated. These experiments have been omitted from the table.

Lambert (1969) irradiated mouse testes using internally distributed tritium from injected  $^3\text{HTdR}$  and  $^3\text{HOH}$ . The criteria for damage was the inability of spermatogonia to divide twice and produce resting spermatocytes. The reference radiation was 200 kVp X rays, delivered at an exponentially decreasing dose rate over a 72-hour period. The RBE of tritium as tritiated thymidine or tritiated water relative to 200 kVp X rays was in the range of 1.3-2.4. Lambert pointed out that a direct extrapolation of his results to man is not possible; however, this study is particularly important because of its use of a very low dose rate (and total dose), and high sensitivity of the endpoint used as damage criteria.

Richold et al. (1971) reported an RBE of  $0.94 \pm 0.11$  following irradiation of aqueous solutions of ribonuclease with  $^3\text{HOH}$  under anoxic and aerobic conditions. The reference radiation was  $^{60}\text{Co}$   $\gamma$ . Of the studies listed in Table 3.1, this is the only case in which an RBE value of less than 1.0 is reported. However, a relatively high dose rate (100 rads/hr, 1 Gy/hr) was used. It has been shown that exposures to tritium under chronic conditions at low dose rates (comparable to "routine release" exposures) potentially produce more harmful effects than acute, high-level exposures (Dobson et al., 1975), and this "dose rate effect" may have affected the outcome of the studies by Richold et al.

In an animal study, Moskalev et al. (1973) exposed rats to tritium oxide injected intraperitoneally. The calculated dose rate from tritium varied from 27.5 to 1.38 rads/hr (0.28 to 0.014 Gy/hr) and the reference radiation used was  $^{137}\text{Cs}$   $\gamma$  rays. This experiment used several indices for RBE determination including quantitative composition of peripheral blood, change in weight of the spleen, thymus, adrenals, and liver, as well as numerous biochemical indices. The total cumulative dose was 340 rads (3.4 Gy). Calculated values for RBE ranged from 1.4 to 2.17 depending upon the index evaluated. Moskalev et al. (1973) reported another study in which rats were again exposed to  $^3\text{HOH}$  and  $^{137}\text{Cs}$   $\gamma$  rays. The criteria for damage were survival rate, peripheral blood response

and thymic and splenic weight response. Although higher dose rates were used in this study than the previous study, it was concluded that the RBE for tritium oxide was in the range of 1.45-1.93.

Bedford et al. (1975) exposed two mammalian cell lines to  $^3\text{HOH}$  and tritiated thymidine and used  $^{60}\text{Co}$   $\gamma$  rays as a reference radiation. The criterion for damage was cell survival. To prevent cell division during exposures, irradiations were carried out with cells held in the frozen state or, in one case, at  $5^\circ\text{C}$ . For cells irradiated at  $5^\circ\text{C}$ , the efficiency of cell killing by beta particles from tritiated water or tritiated thymidine was not appreciably different, but both were more efficient than gamma radiation. For a dose rate of 20 rad/hr (0.2 Gy/hr), the relative biological effectiveness of tritium beta particles compared to  $^{60}\text{Co}$   $\gamma$  rays was estimated to be between 1.7-1.9.

Weanling mice were exposed to  $^3\text{HOH}$  by Dobson et al. (1975) at low dose rates (approximately 5.2 rads/day, 0.052 Gy/day). Surviving primary oocytes were counted microscopically in ovaries and compared to controls. Other weanlings were exposed to  $^{60}\text{Co}$   $\gamma$  rays at 5.9 rads/day (0.059 Gy/day). It was concluded that since the exposure was protracted, more effective microdistribution of tritium atoms may have occurred resulting in an RBE of 1.1-1.7. The investigators emphasized the significance of their results showing that  $^3\text{HOH}$  becomes more damaging compared to gamma rays as the low-level exposure is protracted over longer periods of time. This finding is particularly important relative to exposures from tritium found in the environment. These data were verified in another study by Dobson and Kwan (1976). Again, developing mice were used and survival of primary oocytes was observed. The dose rate was varied to as low as 0.44 rads/day (0.0044 Gy/day) for both  $^3\text{HOH}$  and the reference  $^{60}\text{Co}$   $\gamma$  rays. At effective gamma-ray doses of about 40 rads (0.4 Gy), the RBE was calculated to be 1.6. However, with lower dose rates (giving effective total doses of only a few rads), the RBE for tritium rises to approximately 3. These studies by Dobson et al. (1975) and Dobson and Kwan (1976) are very important since they used extremely sensitive criteria for damage and observed noticeable effects at exceptionally low dose rates and total dose.

Of particular importance is the recent study by Russell et al. (1979) in which transmitted gene mutations induced by tritium were observed in mice. Male mice were injected with tritiated water to give a dose rate of 0.8 rad/min (0.008 Gy/min). A specific-locus-mutation test was used to determine biological damage. This procedure is considerably more sensitive than earlier methods reported for determining RBE. Russell et al. point out that various uncertainties are involved in arriving at a precise value for RBE; however, they recommend that for the purpose of risk estimation, it seems more prudent to use the RBE value of 2 as the best point estimate computed from their data.

In general, the values of relative biological effectiveness for tritium lie in the range 1.0 to 2.4 with more values nearer 2.0 than 1.0. The data summarized in Table 3.1 seem to indicate that, as experimental procedures are progressively refined and become more sensitive, a quality factor between 1.0 and 2.0 could certainly be supported by published data and a return to the original value of 1.7 would be more in line with the current "conservative" approach to assessment of dose.

In summary, recent experimental data on the RBE for tritium and potential, long-term, genetic effects from chronic, low-level exposures lead us to conclude that we may be underestimating the radiological significance of internally deposited tritium. A quality factor between 1.0 and 2.0 can certainly be justified for tritium beta particles, with an optimum value probably closer to 2.0 than 1.0. In view of this conclusion, it seems reasonable to suggest use of the quality factor value of 1.7 originally recommended by the ICRP, particularly for purposes of environmental assessment of routine tritium releases.

The NCRP (1979b) has recently reviewed the tritium quality factor issue, concluding that "...there is ample evidence to ascribe to the tritium beta an RBE of 1 *provided the reference radiation is in the order of 60-80 kVp x-rays*" (italics ours). Because the reference radiation utilized for determinations of quality factor for tritium is ordinarily 220-250 kVp  $\gamma$  rays, or  $^{60}\text{Co}$  rays, it appears that some ambiguity exists within the statements leading to the NCRP recommendation. Given the need for a conservative approach toward dose calculations for chronic,

low-level exposures, use of the 1.7 Q value during continued analysis of the question appears to be a prudent decision.

### 3.2 Biological Half-Life and Organic Uptake of Tritium

The uptake of tritium into organic tissues as bound hydrogen, and the resulting increase in the body burden during low-level exposures to tritiated water, are important in the evaluation of environmental releases of tritium. Young et al. (1975), Strand and Thompson (1976), Rohwer (1976), and the NCRP (1979 a and b) represent some of the more recent reviews published on the subject. Rohwer (1976) summarizes excretion data and finds three elimination components: 8.7 days for body water, 34 days for exchangeable tritium in organic materials, and 300-600 days for nonexchangeable tritium in organic materials. He concludes that dose estimates that include only the body water tritium contribution could be increased by perhaps 20% to include the dose contribution due to organically bound tritium. Table 3.2 lists reported values for percent contribution of organically bound tritium to total body dose.

Several papers have been omitted from the above reviews, and these will be discussed briefly. No new research has appeared in the literature to disprove the above conclusions.

Istomina and Moskalev (1972) studied the kinetics of tritium accumulation in adult rats and determined the cumulative tissue dose received by animals with chronic  $^3\text{HOH}$  intake. They found that with chronic intake of varying activities, an equilibrium was established for intake, accumulation, and elimination of  $^3\text{HOH}$  within the organism by day 20. After administration ceased, 98.6-99% of the  $^3\text{HOH}$  was eliminated with a half-life of 3-4.3 days and the remainder eliminated with  $T_{1/2} = 19-28$  days. The authors conclude that the change in tritium level in the aqueous phase of the rat organism, within the range of doses studied, is unrelated to the level of administered activity and is described by a multicomponent exponential decay function. Analysis of killed rats indicated that there had been slower accumulation of tritium in the dried tissue residues than in the extracted aqueous phase, as

Table 3.2 Contribution of organically bound tritium  
to total body dose

Dose (%)	Type of exposure	Organism exposed	Reference
16	Acute	Human	Bennett (1972)
6.7	Chronic	Mice	Hatch & Mazrimas (1972)
2	Chronic	Clam	Harrison & Koranda (1971)
8	Acute	Rat	Lambert & Clifton (1967)
5-10	Acute	Rat	Thompson (1954)
9	Chronic	Rat	Thompson & Ballou (1954)
28	Acute	Human	Sanders & Reinig (1968)
1-2	Chronic	Human	Pinson et al. (1952)
2	Chronic	Human	Bush (1972)
1.6	Chronic	Human	Snyder (1968)
10	Theoretic	Human	Croach (1973)
30	Chronic	Deer	Evans (1969)
2	Acute	Human	Balonov et al. (1974)
7.5-38	Chronic	Rat	Istomina & Moskalev (1972)



well as slower elimination as discussed above. At apparent equilibrium, during chronic exposure, the quantity of tritium in the residue phase constituted 9% of the total activity in the organism. During 64 days of administration, 92.5% of the integrated dose was delivered by tritium in the aqueous phase and 7.5% by tritium in the residue phase. After stopping administration, 62% of the cumulative tissue dose was from the aqueous phase and 38% from the dried residue phase.

Balonov et al. (1974) studied the transport kinetics of body fluids in man after acute intravenous injection, inhalation, and ingestion of  $^3\text{HOH}$ . The injection procedure duration was reported as 10 s. The excretion of incorporated tritium following ingestion was followed for 300 days, with the basic component of excretion ( $T_{1/2} \sim 12$  days) appearing as well as a second component of about 39-76 days. A four-compartment mathematical model is suggested for  $^3\text{HOH}$  metabolism. Their dose estimates show that a 2% increase in the whole-body dose is due to the organic tritium pool, and that 5-30% of the total dose to soft tissues is accounted for by radiation from organic tritium.

The transfer and incorporation of tritium in mammals as studied for the International Atomic Energy Agency (IAEA) Research Programme on the Behavior of Tritium in the Environment is summarized by Van den Hoek (1979). Uptake in organic constituents following administration of  $^3\text{HOH}$  was studied. In cows' milk the ratio of specific activities for organic milk components vs milk water was found to be 0.30 for casein and 0.60 for lactose. Calves fed tritiated milk powder for 28 days were compared with calves fed water containing  $^3\text{HOH}$ , and a much higher incorporation of tritium into organs and tissues was found for those calves fed organically bound tritium. On the average, 15 times more tritium was incorporated and this represented 4.1% of the ingested tritium. These data on uptake of organically bound tritium in foods reported by Van den Hoek are of a preliminary nature but indicate a need for further investigations.

Bogen et al. (1979) report a continuation of their study on tritium distribution in man and his environment. They confirm that the ratios of activity concentrations of tritium in bound vs loose tissue fractions decrease as the trophic levels are ascended. Tritium was found to be

distributed homogeneously within the organs of each individual studied. The concentration in body water of animals and humans was similar to the concentrations found in environmental water sources. The "bound" tritium values were the same for each organ within an animal, but the tritium concentrations were found to be higher than in the "loose" fraction. The ratio of "loose" to "bound" tritium activity concentrations in food was found to be about 4, and Bogen estimates that about 60% of the dietary intake of tritium is from the "loose" fraction. The "loose" to "bound" fraction in human tissues was found to be about 2.

It should be pointed out that these data do not prove an enrichment or accumulation of tritium in man. As discussed in NCRP Report No. 62 (1979a), the longer biological half-life for the organic constituent, combined with the recent reductions in environmental concentrations of weapons-related tritium (resulting in rapidly reduced concentrations in free body water), can temporarily produce what appears to be an enrichment in the organic tritium component in man and other organisms.

It appears from the range of values reported for the total body dose contribution from organically bound tritium that the 20% increase in dose recommended by Rohwer (1976) is a reasonable estimate. The NCRP (1979a) also suggests multiplying the dose to body water by a factor of 1.2 to account for "combined" tritium in tissue, and this approach is followed in the methodology we are proposing. This estimate is based on a dose model which assumes a biological half-life of 9, 30, and 450 days for tritium in the body (Bennett, 1973), representing the "body water," and two "organic" compartments, respectively.

### 3.3 Regional and Site-Specific Absolute Humidity Data for Use in Tritium Dose Calculations

As mentioned in Chap. 2, absolute humidity is a key parameter in the calculation of dose from environmental releases of tritium. Due to dilution of released tritium by airborne water vapor, dose is found to be inversely proportional to absolute humidity. The comparison of methodologies presented in that chapter assumed a value of  $6 \text{ g H}_2\text{O/m}^3$ ;

however, actual values may vary considerably depending upon the geographical region in which dose is being evaluated. Because of the wide variability of absolute humidity within the United States, it is helpful to take a closer look at regional absolute humidity data from which appropriate values for performing radiological assessments may be estimated.

Absolute humidity was estimated for 218 points across the United States, using information from the 1977 Annual Summary of U. S. Climatological Data (NOAA, 1977). These data points are tabulated elsewhere (Etnier, 1980). Most climatological data are in the form of relative humidity, which we convert to absolute humidity for use in dose calculations.

The relative humidity ( $H_R$ ) is the ratio of the actual to saturation water vapor pressure at a given temperature, (i.e.,  $H_R = P_a/P_s$ ). The absolute humidity ( $H_a$ ) is the actual vapor content expressed in grams per cubic meter, [i.e.,  $H_a = (n/v)m$ , where  $(n/v)$  is the moles of water per unit volume and  $m$  is the molecular weight of water (18 g  $H_2O$ /mole)].

From the ideal gas law, we find:

$$\frac{n}{v} = \frac{P_a}{RT}, \quad (3.2)$$

where

$P_a$  = actual water vapor pressure in atmospheres,

$R$  = gas constant in atmospheres·m<sup>3</sup>/mole·degree K ( $8.2057 \times 10^{-5}$ ),

$T$  = absolute temperature in degrees K.

Therefore, we can calculate absolute humidity ( $H_a$ ) in grams per cubic meter in terms of  $H_R$  as:

$$H_a = \frac{P_s}{RT} m H_R \quad (3.3)$$

where

$P_s$  = the saturation water vapor pressure at temperature  $T$ .

Calculated absolute humidity data for the continental United States were arbitrarily grouped in the following ranges ( $\text{g}/\text{m}^3$ ): 3.0-5.5, 5.6-7.5, 7.6-9.5, 9.6-11.5, 11.6-16.5. Data points were plotted on a U.S. map, and topography and major river systems were utilized to help delineate areas falling within the various ranges. Figure 3.1 shows the areas in which the selected ranges fall. The mean value for each range is listed on the figure.

The U. S. Nuclear Regulatory Commission Guide 1.109 (USNRC, 1977) currently recommends using a default value based on growing season of  $8 \text{ g H}_2\text{O}/\text{m}^3$  in lieu of site-specific data for absolute humidity, while the National Council on Radiation Protection and Measurements (NCRP, 1979a) uses a value of  $6 \text{ g H}_2\text{O}/\text{m}^3$ , which is a reported Northern Hemisphere mid-latitude mean value (NOAA, 1976). The range of values for the United States is 3.0-16.5  $\text{g}/\text{m}^3$ ; use of the NRC default value of  $8 \text{ g H}_2\text{O}/\text{m}^3$ , for example, would thus yield dose estimates which are within a factor of 3 of the extremes of individual site-specific data.

The data presented in Fig. 3.1 allow choice of a regional estimate of absolute humidity when appropriate for use in tritium dose calculations. Section 4.3.5 of this report presents data indicating the relative impact of variations in absolute humidity values upon doses estimated for a typical nuclear facility.

### 3.4 Calculation of Tritium Drinking Water Activity Dilution Coefficients for Use in Estimation of Radiological Dose

In certain specific cases, choice of a dilution coefficient for tritium entering a drinking water supply can significantly influence total calculated doses for a fuel cycle facility. While a default value for the activity dilution coefficient is often used in dose calculations, application of a more site-specific factor is appropriate when data are available. It is not difficult to calculate an activity dilution coefficient for tritium entering a lake at a given rate.

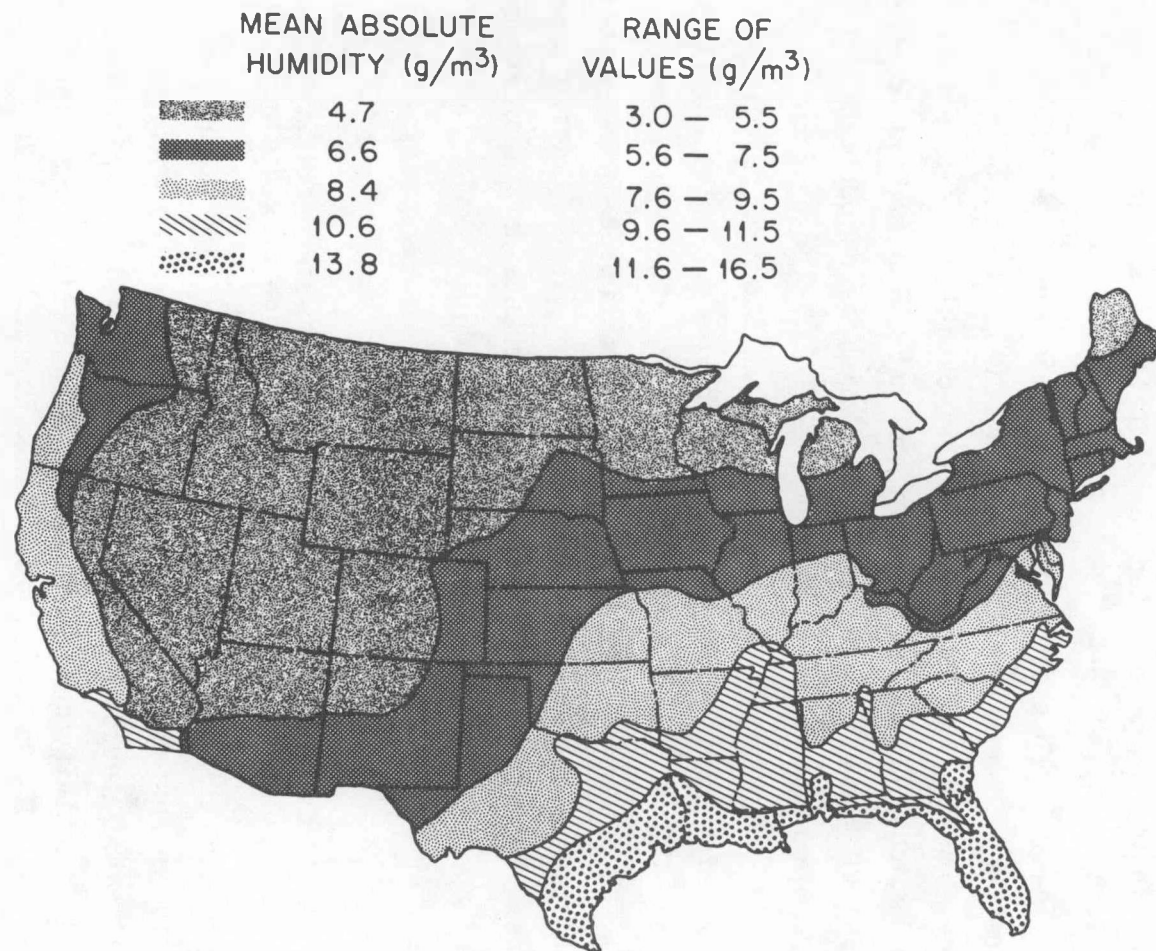


Fig. 3.1 Absolute humidity by geographical region

### 3.4.1 Aquatic releases

Peterson et al. (1969) describe a chain lake model to be used in calculating tritium dilution in lakes when the tritium input rate is known. The time variation of tritium concentration  $[C(t)]$  is described by the expression

$$C(t) = \frac{R}{v\lambda} (1 - e^{-\lambda t}), \quad (3.1)$$

where\*

$R$  = tritium input to lake in Ci/year,

$v$  = lake volume ( $m^3$ ),

$\lambda = \lambda_r + \lambda_e$ ,

$\lambda_r$  = tritium decay constant (0.056/year),

$\lambda_e = q/v$  = physical removal constant ( $year^{-1}$ ),

$q$  = rate of water flow through lake ( $m^3/year$ ) (outflow plus evaporation),

and complete mixing of the lake is assumed.

Equation (3.1) may be used to calculate tritium concentrations for a variety of conditions, representing a range of lake sizes and charge/discharge rates. For our purposes, tritium concentrations are calculated for three input periods: 1, 5, and 20 years. The range of lake volumes chosen includes small reservoirs ( $1000\text{ m} \times 1000\text{ m} \times 10\text{ m}$ ;  $10^7\text{ m}^3$ ) to large lakes  $[(4 \times 10^5\text{ m}) \times (4 \times 10^5\text{ m}) \times 30\text{ m}$ ;  $5 \times 10^{12}\text{ m}^3]$ . The physical removal constant,  $\lambda_e$ , is determined both by rate of water outflow per year and the rate of evaporation assumed for a given site. We chose a range of water outflow of 0 to 5 lake volumes per year (Peterson et al., 1969) as representative of the majority of lakes. Based on U. S. Department of Commerce data (1968), we chose a range of lake evaporation coefficients for the United States as 0.1 to 2 m/year, resulting in ranges of evaporative loss rates of from  $1 \times 10^5$  to  $2 \times 10^6$

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\*1 Ci = 37 gBq.

m<sup>3</sup>/year (small lakes), to  $1.6 \times 10^{10}$  to  $3.2 \times 10^{11}$  m<sup>3</sup>/year (large lakes). The value of  $\lambda_e$  therefore ranges from

$$[(0 + 1 \times 10^5) \div 1 \times 10^7] \text{ year}^{-1} = 10^{-2}/\text{year}, \text{ to}$$

$$[(5 \times 10^7 + 2 \times 10^6) \div 1 \times 10^7] \text{ year}^{-1} = 5.2/\text{year}$$

for small lakes, and from

$$[(0 + 1.6 \times 10^{10}) \div 5 \times 10^{12}] \text{ year}^{-1} = 3.2 \times 10^{-3}/\text{year}, \text{ to}$$

$$[(5 \times 10^{12} + 3.2 \times 10^{11}) \div 5 \times 10^{12}] \text{ year}^{-1} = 1.06/\text{year}$$

for large lakes.

For our purposes,  $\lambda_e$ 's ranging from 0.01/year to  $\sim 10$ /year suffice to approximate the range of physical removal constants for all lake sizes.

Table 3.3 lists activity dilution coefficients for the above conditions. The tabulated  $\lambda$  values equal  $\lambda_e + \lambda_r$ . To calculate tritium activity in drinking water, multiply the appropriate activity dilution coefficient by a tritium input rate, in Ci/year. Table 3.3 indicates that, even for a relatively small lake ( $10^7$  m<sup>3</sup>) with no outflow and a low evaporation rate ( $\lambda = 6.6 \times 10^{-2}/\text{year}$ ), and for a long duration of tritium release ( $\tau = 20$  years), the dilution coefficient is found to be quite small ( $\sim 10^{-6}$  year/m<sup>3</sup>). For large lakes and less restrictive conditions, factors to  $10^{-13}$  are calculated. It is recommended that, when applicable, site-specific water-supply data be utilized, in conjunction with coefficients such as those listed in Table 3.3, to estimate dilution of tritium entering drinking water supplies via liquid releases from nuclear facilities.

As discussed in Chap. 2 of this report, a code such as AIRDOS-EPA (Moore et al., 1979) estimates the dose due to tritium in drinking water as being proportional to the total tritium activity taken into the body via this mode. Thus, the dose is proportional to the concentration of tritium in drinking water, multiplied by the quantity of water ingested per unit time. Killough et al. (1978) recommend use of a total body dose conversion factor (DCF) for tritium ingestion of  $8.3 \times 10^{-5}$  rem/ $\mu\text{Ci}$  ( $2.2 \times 10^{-11}$  Sv/Bq). If the tritium input rate to a drinking water reservoir of known size is available, Table 3.3 and the above DCF can be

Table 3.3. Dilution coefficients for prolonged tritium release into lakes (year/m<sup>3</sup>)

Lake Volume (m <sup>3</sup> ):		10 <sup>7</sup>	10 <sup>8</sup>	10 <sup>9</sup>	10 <sup>10</sup>	10 <sup>11</sup>	10 <sup>12</sup>
$\lambda(\text{year}^{-1})^a$	$\tau(\text{year})^b$						
6.6 E-2 <sup>c</sup>	1	9.68E-8	9.68E-9	9.68E-10	9.68E-11	9.68E-12	9.68E-13
6.6 E-2	5	4.26E-7	4.26E-8	4.26E-9	4.26E-10	4.26E-11	4.26E-12
6.6 E-2	20	1.11E-6	1.11E-7	1.11E-8	1.11E-9	1.11E-10	1.11E-11
0.156	1	9.26E-8	9.26E-9	9.26E-10	9.26E-11	9.26E-12	9.26E-13
0.156	5	3.47E-7	3.47E-8	3.47E-9	3.47E-10	3.47E-11	3.47E-12
0.156	20	6.13E-7	6.13E-8	6.13E-9	6.13E-10	6.13E-11	6.13E-12
0.056	1	6.18E-8	6.18E-9	6.18E-10	6.18E-11	6.18E-12	6.18E-13
0.056	5	9.42E-8	9.42E-9	9.42E-10	9.42E-11	9.42E-12	9.42E-13
0.056	20	9.47E-8	9.47E-9	9.47E-10	9.47E-11	9.47E-12	9.47E-13
5.06	1	1.97E-8	1.97E-9	1.97E-10	1.97E-11	1.97E-12	1.97E-13
5.06	5	1.98E-8	1.98E-9	1.98E-10	1.98E-10	1.98E-12	1.98E-13
5.06	20	1.98E-8	1.98E-9	1.98E-10	1.98E-11	1.98E-12	1.98E-13
10.06	1	9.94E-9	9.94E-10	9.94E-11	9.94E-12	9.94E-13	9.94E-14
10.06	5	9.94E-9	9.94E-10	9.94E-11	9.94E-12	9.94E-13	9.94E-14
10.06	20	9.94E-9	9.94E-10	9.94E-11	9.94E-12	9.94E-13	9.94E-14

<sup>a</sup> $\lambda$  = (<sup>3</sup>H decay constant) + (lake water physical removal constant).

<sup>b</sup> $\tau$  = duration of <sup>3</sup>H release.

<sup>c</sup>Read as 6.6 × 10<sup>-2</sup>.



employed to calculate dose. If such information is not available, or if a generic study employing hypothetical facilities and populations is involved, another approach to the problem of estimating dose due to intake of drinking water may be necessary.

#### 3.4.2 Atmospheric releases

When calculating dose to the maximally exposed individual, a number of conservative, or dose-maximizing, assumptions may be made to compensate for uncertainties in data and modeling. With respect to dose from tritium in drinking water, it might be conservatively assumed that the maximally exposed individual drinks water in which the ratio of tritium to hydrogen is the same as that for atmospheric moisture. Dose is then simply proportional to the concentration of tritium in atmospheric moisture (as estimated via a plume dispersion/depletion model) and to the quantity of drinking water ingested by an individual. The assumption of equal activity concentrations implies that; (1) the contents of the drinking water reservoir come entirely from local atmospheric moisture as rain, and (2) this rain is at activity equilibrium with tritium in the atmospheric "plume" from the nearby tritium-releasing facility under study. Under such assumptions, the atmosphere/drinking water tritium dilution coefficient (ADDC – tritium activity concentration in atmospheric moisture  $\div$  the tritium activity concentration in rainwater) would equal one. However, assumptions leading to an ADDC of one are unnecessarily conservative. A maximally exposed individual residing in, and deriving sustenance from, an area typically 1000 - 2000 m from a tritium-releasing facility (Tennery et al., 1976; Tennery et al., 1978), will drink water not only from the contaminated plume, but also from noncontaminated clouds above the plume, and imported from uncontaminated sources. Estimates as to the dilution of "plume" tritium due to washout by uncontaminated rainwater may be made using data from the Savannah River Laboratory, a facility regularly releasing significant quantities of  $^3\text{HOH}$  to the atmosphere.

Tadmor (1973) reports measured maximum concentrations of tritium in the atmosphere and in rainwater at the perimeter of the Savannah River

Laboratory (SRL) as  $6.8 \times 10^{-9}$  and  $1.8 \times 10^{-4}$  uCi/cm<sup>3</sup>, respectively. The main SRL releases are assumed to occur at the center of the site, about 10 km from the perimeter. Using a mean absolute humidity value of 10.7 g/m<sup>3</sup> for the SRL area (extracted from Fig. 3.1), the tritium concentration in atmospheric moisture for the SRL air concentration reported by Tadmor is:

$$6.8 \times 10^{-9} \frac{\mu\text{Ci}}{\text{cm}^3} \times 1 \times 10^6 \frac{\text{cm}^3}{\text{m}^3} \times \frac{1 \text{ m}^3}{10.7 \text{ g}} = 6.4 \times 10^{-4} \frac{\mu\text{Ci}}{\text{g}}$$

$$(2.4 \times 10^{-2} \text{ kBq}),$$

and the ratio of tritium concentration in atmospheric moisture to that in rainwater is

$$(6.4 \times 10^{-4} \frac{\mu\text{Ci}}{\text{g}}) / 1.8 \times 10^{-4} \frac{\mu\text{Ci}}{\text{cm}^3} \times \frac{1 \text{ cm}^3}{\text{g}} = 3.5 ,$$

which we have defined as the ADDC (above).

Ashley et al. (1979) report equations describing average annual <sup>3</sup>HOH concentrations in atmospheric and rainwater, based on observations at 31 monitoring stations at distances from 3 to 40 km from a "median" central release point on the SRL site (actual releases occur from at least seven locations on site). While a (statistically unverified) trend toward increased dilution with distance from the source is noted, the average ADDC calculated from the Ashley et al. report is 2.2 (range 1.7-2.8 for the 1975-77 results reported).

Section 4.5 of this report describes the impact of the ADDC on the estimated total-body dose to a maximally exposed individual, under a specific set of conditions. For ADDC's of up to approximately 35, ingestion of tritium via the drinking water pathway can contribute significantly to dose. It is clear that the choice of a tritium dilution coefficient value for a specific environmental assessment represents a potentially significant contribution to the error associated with overall estimated dose.



## 4. CALCULATION OF LOCAL DOSE – THE IMPACT OF TRITIUM-RELATED VARIABLES

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### 4.1 The Relative Importance of Tritium in Calculating Local Doses from Fuel Reprocessing

A portion of the Fast Reactor Safety Program has involved engineering and environmental analysis of routine releases of radionuclides from nuclear fuel cycle facilities. This research has been performed by members of ORNL's Health and Safety Research, Metals and Ceramics, and Chemical Technology Divisions. Two studies of the environmental radiological impact of reprocessing advanced fuels have been performed to date (Tennery et al., 1976; Tennery et al., 1978), using methodologies described in Sect. 4.2. In both cases, tritium contributed the majority of estimated dose to total body and most individual organs for the maximally exposed individual, with the assumption that no containment of tritium was employed at the reprocessing plants. In addition, tritium is consistently found to contribute a significant part of the dose from reprocessing of light water reactor fuel (Finney et al., 1977) and high temperature gas-cooled reactor fuel (Davis et al., 1976). Kaye and Till (1978) have pointed out the need for renewed consideration of tritium in nonproliferative fuel cycles. The importance of tritium with respect to radiological dose calculations is one of the driving forces behind the preparation of this document.

### 4.2 Baseline Case Methodology

This section considers the impact of tritium releases from a nuclear facility, focusing on doses to a hypothetical maximally exposed individual, and analyzing the effect of variations in several key parameters which influence calculated radiological doses. In order to consider the impact of tritium releases for a representative situation, we have performed a number of calculations using the AIRDOS-EPA methodology (Moore et al., 1979) to estimate the environmental transport and uptake by man of a typical spectrum of radionuclides released from a hypothetical nuclear fuel reprocessing facility. A detailed discussion of the

methods involved in such a design effort, detailed estimates of the airborne release rates (or source term) of the various radionuclides handled at the facility, and calculations of consequent environmental dispersion and dose to man, are available in Tennery et al. (1978). A summary of these procedures is presented here.

An analytical model for calculating reprocessing facility radiological impact requires several types of information: (1) reactor core description and fuel loading masses, isotopic composition of the fuel charge, core neutronics, and fuel management data, including spent fuel mass charged to the reprocessing plant; (2) spent fuel isotopic composition; (3) flow sheets identifying the quantities of various isotopes released to the environment during reprocessing; and (4) meteorology, population, and animal and food crop data for the facility's location.

Reactor fuel loading data were estimated for a 1200 MW(e) LMFBR design reported by Combustion Engineering (Caspersson et al., 1978). The ORIGEN computer code (Bell, 1973) was used to calculate isotopic distributions for spent fuel recycled to near equilibrium. Spent fuel was assumed to be stored for one year prior to reprocessing. The reactor plant was assumed to generate 1200 MW(e) for 511 full-power days, operating 70% of the time, resulting in an annual core and blanket refueling fraction of 0.5. The radial blanket annual refueling fraction was 0.2.

Conceptual flow diagrams were developed to estimate airborne radioactive releases during routine operation of the reprocessing facility handling spent fuel from such reactors. Several guidelines were assumed for the model facility: (1) the spent fuel had generated 50 GW(e)-years of electricity at a burnup of 25,500 MW(thermal)·d/MT heavy metal and a thermal/electric conversion efficiency of 35.5%; (2) feed to the reprocessing plant was a blended mixture of the core and axial and radial blankets; (3) radionuclide confinement factors (quantity of a radionuclide released to the atmosphere divided by the quantity of that isotope processed) shown in Table 4.1 were identical to those used in earlier studies, including an assumption of no confinement of tritium at the plant (Tennery et al., 1976; Tennery et al., 1978); (4) spent fuel

Table 4.1 Radionuclide release rates for a hypothetical LMFBR fuel reprocessing facility<sup>a</sup>

Radionuclide	Confinement factor <sup>b</sup>	Release rate (Ci/year) <sup>c</sup>
<sup>3</sup> H	1	8.26E4 <sup>d</sup>
<sup>14</sup> C	1E2	38
<sup>85</sup> Kr	1E2	3.3E5
<sup>90</sup> Sr	5E9	3.7E-2
<sup>106</sup> Ru	1E9	6.8E-2
<sup>129</sup> I	1E4	7.9E-3
<sup>137</sup> Cs	5E9	3.5E-2
<sup>144</sup> Ce	5E9	2.3E-1
<sup>212</sup> Pb (daughter of <sup>220</sup> Rn)	1.2E7 <sup>e</sup>	3.8
<sup>228</sup> Th	5E8	6.6E-3
<sup>232</sup> U	5E8	1.2E-2
<sup>233</sup> U	5E8	2.3E-3

<sup>a</sup>Tennery et al. (1978); 2013 Gg heavy metal reprocessed per year to support a 50 GW(e) reactor system.

<sup>b</sup>Activity processed/activity released to atmosphere.

<sup>c</sup>1 Ci = 37 GBq.

<sup>d</sup>Read as 8.26 x 10<sup>4</sup>.

<sup>e</sup><sup>220</sup>Rn decays to <sup>212</sup>Pb with T<sub>1/2</sub> = 55.6 s.

<sup>f</sup>Assumes complete decay of <sup>220</sup>Rn → <sup>212</sup>Pb.

contained 10% of the total tritium generated at the reactors (NCRP, 1979a); (5) 0.69% of the fuel cladding (stainless steel) was dissolved during reprocessing; and (6) the fuel's nitrogen impurity (principal source of  $^{14}\text{C}$ ) was 300 ppm. Based on these assumptions, release rates for the 12 radionuclides contributing significantly to dose were calculated and are listed in Table 4.1. These radionuclides are assumed to be released on a routine basis from a 100-m stack.

The radiological impact to man resulting from operation of the model reprocessing plant is assessed by calculating the dose, in millirems\*, to a maximally exposed individual residing 1 km from the facility. Radionuclides released to the atmosphere reach man through one or more pathways (air, soil, or water), with varying degrees of transfer by food, animals, and crops. Pathways of human exposure include inhalation, ingestion, immersion in air, land surface contamination (exposure to  $\gamma$  or  $\beta$  emitters depositing on land surfaces), and submersion in water (swimming). Ultimately, energy is deposited in human tissue by decay of the radionuclides, via internal ( $\alpha$ ,  $\gamma$ ,  $\beta$ ) or external ( $\gamma$ ,  $\beta$ ) exposure.

Radiological impact is routinely assessed as the 50-year dose commitment resulting from one year's operation of a facility. For certain radionuclides which are retained for long periods in bone, (e.g.,  $^{232}\text{U}$ ,  $^{233}\text{U}$ ) dose from one year's exposure will continue to accrue, at a declining rate, for the lifetime of an individual (Till et al., in press). For many radionuclides, including tritium, essentially all of the 50-year dose commitment will accrue during the first year after exposure. Dose commitments are calculated using implementations of various metabolic models as discussed in Moore et al. (1979), Killough et al. (1978), Dunning et al. (1979), and Kocher (1979).

Utilizing the radionuclide source term and dose estimation methodologies described above, it is possible to determine the sensitivity of the estimated dose to variation of several key parameters related to tritium production and environmental transport. The parameters to be analyzed in detail in the remainder of this section are: (1) the

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\*1 millirem = 10  $\mu\text{Sv}$ .

effects of increased production rates of tritium in LMFBRs; (2) the impact of variation of the tritium quality factor; (3) the impact of variations in the tritium dilution coefficient for drinking water; and (4) the impact of choice of average absolute humidity values. To provide a basis for evaluation of the impact on dose of each of these parameters, the fuel reprocessing plant source term developed above (Table 4.1) is first analyzed utilizing the AIRDOS-EPA methodology (Moore et al., 1979).

It should be emphasized that containment of tritium to a significant degree within a reprocessing plant is feasible, as mentioned earlier in this report. For purposes of comparison, however, and because such containment procedures would require justification for large capital expenditures, tritium doses calculated in this section assumed no tritium confinement technology applied at the reprocessing facility.

The AIRDOS-EPA computer code (Moore et al., 1979) requires user choice of a number of variables, allowing "tailoring" of the calculation to represent more closely a specific site. In order to allow comparison of the following results with previous studies of a similar type (Tennery et al., 1978; Tennery et al., 1976; USAEC, 1974b), the meteorological summaries input to the code are identical to those described in a USAEC study (1974b). Conditions used in estimating the expected ground-level air concentrations of radionuclides were obtained by averaging meteorological data taken from 18 stations in the continental United States. Table 4.2 summarizes key data input to the AIRDOS-EPA code to calculate the radiological doses presented in the remainder of Chap. 4.

Radiological doses calculated with the AIRDOS-EPA methodology are presented in Table 4.3. Percent contributions to dose for this "baseline case" are listed in Table 4.4. This methodology and set of input parameters deliver maximum dose to an individual residing at the plant boundary, 1000 m from the reprocessing plant stack. This hypothetical individual is assumed to derive all of his food from crops and animals raised near his residence, and to drink water from a stream or reservoir which dilutes the tritium activity concentration by a factor of 100 from that in air. Doses are calculated to be in the range of 4 to 6.5



Table 4.2 Selected parameters input to the AIRDOS-EPA computer code<sup>a</sup>

Parameter	Value
Average air temperature	286.1°K
Rainfall rate	0.762 m/y
Stack height <sup>b</sup>	100 m
Effluent velocity <sup>c</sup>	20 m/s
Tritium deposition velocity <sup>c</sup>	$1.0 \times 10^{-3}$ m/s
Tritium scavenging coefficient	$1.6 \times 10^{-5}$ /s

<sup>a</sup>Moore et al. (1979).

<sup>b</sup>Height of reprocessing plant stack discharging process air to atmosphere.

<sup>c</sup>Coefficients influencing rate of removal of tritium from an air-borne plume as it moves downwind.

Table 4.3 Baseline case: 50-year dose commitment<sup>a</sup> to maximally exposed individual (no tritium confinement)

Radionuclide	Dose (millirem <sup>b</sup> )				
	Total body	Bone	Lungs	Thyroid	GI tract
<sup>3</sup> H-food <sup>c</sup>	2.7	2.7	2.7	2.7	2.7
<sup>3</sup> H-water <sup>c</sup>	0.02	0.02	0.02	0.02	0.02
<sup>3</sup> H-inhalation <sup>c</sup>	<u>0.44</u>	<u>0.34</u>	<u>0.44</u>	<u>0.43</u>	<u>0.47</u>
<sup>3</sup> H-subtotal	3.16	3.06	3.16	3.15	3.19
<sup>14</sup> C	0.24	0.38	0.10	0.11	0.18
<sup>85</sup> Kr	0.02	0.03	0.04	0.02	0.02
<sup>90</sup> Sr	0.10	0.90	0.00	0.01	0.08
<sup>106</sup> Ru	0.02	0.02	0.02	0.02	0.23
<sup>129</sup> I	0.00	0.00	0.00	1.2	0.00
<sup>137</sup> Cs	0.20	0.27	0.17	0.19	0.13
<sup>144</sup> Ce	0.01	0.01	0.03	0.00	0.66
<sup>220</sup> Rn <sup>d</sup>	0.03	0.05	0.20	0.02	0.02
<sup>228</sup> Th	0.03	0.52	0.10	0.00	0.02
<sup>232</sup> U	0.06	0.83	0.23	0.01	0.02
<sup>208</sup> Tl <sup>e</sup>	0.15	0.17	0.14	0.17	0.13
<sup>212</sup> Bi <sup>e</sup>	0.03	0.03	0.02	0.02	0.02
<sup>216</sup> Po <sup>e</sup>	0.01	0.23	0.00	0.00	0.10
<sup>224</sup> Ra <sup>e</sup>	0.00	0.01	0.00	0.00	0.00
<sup>233</sup> U	<u>0.00</u>	<u>0.00</u>	<u>0.03</u>	<u>0.00</u>	<u>0.00</u>
Total	4.03	6.49	4.23	4.89	4.77

<sup>a</sup>From one year's operation of reprocessing facility; 2013 Gg heavy metal/year throughput.

<sup>b</sup>1 millirem = 10 mSv.

<sup>c</sup>Dose from <sup>3</sup>H exposure modes: food intake, drinking water (dilution coefficient = 100), inhalation.

<sup>d</sup>Dose from <sup>220</sup>Rn daughters.

<sup>e</sup>Built-up on ground surfaces from decay of parent nuclides.

Table 4.4 Baseline case: Percent contributions to 50-year dose commitment<sup>a</sup> to maximally exposed individual (no tritium confinement)

Radionuclide	Percent of dose				
	Total body	Bone	Lungs	Thyroid	GI tract
<sup>3</sup> H-food <sup>b</sup>	67	41.6	63.8	55.2	56.6
<sup>3</sup> H-water <sup>b</sup>	0.5	0.3	0.5	0.4	0.4
<sup>3</sup> H-inhalation <sup>b</sup>	<u>10.9</u>	<u>5.2</u>	<u>10.4</u>	<u>8.8</u>	<u>9.9</u>
<sup>3</sup> H subtotal	78.4	47.1	74.7	64.4	66.9
All other radionuclides	21.6	52.9	25.3	35.6	33.1

<sup>a</sup>One year reprocessing facility operation; 2013 Gg heavy metal/year throughput.

<sup>b</sup>Fractions from <sup>3</sup>H individual exposure modes.

millirem (40 to 65  $\mu\text{Sv}$ ), with 47 to 78% of the total radiological dose to a specific organ due to intake of tritium.

### 4.3 Analysis of the Impact of Potentially Increased Tritium Production Rates in LMFBR Fuels

#### 4.3.1 Revised ternary fission yield data for fast reactor fuels

One reason for renewed emphasis on tritium is the important data published by Buzzelli et al. (1976), involving provisional experimental values for the ternary fast fission yield in the EBR-II reactor of the fertile isotopes  $^{232}\text{U}$  and  $^{238}\text{U}$ , indicating increased tritium yields. Prior to these data, no fast fission tritium yields for these species had been determined experimentally. Additionally, Buzzelli and Langer (1977) reported new information on the tritium yield from fast fission of  $^{233}\text{U}$ ,  $^{235}\text{U}$ , and  $^{239}\text{Pu}$ . Assessments of tritium production, transport, and release from fast breeder reactors have until recently been based on estimates of yield, because of lack of experimental data concerning ternary fast fission tritium yield of the two primary fissile materials,  $^{235}\text{U}$  and  $^{239}\text{Pu}$ . We have summarized the most recent findings of Buzzelli and Langer in Table 4.5. The yield for tritium from ternary fission of  $^{233}\text{U}$ ,  $^{238}\text{U}$ , and  $^{239}\text{Th}$  in a fast spectrum is much higher than the values listed for the ENDF/BIV (1974) library which have been used in previous radiological assessments investigating advanced uranium-plutonium fast breeder reactor fuels (Tennery et al., 1976; Tennery et al., 1978). Buzzelli and Langer (1977) emphasize the preliminary nature of their observations and the need for further studies. Additional EBR-II samples will be analyzed under the Oak Ridge National Laboratory (ORNL) Fast Reactor Safety Program.

The significance of these relatively high, ternary-fast-fission-yield values can be evaluated in terms of the amount of tritium that could ultimately be released to the environment. Table 4.6 lists estimated source terms for tritium released to the atmosphere during reprocessing of spent (U, Pu) carbide and (Th, U) carbide fuels using fission yields from the ENDF/BIV (1974) library and the experimental values

Table 4.5 Tritium yield from fast neutron  
ternary fission

Isotope	10 <sup>-4</sup> Tritons/Fission	
	Buzzelli et al. <sup>a</sup>	ENDF/BIV Library <sup>b</sup>
<sup>232</sup> Th	6	2.0
<sup>238</sup> U	9	2.3
<sup>235</sup> U	1.5	1.1
<sup>233</sup> U	15	1.4
<sup>239</sup> Pu	20	1.8

<sup>a</sup>Based on irradiations in the fast flux of EBR-II (Buzzelli et al., 1976; Buzzelli and Langer, 1977).

<sup>b</sup>ENDF/BIV library-tapes 401-411 and 414-419, National Neutron Cross Section Center, Brookhaven National Laboratory (Dec. 1974).

Table 4.6 Tritium source terms for reprocessing spent (U, Pu)C and (Th, U)C fuels using ternary fission yields in the ENDF/BIV library<sup>a</sup> and those reported by Buzzelli et al.<sup>b, c</sup>

Fuel type	10 <sup>-4</sup> Tritons/Fission		Tritium released <sup>d</sup> during reprocessing (Ci/Mg) <sup>e</sup>	Blended fuel equivalent in 50 GW(e)-year (Mg)	Tritium released per 50 GW(e)-year equivalent of spent fuel (Ci) <sup>e</sup>
	<u><sup>238</sup>U</u>	<u><sup>239</sup>Pu</u>			
(U, Pu)C <sup>f</sup>					
Values from Tennery et al. (1976)	2.3	1.8	6.5 x 10 <sup>1</sup>	1415	9.2 x 10 <sup>4</sup>
Revised with Buzzelli et al. (1976, 1977) yields	9	20	6.7 x 10 <sup>2</sup>	1415	9.5 x 10 <sup>5</sup>
(Th, U)C <sup>g</sup>					
	<u><sup>232</sup>Th</u>	<u><sup>233</sup>U</u>			
Values from Tennery et al. (1978)	2.0	1.4	4.1 x 10 <sup>1</sup>	2013	8.3 x 10 <sup>4</sup>
Revised with Buzzelli et al. (1976, 1977) yields	6	15	4.3 x 10 <sup>2</sup>	2013	8.7 x 10 <sup>5</sup>

<sup>a</sup>See footnote 6, Table 4.5.

<sup>b</sup>Buzzelli et al. (1976).

<sup>c</sup>Buzzelli and Langer (1977).

<sup>d</sup>Assumes only 10% of tritium generated during irradiation remains with the fuel transported to the reprocessing facility; assumes all tritium entering the reprocessing facility is released to the atmosphere (confinement factor for tritium equals 1).

<sup>e</sup>1 Ci = 37 GBq.

<sup>f</sup>Using ANL 2050 MW(e) design provided by Barthold as reported in ORNL-5230 (Tennery et al., 1976).

<sup>g</sup>Using CE 1200 MW(e) design provided by Caspersson as reported in ORNL/TM-6493 (Tennery et al., 1978).

reported by Buzzelli et al. (1976), and Buzzelli and Langer (1977). These data indicate an increase in the production rate of tritium of approximately one order of magnitude.

#### 4.3.2 Impact on dose of increased tritium production rates

All else being equal, increased production rates as discussed above would be directly reflected in increased tritium release rates at an LMFBR-fuel reprocessing facility, resulting in increased estimated radiological doses to exposed individuals. While verification of the initial results of Buzzelli et al. is necessary prior to final evaluation of the consequences of such increased tritium production rates, it is of interest to provide initial estimates for planning purposes. To allow comparison to the data presented in Tables 4.3 and 4.4, the reprocessing facility radionuclide source term listed in Table 4.1 is again used as input to the AIRDOS-EPA code, holding all input variables constant with the exception of the tritium source term component. A range of tritium release rates, from  $1.6 \times 10^5$  Ci/year ( $5.9 \times 10^6$  GBq/year) to  $8.7 \times 10^5$  Ci/year ( $3.2 \times 10^7$  GBq/year) from the Buzzelli and Langer (1977) data was chosen for these calculations. Table 4.7 lists estimated doses to a maximally exposed individual from exposure to routine releases of a nuclear fuel reprocessing plant as described earlier in this chapter. Table 4.8 lists percent contribution of tritium to total dose for the reprocessing plant releases as a function of increasing tritium release rates. Baseline case data are also included in Table 4.8 to facilitate comparison of the effects of the four tritium release rates (baseline source term plus the three new source term values) considered.

It is apparent from this analysis that potential increased production of tritium in fast breeder reactors leads to parallel increases in reprocessing facility dose estimates. If a federal radiation protection standard such as the EPA's 40 CFR 190 (USEPA, 1977) were applied to the LMFBR fuel cycle (this standard applies only to light water reactor systems), Case III (Table 4.7) doses would exceed the 25 millirem (250

Table 4.7 The radiological impact of increased tritium production rates: 50-year dose commitment<sup>a</sup> to a maximally exposed individual (no tritium confinement)

Radionuclide	Dose (millirem <sup>b</sup> )				
	Total body	Bone	Lungs	Thyroid	GI tract
CASE I					
<sup>3</sup> H release rate:					
1.6 x 10 <sup>5</sup> Ci/year <sup>c</sup>					
<sup>3</sup> H-food <sup>d</sup>	5.21	5.21	5.21	5.21	5.21
<sup>3</sup> H-water <sup>d</sup>	0.05	0.05	0.05	0.05	0.05
<sup>3</sup> H-inhalation <sup>d</sup>	0.85	0.67	0.85	0.84	0.90
<sup>3</sup> H Subtotal	<u>6.11</u>	<u>5.93</u>	<u>6.11</u>	<u>6.10</u>	<u>6.16</u>
All other radionuclides	<u>.90</u>	<u>3.46</u>	<u>1.10</u>	<u>1.77</u>	<u>1.61</u>
Total	7.01	9.39	7.21	7.87	7.77
CASE II					
<sup>3</sup> H release rate:					
4.0 x 10 <sup>5</sup> Ci/year <sup>c</sup>					
<sup>3</sup> H-food	13.04	13.04	13.04	13.04	13.04
<sup>3</sup> H-water	0.12	0.12	0.12	0.12	0.12
<sup>3</sup> H-inhalation	2.12	1.67	2.12	2.10	2.25
<sup>3</sup> H Subtotal	<u>15.28</u>	<u>14.83</u>	<u>15.28</u>	<u>15.26</u>	<u>15.41</u>
All other radionuclides	<u>0.89</u>	<u>3.45</u>	<u>1.09</u>	<u>1.76</u>	<u>1.61</u>
Total	16.05	18.28	16.37	17.02	17.02
CASE III					
<sup>3</sup> H release rate:					
8.7 x 10 <sup>5</sup> Ci/year <sup>c</sup>					
<sup>3</sup> H-food	28.35	28.35	28.35	28.35	28.35
<sup>3</sup> H-water	0.26	0.26	0.26	0.26	0.26
<sup>3</sup> H-inhalation	4.61	3.63	4.61	4.57	4.90
<sup>3</sup> H Subtotal	<u>33.22</u>	<u>32.24</u>	<u>33.22</u>	<u>33.18</u>	<u>33.51</u>
All other radionuclides	<u>0.90</u>	<u>3.46</u>	<u>1.10</u>	<u>1.76</u>	<u>1.61</u>
Total	34.12	35.70	34.32	34.94	35.12

<sup>a</sup>From one year's operation of reprocessing facility.

<sup>b</sup>1 millirem = 10 mSv.

<sup>c</sup>1 Ci = 37 GBq.

<sup>d</sup>Dose from tritium exposure modes: food ingestion, drinking water intake, inhalation.



Table 4.8 The impact on dose of increased tritium production rates: percent contribution to dose<sup>a</sup> to a maximally exposed individual (no tritium confinement)

Radionuclide	Percent of dose				
	Total body	Bone	Lungs	Thyroid	GI tract
CASE I					
<sup>3</sup> H release rate: 1.6 x 10 <sup>5</sup> Ci/year <sup>b</sup>					
<sup>3</sup> H	87	63	85	78	79
All other radionuclides	13	27	15	22	21
CASE II					
<sup>3</sup> H release rate: 4.0 x 10 <sup>5</sup> Ci/year <sup>b</sup>					
<sup>3</sup> H	95	81	93	90	91
All other radionuclides	5	19	7	10	9
CASE III					
<sup>3</sup> H release rate: 8.7 x 10 <sup>5</sup> Ci/year <sup>b</sup>					
<sup>3</sup> H	97	90	97	96	95
All other radionuclides	3	10	3	4	5
BASELINE CASE (FROM TABLE 4.1)					
<sup>3</sup> H release rate: 8.26 x 10 <sup>4</sup> Ci/year <sup>b</sup>					
<sup>3</sup> H	78	47	75	64	67
All other radionuclides	22	53	25	36	33

<sup>a</sup>From one year's facility operation.

<sup>b</sup>1 Ci = 37 GBq.

$\mu\text{Sv}$ ) limit for total body, bone, lungs, and GI tract [under this standard, the allowable dose to thyroid is 75 millirem (750  $\mu\text{Sv}$ )], implying the need to utilize tritium confinement technology to ensure compliance.

#### 4.4 Impact on Dose of an Increase in the Tritium Quality Factor

Section 3.1 of this report analyzes recent data pertaining to the relative biological effectiveness of tritium beta particles, with respect to reference 250 kVp x-radiation. While currently available data do not allow us to make a firm recommendation regarding choice of tritium quality factor, it appears that a conservative approach to calculation of radiological dose implies the use of a quality factor of 1.7, as opposed to the currently recommended value of 1.0 (NCRP, 1979b). Given this possibility, it is of interest to analyze the impact on estimated dose of such a modification. For purposes of comparison, the data presented in Tables 4.3 and 4.4 ("baseline" data), developed from the fuel reprocessing plant radionuclide source term listed in Table 4.1, are modified to reflect the increased dose commitments associated with the higher quality factor value. Table 4.9 presents increased dose commitments to a maximally exposed individual under such modified conditions. These increases range from 33 to 55%, indicating that the impact of such a modification is significant with respect to compliance with regulatory standards.

#### 4.5 Impact on Dose of Variations in the Drinking Water Dilution Coefficient

Section 3.4 of this report presents data defining a range of tritium dilution coefficients in drinking water. These coefficients can be applied on a site-specific basis to estimate dose from drinking water contaminated by tritium released initially to the atmosphere or to streams and rivers flowing into reservoirs and lakes. In this section, we use the AIRDOS-EPA methodology to analyze the impact on radiological dose of variations in the atmosphere/drinking water tritium dilution coefficient (ADDC = tritium activity concentration in atmospheric

Table 4.9. The impact on dose of an increase in the tritium quality factor: 50-year dose commitment to a maximally exposed individual<sup>a</sup> (no tritium confinement)

Radionuclide	Dose (millirem) <sup>b</sup>				
	Total body	Bone	Lungs	Thyroid	GI tract
<u>Q = 1.7</u> <sup>c</sup>					
<sup>3</sup> H-food <sup>d</sup>	4.6	4.6	4.6	4.6	4.6
<sup>3</sup> H-water <sup>d</sup>	0.03	0.03	0.03	0.03	0.03
<sup>3</sup> H-inhalation <sup>d</sup>	<u>0.75</u>	<u>0.58</u>	<u>0.75</u>	<u>0.73</u>	<u>0.80</u>
<sup>3</sup> H Subtotal	5.38	5.21	5.38	5.36	5.43
All other radionuclides	0.90	3.45	1.08	1.77	1.61
Tritium contribution total dose	86%	60%	83%	75%	77%
<u>Q = 1</u> <sup>c</sup>					
<sup>3</sup> H-food <sup>d</sup>	2.7	2.7	2.7	2.7	2.7
<sup>3</sup> H-water <sup>d</sup>	0.02	0.02	0.02	0.02	0.02
<sup>3</sup> H-inhalation <sup>d</sup>	<u>0.44</u>	<u>0.34</u>	<u>0.44</u>	<u>0.43</u>	<u>0.47</u>
<sup>3</sup> H Subtotal	3.16	3.06	3.16	3.15	3.19
All other radionuclides	0.90	3.45	1.08	1.77	1.61
Tritium contribution total dose	78%	47%	75%	64%	67%
Total dose increase due to <sup>3</sup> H quality factor increase	55%	33%	52%	45%	47%

<sup>a</sup>From one year's reprocessing facility operation (Baseline case - see Table 4.4).

<sup>b</sup>1 millirem = 10 mSv.

<sup>c</sup>Q = quality factor.

<sup>d</sup>Doses from tritium exposure modes.

moisture ÷ tritium activity concentration in rainwater), over a range between 1 (no dilution, i.e., tritium in drinking water at the same activity concentration as in the atmosphere) and 100. As in previous analyses in this chapter, data from Tables 4.1, 4.3, and 4.4 form the basis for the comparisons.

Table 4.10 presents data for four dilution coefficients: 1, 10, 35 and 100. A factor of 100 is commonly used as a "default value" in AIRDOS-EPA computer code calculations; the "baseline" dose estimates (Tables 4.3 and 4.4) incorporate this dilution coefficient, and are listed as "Case IV" in Table 4.10. For the specific conditions of methodology and radionuclide source term used in these calculations, choice of a tritium dilution factor larger than approximately 35 results in no significant change in estimated dose. At the level of dilution (100) commonly used in AIRDOS-EPA analyses, the contribution to dose of tritium in drinking water is small, of the order of 0.5%. Only in circumstances in which site-specific conditions indicate use of a dilution coefficient of less than 35 will the drinking water pathway contribute greatly to dose, under the conditions stated.

#### 4.6 Impact on Dose of Variations in Average Absolute Humidity

Data presented in Sect. 3.3 indicate that average absolute humidity values over the United States range from 3-16.5 g/m<sup>3</sup>. A default value of 8 g/m<sup>3</sup> (based on average growing season absolute humidity values) is often used in radiological dose calculations to estimate dose resulting from routine operations of nuclear facilities (Moore et al., 1979; USNRC, 1977). Using the methodologies discussed in this section, and the reprocessing plant data from Table 4.1, we estimate the impact on dose of the use of absolute humidity values other than 8 g/m<sup>3</sup>. Table 4.11 lists dose commitments, to a maximally exposed individual, for absolute humidity values of 4, 8, and 16 g/m<sup>3</sup>. The dose estimates using 8 g/m<sup>3</sup> are taken from Table 4.3. Variation of absolute humidity over the given range results in variations in estimated total dose of up to 78%. It is apparent from these data that choice of site-specific

Table 4.10 The impact on dose of variations in the atmosphere/drinking water dilution coefficient (ADDC)<sup>a</sup>—50 year dose commitment to a maximally exposed individual<sup>b</sup> (no tritium confinement)

	Dose (millirem) <sup>c</sup>				
	Total body	Bone	Lungs	Thyroid	GI tract
CASE I: <sup>3</sup> H ADDC = 1) <sup>a</sup>					
<sup>3</sup> H-food	2.7	2.7	2.7	2.7	2.7
<sup>3</sup> H-water	2.5	2.5	2.5	2.5	2.5
<sup>3</sup> H-Inhalation	0.44	0.34	0.44	0.43	0.47
All other radio-nuclides <sup>d</sup>	0.9	3.5	1.1	1.8	1.6
Tritium contribution to total	86%	61%	84%	76%	78%
CASE II: <sup>3</sup> H ADDC = 10) <sup>a</sup>					
<sup>3</sup> H-food	2.7	2.7	2.7	2.7	2.7
<sup>3</sup> H-water	0.29	0.29	0.29	0.29	0.29
<sup>3</sup> H-inhalation	0.44	0.34	0.44	0.43	0.47
All other radio-nuclides <sup>d</sup>	0.9	3.5	1.1	1.8	1.6
Tritium contribution to total	79%	49%	76%	66%	68%
CASE III: <sup>3</sup> H ADDC = 35) <sup>a</sup>					
<sup>3</sup> H-food	2.7	2.7	2.7	2.7	2.7
<sup>3</sup> H-water	0.07	0.07	0.07	0.07	0.07
<sup>3</sup> H-inhalation	0.44	0.34	0.44	0.43	0.47
All other radio-nuclides <sup>d</sup>	0.9	3.5	1.1	1.8	1.6
Tritium contribution to total	78%	47%	74%	64%	67%
CASE IV: <sup>3</sup> H ADDC = 100) <sup>a</sup>					
<sup>3</sup> H-food	2.7	2.7	2.7	2.7	2.7
<sup>3</sup> H-water	0.02	0.02	0.02	0.02	0.02
<sup>3</sup> H-inhalation	0.44	0.34	0.44	0.43	0.47
All other radio-nuclides <sup>d</sup>	0.9	3.5	1.1	1.8	1.6
Tritium contribution to total	78%	47%	74%	64%	67%

<sup>a</sup>ADDC = atmosphere/drinking water tritium activity concentration dilution coefficient.

<sup>b</sup>From one year's reprocessing facility operation.

<sup>c</sup>1 millirem = 10 μSV.

<sup>d</sup>From Table 4.3 (sum of organ doses for radionuclides other than <sup>3</sup>H).

Table 4.11 The impact of variations in average absolute humidity:  
50-year dose commitment to a maximally exposed individual<sup>a</sup>  
(no tritium confinement)

	Dose (millirem) <sup>b</sup>				
	Total body	Bone	Lungs	Thyroid	GI tract
Absolute humidity 4 g/m <sup>3</sup>					
<sup>3</sup> H-food	5.4	5.4	5.4	5.4	5.4
<sup>3</sup> H-water	0.04	0.04	0.04	0.04	0.04
<sup>3</sup> H-inhalation	0.88	0.68	0.88	0.86	0.96
<sup>3</sup> H Subtotal	6.32	6.12	6.32	6.30	6.38
All other radio-nuclides <sup>c</sup>	0.9	3.45	1.08	1.77	1.61
<sup>3</sup> H contribution to total dose	88%	64%	85%	78%	80%
Absolute humidity 8 g/m <sup>3</sup>					
<sup>3</sup> H-food	2.7	2.7	2.7	2.7	2.7
<sup>3</sup> H-water	0.02	0.02	0.02	0.02	0.02
<sup>3</sup> H-inhalation	0.44	0.34	0.44	0.43	0.47
<sup>3</sup> H Subtotal	3.16	3.06	3.16	3.15	3.19
All other radio-nuclides <sup>c</sup>	0.9	3.45	1.08	1.77	1.61
<sup>3</sup> H contribution to total dose	78%	47%	74%	64%	67%
Absolute humidity 16 g/m <sup>3</sup>					
<sup>3</sup> H-food	1.35	1.35	1.35	1.35	1.35
<sup>3</sup> H-water	0.01	0.01	0.01	0.01	0.01
<sup>3</sup> H-inhalation	0.22	0.17	0.22	0.22	0.23
<sup>3</sup> H Subtotal	1.58	1.53	1.58	1.58	1.59
All other radio-nuclides <sup>c</sup>	0.9	3.45	1.08	1.77	1.61
<sup>3</sup> H contribution to total dose	64%	31%	59%	47%	50%

<sup>a</sup>From one year's facility operation.

<sup>b</sup>1 millirem = 10  $\mu$ Sv.

<sup>c</sup>From Table 4.3.

average absolute humidity data can have a significant impact on estimated dose commitments.

#### 4.7 Summary of the Impact of Tritium-Related Variables on Dose

Data presented in this chapter indicate the relative importance of several key parameters which should be considered during the estimation of dose for a facility releasing large quantities of tritium. It is important to recognize that the variables discussed individually in the section could interact in a multiplicative fashion. The possibility of encountering such cases, with associated high estimated tritium doses, emphasizes the need for careful choice of data or default values utilized in a particular radiological dose assessment. Many of the observations made in this chapter will apply with similar force to a light water reactor fuel cycle involving fuel reprocessing.

## 5. CALCULATION OF THE GLOBAL DOSE

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### 5.1 Scenarios for the Release of Tritium to the Environment by Man

#### 5.1.1 Consumer products

Limited data exist on the historical use of tritium in consumer products (Sect. 1.2.2). For this report, data regarding the distribution of light sources using tritium in sealed glass tubes in the United States for the period 1976 to 1979 were taken from USNRC licensing files; estimates for the U.S. luminous compound industry were derived from the same files. Krejci and Zeller (1979) estimated Swiss use of tritium in the watch industry. Taylor and Webb (1978) made an estimate for the quantity of tritium used in luminous watches in the United Kingdom (U.K.), although no estimates for back-lit watches are available. Making certain licensing assumptions, Wehner (1979) estimated tritium use rates in the Federal Republic of Germany (FRG) for self-luminous wristwatches. Table 5.1 summarizes the above estimates.

The lifetime of a consumer product may be divided into 5 stages: production, distribution, use, repair, and disposal. Assuming reasonable controls, little tritium is released to the atmosphere during normal production, distribution and use (McDowell-Boyer and O'Donnell, 1978b). The critical stage when considering global releases appears to be disposal.

In order to estimate a global source term, several assumptions have been made regarding the use and disposal of wristwatches containing tritium. First of all, it is assumed that production rates of back-lit watches will remain constant from 1980 through 2020. Second, we assume that the average lifetime of a watch is 5 years. Finally, it is assumed that the ultimate disposal of all watches will be by burial in landfills or by incineration. Products buried in landfills are assumed to contain the enclosed tritium for its effective lifetime; however, incineration



Table 5.1 Quantity of tritium used in the watch industry (MCi)<sup>a</sup>  
1976 to 1979

Country	1976	1977	1978	1979
FRG <sup>b</sup>	0.024	0.024	0.024	0.6
U.K. <sup>c</sup>	0.025	0.025	0.025	0.025
Switzerland <sup>d</sup>	0.35	0.35	0.35	0.35
U.S. <sup>e</sup>	0.008	0.013	0.40	0.50
Total	0.41	0.41	0.80	1.48

<sup>a</sup>1 MCi = 37 PBq.

<sup>b</sup>Wehner (1978).

<sup>c</sup>Taylor and Webb (1978); only luminous compounds included.

<sup>d</sup>Krejci and Zeller (1979).

<sup>e</sup>USNRC licensing files.

would release all of the tritium, oxidized to  $^3\text{HOH}$ , to the atmosphere. We assume that 50% of all discarded watches will be incinerated (McDowell-Boyer and O'Donnell, 1978b). Table 5.2 lists the quantity of tritium projected to be released worldwide to the atmosphere under these assumptions.

#### 5.1.2 World nuclear power industry

The projection for nuclear energy growth was divided into two periods, the first period covering the 12 years from 1975 through 1986 and the second extending from 1987 to 2020. Nuclear energy growth during the first period is relatively easy to predict because nuclear plants that will be operating during this time either have already been completed, are currently under construction, or are scheduled for construction. Data for this period were taken from recently published listings (USERDA, 1976b; Kee et al., 1976; IAEA, 1975; AIF, 1976). Included in the listings are descriptions of reactor type, generating capacity, date of availability, and location by country.

The forecast for nuclear growth beyond 1986 incorporates assumptions and data published by the Organization for Economic Cooperation and Development and the International Atomic Energy Agency (OECD, 1975), and by Hanrahan et al. (1976). Projections in our study also incorporate estimates of the contribution from Eastern Bloc countries and China, based on data supplied by the Energy Research and Development Administration (USERDA, 1975).

5.1.2.1 Reactor types and fuel reprocessing. The types of reactors considered in this study include pressurized-water reactors (PWR), boiling-water reactors (BWR), water-cooled graphite-moderated reactors (GMR), fast breeder reactors (FBR), high-temperature gas-cooled reactors (HTGR), and other reactors not included in these categories (e.g., advanced gas reactors and heavy-water-moderated reactors). Among the light-water-cooled reactors (LWR) constructed after 1986, 65% are assumed to be PWRs and 35% BWRs.

Table 5.2 Quantity of tritium released from the incineration of tritium back-lighted watches

Year	Tritium released (MCi/year) <sup>a</sup>
1981	0.16
1982	0.17
1983	0.3
1984	0.55
1985-2020	0.76

<sup>a</sup> 1 MCi = 37 PBq.

Figure 5.1 shows plots of the scenario developed for world nuclear electrical generating capacity by reactor type between 1975 and 2020. Light-water reactors (PWRs and BWRs) will continue to dominate during most of this period, but FBRs will play a more important role during the last decade of the 45-year period as the contribution from LWRs diminishes. Total installed nuclear capacity eventually approaches a steady-state scenario.

Reprocessing of the spent nuclear fuel from the reactor scenario in Fig. 5.1 is projected in Fig. 5.2. Because of the uncertainty associated with fuel reprocessing at this writing, there are no reliable published data that project worldwide fuel reprocessing needs. We estimate the fuel reprocessing requirements on the basis of the factors shown in Table 5.3. We have relied on the estimates of Kee et al. (1976) for LWR and HTGR cycles and those of Tennery et al. (1976) for LMFBRs. It is further assumed that commercial reprocessing of LWR fuel begins in 1980, GMR fuel in 1990, HTGR fuel in 1995, FBR fuel in 2000, and fuel from other reactor types (excepting the HWR) in 1985. Our scenario implies that in 2020 fuel reprocessing reaches equilibrium with nuclear reactor spent fuel output.

5.1.2.2 Source terms and projections for the release of tritium to the atmosphere. Table 5.4 lists the source terms used in this study for tritium releases to the atmosphere by the nuclear industry. The most important site of atmospheric release is at fuel reprocessing plants, although the total release by reactors contributes significantly to the atmospheric global inventory and may exceed that of reprocessing plants when confinement of tritium is used during reprocessing.

It is important to recognize that these source terms account for only that tritium which is released as a gaseous effluent. The radiological impact of tritium released to liquid effluents will be discussed in Sect. 5.3 in some detail. Liquid release rates are considered in Sect. 5.1.2.3.

The atmospheric source terms are listed in Table 5.4 for tritium at the reactor, and at the reprocessing plant as a function of the confinement factor (CF). The confinement factor is defined as the number of

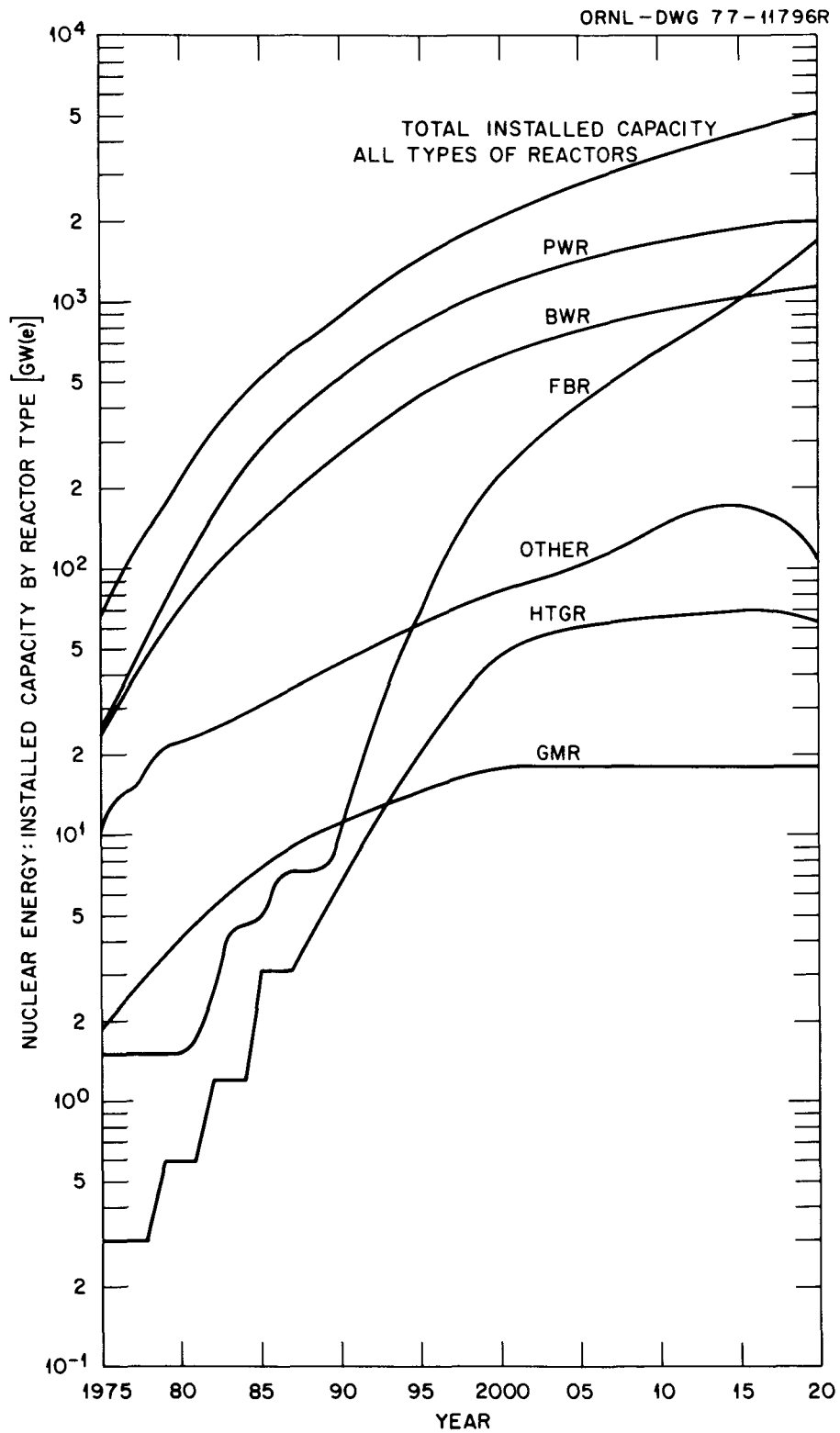


Fig. 5.1 Scenario projecting the growth of nuclear energy world-wide

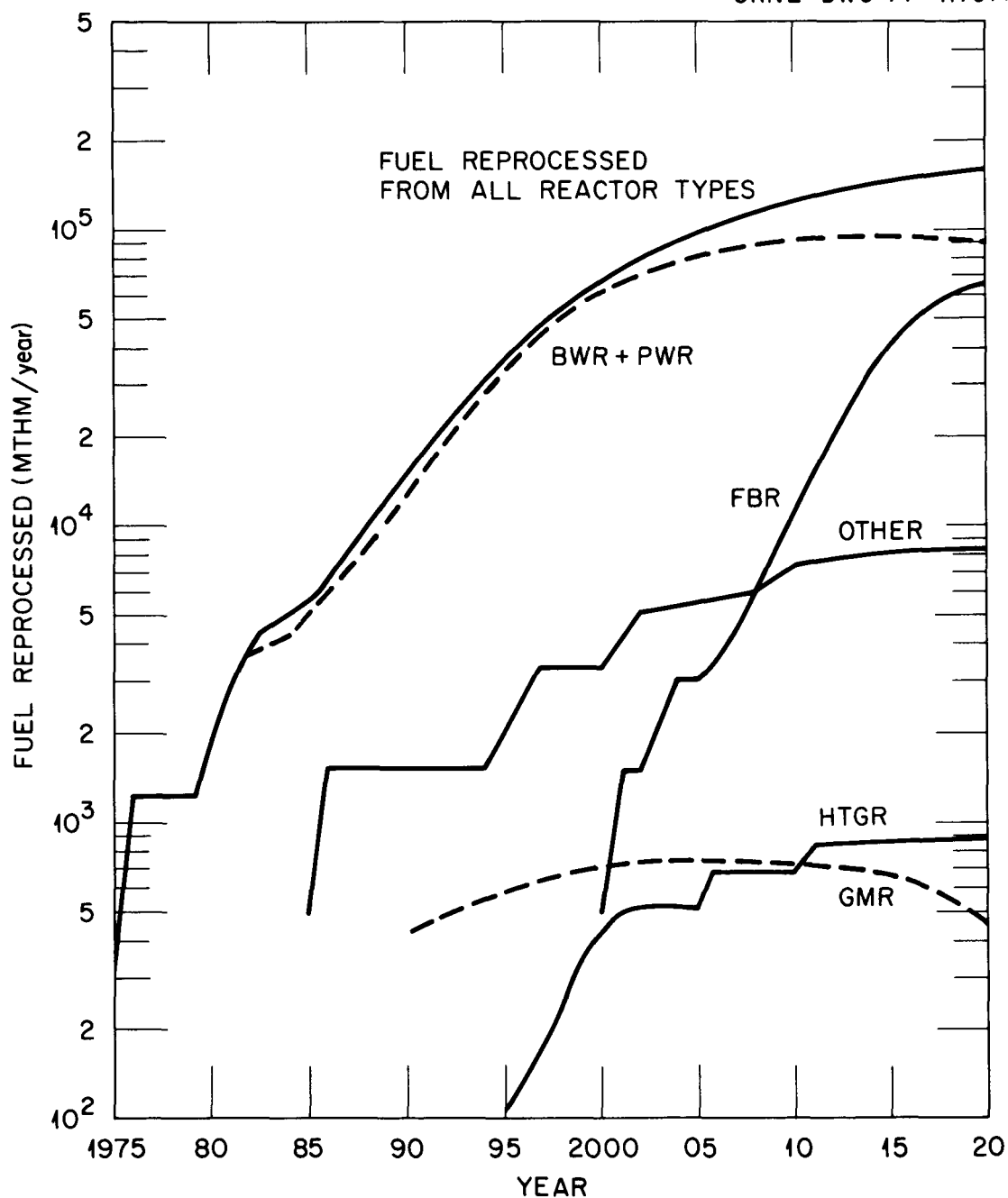


Fig. 5.2 Scenario projecting fuel reprocessing worldwide

Table 5.3 Fuel reprocessing by reactor type

Reactor	Reprocessed fuel [MTHM/GW(e)-year]
PWR	33.5 <sup>a</sup>
BWR	40.2 <sup>a</sup>
GMR	40.0 <sup>b</sup>
FBR	36.8 <sup>c</sup>
HTGR	10.1 <sup>a</sup>
Others	40.0 <sup>b</sup>

<sup>a</sup>Kee et al. (1976).

<sup>b</sup>This is an assumed value for this study.

<sup>c</sup>Tennery et al. (1976).

Table 5.4 Source terms for tritium releases to the atmosphere by the world nuclear power industry

Type of reactor or fuel being reprocessed	Content of $^3\text{H}$ in fuel one year after removal from reactor <sup>a</sup> (Ci/HTHM)	Release rate of $^3\text{H}$ to the atmosphere, Ci/GW(e)-year			
		Reactor	Reprocessing plant confinement factor for $^3\text{H}$		
			1	10	100
PWR	$5.1 \times 10^2$ <sup>a, b</sup>	35 <sup>c</sup>	$1.7 \times 10^4$	$1.7 \times 10^3$	$1.7 \times 10^2$
BWR	$5.1 \times 10^2$ <sup>b</sup>	20 <sup>c</sup>	$1.7 \times 10^4$	$1.7 \times 10^3$	$1.7 \times 10^2$
GMR	$5.1 \times 10^2$ <sup>d</sup>	35 <sup>d</sup>	$1.7 \times 10^4$	$1.7 \times 10^3$	$1.7 \times 10^2$
FBR	$6.7 \times 10^1$ <sup>e</sup>	60 <sup>c</sup>	$2.5 \times 10^3$	$2.5 \times 10^2$	$2.5 \times 10^1$
HTGR	$3.2 \times 10^3$ <sup>f</sup>	20 <sup>d</sup>	$1.3 \times 10^4$	$1.3 \times 10^3$	$1.3 \times 10^2$
Other					
Advanced gas reactors	$6.7 \times 10^2$ <sup>e</sup>	35	$2.7 \times 10^3$	$2.7 \times 10^2$	$2.7 \times 10^1$
Gas-cooled reactors	$6.7 \times 10^1$ <sup>e</sup>	35	$2.7 \times 10^3$	$2.7 \times 10^2$	$2.7 \times 10^1$
Heavy water reactors	NA <sup>g</sup>	$5 \times 10^3$			

<sup>a</sup> Assuming 33,000 MWD(thermal)/MTHM burnup for each type of fuel.

<sup>b</sup> Finney et al. (1977).

<sup>c</sup> NCRP Report No. 62 (1979a).

<sup>d</sup> Assumed value for this study.

<sup>e</sup> Tennery et al. (1976) - using pre-1976 tritium yield data.

<sup>f</sup> Davis et al. (1976).

<sup>g</sup> It is assumed there would be no reprocessing of heavy water reactor fuel.



curies of tritium processed divided by the number of curies released. For example, a confinement factor of 10 implies that 90% of the tritium processed would be contained at the reprocessing plant by a retention system, while a confinement factor of 100 implies that 99% is contained. For most reactor types, tritium confinement does not have meaning in the practical sense because retention systems at the reactor designed to trap tritium would not be economically justifiable. The LMFBR is an exception, since tritium can readily escape from the fuel elements through the cladding and will be collected in cold traps before it can escape into the environment (USAEC, 1974b).

Generally, source terms for gaseous releases at reactors are relatively small compared to reprocessing plants that have no containment of tritium. The heavy-water-moderated reactor, however, such as the CANDU type, is expected to release to the atmosphere approximately 5000 Ci/year (185 TBq/year) per GW(e) (NCRP, 1979a; Gorman and Young, 1979). This input cannot be neglected as it is assumed in our scenario that heavy-water reactors contribute about 4% to the total installed generating capacity between 1985 and 2020. Since the reactors of the CANDU type are fueled with natural uranium, it is assumed there will be no reprocessing of HWR fuel.

In this scenario, a capacity factor of 0.75 is assumed, where the capacity factor is defined as actual power generated [GW(e)-year] divided by the power generated assuming continuous reactor operation [GW(e)-year]. This capacity factor is slightly greater than that assumed by some investigators (Hanrahan et al., 1976; Kelly et al., 1975); however, the use of the 0.75 value in this chapter is intended to reflect expected improvements in reactor technology and reliability.

The release of tritium from reactors was calculated by multiplying the total installed capacity by the corresponding tritium inventory from Table 5.4 and by the capacity factor. For reprocessing plants, the number of metric tons of heavy metal reprocessed each year for each type of reactor fuel was multiplied by the tritium inventory in curies per metric ton heavy metal (MTHM) to give the curies per year of tritium available for release to the atmosphere. The confinement factor is

applied only to reprocessing plants, and accounts for proposed tritium effluent treatment technology.

We have focused our attention on three scenarios of tritium release, which are distinguished from one another by choice of the confinement factor applied to determine atmospheric releases during reprocessing. For convenience, the scenarios are designated according to the following scheme:

- Scenario M (minimal) assumes no containment of tritium during reprocessing ( $CF = 1$ ).
- Scenario A (advanced) is based on 99% containment of tritium during reprocessing ( $CF = 100$ ).
- Scenario I (intermediate) incorporates a phased improvement in effluent treatment technology for tritium at the reprocessing plant ( $CF$  increases to 10 in 1990, 100 in 2000).

Figures 5.3, 5.4, and 5.5 are graphs of each of the three scenarios. In each case the rate of natural production of tritium in the earth's atmosphere is plotted for comparison. As discussed earlier, we assumed a natural background production rate of 4.0 MCi/year (150 PBq/year) based on the value recommended by the NCRP (1979a).

According to scenario "M" (Fig. 5.3), the total release rate to the atmosphere from the nuclear power industry would exceed the natural background production rate before 1990 and would approach a constant rate around 2010 that is approximately  $5 \times 10^7$  Ci/year (1.9 EBq/year). Essentially all of the tritium released to the atmosphere by the nuclear fuel cycle would come from reprocessing nuclear fuel; the contribution from reactors would remain below the natural production rate.

Scenario "A" (Fig. 5.4) indicates that the combined source terms for reactors and reprocessing plants do not exceed the natural production rate and approach a level slightly less than 2 MCi/year (74 PBq/year). This scenario indicates that the contribution from reactors is greater than the contribution from reprocessing facilities. Since effluent treatment technology for tritium at the reactor site does not

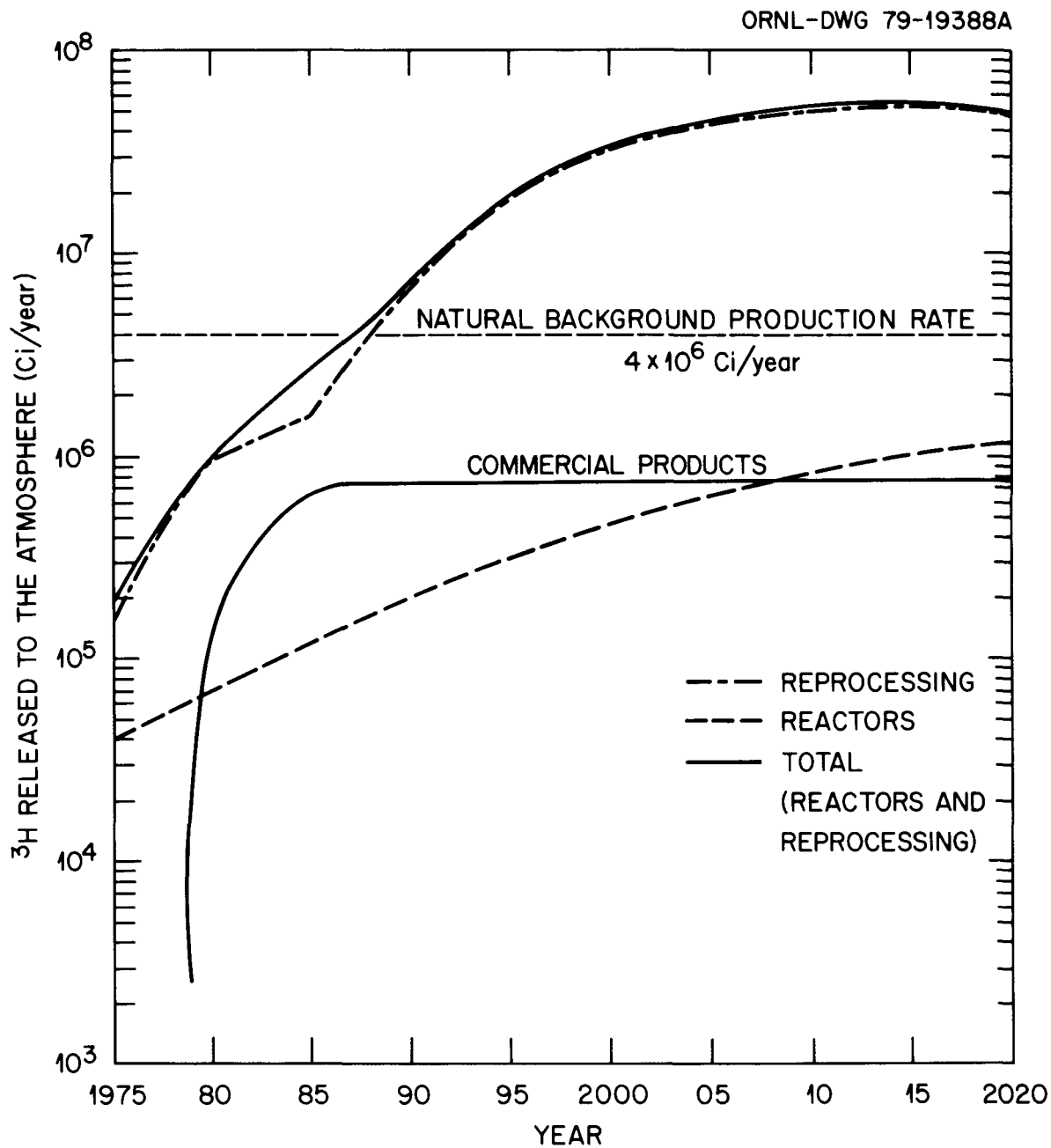


Fig. 5.3 Scenario M (minimal): Tritium released to the atmosphere by the nuclear industry. The reprocessing plant confinement factor = 1

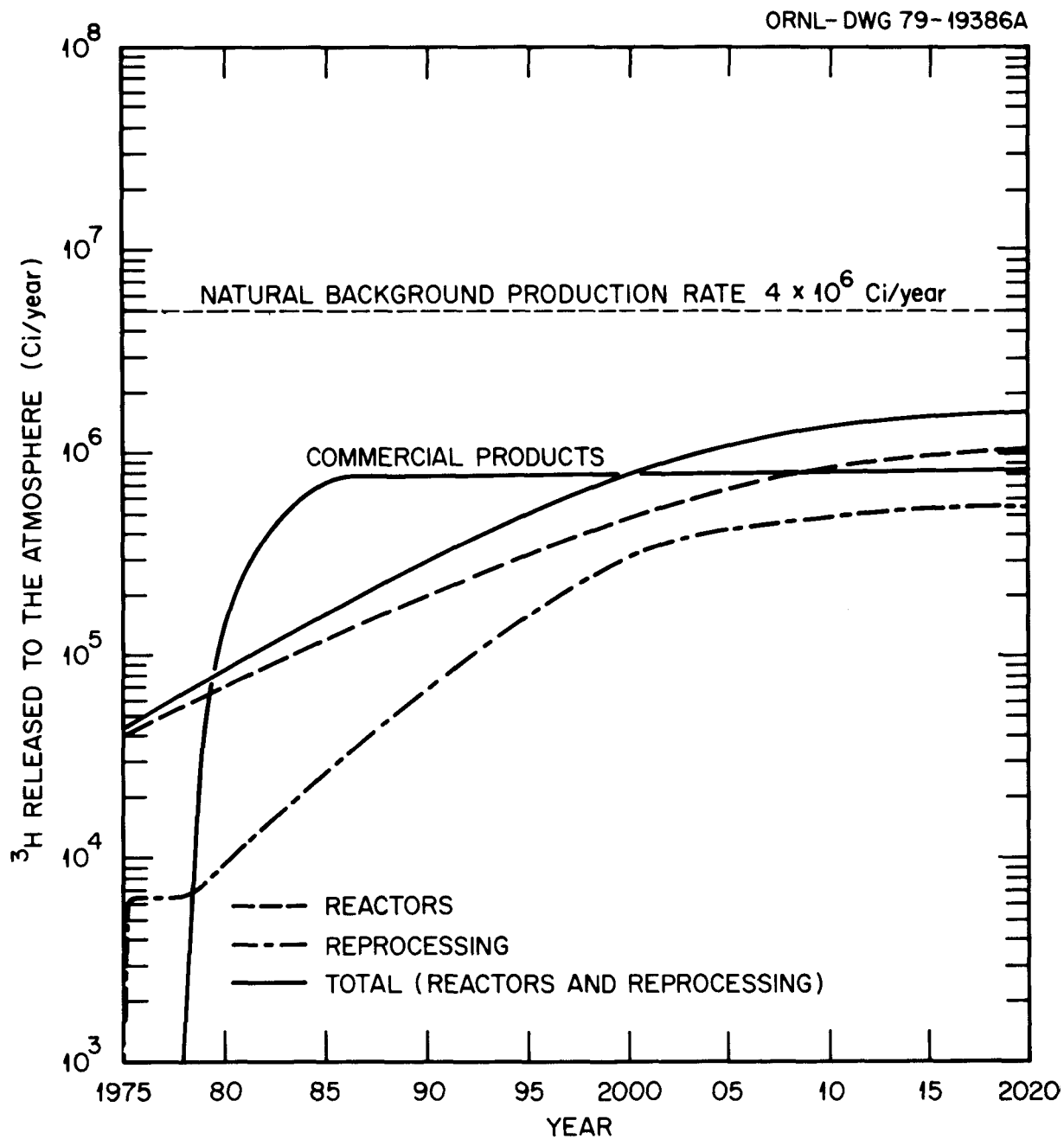


Fig. 5.4 Scenario A (advanced): Tritium released to the atmosphere by the nuclear industry. The reprocessing plant confinement factor = 100

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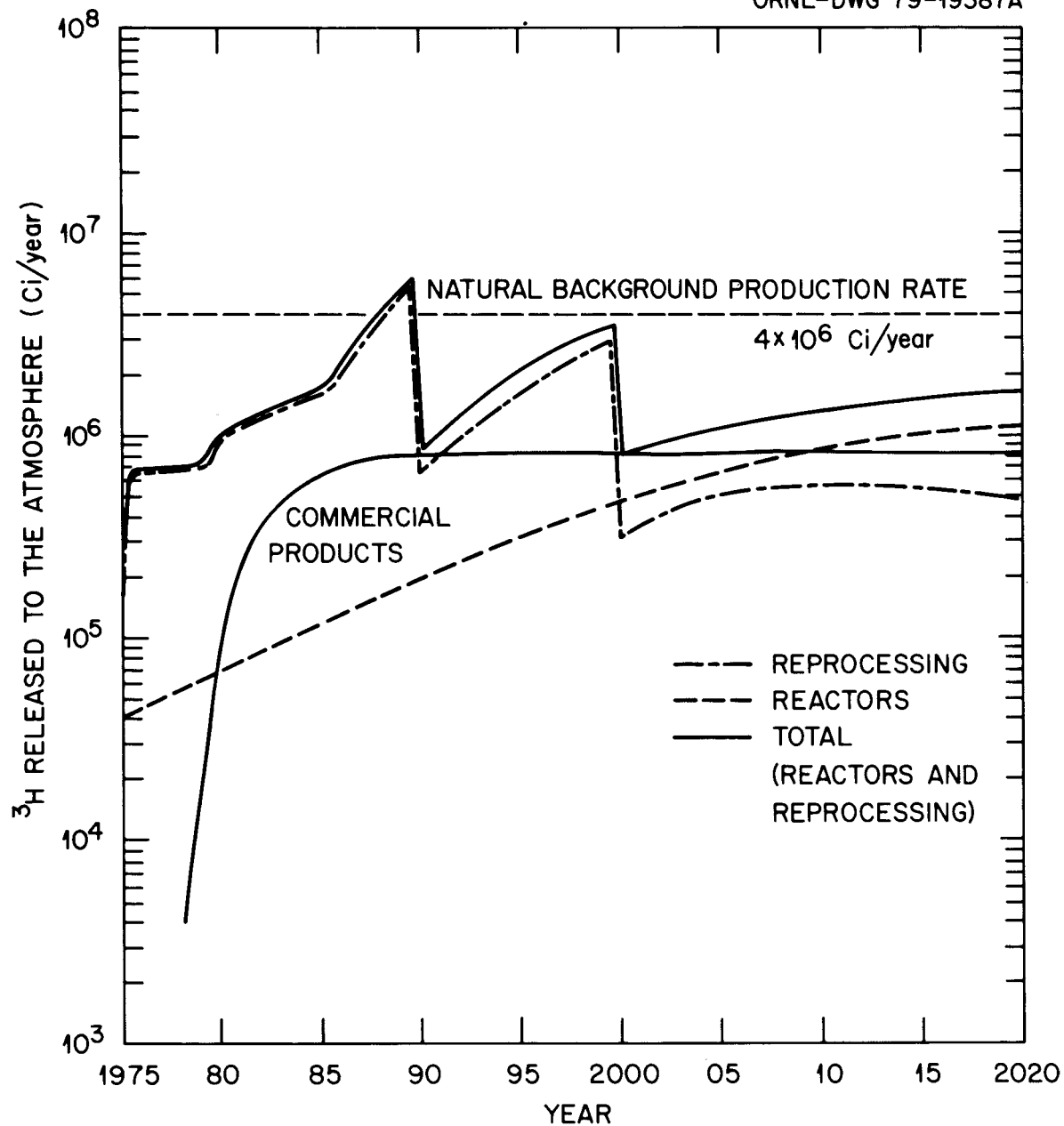


Fig. 5.5 Scenario I (intermediate): Tritium released to the atmosphere by the nuclear industry. The reprocessing plant confinement factor increases in steps to simulate improvements in tritium control technology

currently appear to be economically feasible, this scenario suggests that a confinement factor at reprocessing plants of slightly less than 100 is justifiable. For example, a CF for tritium of 50 instead of 100 would increase the total steady-state production rate in 2020 by about 30% [from 1.7 MCi (63 PBq) to 2.2 MCi (81 PBq) per year]. Obviously, policymakers must maintain an awareness of the diminishing returns of a vigorous effort for tritium containment at reprocessing plants without comparable reductions at the reactors.

Scenario "I" (Fig. 5.5) includes implementation of tritium effluent controls at reprocessing plants in 1990 to provide a CF of 10, and in 2000 to provide a CF of 100. Some improvement is to be expected in the technology for tritium containment even though these improvements may not come in distinct steps as we have shown. This scenario, in its general effect, is perhaps the most plausible of the three.

5.1.2.3 Source terms and projections for the release of tritium to the aquatic environment. Table 5.5 lists the source terms used in this study to predict the release of tritium into the aquatic environment. The only release point in the nuclear fuel cycle would be at the reactor, since it is assumed that no radioactive liquids escape from fuel reprocessing plants during routine operation. Increased release of tritium by PWRs over BWRs is due to the higher PWR production of tritium created by the use of boron to control core reactivity. The source term for tritium released by GMRs is assumed to be similar to that for PWRs.

Among the liquid source terms listed in Table 5.5, the heavy-water-moderated reactor is again the exception. This type of reactor is assumed to release 50,000 Ci/GW(e)-year [1.85 PBq/GW(e)-year] of tritium via liquid effluent (NCRP, 1979a; Gorman and Young, 1979). Although the contribution of heavy-water reactors to the total energy generated by nuclear plants is assumed to be small, this source of tritium cannot be neglected in the scenario.

Figure 5.6 shows tritium released to the aquatic environment by nuclear reactors as a function of reactor type between 1975-2020. The contribution from heavy-water reactors is significantly greater than from any other reactor type. No consideration is given to the possible

Table 5.5 Source terms for tritium released in liquid effluents of nuclear reactors<sup>a</sup>

Type of nuclear reactor	Release rate of <sup>3</sup> H to the environment in liquid effluent [Ci/GW(e)-year <sup>b</sup> ]
BWR	45 <sup>c</sup>
PWR	800 <sup>c</sup>
GMR	800 <sup>d</sup>
FBR	60 <sup>c</sup>
HTGR	50 <sup>d</sup>
Others	
HWR	50,000 <sup>c</sup>
AGR	50 <sup>d</sup>
GCR	50 <sup>d</sup>

<sup>a</sup>It is assumed that no tritium would escape in liquid effluent from reprocessing plants.

<sup>b</sup>1 Ci = 37 GBq

<sup>c</sup>NCRP Report 62 (1979a).

<sup>d</sup>Assumed value for this study.

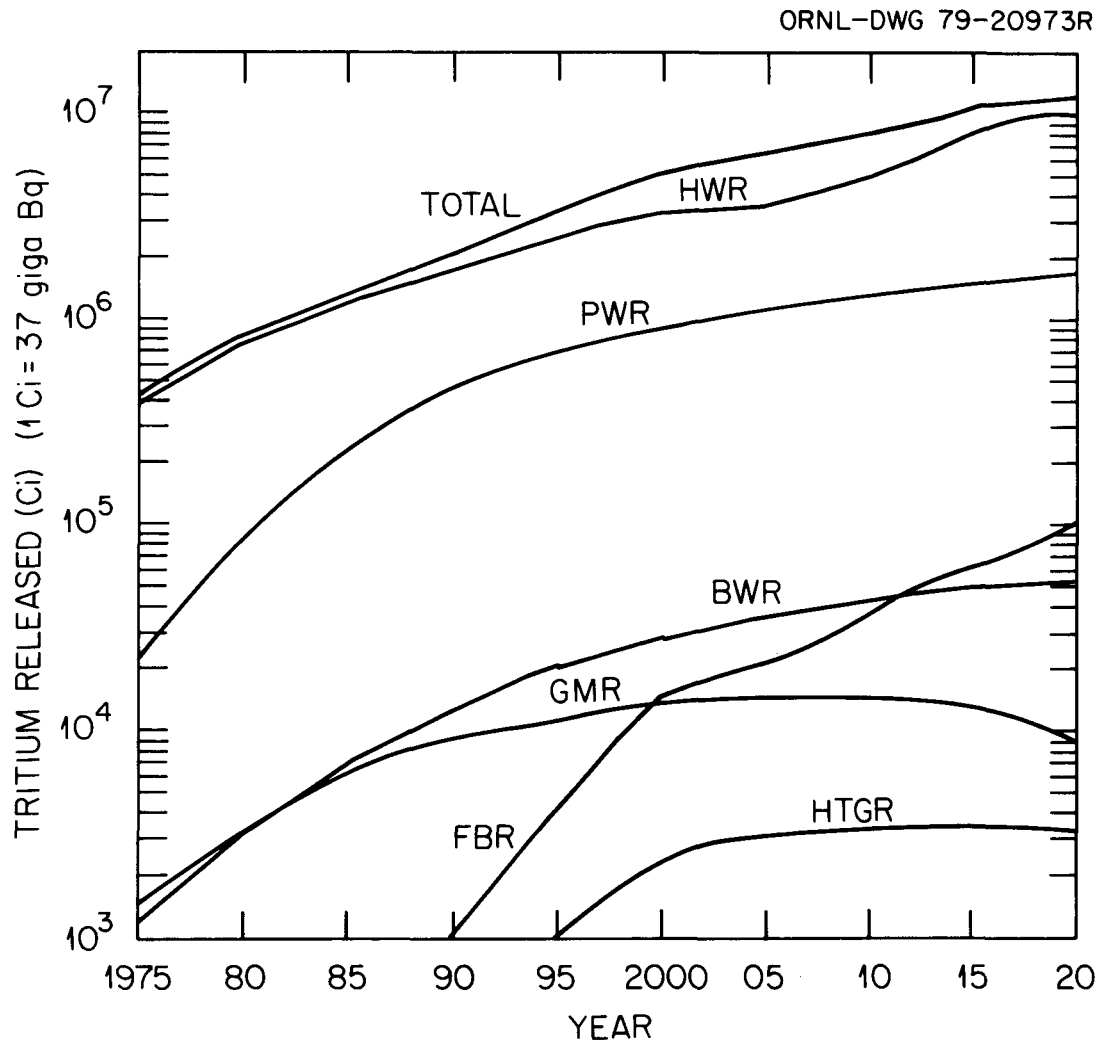


Fig. 5.6 Release of tritium from nuclear reactors to the aquatic environment



selective containment of  $^3\text{H}$  in liquid effluent during this scenario, although treatment of liquid effluent, particularly in heavy-water reactors, may occur in the future.

## 5.2 Global Tritium Modeling

Because of the ubiquitous nature of tritium as  $^3\text{HOH}$ , global population dose estimates due to specific tritium sources have received considerable attention. However, estimates of annual global dose commitments arising from a 1 Ci/year (37 GBq/year) atmospheric release are found to range from an EPA estimate (USEPA, 1973) of  $4 \times 10^{-4}$  man-rem/year, ( $4 \times 10^{-6}$  man-Sv/year) to  $2.2 \times 10^{-2}$  man-rem/year ( $2.2 \times 10^{-4}$  man-Sv/year) obtained by Soldat and Baker (1979). The calculation of global dose requires the estimation of environmental tritium concentrations and their potential biological hazard to an individual. It can be shown (Sect. 2) that, for a given atmospheric concentration of tritium, several dosimetry models yield very similar dose estimates when normalized for comparison. Consequently, we must focus our attention on the estimates of environmental tritium concentrations to explain the range of global dose estimates cited above. In the following we review the basic global tritium distribution models, discuss the uncertainties associated with these models, and present a "reasonable" bound for concentrations obtained from simple assumptions. This discussion excludes the additional consideration of the influence of tritiated gas,  $^3\text{HH}$ , on global dose estimates.

### 5.2.1 Bases of global models

For the purpose of estimating global dose commitments due to specific tritium sources, a global distribution model is assumed to reach a semi-equilibrium state. Thus, no local, regional, or even global first-pass influences are considered. The accuracy of any dose estimates will depend on the accuracy with which the distribution model estimates the tritium concentrations in those environmental pools that directly contribute to individual exposure. Therefore, even though the role of deep

ocean water as a tritium sink may be important to global dynamics, the deep ocean concentration need not be known to estimate doses. The relative isolation of the hydrologic cycles of the northern and southern hemispheres, coupled with the demographic concentration of almost 90% of the world population in the northern hemisphere, justifies the restriction of most models to consideration of northern hemispherical tritium circulation, even though the models are referred to as "global."

The basic assumption of global tritium models is that tritium follows the hydrologic cycle without discrimination. The standard approach to describing tritium distribution is to consider the primary water pools as compartments and to describe equilibrium tritium concentrations in each compartment. The simplest model (USEPA, 1973) treats the entire circulating surface water of the northern hemisphere as a single compartment, ignoring atmospheric water due to its relatively small volume. Bonka (1979) employs a two-compartment box model with an ocean mixed layer and an ocean deep layer.

The difficulty with these simple models is that they have large compartments, in which the tritium concentrations are assumed to be uniform. In nature, tritium concentrations vary considerably, even in the equilibrium state, between various aqueous pools and geographical locations.

To construct a model that accurately reflects environmental equilibrium concentrations, a natural step is to increase the number of compartments to reflect the identifiably distinct aqueous pools. A simple three compartment model discussed by NCRP (1979a) distinguishes atmospheric water, circulating ocean surface water, and land surface water. Under equilibrium assumptions this model differs little from the one compartment model, except to give a slightly more accurate total water volume for dilution. The most reasonable model, in terms of accounting for distinct accumulation pools, is of the type proposed by Easterly and Jacobs (1975) with seven compartments, and that of Bergman et al. (1979) with eight compartments. For these more refined models, equilibrium assumptions are inappropriate; instead, transfer coefficients between compartments are estimated and the system is simulated until it reaches

a pseudo-equilibrium state. This enables the comparison of projections with different initial conditions and transfer coefficients. The transfer coefficients are usually estimated on the basis of hydrogen transport studies and gross meteorological data, but in some cases are simply chosen to balance the system. Their accuracy is thus uncertain and as the complexity of a model is increased, the combined effect of the additional parameter uncertainty could result in reduced predictive reliability.

Another obvious problem with global circulation models is that, within a physically distinct compartment such as the atmosphere, there is not uniformity even in the steady state due to the dynamics of the hydrologic cycle. However, the major hydrologic flow is in a west-east direction with diffusion from the poles toward the equator. The result is that actual tritium concentrations within a given pool at fixed latitudes are relatively constant, with primary differences appearing between latitude levels. (Actually, Atlantic Ocean surface concentrations are consistently somewhat greater than concentrations at the same latitude in the Pacific Ocean.) Bander et al. (1979) have introduced 10° latitudinal bands into a two-compartment tritium model, including surface water and atmospheric water. Their model requires 104 transfer coefficients, compared to the 19 parameters in the Easterly-Jacobs model (1975), and thus, although only a generalized two-compartment model, the Bander model has a higher factor of parameter uncertainty than the seven-compartment model.

### 5.2.2 Reasonable concentration bounds

In light of the increased parameter uncertainty associated with further refinements of global tritium models, it seems appropriate to reconsider basic tritium distribution data and to establish "reasonable" bounds for estimates of environmental tritium concentrations. Such bounds may then provide a basis for making usable global dose estimates. They may also be used to identify estimates from more refined global models that appear to be too large, indicating a need for careful uncertainty analysis of such models' parameter bases.

We will now establish upper and lower bounds for estimated global tritium dose based on empirical evidence of actual tritium distributions. Schell et al. (1973) considered troposphere tritium distribution in the northern hemisphere and found that it could be described by a function of the form

$$y(\theta) = \begin{cases} y_0 e^{\alpha\theta} & \text{if } 0^\circ \leq \theta \leq 70^\circ \\ y_0 e^{\alpha \cdot 70^\circ} & \text{if } 70^\circ \leq \theta < 90^\circ \end{cases} \quad (5.1)$$

(see Fig. 5.7), where  $\theta$  represents the latitude and  $y(\theta)$  the concentration of tritium in water. The surprising result was that the exponent coefficient  $\alpha$  remained relatively constant at  $\alpha = 0.0538$  when the total tritium inventory varied. The Atlantic Ocean surface tritium profile and precipitation data of Weiss et al. (1979) fit a value of  $\alpha = 0.044$ . We choose the higher  $\alpha$  value of 0.0538 for our calculations as this yields higher dose estimates, a prudent course of action. The total tritium  $M$  can be represented by an integral of the form

$$M = \int_{0^\circ}^{90^\circ} y(\theta) dV(\theta), \quad (5.2)$$

where  $y(\theta)$  is as above and  $dV(\theta)$  is the incremental rate of change of the circulating water volume as a function of latitude  $\theta$ . The value of  $y_0$ , as a function of total tritium, is given by

$$y_0 = \frac{M}{\int_{0^\circ}^{70^\circ} e^{\alpha\theta} dV(\theta) + \int_{70^\circ}^{90^\circ} e^{\alpha \cdot 70^\circ} dV(\theta)} \quad (5.3)$$

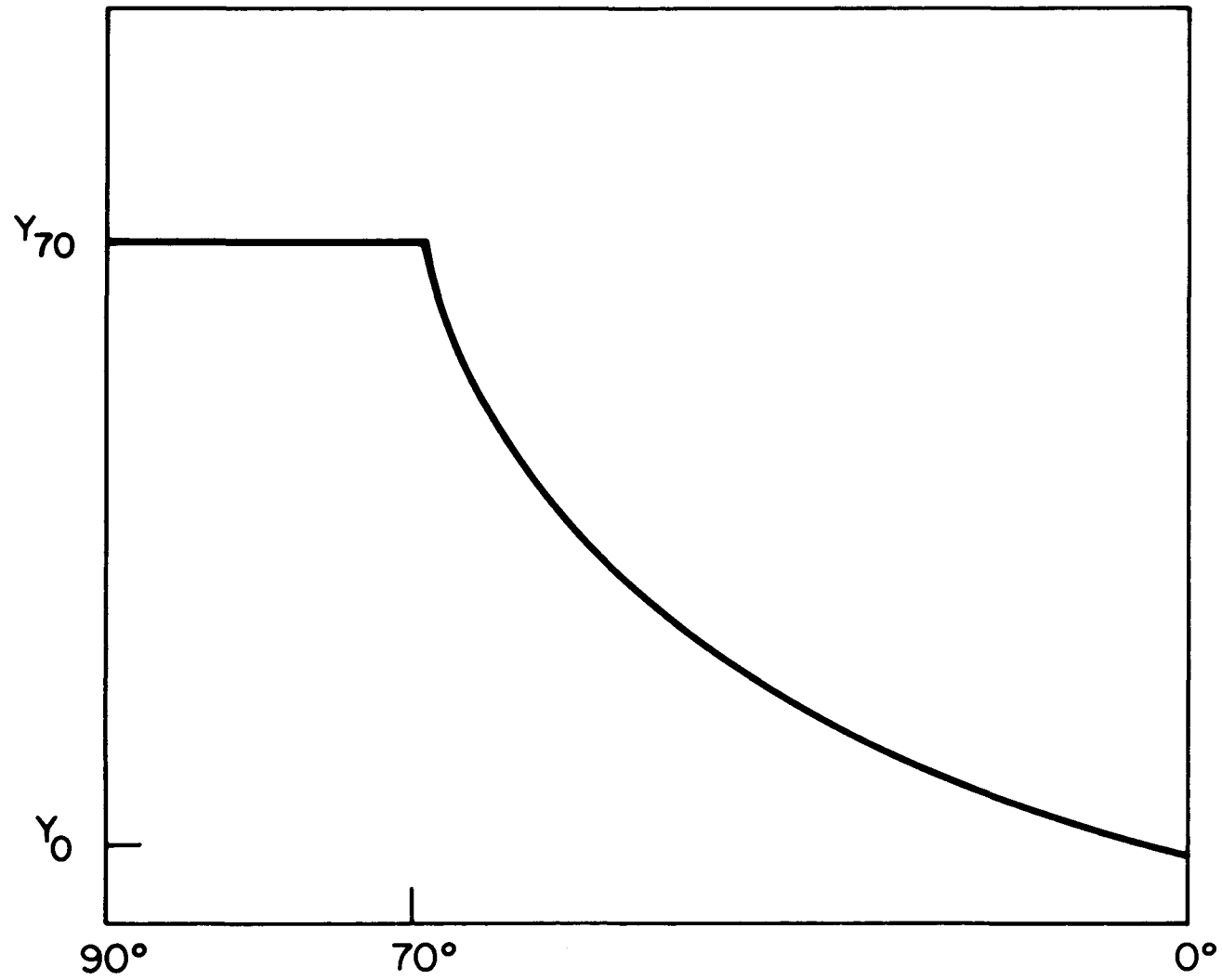


Fig. 5.7 Model of tritium concentration in the Northern Hemisphere troposphere and ocean surface as a function of latitude

Using 5° latitudinal values of ocean and land distribution (Baumgartner and Reichel, 1975), a 75-m ocean layer and a 0.5-m surface-water layer, the functional measure  $dV(\theta)$  and the above integrals may be approximated. With  $\alpha = 0.0538$ , the resulting value of  $y_0$  corresponding to a 1 Ci/year (37 GBq/year) release of tritium is  $1.66 \times 10^{-16}$  Ci/m<sup>3</sup> ( $6.14 \times 10^{-6}$  Bq/m<sup>3</sup>).

The tritium concentration measured over continental land masses increases in an easterly direction, with an average increase of about 3.0 times marine values (Schell et al., 1973). As the above value of  $y_0$  represents marine concentration, it is increased by a factor of 3.0 to calculate doses. An upper bound for the global population dose is obtained by assuming that the total population of the northern hemisphere is exposed to the maximum concentration level  $3.0 \times y(70) = 1.79 \times 10^{-16}$  Ci/m<sup>3</sup> ( $6.62 \times 10^{-6}$  Bq/m<sup>3</sup>). Substituting this value into the specific activity dosimetry model (USEPA, 1973) with the same population ( $3.0 \times 10^9$  persons) employed by Soldat and Baker (1979) yields a "reasonable" upper bound for the global population dose of  $5.5 \times 10^{-3}$  man-rem/year ( $5.5 \times 10^{-5}$  man-Sv/year). Table 5.6 indicates the dose estimates of the more recently published models. The only estimate that is greater than our upper bound is the  $2.2 \times 10^{-2}$  man-rem/year ( $2.2 \times 10^{-4}$  man-Sv/year) estimate of Soldat and Baker. Their estimate appears to be too high due to reliance on the higher tritium concentration estimates of Bander et al. (1979). The reason for the high estimates is probably a combination of the greater parameter uncertainty inherent in the Bander model and the omission of a deep-ocean tritium sink.

A lower bound for the global population dose is obtained by assuming the population is exposed to the lower limit of environmental concentrations,  $3.0 \times y_0$ . This gives a dose estimate of  $1.28 \times 10^{-4}$  man-rem/year ( $1.28 \times 10^{-6}$  man-Sv/year), which is below even the USEPA (1973) estimate. An "average" concentration is given by

$$\bar{y} = \frac{1}{70} \int_{0^\circ}^{70^\circ} y(\theta) d\theta = 1.86 \times 10^{-15} \text{ Ci/m}^3. \quad (5.4)$$

Table 5.6 Global dose estimates using several recent models

Model	Environmental concentration per 1 Ci/year atmospheric release ( $\text{pCi}/\text{m}^3 \text{ H}_2\text{O}$ ) <sup>a</sup>	Global dose per 1 Ci/year atmospheric release (man-rem) <sup>b</sup>
Soldat and Baker (1979)	$1.7 \times 10^{-1}$	$2.2 \times 10^{-2}$
Bergman et al. (1979)		$2.2 - 3.5 \times 10^{-3}$
Jacobs et al. (1979)	$2.4 \times 10^{-3}$	$1.4 \times 10^{-3}$ <sup>c</sup>
USEPA (1973)	$1.3 \times 10^{-3}$	$4 \times 10^{-4}$

<sup>a</sup>1 Ci = 37 GBq.

<sup>b</sup>1 rem = 0.01 Sv.

<sup>c</sup>From Table 5.9, this report.

This concentration results in a global population dose estimate of  $1.43 \times 10^{-3}$  man-rem/year ( $1.43 \times 10^{-5}$  man-Sv/year). This value presumes a uniformly distributed population.

The global dose estimates of Sect. 5.3 are slightly different in the sense that they are calculated for varying tritium release strategies. However, using data from Page 101, with constant population levels, results in an estimate of global population doses resulting from 1 Ci/year (37 GBq/year) release, of  $1.42 \times 10^{-3}$  man-rem/year ( $1.42 \times 10^{-5}$  man-Sv/year), a value within our bounds.

### 5.3 Global Component of Tritium Dose to the World Population

The collective dose commitment  $H_{\infty}$  to the world population due to a globally dispersed release of tritium is based on the equation

$$H_{\infty} = \int_{t_0}^{t_{\infty}} N(t) \cdot \dot{D}(t) \cdot dt \text{ man-rem} , \quad (5.5)$$

where  $N(t)$  denotes the number of individuals in the population at time  $t$  and  $\dot{D}(t)$  is the dose rate ( $\text{rem} \cdot \text{year}^{-1}$ ) to an average individual at time  $t$ . The time  $t_0$  is taken as the beginning of the release.

Estimation of the individual dose rate  $\dot{D}(t)$  is accomplished as follows:

$$\dot{D}(t) = (\text{DRF}) \cdot F_w \cdot C_m(t) \text{ rem} \cdot \text{year}^{-1} , \quad (5.6)$$

where (DRF) is a dose-equivalent rate factor for body water ( $\text{rem} \cdot \text{year}^{-1}$  per  $\text{g } ^3\text{H m}^{-3}$ ),  $F_w$  is the average fraction of body tissue that is water,  $\sim 0.75$  (ICRP, 1975), and  $C_m(t)$  is an estimate of the concentration of tritium in the body water of an average member of the population ( $\text{g } ^3\text{H m}^{-3}$ ) at time  $t$ . With 1 g tritium in each  $\text{m}^3$  of body water ( $1 \text{ m}^3 = 10^6 \text{ g H}_2\text{O}$ ), we have



$$\begin{aligned}
 (\text{DRF}) &= \frac{(51.2)(5.7 \times 10^{-3} \text{ MeV dis}^{-1})}{10^6 \text{ g body water}} \times 1 \text{ g } ^3\text{H} \times 9.606 \\
 &\times 10^9 \text{ } \mu\text{Ci}(\text{g } ^3\text{H})^{-1} \times 365 \text{ d year}^{-1} \\
 &= 1.02 \times 10^6 \text{ rem } (1.02 \times 10^4 \text{ Sv}) \text{ year}^{-1} \text{ per g } ^3\text{H m}^{-3}, \quad (5.7)
 \end{aligned}$$

where

$$\begin{aligned}
 51.2 &= \frac{(3.7 \times 10^4 \text{ dis s}^{-1} \mu\text{Ci}^{-1})(1.6 \times 10^{-6} \text{ erg MeV}^{-1})(1 \text{ rad})}{100 \text{ erg (g tissue)}^{-1}} \\
 &\times 86,400 \text{ s d}^{-1}, \text{ a unit conversion factor.} \quad (5.8)
 \end{aligned}$$

Eq. (5.7) is based on the well known internal dose rate formula (ICRP, 1959)

$$\text{dose-rate} = \frac{51.2\varepsilon}{m} \cdot q \text{ rad d}^{-1}, \quad (5.9)$$

where  $\varepsilon(\text{MeV dis}^{-1})$  is the absorbed energy,  $m$  is the mass (g) of the absorbing medium, and  $q$  is the activity ( $\mu\text{Ci}$ ) assumed to be distributed uniformly throughout the medium. If  $\varepsilon$  is assumed to contain a quality factor appropriate to the decay radiations, the formula of Eq. (5.9) may be considered to represent dose equivalent rate in units of  $\text{rem d}^{-1}$ . For the 5.7 keV  $\beta^-$  emissions of tritium, a quality factor of unity is assumed for the present section; contrary indications are discussed in Chap. 3 of this report.

The factor  $C_m(t)$  of Eq. (5.6) is estimated from a dynamic simulation of concentrations of released tritium in the water of several reservoirs of the global hydrologic cycle, in proportion as the water from these sources is taken in by man. With assumptions similar to those of NCRP Report No. 62 of the NCRP (1979a), we write for the partition of  $C_m$ ,

$$C_m = \frac{0.99}{3.0} C_{\text{air}} + \frac{1.99}{3.0} C_{\text{water}} + \frac{0.02}{3.0} C_{\text{ocean}} \quad (5.10)$$

where the variables on the right denote concentrations of tritium ( $\text{g m}^{-3}$ ) in the media indicated by the subscripts. The first term is assumed to contribute to the concentration in body water by inhalation ( $0.13 \text{ l d}^{-1}$ ), absorption through the skin ( $0.09 \text{ l d}^{-1}$ ), and to one-half of the concentration in the water taken in food ( $0.77 \text{ l d}^{-1}$ ). The second term expresses the assumed contribution of the land waters through the remaining half of water content in food and through drinking water ( $0.77$  and  $1.22 \text{ l d}^{-1}$ ). In the third term, a small contribution due to eating fish is taken into account. The total water intake for the reference individual is  $3.0 \text{ liter d}^{-1}$ .

Simulations of dynamic exchange of tritium among global reservoirs were carried out with our implementation of a seven-reservoir model defined in NCRP Report No. 62 (1979a), which is itself a variant of a model of Easterly and Jacobs (1975) (see Sect. 5.2). Exchange rates are assumed to be proportional to the tritium levels in the donor compartments, with the transfer coefficients being derived from estimated steady-state fluxes of the global hydrologic cycle. Table 5.7 characterizes the structure and parameterization of the model; the reader is referred to NCRP (1979a) and Easterly and Jacobs (1975) for further details and references.

The seven differential equations that express the rates of change of tritium levels in the world reservoirs are solved with exogeneous functions that represent inputs of tritium into several of the reservoirs as a result of the releases being simulated. In the case of releases from the nuclear power industry, the receiving compartments are atmosphere and water: for aquatic releases, approximately 80% is assumed to be discharged to surface streams and freshwater lakes, while the remaining 20% goes into the surface waters of the ocean. In simulations of transport of nuclear weapons-produced tritium, the total input is assumed to enter the system through the atmosphere. In all cases, the dynamic levels of tritium in the reservoirs that are available to man are diluted in the reservoir water volumes (Table 5.7) and the resulting concentrations inserted in Eq. (5.10). We have retained the NCRP (1979a) assumption that 80% of drinking water is from streams and

Table 5.7 Reservoir model of the global hydrological cycle  
applied to the transport of tritium

	Transfer coefficients <sup>a</sup> (year <sup>-1</sup> )							Water Volume (m <sup>3</sup> )
	A	OS	DO	SW	GW	FW	SL	
A		24.62		7.638		0.4615E-1	0.7692E-2	1.3E13
OS	0.129E-1 <sup>b</sup>		0.5926E-1					2.7E16
DO		0.1240E-2						1.29E18
SW	1.022				0.3806	0.4388		6.7E13
GW				0.2874E-2		0.1198E-3	0.4790E-4	8.35E15
FW	0.7937E-2	0.2381						1.26E14
SL	0.4808E-2							1.04E14

<sup>a</sup>Abbreviations: A = atmosphere, OS = ocean surface (75 m), DO = deep ocean, SW = surface soil water, GW = deep ground water, FW = surface streams and fresh water lakes, and SL = saline lakes and inland seas.

<sup>b</sup>Read as  $0.129 \times 10^{-1}$ .

freshwater lakes, with the remaining 20% being drawn from deep wells (deep groundwater). In our calculation,  $C_{\text{water}}$  of Eq. (5.10) is a composite of the tritium concentrations in these two compartments; in the NCRP calculation, however, the surface soil water compartment appears to be included as well.

The world population scenario,  $N(t)$ , [Eq. (5.5)] is based on the "medium variant" projection published by the United Nations (1974) through the year 2075. Subsequent to that date we have assumed the population to be stationary at the 2075 level. Table 5.8 shows the projected population values at 25-year intervals.

Table 5.9 displays the collective dose commitments estimated in the manner described above for releases of tritium from consumer products and the nuclear power industry 1975 to 2020 under scenarios A, I, and M (Sect. 5.2), together with the estimated collective dose commitment due to the estimated tritium production from nuclear weapons 1940 to 1975. Each of these total collective dose commitments is broken down to show components associated with the several environmental exposure media (atmosphere, deep groundwater, freshwater lakes and streams, and ocean surface). These components do not constitute an analysis of pathways relative to modes of release, but rather indicate the extent to which man's exposure to each of the respective environmental compartments contributes to dose for the combinations of release modes assumed for the power scenarios, weapons testing, and natural tritium.

The two columns of Table 5.9 that give collective dose commitments from the natural source of tritium deserve some comment. The first and larger total ( $1.05 \times 10^6$  man-rem or  $1.05 \times 10^4$  man-Sv) is based on integrating the product of the constant individual dose rate of  $7.3 \times 10^{-4}$  millirem $\cdot$ year $^{-1}$  ( $7.3 \times 10^{-6}$  mSv $\cdot$ year $^{-1}$ ) (caused by the naturally produced tritium that is in steady-state in the environment) and the population,  $N(t)$ , over a period comparable to the nuclear power scenarios: 1975 to 2020 (release period) plus 100 years (approximately 8 half-lives of tritium). The second total is the result of treating the estimated natural rate of production ( $4.0$  MCi $\cdot$ year $^{-1}$  or  $0.15$  EBq $\cdot$ year $^{-1}$ ) in the same manner as a nuclear industry scenario and integrating the consequent collective dose rate from 1975 to 2020; then with this source

Table 5.8 World population history and scenario<sup>a</sup>

Year	1925	1950	1975	2000	2025	2050	2075 <sup>b</sup>
World total (billions)	1.96	2.51	3.99	6.41	9.07	11.16	12.21

<sup>a</sup>United Nations (1974).

<sup>b</sup>Our scenario assumes a stationary population of 12.21 billion after 2075.

Table 5.9 Global component of collective dose commitment (man-rem<sup>a</sup>) to the world population from man-made and natural sources of tritium

Exposure medium	Nuclear power scenarios plus consumer products			Nuclear weapons	Natural source	
	A	I	M		Steady-state	Produced 1975 to 2020
Atmosphere	5.01E4 <sup>b</sup>	6.28E4	7.19E5	4.83E5	4.57E5	9.14E4
Deep ground-water <sup>c</sup>	9.86E2	1.26E3	1.43E4	1.11E4	8.22E4	1.90E3
Freshwater lakes and streams <sup>d</sup>	1.60E6	1.61E6	2.31E6	5.24E5	5.2E5	9.85E4
Ocean surface	3.57E2	3.86E2	1.85E3	1.13E3	9.47E2	2.08E2
Total	1.65E6	1.68E6	3.05E6	1.02E6	1.05E6	1.92E5

<sup>a</sup> 1 man-rem = 0.01 man-Sv.

<sup>b</sup> Read as 5.01 × 10<sup>4</sup>.

<sup>c</sup> Contributes 20% of drinking water.

<sup>d</sup> Contributes 80% of drinking water.

switched off, the integration is continued to infinity. Comparing collective dose commitments from the power scenarios to the latter number is analogous to comparing release rates to the natural production rate.

The estimate of collective dose commitment due to nuclear weapons testing is less than that of any power scenario, even though the cumulative weapons source term is greater than those of the power scenarios (e.g., 1890 MCi vs 1464 MCi for scenario M) (70 EBq vs 54 EBq). Two factors are involved in this relation: first, the population is greatest when the release rate is maximum in each of the power scenarios, and that population exceeds the levels of the 1950s and 1960s by a factor greater than 2; and second, the power scenarios involve significant releases to the freshwater aquatic environment (Sect. 5.1.2.3) which contribute substantially to dose through food and drinking water, whereas the weapons releases are to the atmosphere (undersea bursts have been ignored) and contaminate the freshwater environment only indirectly. In particular, the dose commitment to a stationary population from a unit release to the compartment representing freshwater lakes and streams is about four times that for a unit release to the atmosphere.

Such comparisons of collective dose commitments are premature, however, until satisfactory estimates of first-pass and regional components can be estimated. There are some indications (Kelly et al., 1975) that these components could account for a significant fraction of the collective dose commitment and further analyses of this problem are currently being conducted.

## 6. CONCLUSIONS

This study of key parameters related to the assessment of tritium released to the environment leads to several conclusions. It is evident that the production and use of tritium and its release to the biosphere will continue and that tritium is an important contributor to dose from nuclear fuel cycle facilities. Although methodologies exist for evaluating man-made tritium entering the environment, these methodologies vary considerably in complexity and generally apply to chronic exposure conditions only. A new model has been proposed, based primarily upon NCRP methodology, that maintains both simplicity and the ability to incorporate dose from tritium in food products grown at several locations.

Our analysis of recent experimental data leads to the conclusion that a reevaluation of the quality factor for tritium is needed and that a value of 1.7 for  $Q$  is more representative of data published since 1968. Estimated dose from tritium is linearly dependent upon quality factor; given the current "conservative" approach to radiation protection, we recommend returning to a  $Q$  value of 1.7 for tritium betas.

General agreement appears to exist among researchers that organically bound tritium in the body must be incorporated into the calculation of dose. Neglecting this fraction can result in underestimation of dose by approximately 20%.

Absolute humidity and drinking water dilution coefficients are important factors in the calculation of dose from environmental releases of tritium. We present data allowing choice of a regional estimate of absolute humidity, rather than the default value of  $8 \text{ g H}_2\text{O/m}^3$  recommended by the NRC when site-specific data are not available. Dilution coefficients for drinking water have also been recommended for a number of cases, although it is suggested that site-specific water supply data be used when available.

Several models exist which calculate the buildup of tritium in the atmosphere and dose to the global population. Our estimates for the release of man-made tritium to the environment and prediction of collective dose commitment to man suggest that the dose from nuclear weapons



testing will be less than that from proposed nuclear power scenarios even though the cumulative weapons source term is greater than that for any of our energy scenarios.

In summary, tritium continues to be a radionuclide of interest because of the large quantities released to the environment from man-made sources and because of tritium's potentially large contribution to dose around nuclear facilities. This study has reviewed new theoretical and experimental data that affect the assessment of environmental releases of tritium and has analyzed the significance of this recent information in terms of the dose to man.

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