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Paper

OVERVIEW OF TRITIUM: CHARACTERISTICS, SOURCES, AND PROBLEMS

Shigefumi Okada* and Noriyuki Momoshima†

Abstract—**Tritium has certain characteristics that present unique challenges for dosimetry and health-risk assessment. For example, in the gas form, tritium can diffuse through almost any container, including those made of steel, aluminum, and plastics. In the oxide form, tritium can generally not be detected by commonly used survey instruments. In the environment, tritium can be taken up by all hydrogen-containing molecules, distributing widely on a global scale. Tritium can be incorporated into humans through respiration, ingestion, and diffusion through skin. Its harmful effects are observed only when it is incorporated into the body. Several sources contribute to the inventory of tritium in our environment. These are 1) cosmic ray interaction with atmospheric molecules; 2) nuclear reactions in the earth's crust; 3) nuclear testing in the atmosphere during the 1950s and 1960s; 4) continuous release of tritium from nuclear power plants and tritium production facilities under normal operation; 5) incidental releases from these facilities; and 6) consumer products. An important future source will be nuclear fusion facilities expected to be developed for the purpose of electricity generation. The principal health physics problems associated with tritium are 1) the determination of the parameters for risk estimation with further reduction of their uncertainties (e.g., relative biological effectiveness and dose-rate dependency); 2) risk estimation from complex exposures to tritium in gas form, tritium in oxide form, tritium surface contamination, and other tritium-contaminated forms, with or without other ionizing radiations and/or nonionizing radiations; 3) the dose contributions of elemental tritium in the lung and from its oxidized tritium in the gastrointestinal tract; 4) prevention of tritium (in oxide form) intake and enhancement of tritium (oxide form) excretion from the human body; 5) precise health effects information for low-level tritium exposure; and 6) public acceptance of tritium leakage and waste disposal from reactors and fuel reprocessing plants.**
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Key words: tritium; health effects; dosimetry; environmental impact

INTRODUCTION

TRITIUM is a radioactive isotope of hydrogen with an atomic number of 1 and atomic weight of 3. In this paper, tritium is expressed as either ^3H or T. It decays with emission of an electron [mean energy of 5.685 keV (NCRP 1985)] to form ^3He:

$$^3H \rightarrow {}^3He + e^- . \quad (1)$$

Its half-life is 12.35 y (NCRP 1985). This means that tritium cannot be stored in pure form for extended time periods because 5.6% of the tritium decays annually (Table 1).

CHARACTERISTICS OF TRITIUM

Physical and chemical characteristics of tritium are almost the same as hydrogen: Tritium gas burns in

Table 1. Characteristics of tritium.

Properties	Value	Reference
Half-life	12.35 y	NCRP 1985
Decay constant	5.6×10^{-2} y^{-1}	
	1.780×10^{-9} s^{-1}	
Nuclear recoil (^{3}He)		
Recoil energy	0–3 eV	Feinendegen 1967
Excitation energy	~11 eV	Feinendegen 1967
Characteristics of β^-		
Average energy	5.685 keV	NCRP 1985
Average track length	0.56 μm (water)	Carsten 1979
Maximum energy	18.600 keV	NCRP 1985
Maximum track length	6.0 μm (water)	Carsten 1979
Maximum track length	5 mm (air)	Silini et al. 1973
Lineal energy (peak)	~3 keV μm^{-1}	Ellet and
(range)	0.03–8 keV μm^{-1}	Braby 1972
Bremstrahlung x rays	1.12×10^{-4} keV	Browne and Firestone 1986
Effective half-life (human)		
First component	9 d	NCRP 1979
Second component	30 d	NCRP 1979
Third component	450 d	NCRP 1979
Tritiated water		
Annual limit of intake	3×10^9 Bq	ICRP 1978
	1×10^9 Bq	ICRP 1991c
Derived air concentration	8×10^5 Bq m^{-3}	ICRP 1978
Elementary form of tritium		
Derived air concentration	2×10^{10} Bq m^{-3}	ICRP 1978
Surface dose density limit	40 Bq cm^{-2}	JIA 1989
Radiation weighting factor (Wr)	1	ICRP 1991b
Quality factor (Q)	2	ICRU 1986

* Shibayama 1–25–8, Funabashi, Chiba 274, Japan; †Department of Chemistry, Faculty of Science, Kyushu University, Hakozaki, Higashi-ku, Fukuoka 812, Japan.
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oxygen to form tritiated water. It exchanges with hydrogen atoms of organic compounds. Tritium gas is known to diffuse through container walls (especially plastic walls). Because tritium atoms are three times heavier than hydrogen (^1H), the specific gravity of T_2O is heavier than that of H_2O, and its boiling point is higher than that of H_2O. The diffusion rates and chemical reaction rates of tritium compounds are often slower than those of hydrogen compounds ("isotope effect").

The maximum energy of electrons emitted from tritium decay is 18.6 keV (NCRP 1985), which gives a maximum track length of 6 μm in water (nearly equivalent in human tissue) (Table 1). Human skin is composed of the epidermis, 20–100 μm thick, and the dermis, 1–3 mm thick (ICRP 1991a). The target cells for skin cancers and skin damage of other types are present at the basal layer of the epidermis and in the dermis. Thus, electrons emitted from tritium outside of the body could never reach these targets. In other words, electrons from tritium can inflict damage on humans only when tritium is present inside the body. This also implies that most commonly used monitors such as film badges, thermoluminescence dosimeters or pocket ionization chambers are ineffective for detection of tritium in the body and that bioassay of urine, blood, or water vapor from expired air by liquid scintillation counting is the means to monitor tritium in the body.

The most abundant chemical form of tritium is tritiated water. Incorporation of tritiated water into the human body can occur by three pathways: 1) tritiated water vapor in air can be taken into the body via the lung through respiration, 2) tritium in fluids and foods can be ingested orally and absorbed through the gastrointestinal tract, and 3) tritiated water or its vapor can be absorbed through the skin. Tritium stays in the body in tritiated water and tritium-labeled organic molecules. Tritium is eliminated mostly from the body with a biological half-life of 9.7 d and partly with half-lives of 30 d and 450 d (NCRP 1979). Decay of tritium in a molecule would result in emission of a beta particle and in transmutation to a chemically inert helium nucleus. Since the energy (0–11 eV) associated with transmutation is far less than the 5,685 eV of the emitted electron, damage inflicted by tritium decay can be said to be attributable mostly to the electron emission (Table 1).

For the purpose of radiation protection, ICRP (1978) recommended the use of an annual limit of intake (ALI) and a derived air concentration (DAC), replacing the earlier term used—maximum permissible concentration (MPC) (ICRP 1959). Recently, ICRP (1991b) recommended the reduction of the committed effective dose limit for workers from 0.05 Sv to the average annual limit of 0.02 Sv. With this consideration, ICRP (1991c) now recommends an ALI for occupational exposure to tritiated water to be 2/5 of the ALI in the ICRP (1978) (Table 1).

Table 1 summarizes the characteristics of tritium and Table 2 provides useful tritium units and conversion factors.

Table 2. Table of tritium units.

Unit	Equivalent units[a]
1 g of T_2	3.59×10^{14} Bq, 359 TBq 9.7×10^3 Ci
1 TU[b] or 1 TR[c]	1 T atom $(10^{18}$ hydrogen atoms$)^{-1}$ 0.118 Bq HTO L^{-1} of water 3.2 pCi HTO L^{-1} of water
1 mCi HTO g^{-1} of water	37 GBq g^{-1} of water, 37 TBq kg^{-1} of water 3.4×10^{11} TU 0.12144 Gy h^{-1} 0.2023 cGy m^{-1}, 0.002023 Gy m^{-1} 12.14 rad h^{-1}, 0.2023 rad m^{-1}
1 PBq HTO kg^{-1} of water	1 TBq g^{-1} of water 9.189×10^{12} TU 3.281 Gy h^{-1} 5.468 cGy m^{-1}, 0.05468 Gy m^{-1}

[a] 1 GBq = 10^9 Bq
1 TBq = 10^{12} Bq
1 PBq = 10^{15} Bq
1 Gy = 1 J kg^{-1} = 100 rad = 100 cGy
1 cGy = 1 rad = 100 erg g^{-1}
1 Ci = 37 GBq
1 mCi = 10^{-3} Ci = 37 MCi
1 pCi = 10^{-12} Ci = 0.037 Bq.
[b] TU = tritium unit.
[c] TR = tritium ratio.

SOURCES OF TRITIUM

Tritium in nature

Discovery of "natural" tritium is relatively new. The search for the origin of ^3He in the atmosphere led to the discovery of cosmic-ray-produced tritium. Tritium as free hydrogen in dry air was detected by Harteck and Faltings (1950), and tritium in surface water was found in heavy water, concentrated from lake water in Norway (Grosse et al. 1951).

It is interesting that tritium levels in vintage wines, bottled before the beginning of hydrogen bomb testing in 1954, decayed with a half-life of about 12 y in relation to the time elapsed (Kaufmann and Libby 1954). However, the continuous presence of tritium in the earth's environment (e.g., natural waters) means that tritium could be produced continuously in the environment, counterbalancing the loss of tritium by its physical decay.

Major tritium production in nature is from nuclear reactions of atmospheric atoms with cosmic rays in the upper atmosphere (Kaufman and Libby 1954), as follows:

$$\begin{aligned} {}^{14}N + n &\rightarrow {}^3H + {}^{12}C \\ {}^{16}O + n &\rightarrow {}^3H + {}^{14}N \\ {}^{14}N + p &\rightarrow {}^3H + \text{products} \\ {}^{16}O + p &\rightarrow {}^3H + \text{products.} \end{aligned} \quad (2)$$

A very minor fraction of natural tritium is produced in the earth's crust from the neutron capture reaction of ^6Li in rocks, where neutrons would be provided by spontaneous fission of uranium, and by the (α,n) reactions (Kaufmann and Libby 1954).

UNSCEAR (1977a) estimated the tritium inventory in natural occurrence to be about 1.0–1.3 EBq by assuming near-equilibrium. The global inventory of tritium in 1990, including manufactured sources, is illustrated in Fig. 1, where its quantity and annual rate are shown in units of EBq (10^{18} Bq) and EBq y^{-1}, respectively.

Injection of tritium by nuclear bomb tests into the earth's environment

Since the first bomb test by the U.S. in 1945, various radionuclides have been released into the atmosphere, one of them being manufactured tritium. A series of hydrogen-bomb tests beginning in 1954 released especially large quantities of tritium each year until the cessation of atmospheric bomb tests in 1963. The total amount of tritium injected into the earth's atmosphere by 1963 was manufactured 185–240 times the natural tritium (UNSCEAR 1977a). UNSCEAR (1982) estimated that the amount of tritium released into the environment by 1982 was 240 EBq (6,500 MCi) by fusion and 0.0056 EBq (0.15 MCi) by fission explosions. If tritium is assumed to decrease only by radioactive decay during subsequent 30 y, the tritium level from the nuclear tests would be about 40–52 times the natural tritium in 1990 (Fig. 1).

Bomb tests resulted in radioactive contamination of the earth's environment, but gave us opportunities to use tritium in oxide form (HTO), tritium in gas form (HT), and T-methane as radioactive tracers for the study of the kinetics of water, hydrogen, and methane in the environment on a global scale (Takashima 1991). For example, movements of ocean currents and their vertical mixing, retention time of groundwater or lake water, mixing of groundwater with rain water (NCRP 1979; Takashima 1991), and the exchange rate of atmospheric water vapor between northern and southern hemispheres (Mason 1977) have been investigated.

Release of tritium from nuclear facilities during normal operation

Nuclear facilities that release tritium into the environment are nuclear power stations, nuclear fuel reprocessing plants, and tritium production plants.

In 1987, 417 nuclear power stations in 26 countries generated 298 GW of electricity (UNSCEAR 1988). In light water reactors, tritium were produced by ternary fission of the nuclear fuel and from neutron activation of lithium and boron. In heavy water reactors, neutron activation of deuterium was the main source of tritium, exceeding that from ternary fission. The annual production of tritium would be calculated to be about 0.22 EBq (5.9 MCi) by the ternary fission and 0.37 EBq (10 MCi) from the deuterium activation (UNSCEAR 1988). Tritium released into the environment from these reactors was estimated to be 0.023 EBq (0.62 MCi). Fig. 2 shows the annual amounts of released tritium from four types of nuclear plants per unit power production (GW) (UNSCEAR 1988). A Sellafield fuel reprocessing plant released tritium in quantities similar to a heavy-water reactor (UNSCEAR 1988).

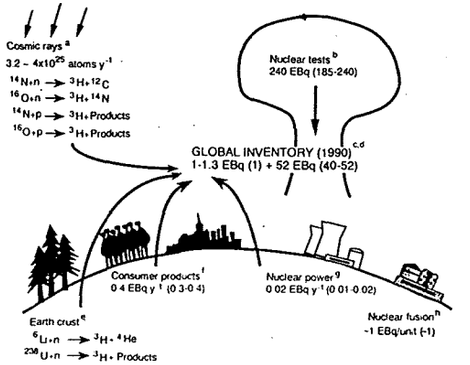

Fig. 1. Global inventory of tritium. Figures in parenthesis are relative values in relation to the tritium inventory in nature (1977a). (a) Annual production of tritium by cosmic rays, calculated from tritium production rates (0.2–0.25 atoms cm^{-2} s^{-1}) (Craig and Lal 1961; Teegarden 1967). (b) Total amount of tritium released from nuclear tests in the 1950s and 1960s (UNSCEAR 1982). (c) Tritium inventory in natural occurrence (UNSCEAR 1977a). (d) Amount (52 EBq) of tritium in 1990 calculated by considering radioactive decay of 240 EBq of the bomb tests (UNSCEAR 1982a) from 1963. (e) Contribution of tritium production from the earth's crust to the tritium inventory is very small (Kaufman and Libby 1954). (f) Annual amount of tritium used for consumer products in the U.S. in 1978 (Brown 1989). (g) Annual rate of tritium released into the environment from nuclear power stations under routine operation (UNSCEAR 1988), excluding those from tritium production facilities and their incidental release. (h) Tritium inventory of a 1.2-GW fusion facility of the future (IAEA 1990).

Chemical forms of tritium released from nuclear establishments are mostly hydrogen gas, water, or hydrocarbons. The gaseous forms are released to the atmosphere from one or more stacks and the liquid form through water pipes to rivers or the sea. Tritium activity of environmental samples (i.e., poplar leaves) around Chalk River Laboratory in Canada were found to decrease with distance (Brown 1979).

Around the Tokai nuclear facilities in Japan (Inoue et al. 1988 and 1989), tritium levels in rain water, groundwater, pine needles (leaves), and soil water were two to 30 times greater than the background levels and declined with distance. However, tritium concentrations of atmospheric hydrogen near the facilities were

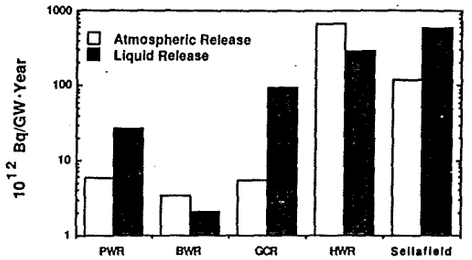

Fig. 2. Amount of tritium released per GW electricity production from various types of nuclear power stations and from the Sellafield nuclear fuel reprocessing plant, averaged from 1980 to 1985 (UNSCEAR 1988). PWR = pressurized water reactor. BWR = boiling water reactor. HWR = heavy water reactor. GCR = gas-cooled reactor. Sellafield : Sellafield nuclear fuel recycling plant.

similar to that of nonnuclear areas (Katagiri et al. 1989). Since liquid wastes from the Tokai facilities were released into the "Kuroshio" current of the Pacific Ocean, the most likely source of environmental tritium around the facilities is tritiated water vapor released from the stacks, which was trapped in rain and accumulated in the ground. The exposure dose to humans, estimated from the maximum tritium levels of pine needles, soil water, and groundwater, is about 0.0005 mSv y^{-1}. This was 0.05% of the annual limit for the public (ICRP 1991b).

Environmental tritium levels at the nuclear waste disposal area in Oak Ridge National Laboratory in the U.S. were studied by Amano et al. (1987). A general trend of tritium movement in the environment was from the disposal area to soil water, to streams, to rivers. However, tritium levels in the area were found to vary greatly from place to place, dependent upon their geological structures and hydrological conditions. Tritium concentrations in free water of pine needles reflected tritium levels of surrounding atmosphere and soil water. Annual exposure dose was estimated to be 0.65 mSv; about 3.3% of the annual dose-equivalent limit (20 mSv) for occupational exposure (ICRP 1991b).

Incidental release of tritium from nuclear facilities

Incidental releases of tritium have been reported from Lawrence Livermore Laboratory (LLL) (Myers et al. 1971) and Savannah River Plant (Marter et al. 1974; Jacobsen et al. 1976; Garrett et al. 1981; Garrett et al. 1983; Evans et al. 1985; Kurzeja et al. 1988).

When an incidental release occurred, a facility would use one or more computer codes to simulate a tritium dispersion pattern, to confirm the pattern from environmental samplings, and to estimate exposure to the workers in the facility and to inhabitants outside the facility.

Examples of monitoring and exposure data from release incidents of relatively large scale are shown in Table 3. Chemical forms of released tritium were either gas (T_2 or DT or HT) or water (HTO) (including vapor) or a mixture of gas and water. The released amounts ranged from 0.28–18 PBq. The environmental monitoring was mostly carried out on HTO, a major contributing factor to human exposure. In the six cases of major incidental releases (Table 3), the maximal exposure to workers within the facility was 0.14–3.0 μSv, which was 0.0008–0.015% of the worker's committed effective dose limit of 20 mSv (ICRP 1991b).

In the Chernobyl Accident (26 April—6 May 1987), tritium was released together with other radionuclides (e.g., ^{131}I and ^{137}Cs). However, the amount of tritium was so small that only a few investigators were able to detect it. For example, Salonen (1987) found 76–350 Bq L^{-1} of HTO in the air from 29 April to 1 May 1986 at Helsinki, Finland. This was about 100 times the background level.

The conversion of HT into HTO can occur via various processes (Noguchi and Kato 1985): 1) oxidation in air; 2) exchange reactions with H_2O; 3) photochemical oxidation in air; 4) oxidation in soil; and 5) oxidation in plants. Table 4 shows that the relative rates involving biological systems are far greater than those of nonbiological systems.

Since HTO is 25,000 times more radiotoxic to humans than HT (ICRP 1978), the following questions need to be answered regarding an incidental HT release: 1) How much and in what way would released HT be converted to HTO, and 2) Which would contribute more to human exposure, HT or HTO? The first field experiment to settle these questions was carried out in France by releasing tritium gas in the amount of 2.6×10^{14} Bq (Canadian Fusion Fuels Technology Project 1988a); a second experiment was conducted in Canada where 3.54×10^{12} Bq of HT was released (CFFTP 1988b). It was found that little conversion of HT to HTO occurred in air and plants. Major conversion occurred in the surface layer of soil. Then, the converted HTO was gradually released as water vapor into the air from the soil. Also, the soil HTO was taken up by plants and released into the air. Consequently, the major contributing factor to human exposure was HTO (Fig. 3) (CFFTP 1988a,b).

Oxidation of HT in the soil layer was attributed to microorganisms because autoclaving of soils abolished HT oxidation. Ichimasa et al. (1988) identified them as aerobic bacteria of the family *Streptomyces*. Their two major strains were *Streptomyces exfoliatus* and *Streptomyces rochei*. It is noted that some lichens and mosses on the surface of tree trunks or on the ground showed oxidation capacities of 100–1,000 times higher than those of pine needles. Ichimasa et al. (1989a) suggested 1) that lichens and mosses of high oxidation capacities could be used as environmental monitoring plants to

Table 3. Maximum tritium activities of environmental samplings and maximum human exposure inside a plant in tritium release incidences.[a]

Place	Date (y,mo,d)	Cause of leakage	Released tritium Amount	HTO (%)	Vapor (Bq m^{-3})	Milk (Bq mL^{-1})	Grass (Bq mL^{-1})	Human urine (Bq mL^{-1})	Human exposure[b] (μSv)
SRP[c]	74–05–02	Process valve	18 PBq	<1	7.0	0.85	170	<37	0.18
SRP	75–12–31	Vacuum meter	6.7 PBq	0.6	2.2	0.33	3.4	<37	0.14
SRP	81–03–27	Pipe joint	1.2 PBq	>99	350	0.41	10	0.78	3.0
SRP	83–07–16	Pipe joint	2.1 PBq	1	0.74	0.15	4.1	0.13	0.40
SRP	84–03–23	—	0.28 PBq	70	—	2.6	51	0.1	—
LLL[d]	70–08–06	—	11 PBq	<1	52	0.30	37	<190	0.25 0.7[e]

[a] The table by Sakuma (1988) is modified by the authors.
[b] Maximum human exposure inside a plant.
[c] SRP = Savannah River Plant.
[d] LLL = Lawrence Livermore National Laboratory.
[e] Maximum exposure of children outside the plant, estimated by assuming that they drank milk from cows that were fed with maximally contaminated grass (37 Bq mL^{-1}).

detect the path of an elemental tritium "puff" (i.e., a small cloud formed from rapidly released tritium gas) (Fig. 3), and 2) that the ground in a fusion facility could be covered by lawns of low oxidation capacity (Table 4).

Consumer products

The amount of tritium used for self-luminous light sources in the U.S. was about 400 PBq in 1978, equivalent to tritium produced by light-water power reactors of the country (Brown 1989) (Fig. 1).

The tritium content of dial paint per timepiece was up to 740 kBq (NCRP 1977). The minimum thickness of a watch case was sufficient to absorb beta particles from tritium. However, tritiated water or tritiated organic compounds evolved slowly from the paints and could be absorbed by humans through inhalation or skin absorption (UNSCEAR 1977b). Exposure to the wearer of such a watch was estimated to be an average of 6 μSv y^{-1} (NCRP 1977). (It should be noted that all the tritium in consumer products would eventually be released to the environment.)

In the 1960s, seven paint makers, some of whom handled 37–100 TBq of tritium annually, were found to show tritium activities in their urine. Two heavily contaminated persons died of panmyelophthisis (aplastic anemia) (Seelentag 1973). Because of their previous records of handling other radioisotopes (radium and strontium), it was not possible to attribute their cause of death solely to tritium. Hirashima (1988), when considering a decrease of hematopoietic stem cells found in Japanese thorium-oxide-treated patients, suggested that continuous irradiation of bone marrow by incorporated radioisotopes may damage their hematopoietic stem cells and eventually cause the development of aplastic anemia. In Japan, tritium is used at a rate of about 4 TBq y^{-1}, but no longer for luminescent paints.

In laboratories, tritium-labeled molecules are used as tracers in life science research and other areas.

Nuclear fusion reactors

Radiation protection problems for fusion reactors were first described in the Tokamak Fusion Test Reactor (TFTR) at Princeton (Stencel et al. 1989), where a *D-D* fusion reaction was produced

$$D + D \rightarrow \begin{cases} p + T \\ n + {}^3He. \end{cases} \quad (3)$$

In 1987, *D-D* fusion reactions produced 3×10^{18} neutrons and were accompanied by the production of about 5.4 GBq (145 mCi) of tritium. When the reaction vessel was opened, tritium activity in air was measured to be 1 MBq m^{-3}, which was already over the derived air concentration (0.8 MBq m^{-3}) of tritiated water (ICRP 1978). A differential tritium sampler gave its composition to be a mixture of HTO and HT at its ratio of 57:1. When the vessel was temporarily closed and reopened, the tritium level increased through release of tritium absorbed on graphite plates in the vessel. Tritiated powders, formed from graphite, were one of the major sources of radioactive contamination.

Tritiated water was also found in vacuum pump oil, originating from the cryopanels of the neutral beam injector. In addition to HT and HTO, tritiated methane could have been formed, but the amount has not yet been estimated.

Environmental tritium monitoring of surface waters, shallow groundwaters, plants, and soil around the TFTR were shown to be about 3 Bq L^{-1}, not different from the background levels of tritium. However, if a *D-D* reaction in larger scale were conducted, environmental tritium could become a radiation protection problem.

The first *D-T* fusion reaction with 0.2 g (70 TBq or 2,000 Ci) of *T* was carried out by the Joint European Torus machine on November 1991 at Oxfordshire

Table 4. Reaction rates of HTO formation from elemental tritium.[a]

Type of reaction	Conditions	Oxidation rate[b]	Reference
Oxidation in dry air	In the presence of:		
	No metal	0.03% d^{-1}	Eakins and Hutchinson (1973)
	Brass	0.03% d^{-1}	
	Soft steel	0.2% d^{-1}	
	Aluminum	0.2% d^{-1}	
	Platinum	1.1% d^{-1}	
Exchange reaction with H_2O in air[c]	In the presence of:		
	No metal	0.1% d^{-1}	Eakins and Hutchinson (1973)
	Brass	3.9% d^{-1}	
	Soft steel	2.7% d^{-1}	
	Aluminum	0.3% d^{-1}	
	Platinum	28% d^{-1}	
Photochemical reaction (OH reaction)		0.06% d^{-1}	Hoguchi and Kato (1985)
Oxidation in soil	Water content 140%	0.2–1.1% min^{-1}	McFarlane et al. (1979)
Forest soil			
0–5 cm	Water content: 32.3%	20% min^{-1}	Ichimasa et al. (1988)
5–10 cm	27.6%	4.5% min^{-1}	
10–15 cm	28.0%	2.4% min^{-1}	
Oxidation in plants			Ichimasa et al. (1989a)
Pine needles		0.001% min^{-1}	
Pine branches		0.008% min^{-1}	
Pine bark		0.008% min^{-1}	
Pine roots		50.7% min^{-1}	
Soil surrounding roots		8.5% min^{-1}	
Zelkova tree leaves		0.001% min^{-1}	
Zelkova roots		5.7% min^{-1}	
Lawn grass		0.1% min^{-1}	

[a] This is a modified form of the table by Noguchi and Kato (1985).
[b] Oxidation rate (% d^{-1} g^{-1}) = (amount of oxidized HT)/(initial amount of HT) × 100% × (reaction time in d)$^{-1}$ × (sample weight in g)$^{-1}$. The rates are calculated under specified experimental conditions from the data given in references column.
[c] Exchange reaction rates are calculated from the difference of oxidation rates in wet and dry air (Eakins and Hutchinson 1973).

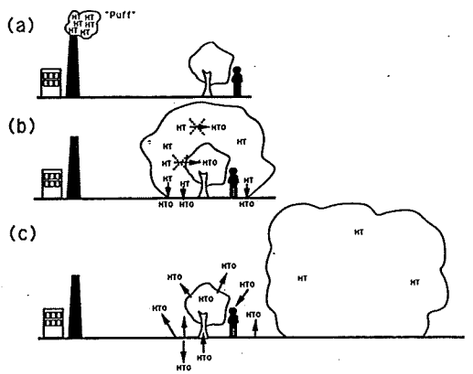

Fig. 3. A simplified scheme of incidental release of a puff of tritium gas from a tritium-handling facility (a nuclear fusion facility or a tritium production plant). The figure is modified from Fig. 1 of CFFTP (1988b). Crosses indicate that conversion rates would be negligibly small.

$$D + T \rightarrow \mathrm{He} + n. \quad (4)$$

It produced a megawatt of energy for 2 s. A full-power *D-T* reaction is now scheduled in 1996, after which the radioactively contaminated machine is planned to be decommissioned (Pease 1991).

Currently, Japan, U.S., Europe, and Russia are in the engineering design stage of the International Thermonuclear Experimental Reactor of 1,000 MW power. Fig. 4 shows a scheme of a *D-T* fusion reactor (IAEA 1990). The *D-T* fusion reaction will produce 3.5-MeV alpha particles (helium nucleus) and 14-MeV neutrons. When the alpha particles slow down within the plasma, they heat it to the required plasma temperature for continuation of the fusion reaction. A 14-MeV neutron escapes the plasma region; carries the remaining 80% of the energy, and releases it into the blanket. Coolant circulates inside the blanket and transfers the heat out of the reactor to produce steam for generation of electricity. Neutron bombardment breeds tritium in the lithium-containing blanket, the tritium will then be used as the fuel for the continuing fusion reaction.

How much tritium may be used in a fusion power plant? The IAEA (1990) stated that a Tokamak power plant of 1.2 GW(e) would have several kilograms (a few EBq) of the tritium inventory on the site and that

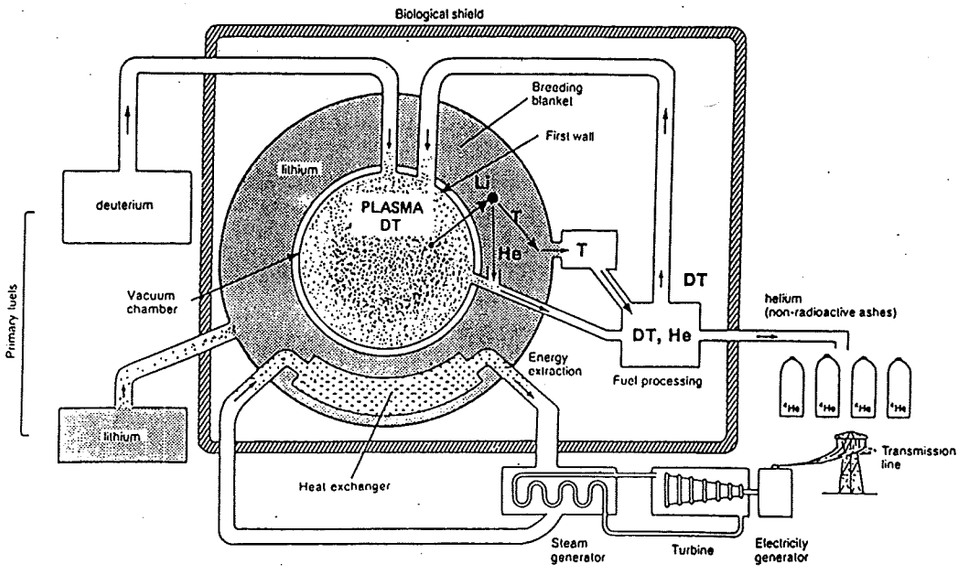

Fig. 4. A *D-T* fusion reactor (IAEA 1990). This is a copy reproduced with IAEA's permission from Fig. 1 of the IAEA report (IAEA 1990).

partitions of the facility would make it vulnerable to release only a fraction of the tritium inventory under accident conditions.

Tritium in humans and their environment

Since recent tritium data on humans and their surrounding environment have been accumulated in Japan, a relationship of environmental tritium to human tritium will be described mostly with these data.

The atmospheric nuclear tests of 1950–1960 injected tritium at a level of about 185–240 times the amount of natural tritium into the atmosphere. The cessation of the tests in 1963 resulted in a gradual decrease of atmospheric tritium levels. The latest estimates made in 1984–1990 at Fukuoka City showed that tritium levels of HT, HTO, and T-methane per cubic meter of air are of similar magnitude, but specific activities (in tritium units) of T-methane and HT are 1,000–100,000 times higher than that of HTO vapor (Table 5) (Takashima 1991).

The tritium activity of HTO in precipitation is slightly lower than that of HTO vapor in air, being diluted by water vapor from the surface waters of low specific activities (Table 5 and 6). Their HTO activities (0.1–3.4 Bq L^{-1}) are getting close to those values present before the bomb tests (e.g., 0.14–7.9 Bq L^{-1} for Chicago rain water, 0.16–0.21 Bq L^{-1} for Lake Michigan, and

Table 5. Tritium in air at Fukuoka City of Japan from 1984–1990 (Takashima 1991).

Chemical form	Tritium concentration (mBq m^{-3} air)	Specific activity (TU)	Tritium activity (Bq L^{-1})
HTO vapor	19–23	15–17	1.8–2.0
HT	33–47	$0.69–1.01 \times 10^6$	—
Methane-T	13–15	$3.6–4.6 \times 10^4$	—

Table 6. HTO in waters of various types on the Japanese environment from 1982–1989.

Samples	Location	Tritium activity (Bq L^{-1})
Precipitation[a]	Fukuoka City	0.1–3.4 (0.9)[b]
	All Japan	0.5–1.7
Groundwater[a]	Aichi Prefecture	0.1–10.7
	Toyama Prefecture	0–4.8
River water[a]	All Japan	—(1.9)[b]
Coastal sea[c]	All Japan	0.4–1.1 (0.7)[b]
Ocean[c]	Pacific Ocean	0–0.6
	Japan Sea	0–0.6

[a] Momoshima et al. (1990).
[b] Figures in parentheses are average values.
[c] Takashima (1991).

0.30–0.77 Bq L^{-1} for the Mississippi River) (Kaufmann and Libby 1954). The tritium levels of tap water (drinking water) in Japan, originating from rivers, lakes, or groundwater, now range from 0.3–3.6 Bq L^{-1} (Table 7).

When comparing tritium levels of rice and tap water in Akita City (Table 8) with those of all Japan (Table 7), the Akita levels are within the range of all Japan. Thus, levels observed at Akita could be considered to be similar to those of all Japan.

In Tables 7 and 8, tritium levels are expressed in tissue free water tritium (TFWT) and organically bound tritium (OBT): TFWT represents tritium activity in the "free water portion" of food (or tissue) in which free water is collected by drying a tissue. OBT is estimated in water, which is collected by oxidizing (or burning) the dried tissue. Specific activity ratio (SAR) is a ratio of OBT:TFWT and is assumed to be an indicator for the ability of the biological system to concentrate tritium into organic fractions (e.g., Moghissi et al. 1987). In addition, SAR could sometimes reflect a direct pathway from food OBT to tissue OBT in the system (Etnier et al. 1984; Travis et al. 1984; Takeda et al. 1985), or it could reflect longer retention of OBT fractions and quicker turnover of TFWT fractions in the biological systems (Myers and Johnson 1987).

To examine the "concentration" hypothesis, Moghissi et al. (1987) fed rabbits with alfalfa and water with a SAR of 1 for three generations over 3 y. Rabbits were sacrificed at various intervals. After estimation of TFWT and OBT in six organs, the calculated SAR was found to be 1 in each of six organs. The authors concluded that there is no evidence of tritium concentration in OBT under the "static equilibrium condition."

As shown in Table 8, TFWT in food samples ranged from 0.5–2.5 Bq L^{-1} while their OBT varied from 1.0–2.1 Bq L^{-1}. It is mentioned that tritium in free water of foods was easily influenced by atmospheric HTO (Inoue et al. 1991). This may be one reason for wider variation in TFWT concentration in foods. When pooled portions (daily foods in Table 8) of breakfast, lunch, and supper for 5–6 persons were analyzed, the total TFWT and OBT were similar to TFWT and OBT of three organs in humans. Also, the SAR of 1.2 in the daily foods was the same as three organs of humans. This seems to indicate that tritium in the environment, foods, and humans are nearly in equilibrium in Japan.

The total daily tritium uptake of the Japanese is 4.35 Bq, of which foods contribute 1.71 Bq, water vapor in air contributes 0.40 Bq, and drinking water contributes 2.25 Bq. This shows that tritium intake is 39% from food, 52% from drinking water, and 9% from air (Hisamatsu et al. 1987, 1989b; Okada et al. 1990).

If a total tritium uptake from Japanese food materials is taken as 100%, contribution of cereals constitutes 26% (rice 16%); vegetables 18%; milk and dairy products 14%; fruits 11%; and fish and meats 9% (Hisamatsu et al. 1989b; Okada et al. 1990).

An annual dose equivalent from tritium exposure can be calculated from tritium concentrations in various organs of humans by assuming that tritium in soft tissues is the major contributing factor (ICRP 1978). In an ICRP Reference Man (70 kg of body weight), soft tissue is 63 kg, in which body fluid (water) is 42 kg and other organic matters are 21 kg (ICRP 1975).

In Table 8, TFWT in human organs is 1.6 Bq L^{-1} and it's OBT is 1.8 Bq L^{-1}. Assuming density of soft tissue to be 1 kg L^{-1}, a total tritium content in soft tissues will be 1.6 Bq L^{-1} × 42 kg + 1.8 Bq L^{-1} × 21 kg = 105 Bq.

If 105 Bq of tritium is uniformly distributed in 63 kg of soft tissues, its tritium concentration will be 105 Bq/63 kg = 1.67 Bq kg^{-1}.

The dose equivalent for 1 y from decay of tritium at this constant concentration in equilibrium will be 1.67 Bq kg^{-1} × (28.8 nSv y^{-1}) Bq^{-1} kg^{-1} = 0.0481 μSv y^{-1}. Japanese people, in the present environment, would annually receive 0.048 μSv from tritium decay, which is 0.0034% of their annual natural radiation dose (1.4 mSv) (Abe 1989).

Table 7. Tritium in pine needles, polished rice, and tap water in all Japan.

Samples	TFWT[a] (Bq L^{-1})	OBT[b] (Bq L^{-1})	SAR[c] (average)	References
Pine needles	0.8–3.1	1.4–3.6	0.9–2.1 (1.7)[d]	Takashima (1991)
Polished rice	0.0–4.7	0.2–4.3	0.1–1.6 (0.83)[d]	Inoue and Iwakura (1990)
Tap water	0.3–3.6	—	—	Yamada et al. (1986)

[a] TFWT = tissue free water tritium.
[b] OBT = organically bound tritium.
[c] SAR = specific activity ratio = OBT:TFWT.
[d] Figures in parentheses are average values.

TRITIUM PROBLEMS

Radiation quality and dose rate dependency

Because few data on tritium-induced health injury in humans are available, risk estimation has to be based on animal and cellular experiments with tritium. Such experiments show that acute exposure to tritiated water induced carcinogenesis, fetus malformation, mutagenesis, animal death, or killing of hematopoietic stem cells. The injuries were qualitatively similar to injuries caused by low-LET radiations (e.g., ^{137}Cs or ^{60}Co gamma rays, and x rays). The quantitative difference between tritium beta and a standard low-LET radiation is expressed by relative biological effectiveness (RBE), an inverse ratio of the doses of two kinds of radiation that produce the same biological effect. RBE of tritium beta ranged mostly from 1 to 3 (Sawada and Okada 1990; Straume 1991; Straume and Carsten 1993), and are influenced by various factors (Okada et al. 1986): 1) different RBEs were obtained in the same tritiated-water treated cells, depending upon types of biological endpoints; 2) RBEs for the same endpoint (e.g., killing) were dependent upon cell types; 3) RBEs were sometimes dependent upon dose or dose rate; 4) RBEs for the same endpoint were dependent upon types of tritium compounds (e.g., HTO vs. T-thymidine) (e.g., Ueno et al. 1989); and 5) RBEs are dependent on type of a standard radiation (e.g., x rays, ^{137}Cs gamma rays, and ^{60}Co gamma rays).

RBE values have been a basis for assigning a radiation weighting factor, Wr (ICRP 1991b), which is a correction factor to each of a different quality of radiation, necessary for conversion of an absorbed dose (Gy) to a dose equivalent (Sv) for the purpose of radiation protection. Wr was previously called a radiation quality factor (Q) (ICRP 1978). When the latest ALI of tritium is derived (ICRP 1991c), Wr of 1 (ICRP 1978, 1991b) has been used for the tritium beta. It is added that a joint task group of ICRP and ICRU recommended a Q of 2 for tritium beta from microdosimetric considerations (ICRU 1986) and that radiobiological experiments gave a RBE of 1–3 (e.g., Sawada and Okada 1990). The question of what would be the most appropriate Wr value of tritium beta rays for radiation protection still remains to be decided.

Most radiobiological experiments with tritium have been carried out under acute exposure conditions using tritiated water. One area of study needed now is chronic exposure to tritium by workers in tritium-handling facilities. One of a few studies on this subject was done by Carsten (Carsten et al. 1989), who maintained mice in drinking water at 10×, 33×, 100×, ... of the MPC (1.11 kBq mL^{-1}) of HTO. At 10× MPC, no significant biological effects were found. However, when the HTO concentration was ≥33×, marrow stem cells were slightly reduced. At the higher concentrations of HTO, chromosomal damage in liver and micronuclei formation in red blood cells increased. Carcinogenesis in these mice has not yet been studied. From a literature survey, Ujeno (1983) suggested that RBE would increase with lowering dose rate. Because there are few experimental data available, further studies are definitely needed for assessing health risks from tritium under continuous exposure.

Complex exposures

In tritium-handling facilities and fusion research facilities, tritium in large quantities creates contamination problems on maintenance, clean-up operations, or incidental release situations. Even one of the least

Table 8. Tritium in foods and humans at Akita City, Japan.

Samples	TFWT[a] (Bq L^{-1})	OBT[b] (Bq L^{-1})	Daily uptake of tritium[c] (Bq d^{-1})	SAR[d]	Reference
Uncooked foods[e]					
Rice	2.5	1.8	0.279	0.72	Hisamatsu et al. (1987, 1989b)
Green vegetables	1.4	1.7	0.078	1.19	Hisamatsu et al. (1987, 1989b)
Other vegetables	1.0	1.8	0.229	1.75	Hisamatsu et al. (1987, 1989b)
Fishes/clams	0.49	1.0	0.055	2.11	Hisamatsu et al. (1987, 1989b)
Meats	1.3	1.8	0.094	1.33	Hisamatsu et al. (1987, 1989b)
Eggs	1.7	2.1	0.067	1.20	Hisamatsu et al. (1987, 1989b)
Milk/dairy products	2.0	2.1	0.241	1.04	Hisamatsu et al. (1987, 1989b)
Tap water	1.9	—	2.25[f]	—	Hisamatsu et al. (1987, 1989b)
Daily foods[g]	1.7	1.9	4.2	1.19	Hisamatsu et al. (1987, 1989b)
Human					
Brain	1.6	1.7	—	1.1	Hisamatsu et al. (1989a)
Lung	1.6	1.9	—	1.3	Hisamatsu et al. (1989a)
Liver	1.6	1.8	—	1.1	Hisamatsu et al. (1989a)

[a] TFWT = tissue free water tritium.
[b] OBT = organically bound tritium.
[c] SAR = specific activity ratio = OBT:TFWT.
[d] The table of daily uptake (g) of each food sample from the National Nutrition Survey (Ministry of Health and Welfare 1987) is used to obtain daily tritium uptake (Bq d^{-1}).
[e] Uncooked but edible portion of food materials are used for tritium measurement.
[f] Okada et al. (1990).
[g] Daily foods (from breakfast, lunch, and dinner) consumed by 5–6 persons are collected and analyzed. These exclude beverage taken between meals.

are the mortality studies of the workers at the UK Atomic Energy Authority (Beral et al. 1985) and the UK Atomic Weapons Establishment (Beral et al. 1988). Both studies showed that an increase of prostatic cancer death might be related to internal exposure to tritium. However, the number of deaths are still so small that further extensive studies are needed to confirm these findings.

Tritium leakage, tritium waste disposal, and public acceptance

Tritium is easily spread, and enters into the environment and humans. Several incidences of tritium leakage from tritium facilities showed that tritium of several PBq imposed little health risk (Table 3). However, if tritium leakage of a much larger quantity would occur, health risk would become a problem (Kulcinski 1988).

The other problem is waste disposal (Morikawa et al. 1989). Some waste (e.g., tritiated water) of high activity may be subjected to concentration, and reutilization. That cannot be reutilized may be solidified for permanent storage. Other waste (e.g., tritium absorbents, tritium-contaminated powders and materials, and tritium-contaminated parts of machines and instruments) which occupies a large volume with low tritium activity would need to be stored under constant surveillance. Tritiated water could be diluted and released into the ocean. All types of disposal will become a problem perhaps not because of health risk but because of lack of public acceptance. We should be prepared to meet these issues of public perception of risks for the future use of fusion energy.

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EPIDEMIOLOGICAL STUDIES
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Guest Editorial

PREFACE: TRITIUM DOSIMETRY, HEALTH RISKS, AND ENVIRONMENTAL FATE

RESEARCH during the past decade or so has provided substantial new information pertinent to the dosimetry, health risks, and environmental fate of tritium. The last detailed reviews of the tritium data were made during the 1970s (Moghissi and Carter 1973; NCRP 1979a,b). More recent work has been presented at a variety of workshops, seminars, and meetings, including the Workshop on Tritium Radiobiology and Health Physics in 1981 in Chiba, Japan; the European Seminar on the Risks from Tritium Exposure in 1982 in Mol, Belgium; the Second Workshop on Tritium Radiobiology and Health Physics in 1984 in Chiba, Japan; the Third Japan-U.S. Workshop on Tritium Radiobiology and Health Physics in 1988 in Kyoto, Japan; and informal reviews prepared for the U.S. Department of Energy in support of the New Production Reactor Program. Also, considerable tritium research has been done during the past ten to fifteen years in the former Soviet Union, much of which has not appeared in non-Russian publications.

Because most of this new information has not been published in open peer-reviewed journals, we believed that it would be important to make this information available to the world-wide scientific community in *Health Physics* and provide the information in a format that would be particularly useful to health physicists, environmental scientists, risk assessment experts, and the regulatory community.

Invitations were extended to internationally recognized experts in the tritium field to submit papers on selected subject areas. The subject areas were overview of tritium sources and problems; tritium sampling and measurement; metabolism and dosimetry of tritium; microdosimetry of tritium; tritium radiobiology and RBEs; tritium risk assessment; tritium transport and cycling in the environment; organically bound tritium; and recent research in radiobiology of tritium and its compounds in the former Soviet Union.

"Overview of tritium: Characteristics, sources, and problems" by Okada and Momoshima provides a concise overview of the many characteristics of tritium relevant to health physics and environmental assessments. The paper also identifies and discusses the most significant sources contributing to the inventory of tritium in our environment and the principal health physics problems.

"Tritium sampling and measurement" by Wood, McElroy, Surette, and Brown describes the current methods used to sample and measure tritium. This paper identifies several recent improvements in the instrumentation technology for tritium measurement, primarily as a result of fusion-related research during the 1980s. The paper also identifies technology limitations where further research and development are required.

"Metabolism and dosimetry of tritium" by Hill and Johnson presents the physical, chemical, and metabolic characteristics of various forms of tritium as they pertain to performing dose and risk assessments.

"Microdosimetry of tritium" by Morstin, Kopec, Olko, Schmitz, and Feinendegen deals with the energy deposition of tritium beta rays on the μm and nm scale. The energy deposited in such small dimensions appears to be the most relevant for the induction of biological damage by ionizing radiation.

"Tritium radiobiology and relative biological effectiveness" by Straume and Carsten provides a review and analysis of the available data on tritium radiobiology and RBEs. Best-estimate RBE values are provided for use in tritium risk assessment.

"Tritium risk assessment" by Straume provides an approach to estimate tritium risk that incorporates the most recent information from epidemiology and radiobiology. Best-estimate risks are provided for low-level exposures to tritiated water and certain tritiated organic molecules. Uncertainties in risk values are estimated using a Monte Carlo sampling approach.

"Tritium transport and cycling in the environment" by Murphy presents data, approaches, and models for the transport of tritium in environmental media. A particularly useful example is presented describing tritiated water transport and cycling in terrestrial plants.

"Organically bound tritium" by Diabaté and Strack reviews the most important processes of organically-bound tritium (OBT) production and transport through food chains. Quantitative information is presented that should be useful for tritium dosimetry and modeling, e.g., biological half lives and transfer rates for OBT molecules.

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The total excess risk for all genetic diseases that are expected to result from chronic exposure to low-LET) radiation has been estimated. eg. UNSCEAR, 1986; BEIR 1990).

Paper

TRITIUM RISK ASSESSMENT

Tore Straume*

Abstract—**Estimates of the health risks in humans from low-level exposure to tritium are presented. The health risks considered are those for cancer, genetic effects, and developmental abnormalities from exposures *in utero*. Because direct risk information for these effects is not available from human exposures to tritium, the following approach was used. Excess risks for the effects given following low-level exposure to x rays or gamma rays were estimated from available human epidemiological data using appropriate dose-rate effectiveness factors. These human-risk estimates for low-level x rays or gamma rays were then multiplied by the appropriate best-estimate relative biological effectiveness for tritium, taking into account differences in effectiveness of comparison radiations. The resultant lifetime risk coefficients for low-level exposure to tritiated water are as follows. For cancer mortality, the most probable risk (50th percentile) is 81 × 10^{-6} mGy^{-1} and the 90% confidence interval is 38 to 185 × 10^{-6} mGy^{-1}. For genetic effects in the first generation after exposure the risk is 7.9 × 10^{-6} mGy^{-1}, with a 90% confidence interval of 3.8 to 16.3 × 10^{-6} mGy^{-1}. For developmental effects from low-level tritiated water exposures *in utero*, the risk is uncertain but is estimated to be <400 × 10^{-6} mGy^{-1}. The risks from exposure to organically bound tritiated molecules are estimated to range from values that are similar to those for tritiated water to about a factor of 2 higher.**
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Key words: tritium; risk analysis; gamma radiation; health effects

INTRODUCTION

DUE TO the nonexistence of human data for tritium risk, dose rate, and relative biological effectiveness (RBE) for any of the endpoints considered here (i.e., cancer, genetic effects, and developmental effects), extrapolations from other data are required. Fortunately, a wealth of cancer information is available for gamma rays and x rays from human epidemiological studies (e.g., BEIR 1980, 1990; UNSCEAR 1986, 1988). Also, much dose rate and RBE information is available from experimental animals (mostly mice) and from *in vitro* studies for gamma rays, x rays, and tritium. Thus, the approach taken here is to predict tritium risks in humans by combining human-risk estimates for gamma rays and x rays available from recent reports (BEIR 1990; UNSCEAR 1986) with dose-rate effectiveness factors (DREFs) (Straume 1988; BEIR 1990) and tritium RBEs (Straume and Carsten 1993) for relevant experimental endpoints. Uncertainties in risk estimates are calculated using a Monte Carlo sampling approach.

* Lawrence Livermore National Laboratory, University of California, Livermore, CA 94550.
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Estimation of Cancer Risk

Background information

Cancer-risk estimates are generally expressed as the lifetime excess mortality per unit dose in a specified population (e.g., deaths per 10^6 persons per mGy). Lifetime excess mortality is the number of radiation-induced deaths that occur in an exposed population during the remaining lifetime of all individuals in that population. Because there are as yet no human populations for which full lifetime risk expression has been measured, models are required to project the risk into the future based on presently available data. The two most common projection models are the absolute risk and the relative risk models. The absolute risk model is based on the assumption that the baseline risk and the excess risk are additive. The relative risk model is based on the assumption that the baseline risk and the excess risk are multiplicative. From data available at this time, the relative risk model appears to be favored for most cancers (e.g., BEIR 1990).

Several physical and biological factors can modify cancer risk and must be considered. The principal physical factor is radiation dose. The dose received by target tissues and organs can produce lifetime excess cancer-mortality risks in human populations ranging from near zero at very low doses to as high as ~20% at near-lethal doses, inferred from data in BEIR (1990). Because risk estimates are dependent on dose, a high premium is placed on good dosimetry (e.g., see RERF 1987).

The dose rate is also important, especially for low-linear energy transfer (LET) radiations such as gamma rays, x rays, and beta particles (including those from tritium). It has been clearly demonstrated for a variety of radiobiological endpoints that gamma rays and x rays delivered at low dose rates are less effective than when delivered at high dose rates. This is especially the case at relatively high total doses; the many studies on this subject indicate that low dose rates of gamma rays and x rays are about 2 to 10 times less effective than

high dose rates (NCRP 1980). In contrast, at low total doses, the dose-rate effect has been observed to diminish and, in fact, is predicted to disappear at the very low doses of most practical importance for radiation protection; however, it should be noted that the induction of certain cancers appears to have differing dose-rate responses. For example, a dose-rate response is predicted for human leukemia (BEIR 1990) while for breast cancer in women, the risk appears to be independent of dose rate (Land et al. 1980). Differences in dose-rate response for tumors of various sites are also seen in experimental animals (refer to information in the following paragraphs and NCRP 1980). Of particular interest here is the dose-rate effect for tritium which has been measured for several experimental endpoints and found to differ by a factor of about 2 between acute and chronic exposures (Ujeno 1983). Although this is less than the DREFs observed for gamma rays (NCRP 1980), it does indicate that the dose rate must be considered for tritium beta particles as well.

The radiation quality is also an important factor, especially in the case of exposures at low doses and low-dose rates. Even among the so-called low-LET radiations, differences are seen at low doses or low dose rates where, for example, 250-kVp x rays appear to be about twice as effective as ^{60}Co gamma rays (ICRU 1986). The biological effectiveness of tritium beta rays is generally somewhat greater than for x rays and gamma rays.

The principal biologic factors affecting cancer risk are organ or tissue exposed, baseline-cancer incidence, latency period, age at exposure, time since exposure, and sex. It is now clear from both human and animal data that different organs can have very different susceptibilities to cancer induction by ionizing radiation (e.g., Fabrikant 1990; Fry 1981). Baseline-cancer incidence may be important because it appears that the radiation-induced risks for at least some cancers are related to the baseline risk in such a way that they can be expressed as multiples of the natural incidence.

The delay between exposure and cancer detection is called the latency. The minimum latency may be only 2–3 y, as is the case for leukemia, or it may be more than a decade, as is the case for many solid tumors. It appears that the latency varies with tissue or organ as well as age at exposure; it is longer for younger than for older individuals. Indications from experimental animal data are that the latency may also be affected by dose, dose rate, and radiation quality. At the present time, latency periods can only be estimated reasonably well for leukemia induced by high dose-rate low-LET radiation (Fabrikant 1990). Significant uncertainties exist in the estimation of latencies for solid tumors (i.e., nonleukemia cancers), and even greater uncertainties exist for tumors induced by chronic radiation exposures (Fabrikant 1990).

Another factor that can affect cancer risk is age at exposure. Generally, the lifetime excess-cancer risk decreases with age at exposure. This may result from a combination of less time available in older individuals for risk expression and, for certain cancers, higher susceptibility in younger individuals. Estimates of total lifetime excess-cancer risk (leukemia plus nonleukemia) for those exposed as children appear to be about twice as high as the risk for those exposed as adults (BEIR 1990).

The cancer-risk data available for *in utero* exposures are very limited. There are basically two sources of data; those from medical exposures and those from the atom-bomb survivors. Data from the Oxford and New England surveys, the largest studies on children exposed *in utero* for medical reasons, show a positive association between *in utero* exposure and subsequent development of leukemia and other malignancies (UNSCEAR 1986; BEIR 1990). A positive association also appears likely in the most recent follow-up of atom-bomb survivors (BEIR 1990).

The amount of time that has passed after exposure influences the risk of radiation-induced cancer. For leukemias, the excess risk begins to increase after a minimum latency period of ~2 y, then reaches a plateau after 10 to 15 y, and declines to normal preexposure levels by 30 y after exposure. For nonleukemias, excess risk generally does not become apparent until about 10 y after exposure. In contrast to the case for leukemia, the excess risk for most solid tumors continues to increase with time after exposure and appears to be proportional to the increased background incidence with age.

Gender also influences the cancer risk, particularly for breast and thyroid cancer. For example, women are about three times more susceptible to radiogenic thyroid cancer than men (BEIR 1990); however, the overall risk of excess cancer mortality is ~25% less in females than it is in males.

Cancer risks for gamma rays and x rays

The values listed in Table 1 are age-specific point estimates of lifetime-cancer-mortality risks induced by low doses of gamma rays or x rays. The risk estimates for *in utero* exposures are limited to cancers that appear

Table 1. Point estimates of lifetime excess cancer mortality risk for low-level exposures to gamma rays or x rays[a].

	Age at exposure			
	In utero[b]	5 y of age[c]	45 y of age[c]	All ages[c]
Leukemia	~10	9	9	10
Nonleukemia	~10	33	12	17
Total	~20	42	21	27

[a] Deaths per 10^6 persons per mGy.
[b] UNSCEAR (1986) and BEIR (1990); risks during the first 10 y of childhood, only, primarily from x-ray exposures.
[c] From Table 4–3 of BEIR (1990), 1:1 male-to-female; primarily from high-energy gamma-ray exposures; includes DREF of 4 for nonleukemia cancers and an implicit DREF of 2.1 for leukemia.

during the first 10 y of life and are best-estimate summary values from UNSCEAR (1986) and BEIR (1990). The risk estimates for all ages in Table 1 are based on the age distribution of the U.S. population. All postnatal risk estimates were obtained from the values listed in Table 4–3 of BEIR (1990). Risks for leukemia are the arithmetic means of the risks for males and females (assumes a male-female ratio of unity). Risks for nonleukemias are the arithmetic means of risks for males and females divided by the DREF. The total risk is the sum of the risks for leukemia and nonleukemia.

Since we are concerned here with low-level exposures of low-LET radiation, postnatal risk values obtained principally from high dose and high dose-rate data (i.e., mostly from atom-bomb survivors) were converted to the risks that would be expected following low-dose rates; this was accomplished using appropriate DREFs. The DREF is defined as the slope of the linear curve fitted to the high dose/high dose-rate data divided by the slope of the curve fitted to the low dose or low dose-rate data for the same endpoint and the same radiation. The use of DREFs to convert risk factors from acute, high-dose data (e.g., those from atom-bomb survivors) to low dose rates is broadly accepted in radiation risk assessment (e.g., UNSCEAR 1977, 1982, 1986, 1988; NCRP 1980; BEIR 1980, 1990).

Exposures *in utero* involved low doses of x rays to the fetus, a 5–50 mGy average dose from diagnostic radiology procedures (BEIR 1990). Thus, a DREF of 1 was applied to the cancer risk estimates for *in utero* exposures from UNSCEAR (1986) and BEIR (1990), and the results are listed in Table 1. As previously mentioned, the major sources of cancer data following fetal exposures are the Oxford survey of childhood cancers and the New England survey. Results from these large surveys suggest risk coefficients during the first 10 y of life of ~20 per 10^6 persons per mGy. About one-half of the cancers are attributable to leukemias, and one-quarter to tumors of the nervous system (UNSCEAR 1986; BEIR 1990). Although only two cases of childhood cancer have been observed so far among 1,630 *in utero* exposed atom bomb survivors, these results are not inconsistent with the estimates from the Oxford and New England surveys. Epidemiological studies do suggest an association between *in utero* exposure and excess cancer risk in adult life; however, the magnitude of that risk is too uncertain to be quantified (BEIR 1990) and thus is not included in Table 1. Because the cancer risk expressed in adult life (i.e., beyond 10 y of age) as a result of *in utero* exposure is not included, the *in utero* values in Table 1 are probably underestimates.

Postnatal exposures involved principally high doses and high dose rates of high-energy gamma rays from the Hiroshima and Nagasaki atom bombs [the small neutron component of the DS86 dose in Hiroshima contributed less than 10% to the uncertainty in risk estimates for both cities combined when the BEIR V method was used (RERF 1987; BEIR 1990; Brenner 1991)]. The nonleukemia cancer risk values in Table 1 were obtained by dividing the nonleukemia cancer risks from Table 4–3 of BEIR (1990) by the "best-estimate" DREF of 4 suggested by the BEIR V committee for carcinogenesis endpoints (see Table 1–4 of BEIR 1990). For leukemia, the postnatal risk values were obtained directly from Table 4–3 of BEIR (1990) because they already contain an implicit DREF, i.e., the BEIR V committee used a linear-quadratic dose-response function for leukemia. Based on uncertainty analyses in BEIR (1990), we estimate the 90% confidence interval on the total gamma-ray induced cancer-mortality-risk coefficient for all postnatal ages in Table 1 to be 16–46 per 10^6 persons mGy^{-1}. This interval includes the BEIR III estimate for low-level, low-LET radiation of 23 per 10^6 persons mGy^{-1} (BEIR 1980).

Cancer risk estimates for tritium

As previously stated, because cancer-risk information is not available from human exposures to tritium, such estimates for tritium must be obtained using the human-cancer data that are available for gamma rays and x rays together with RBEs and dose-rate information from experimental animal studies. Eqn (1) was employed here to estimate postnatal cancer risk for tritiated water (HTO). The associated uncertainties were calculated by first determining uncertainty distributions for all input parameters to the extent possible and then using a Monte Carlo code[†] to sample from each parameter distribution according to eqn (1):

$$R_{\mathrm{HTO}} = \{[(R_{n,m} + R_{n,f})/2 \times DREF] + (R_{l,m} + R_{l,f})/2\} \times RBE_{HTO}, \quad (1)$$

where

R_{HTO} = Excess lifetime cancer mortality risk from chronic exposure to HTO;

$R_{n,m}$ = Excess lifetime cancer mortality risk in the male (m) for nonleukemias (n) following acute exposures to gamma radiation, assuming a population with an age distribution that is identical to that in the U.S. [from Table 4–2 of BEIR (1990), continuous lifetime exposure]. For uncertainty analysis, the risk distribution for males in Figure 4.1 of BEIR (1990) was used;

$R_{n,f}$ = Excess lifetime cancer mortality risk in the female (f) for nonleukemias (n) following acute exposures to gamma radiation, assuming a population with an age distribution that is identical to that in the U.S. [from Table 4–2 of BEIR (1990); continuous lifetime exposure]. For uncertainty analysis, the risk distribution for females in Figure 4.1 of BEIR (1990) was used;

$DREF$ = ose-rate effectiveness factor for gamma-ray-induced tumors in experimental animals. A

[†] Crystal Ball (Version 2), A Forecasting and Risk Management Program for the Macintosh, Comtech Services Inc., Denver, CO.

cro- or anencephaly, micro- or anophthalmia, extra or omitted digits, growth retardation, sterility, and damage to the central nervous system (e.g., UNSCEAR 1977). A variety of developmental effects have also been observed in humans following prenatal radiation (e.g., Dekaban 1968; Otake and Schull 1983). The kind and severity of the abnormality is usually related to the stage of development at the time of exposure. Large exposures prior to major organ development (zygote through the blastula stage) will usually result in death of the embryo. Large exposures during organ development tend to result in the most severe damage to the organs undergoing rapid cell proliferation and differentiation; this damage is often expressed as serious physical abnormalities in offspring. Exposure after major organogenesis (i.e., during the fetal stage) will tend to produce less severe functional problems. As an example, a classic experiment was performed by Russell (1954) in which mice were irradiated with 200 R of x rays at various times postfertilization and the incidence of abnormalities, prenatal death, and neonatal death were recorded. They found that ~80% of the early embryos died when exposed within a few days after fertilization. Also, they found that the most sensitive period for malformations is between 6 and 13 d postfertilization, which is the time when major organogenesis occurs in the mouse. Exposure to 200 R during this interval resulted in ~100% of the animals having visible malformations at birth, and a substantial fraction of these animals died shortly after birth.

An effect that is of particular concern is central nervous system (CNS) damage because it has been shown to be easily induced by radiation in both animals and humans. Studies with rats have demonstrated that x ray doses as low as 0.03 Gy given during the development of the CNS can result in detectable damage (UNSCEAR 1986). The high CNS sensitivity seen in experimental animals has also been observed in humans. Data now available from the Hiroshima and Nagasaki atom bomb survivors indicate that the most sensitive developmental stage for mental retardation in humans is between 8 and 15 wk of gestation (Otake and Schull 1983; Otake et al. 1987). During this time, exposure to 1.4 Gy resulted in 75% of the children becoming severely mentally retarded; at 0.7 Gy, about 25% became severely retarded. The shape of the dose-response relationship can be described by either a linear-quadratic curve or a linear curve at higher doses with a threshold at about 0.25 Gy. The statistical precision of the data is not sufficient to discriminate between these dose-response models (BEIR 1990).

In addition to data for severe mental retardation, the results from atom bomb survivors also provide information on more subtle CNS damage that may have resulted from exposure to atom bomb radiations. Intelligence tests taken at 10 to 11 y of age by individuals who were exposed prenatally to the atom bomb radiations demonstrate a substantial decrease in test scores in those exposed between 8 and 15 wk of gestation (Schull and Otake 1986). The data suggest a linear dose-response curve without a threshold and a decrease in intelligence of ~25 points per Gy. Another approach to measuring CNS damage in atom bomb survivors exposed *in utero* is school performance. In this case also, the greatest effect was observed for those exposed between 8 and 15 wk of gestation. A regression analysis for scholastic performance vs. dose suggests a linear dose-response curve without threshold.

Taken together, the data on CNS damage in animals and humans are consistent with either linear dose-response relationships or dose-response relations that decrease in effectiveness as the dose diminishes and, in some cases, appear to have a threshold or quasi-threshold at low doses (Otake and Schull 1983; NRC 1985; Schull and Otake 1986; Otake et al. 1987; BEIR 1990).

Risk estimates

UNSCEAR has estimated the total risk for all deleterious effects from *in utero* exposure to x rays and gamma rays (UNSCEAR 1986). Although the uncertainties are large, they conclude that for the small doses of most practical concern in radiation protection the risk is no more than 200 per 10^6 liveborn mGy^{-1}. UNSCEAR also points out that this is small compared with the natural incidence of malformations in nonirradiated populations, which is on the order of 60,000 per 10^6 liveborn in humans (UNSCEAR 1986). These estimates for low-level exposures in humans are corroborated by experimental data available from monkeys exposed to HTO throughout pregnancy. In an experiment by Jones et al. (1980), HTO was given in the drinking water of pregnant squirrel monkeys at levels ranging from 16 to 1,000 times the permissible level for human consumption. Results indicated no effects on the newborn progeny in terms of body weight, body dimensions, organ weights, hematologic patterns, the histology of organs, or tissues, with the exception of the ovaries. The number of immature oocytes decreased markedly, as has been observed by others for squirrel monkeys exposed *in utero* (Dobson et al. 1984, 1986). Functional effects, such as may be caused by damage to the CNS, were unfortunately not explored.

Since the UNSCEAR estimates are based on a combination of high-energy gamma-ray (atom bomb) and orthovoltage x-ray (medical) data for low-level irradiation, reasonable risk estimates for HTO would be about two times higher than the UNSCEAR (1986) values previously cited, that is, 400×10^{-6} per liveborn mGy^{-1} appears to be an upper-limit estimate on the risk for HTO-induced developmental effects *in utero*. Although the actual risk for low-level HTO exposure may be much lower than this due to the possibility of threshold for certain developmental effects at low doses, it should be noted that the range (0 to 400×10^{-6} per liveborn mGy^{-1}) includes risk values that are substantially higher than those for cancer and genetic effects. Further evaluation of the available data could possibly provide some reduction in the uncertainties associated with the estimation of risk for developmental effects *in utero* and should be explored.

CONCLUSIONS

The approach and information presented here, together with the radiobiological and RBE data presented in Straume and Carsten (1993), provide an updated basis for tritium risk assessment. For example, we have used updated tritium RBE values, radiobiologically consistent DREFs, custom distributions for each variable, and a Monte Carlo sampling approach to determine overall uncertainties. The approach presented can also be used to obtain age-specific cancer risk estimates for exposures to tritium. This would be accomplished if age-specific cancer risk values for gamma rays were used in eqn (1). It is recommended that additional analyses of the human and animal data be performed to better quantify the uncertainties associated with *in utero* developmental effects. Also, additional work is recommended to better define the RBEs for OBT molecules.

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Paper

TRITIUM SAMPLING AND MEASUREMENT

M. J. Wood, R. G. C. McElroy, R. A. Surette, and R. M. Brown*

Abstract—**Current methods for sampling and measuring tritium are described. Although the basic techniques have not changed significantly over the last 10 y, there have been several notable improvements in tritium measurement instrumentation. The design and quality of commercial ion-chamber-based and gas-flow-proportional-counter-based tritium monitors for tritium-in-air have improved, an indirect result of fusion-related research in the 1980s. For tritium-in-water analysis, commercial low-level liquid scintillation spectrometers capable of detecting tritium-in-water concentrations as low as 0.65 Bq L^{-1} for counting times of 500 min are available. The most sensitive method for tritium-in-water analysis is still ^{3}He mass spectrometry. Concentrations as low as 0.35 mBq L^{-1} can be detected with current equipment. Passive tritium-oxide-in-air samplers are now being used for workplace monitoring and even in some environmental sampling applications. The reliability, convenience, and low cost of passive tritium-oxide-in-air samplers make them attractive options for many monitoring applications. Airflow proportional counters currently under development look promising for measuring tritium-in-air in the presence of high gamma and/or noble gas backgrounds. However, these detectors are currently limited by their poor performance in humidities over 30%.**

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Key words: tritium; ionization chambers; sampling; proportional counters

INTRODUCTION

SEVERAL previous reviews of tritium sampling and measurement technology remain relevant (Butnitz 1974; NCRP 1976; IAEA 1991). Most of the recent improvements have resulted from improved engineering of established technologies. The commercial market for tritium-monitoring equipment has grown considerably, largely as a result of increased activity in the fusion community. This has encouraged investment in product development which has resulted in a significant improvement in the quality and diversity of commercial tritium-monitoring products, especially ion-chamber-based monitors. This growth of interest in tritium-monitoring technology has been quite evident at the four "Tritium Technology in Fission, Fusion, and Isotopic Applications" conferences held over the last 12 y (in Dayton, OH 1980 and 1985; Toronto, Canada 1988; and in Albuquerque, NM 1991).

This report concentrates on the recent developments in tritium-monitoring technology. It supplements, but does not replace, previously mentioned reviews. In particular, sampling and sample preparation methods that have not changed significantly in the last 10 y are dealt with in more detail in NCRP Report No. 47 (NCRP 1976).

The following nomenclature will be used. Equipment used to collect tritium for subsequent analysis will be referred to as "collectors." Gas-washing bottles (bubblers) and silica gel traps are examples of tritium collectors. Equipment that employ collectors will be referred to as "samplers." The term "monitor" will be reserved for instruments that measure directly and provide an immediate indication of the sample being analyzed. Instruments employing ionization chambers and flow-through gas-proportional counters are examples of tritium monitors.

MONITORING APPLICATIONS

The measurement of tritium can be separated into three measurement ranges—environmental, radiation protection, and process. The term "environmental monitoring" will be used to indicate outdoor monitoring applications, starting with "true background" measurements. Samplers, rather than monitors, must be used for almost all environmental tritium measurement applications because of the low concentrations to be measured. Typical ranges for background tritium concentrations are 0.02–0.5 Bq L^{-1} for surface seawater (Van Scoy et al. 1991) and 10–100 mBq m^{-3} for air (Östlund 1974; Okai and Takashima 1991; Rozanski et al. 1991a).

For radiation protection monitoring, tritium-in-air concentrations must be measured down to a fraction of a derived air concentration (DAC). Different values of the DAC [or maximum permissible concentration in air (MPC_a)] are used in different jurisdictions. Based on the most recent recommendations of the ICRP (1991a,b), the DAC for HTO† is 300 kBq m^{-3} (based on a 20 mSv y^{-1} dose limit).

* AECL Research, Chalk River Laboratories, Chalk River, Ontario K0J 1J0.

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† HTO will be used for all forms of tritiated water vapor, whether it be HTO, T_2O, or DTO. Similarly, HT will be used for all forms of tritiated hydrogen gas, whether it be HT, T_2, or DT (D = ^{2}H, T = ^{3}H).

Most tritium-monitoring programs designed for radiation protection use a combination of tritium samplers and tritium monitors.

Since radiation protection monitoring is often performed in areas with other sources of radiation, tritium-measuring equipment may be required to discriminate against other forms of radiation or other radionuclides.

Doses from tritium are usually assessed by measuring tritium levels in urine. In most cases, this measurement is a much more accurate indication of the dose received from a tritium exposure than a measure of the tritium source. An exception might be very high concentrations of essentially pure HT (McElroy and Johnson 1988). Tritium dose determination from bioassay measurements is discussed in Bioassay Guidelines (Health and Welfare Canada 1983) and in the American Standard for Dosimetry (ANSI 1983).

Monitors, rather than samplers, are used almost exclusively for measuring tritium in process systems. Unlike environmental and radiation protection monitoring applications, process monitors have to be designed to operate under pressure and/or vacuum, and often to strict pressure code requirements.

TRITIUM SAMPLING TECHNIQUES

General considerations

Obtaining a representative sample is a major concern in any sampling program. When measuring tritium in the environment close to a source, the local meteorological conditions must be considered. Significant variations in the tritium-in-air concentration may be present, especially in areas with strong prevailing winds. Concentration gradients may also be notable in lakes and oceans (Fine et al. 1987; Van Scoy et al. 1991).

Tritium-in-water sampling

In most applications, water samples are collected manually in glass or plastic containers (grab sampling). If the purpose of the sampling is routine compliance monitoring, a sample size of a few milliliters is often all that is required. For low-level measurements using low-level liquid scintillation counting or gas counting, the sample should be at least 20 mL (500 mL if isotopic enrichment is to be performed or the sample is to be analyzed by ^{3}He mass spectrometry). It is recommended, however, that as large a sample as practical be collected since the larger sample will dilute any cross contamination that may occur between the time of sample collection and preparation of the sample for counting.

Automatic samplers are sometimes used to monitor liquid effluent (Cooper 1985). Ideally, an automatic sampler collects a constant fraction of the total effluent stream. Such a sampler requires an accurate on-line measurement of the effluent flow rate and appropriate metering of the sample flow. A much simpler solution is to sample a constant volume at regular intervals. As long as the effluent flow rate is reasonably constant, this constant rate automatic sampling will yield a representative sample.

Tritium-in-air sampling

Active tritium-in-air sampling. A basic sampling system for HTO consists of a pump, a sample collector, and a flow-measuring or flow-recording device. Air is drawn through the collector for a measured time period at a monitored flow rate to determine the total volume of air sampled. The total amount of HTO recovered from the collector is divided by the total volume of air sampled to determine the average HTO-in-air concentration of the air sampled. In some sampler types, the specific activity of the water collected is measured and the air concentration is determined from the known or measured humidity. Some common collectors are cold traps, tritium-free water, and solid desiccants such as silica gel, DRIERITE,‡ or molecular sieve.

Cold traps are usually made of glass and consist of cooled collection traps through which sample air flows. The trap is cooled well below the freezing point of water, usually with liquid nitrogen. The water vapor collected is then prepared for analysis, usually by liquid scintillation counting. Phillips and Easterly (1982) have shown that over 95% HTO collection efficiency can be obtained using a single cold trap. Often, a pair of cold traps is used in series, resulting in a collection efficiency in excess of 99%. There is, however, a danger that cold traps of poor design can lose sample by the frost being physically blown out of them.

Gas-washing bottles (i.e., "bubblers") filled with an appropriate collecting liquid, usually tritium-free water, are used quite extensively for collecting HTO from air. HTO in the sample gas stream "dissolves" in the collecting liquid. For the effective collection rate to remain the same as the sample flow rate, the specific activity of the bubbler water must be negligible with respect to the specific activity of the water vapor. Thus, the volume of air that can be sampled is ultimately limited by the volume of water in the bubbler. However, except when sampling under conditions of very high humidity, sample loss (dryout) from the bubbler usually limits collection time rather than the attainment of specific activity equilibrium. Osborne (1973) carried out a thorough theoretical and experimental evaluation of the HTO collection efficiency of water bubblers over a wide range of conditions.

Low-vapor-pressure sorbents, such as ethylene glycol, are often used to avoid the problems of dryout; however, the advantages are not quite as great as may first appear. The water loading of the ethylene glycol will eventually saturate at a value determined by the humidity of the air being sampled. Sampling longer than this then results in sample loss. The effective

‡ DRIERITE, W. A. Hammond DRIERITE Company Ltd., P.O. Box 460, Xenia, OH 45385.

purification is all that is required. If tritiated organics are to be measured, however, the organics have to be converted to water for analysis.

Once a sample is collected, precautions must be taken to prevent exchange between atmospheric water vapor and the sample. Samples should always be stored in sealed containers. Since HTO will diffuse through plastic, glass or metal containers are recommended for long-term storage. However, plastic containers are acceptable for short-term storage (up to a few months). The bottle cap seals should always be checked.

Water recovery

The methods outlined in NCRP Report No. 47 (NCRP 1976), lyophilization, distillation, azeotropic distillation, and dilution are still the most prevalent methods of recovering tissue-free water from environmental samples. For all of these methods, the sample size required depends on the water content of the sample and the amount of water required for analysis. Assuming the water sample is analyzed by liquid scintillation counting, a 5–10-mL sample is ideal. Larger sample sizes are required if the sample is to be electrolytically enriched (see the section on electrolytic enrichment later in this paper).

Lyophilization. Lyophilization (or freeze drying, as it is more commonly known) consists of freezing the sample and then recovering the water under vacuum. The water is recovered in a cold finger located between the vacuum pump and the sample. The disadvantage of this technique is the time required. Several days may be required to obtain enough water from environmental samples for low-level analysis.

Distillation. Distillation is a common method of recovering water from samples. The sample is simply heated and the distillate is collected on a cold finger. If accurate measurements are required (within 5 to 10%), isotopic fractionation should be considered. The possibility of isotopic separation can be eliminated if the sample is completely distilled, but this often results in contamination of the distillate by volatile pyrolytic products from any organic residue.

Azeotropic distillation. Azeotropic distillation is a convenient method of recovering water from vegetation, soil, urine, and animal tissue. In this technique, the water-containing material is placed in an immiscible hydrocarbon, such as toluene or xylene, and the water is boiled out as a constant boiling mixture of fixed water-hydrocarbon composition. The distillate is collected in a receiver where the phases separate and the hydrocarbon returns to the distillation flask. When taken to completion, there is no isotopic fractionation and the distilled water is free of any constituent that would interfere with liquid scintillation counting. The disadvantage of the technique is that the dried organic residue must be free of hydrocarbon if an organically bound tritium measurement is desired on the same samples.

Dilution. Dilution of a sample with a solvent is a simple way to recover HTO. The solvent can simply be tritium-free water. If left long enough, the water in the sample will come into equilibrium with the added water. This exchange can be accelerated by adding heat. Enough solvent should be added to the sample so that the fraction of HTO remaining in the sample is small compared to the amount of HTO in the solvent. The disadvantage of this method is that the sample is diluted by the solvent. This technique is usually reserved for samples with tritium concentrations significantly above background levels.

Urine

Tritium bioassay is used routinely to assess tritium doses received by workers. Liquid scintillation counting of a worker's raw urine is sensitive enough to detect tritium concentrations of any significance for radiation protection purposes. However, in some applications, such as monitoring environmental transport of tritium in an ecosystem, lower tritium-in-urine concentrations need to be measured. In such applications, the reduction of counting efficiency resulting from chemical and color quenching of the raw urine is limiting. Activated charcoal filters have been used to remove the organic fraction (Eakins et al. 1975; Stencel et al. 1988), greatly reducing the sample quench. The azeotropic distillation method previously mentioned is also suitable for reducing the quench in urine samples.

Organically bound tritium

Once the tissue-free water tritium (TFWT) has been removed from a sample, tritium may still exist in the sample in the form of OBT. The OBT can be recovered from the dried sample by combustion. Moghissi et al. (1975) described a method in which the sample is placed in a 2-L steel Parr[1] bomb, pressurized with oxygen to about 1.5–2 MPa and ignited. The bomb is then evacuated through a cold trap to collect the water. This method has the advantage that it is very rapid, permitting quicker turnaround times than conventional combustion methods, and the water obtained usually does not require further purification. It is limited to about 10 g of dried organic material. Larger samples must be combusted by the conventional technique in a quartz tube with carefully controlled oxygen flow (Hisamatsu et al. 1990a).

The water sample obtained from combustion usually requires purification by refluxing with $KMnO_4$ and/or Na_2O_2, distilled to obtain a pure sample for counting (Hisamatsu et al. 1990a). In a recent interlaboratory comparison of OBT measurements (Hisamatsu et al. 1990b), good agreement was found between all six laboratories participating. However, although the same general procedures were used (combustion followed by purification and distillation), the details were not the same for any of the six laboratories.

[1] Parr bomb, Parr Instrument Company, 211 53rd Street, Moline, IL 61265.

Electrolytic enrichment

One of the limiting parameters of liquid scintillation counting (LSC) and internal gas counting is the amount of sample that can be put into the counting system. When the tritium concentration of a water sample is too low for these methods, isotopic enrichment can be used to concentrate the tritium. Electrolytic enrichment factors of up to 100 are obtainable with two-stage electrolysis. This technique, however, is only suitable if large sample volumes are available (Brown and Grummitt 1956; Östlund and Werner 1962; Östlund et al. 1964; Taylor 1981; Kakiuchi et al. 1991).

MEASUREMENT OF TRITIUM IN WATER

Liquid scintillation counting of discrete samples

The most common procedure for measuring tritium in water is liquid scintillation counting of discrete samples. The water samples to be analyzed are mixed with a liquid scintillation solution (cocktail) in a scintillation vial. Standard scintillation vials have a capacity of 20–24 mL, but smaller vials ("mini vials") are also used if sensitivity is not a problem. Typical water:cocktail ratios between 1:5 and 1:15 are used in routine applications; however, for low-level counting, special cocktails are used that permit ratios of up to 10:10. In a liquid scintillation counter, the prepared sample is placed between two photomultiplier tubes. The beta radiation in the sample interacts with the scintillant, resulting in several light photons being eventually emitted. The summed height of the pulses from the two photomultipliers is proportional to the energy of the beta particle. These counters are run in coincidence mode in order to distinguish photon pulses from photomultiplier noise. Most LSC counting systems are based on commercially available counters using commercially available cocktails.

Typical performance of a regular liquid-scintillation counter is a background of 10–15 cpm with a tritium counting efficiency in water of 35 to 55%, depending on sample composition and sample size. The corresponding detection limit is on the order of 200 Bq L^{-1} for a 1-mL water sample counted for 10 min.

Recent developments in background correction and compensation techniques made possible through improved electronics have greatly improved the performance of commercial liquid scintillation spectrometers (Schönhofer and Henrich 1985; Rozanski et al. 1991b).

Chemiluminescence in a sample prepared for liquid scintillation counting results in single photon events that can be detected by the scintillation counter. Although coincidence counting greatly reduces the possibility of these events being mistaken as tritium events, some random coincidences of these single photon events will occur. Circuits to detect and correct for random coincidences are available for many commercial liquid scintillation counters. Although the details of how instrument manufacturers accomplish chemiluminescence correction vary, they are all based on determining the random coincidence component in the tritium channel and subtracting it from the total signal to obtain the true tritium signal.

Tritium decay in a normal liquid scintillator results in single pulses with a pulse width of a few nanoseconds. When a cosmic-ray particle passes through the sample in the liquid scintillation counter, several smaller "afterpulses" follow the initial pulses resulting from the interaction. Canberra Packard[1] uses a burst counting technique (Roessler et al. 1991) to detect the number of after pulses, referred to as the "pulse index," and only accepts pulses with a low pulse index, effectively discriminating against interferences such as cosmic-ray particles.

The Wallac 1220 Quantulus liquid scintillation counter,[2] designed for very low-level counting, uses a combination of passive and active shielding around the sample detector assembly, in addition to a pulse-shape discriminator and a pulse-height comparator (Kaihola 1991). The passive shielding consists of a copper-lined, 630-kg (considerably more massive than that used in conventional LSCs) lead shield. Between this shield and the sample is the active shield, consisting of a cavity filled with a liquid scintillator and a photomultiplier tube on either side. Single photons are detected directly and multiphoton events are detected with coincidence circuits, both in the active shield and in the sample. In addition, events that cause multiphoton events in the active shield and the sample at the same time can be detected (a third coincidence circuit) and subtracted from the tritium channel.

Automatic optimization, in which the counter determines the optimum window settings (energy windows) for the best figure of merit, is available on some machines. This feature eliminates the time-consuming and tedious task of determining optimum window settings by repeated measurements.

These are just a few representative features available on some modern liquid scintillation counters. More complete information can be obtained from manufacturers of specific products. Although some of these techniques have been known for some time, it is only over the last several years that many of them have become available on commercial instruments.

Background count rates as low as 0.4 cpm with a counting efficiency of 28% have been reported for an 8-mL water sample, corresponding to a detection limit of 0.65 Bq L^{-1} for a 500-min count time (Schönhofer

[1] Canberra Packard, Packard Instrument Company, One State Street, Meriden, CT 06450.

[2] Wallac 1220 Quantulus liquid scintillation counter, Wallace Oy, P.O. Box 10, SF-20 101 Turku, Finland.

Table 3. Detection of tritium on surfaces. LSC = liquid scintillation counting. PMT = photomultiplier rule.

Detection limit (Bq cm^{-2})					Method of measurement	Sample preparation time	Measurement time
10^{-3}	10^{-1}	10^{1}	10^{3}	10^{5}			
■	■				swipe—LSC	~ 2 min	~ 10 min
	■	■			swipe—windowless gas-flow proportional counter	~ 2 min	~ 1 min
		■	■		windowless gas-flow proportional counter		~ 1 min
			■	■	air-flow proportional counter		~ 1 min
				■	PMTs/solid scintillator counter		~ 1 min
				■	solid-state detector (APD)		~ 1 min

MA 02172). The sample, when appropriately prepared on either a glass slide or a fiber filter, is placed in a mechanism that locates the sample under the detector for measurement. Yamamoto et al. (1989) also describe a low-energy particle detector using a modified commercial silicon photodiode. With the low-energy discriminator set at 6 keV, the detection efficiency for beta rays was estimated to be 85% for ^{14}C and 37% for ^{3}H.

Solid-state detectors fabricated from HgI_2 have been used to detect low-energy x rays. The limitations to detecting low-energy beta particles is not inherent in the crystal, except with a device structure. Shah et al. (1990) developed a HgI_2 detector with a detection efficiency of about 25% for tritium betas.

Although many reports show the feasibility and usefulness of solid-state detectors for measuring tritium, these devices are still in the research stage. New developments in semiconductor fabrication may make production of suitable monitors readily available.

Bremsstrahlung counting

The use of bremsstrahlung for quantitative determination of radioactive substances with no gamma emission is a well-known procedure and has been extended to the measurement of tritium in aqueous and organic systems (Cameron et al. 1959; Westermark et al. 1960; Rosen et al. 1967; Curtis 1968; Matsuyama et al. 1991). These methods can be used for relatively large amounts of tritium, in the order of tens of MBq (millicuries) and upwards.

The advantage of detecting and measuring the bremsstrahlung emission from the interaction of tritium betas with different substances is that the low-energy betas from tritium do not penetrate the front surfaces of most detectors or the walls of the vessel the tritium is contained in. X rays of similar energy as the tritium betas are somewhat more penetrating and, therefore, can be detected at greater distances from the source and through materials that would normally absorb the tritium betas.

One report (Johnson et al. 1970) described the use of a 7.5-cm-diameter, 2.0-mm-thick sodium iodide crystal with a 25-μm aluminum window. The minimum detectable concentration for this detector was in the order of 40 MBq kg^{-1} of tritium in water. Detection of bremsstrahlung generated by tritium betas has been suggested as a practical method of measuring tritium activity in samples not suitable for measurement by conventional methods, such as liquid scintillation counting. Examples include tritiated charcoal, vacuum pump oil metals, storage beds, colored aqueous wastes, and biological materials (Westermark et al. 1960; Rosen et al. 1967; Curtis 1968; Webb and Jones 1974; McGann et al. 1985).

Bremsstrahlung counting has been used for measuring the amount of tritium gas in glass and alloy tubes (Souers et al. 1981; Matsuyama et al. 1986). Gas-proportional counters were placed external to the vessels containing tritium gas, and the bremsstrahlung fluence was measured. A prototype system consisting of a sampling cylinder, gas-circulating apparatus, a NaI(Ti) detector and associated electronics has been developed (Bingo et al. 1982). Pulses due to bremsstrahlung with energies from 4–17 keV were counted. The minimum detectable concentration of this prototype monitor was 370 MBq m^{-3}.

The design of an on-line tritium monitoring system has also been described (Kishukawa et al. 1984; Matsuyama et al. 1991). The study described tritium absorbed in titanium plated onto a copper disk placed on the wall of a cell through which tritium gas was passed. The sensitivity of this monitor was found to be suitable for deuterium-tritium (D-T) fuel measurements for fusion reactors.

Although bremsstrahlung counting is an effective method for measuring tritium in various materials, the drawback is that the bremsstrahlung yield is in the order of 10^{-4} for most materials. The signal is further reduced by absorption in the vessel walls and the detector window and dead layer.

Hyperpure germanium (HpGe) detectors with very thin windows will detect low-energy x rays down to a few keV, with a resolution of about 300 eV for 5.9 keV ^{55}Fe x rays. Unfortunately, these are very expensive and require liquid nitrogen cooling for operation.

Microcalorimetry

Microcalorimetry has been used to calibrate tritium standards. A good description of microcalorimetry for measuring tritium is given in NCRP Report No. 47 (1976). This technique is not very sensitive to the very low heating resulting from tritium beta decay, 0.9108 μW GBq^{-1} (Martin 1985). Genka and Kobayashi (1987) have recently described a calorimeter for measuring tritium and other pure beta emitters capable of measuring power changes as low as 0.2 μW, which corresponds to a detection limit of 200 MBq of tritium.

SUMMARY

Scintillation counting persists as the most widely used technique for analysis of tritium in water. Improvements in signal processing technology (both hardware and software) have improved the performance of modern scintillation spectrometers. New "biodegradable" liquid scintillation cocktails, which are rapidly replacing their more toxic predecessors, have greatly helped reduce the waste problems associated with scintillation counting.

Ion chambers continue to be used extensively for monitoring tritium in air, and the availability and quality of ion chamber-based instruments has increased noticeably over the last 10 y, as a result of an increased market resulting primarily from fusion research. Improvements in the performance of gas-flow proportional counters designed for measuring tritium in air have also been realized over the last 10 y, mostly due to improved signal conditioning and compensation techniques.

The use of passive samplers for measuring HTO in air is increasing both for workplace monitoring and, to a lesser extent, in environmental monitoring applications. The simplicity of these devices makes them extremely attractive, especially in locations where electrical power is not available and long-term integration is desirable.

Several instruments and techniques are available for measuring tritium on surfaces; however, many of the more sensitive instruments are inherently the most fragile. Also, while the sensitivity of surface monitors will increase with surface area, large-area surface monitors are only useful if the surfaces to be monitored are large and flat. Tritium-swipe monitoring, followed by scintillation counting, continues to be the most widely used method for measuring tritium surface contamination. This technique is much less prone to contamination problems than direct-reading monitors or swipe sampling, followed by swipe counting with a windowless proportional counter.

The extremely low energy of tritium betas makes it essentially impossible to measure the total activity of solids in a nondestructive manner except by ^{3}He mass spectrometry.

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Paper

METABOLISM AND DOSIMETRY OF TRITIUM

Robin L. Hill and John R. Johnson*

Abstract—**This document was prepared as a review of the current knowledge of tritium metabolism and dosimetry. The physical, chemical, and metabolic characteristics of various forms of tritium are presented as they pertain to performing dose assessments for occupational workers and for the general public. For occupational workers, the forms of tritium discussed include tritiated water, elemental tritium gas, skin absorption from elemental tritium gas-contaminated surfaces, organically bound tritium in pump oils, solvents and other organic compounds, metal tritides, and radioluminous paints. For the general public, age-dependent tritium metabolism is reviewed, as well as tritiated water, elemental tritium gas, organically bound tritium, organically bound tritium in foodstuffs, and tritiated methane.**

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Key words: tritium; metabolism; dosimetry; biokinetics

INTRODUCTION

THE uptake of tritium in humans can occur by exposure to air, water, or food containing tritium. Assessment of radiological doses received from such exposures to tritium requires modeling to estimate tritium concentrations in various body tissues. Quantitative estimates resulting from metabolic models are combined with knowledge of the physical characteristics of tritium and of the dose-to-risk relationship to provide estimates of risk from exposure to tritium.

This document is a review of the current knowledge of physical, chemical, and biological transport processes that pertain to tritium metabolic modeling and summarizes the *in vivo* behavior of tritium, which is required for dosimetry and radiological risk assessments. It expands on, and extends, previous reviews (Myers and Johnson 1991; NCRP 1979b).

TRITIUM SOURCES

Tritium (T or ^{3}H) occurs from both natural and manufactured processes. As an isotope of hydrogen (H), tritium has an extremely small natural abundance relative to H and deuterium (D). Naturally occurring tritium is produced in the atmosphere by interaction of high-energy cosmic radiation with oxygen and nitrogen and by ternary fission in geological formations. Tritium in the upper atmosphere is oxidized to tritiated water (HTO)[†] and mixes with the surface waters of the earth.

Since 1954 (UNSCEAR 1982), a major portion of the global inventory of tritium has resulted from the release of large amounts of manufactured tritium into the environment from nuclear weapons testing, nuclear power production, and industrial, commercial, and research uses of tritiated compounds (Table 1). Much of the tritium that remains in the environment from nuclear weapons testing exists as HTO. As HTO, tritium can enter the body by inhalation, ingestion, or skin absorption (Cawley and Lewis 1985). With the elimination by the U.S. and the U.S.S.R. of atmospheric nuclear weapons tests in the early 1960s, the concentration of tritium in the environment has been decreasing.

PHYSICAL AND CHEMICAL CHARACTERISTICS OF TRITIUM

Tritium is an isotope of the element hydrogen which is both naturally occurring and manufactured. Its half-life is 12.26 y (Unterweger et al. 1980), decaying to helium (^{3}He) while emitting a beta particle. The beta particles, while of low energy (18.6 keV maximum, 5.7 keV average), have enough energy to produce ionizations (≃0.03 keV per ion pair) and excitations of molecules in their path. Tritium poses no external hazard since the beta particles released during tritium decay cannot penetrate the outer layer of dead skin cells due to their small range in tissue of <1 μm, and maximum range of only 6 μm (ICRP 1983). Because of the low beta energy, dilution throughout all of the soft tissues, and elimination with an average biological half-life of ≃ 10 d in adults (ICRP 1979–1982), tritium as HTO has a relatively low radiological toxicity when compared to other pure beta emitters, such as ^{32}P or ^{90}Sr, or to common beta-gamma emitters, such as ^{131}I or ^{137}Cs. Of the three hydrogen isotopes, only tritium is radioactive.

The incorporation of tritium in chemical compounds can occur by many different mechanisms. Inclusion of tritium in molecules during synthesis occurs naturally in photosynthesis (Moses and Calvin 1959; Belot et al. 1983) and other biological processes. Intermolecular exchange of tritium with other hydrogen isotopes will also result in assimilation of tritium. For instance, when tritium is combined with the two other hydrogen isotopes (i.e., H and D), many molecular combinations can be formed. Molecular combinations include *hydrogen gas* (as H_2, HD, HT, D_2, DT, and T_2), *water* (as H_2O, HDO, HTO, D_2O, DTO, and T_2O), and *methane* (as CH_4, CH_3D, CH_2D_2, CHD_3, CD_4, CH_3T, CH_2T_2, CHT_3, CT_4, $CHDT_2$, CH_2DT, CD_3T, CD_2T_2, and CDT_3).

When substantial quantities of tritium are present, a radiation effect exists where reactions can occur at room temperature instead of the elevated temperatures required for hydrogen or deuterium reactions. The beta particles from tritium decay provide the necessary activation energy to drive the reactions. The chemical reactivity, diffusion properties, and radioactivity of tritium contribute to it ability to interact with almost all materials.

Although tritium is not considered a particularly toxic radionuclide, it presents a concern since it can become part of the biologically necessary hydrogen pool. If released into the environment, the tritium poses a potential internal radiation hazard since compounds containing tritium undergo various chemical transformations resulting in forms which can enter the body. The majority of tritium in the environment exists as HTO. Because of its mobility in the environment and its biological importance, water is one of the most important compounds of tritium. As HTO, tritium can enter the body by inhalation, ingestion, or diffusion through the skin (Cawley and Lewis 1985). Once inside the body, the HTO diffuses freely and rapidly across cellular membranes, equilibrating throughout the total body water pool. The uniform concentration of HTO will result in the radiation dose being uniformly distributed throughout the body.

On the other hand, the tritium from HTO may exchange with hydrogen atoms and thereby become incorporated into organic molecules. HTO is the primary chemical precursor for other chemical forms of tritium. Essentially any organic molecule can incorporate tritium in this manner. Each will have a specialized metabolism associated with it that may result in inhomogeneous distributions of tritium within the body, within individual organs, and even within individual cells. To better understand the metabolic behavior of organically bound tritium (OBT),[‡] it is important to point out the nature of the chemical bonds of tritium in organic molecules. In most organic compounds, whether inhaled, ingested in foodstuffs, or synthesized *in situ*, the tritium bound to oxygen, nitrogen, phosphorus, or sulfur can readily exchange with hydrogen found in the body water pool and, thus, will have the same metabolism and distribution as for HTO. This has been called the *exchangeable bound tritium fraction*. Tritium may also exchange with hydrogen in carbon-hydrogen (C-H) bonds. Once bound to carbon, tritium will be difficult to remove and, thus, has been named as the *nonexchangeable bound tritium fraction*. Nonexchangeable bound tritium is normally released only as a result of enzymatic breakdown of the molecule containing this carbon-tritium bond (ICRP 1989). Experimental determinations of the proportions of exchangeable and nonexchangeable bound tritium in biological systems have not yet been made but may be estimated if the molecular structure of the molecules are known. The designations of exchangeable and nonexchangeable forms of OBT are only conceptual terms used for modeling purposes and are meant only to represent differences in turnover and hydrogen bonding.

The distribution and rate of metabolic degradation of nonexchangeable bound tritium fraction will vary depending on the type of organic molecule that contains the tritium atoms (Smith 1986; Taylor 1989; Konig 1990; Taylor et al. 1990). In general, the more rapidly a molecule is turned over, the more tritium will be incorporated per unit time and the more rapidly the tritium will be removed from the same molecule. For longer-lived molecules, such as the structural protein, collagen, or the phospholipids of some nerve cells, fewer tritium atoms will be incorporated per unit time and those that are incorporated will be retained for longer periods of time (NCRP 1979b). After degradation of organic molecules containing tritium, a large fraction of tritiated amino acids can directly enter the general body pool or undergo degradation into water and other metabolites that may enter the general body pool to be recycled into other macromolecules or be excreted (NCRP 1979a).

Significant levels of elemental hydrogen gas (HT) can be released into the environment due to manufactured processes (Ichimasa et al. 1986). Compared to HTO, HT[§] is relatively inert in biological systems and usually has a very low uptake into body fluids. Studies have shown that conversion of HT to HTO in the atmosphere is extremely slow (Brown et al. 1990). HTO seen in the atmosphere following release of HT is thought to arise mainly from oxidation by microbial organisms within the soil (McFarlane et al. 1978; Dun-

Table 1. Comparison of tritium sources.[a]

	Current source term (TBq y^{-1})	Current inventory (TBq y^{-1})
Natural production	7.4×10^4	1.3×10^6
Nuclear explosions	—	3.7×10^7
Nuclear fuel cycle	$\sim 1 \times 10^4$	$\sim 4 \times 10^4$
Consumer products[b]	$\sim 1 \times 10^4$	$\sim 1 \times 10^5$

[a] Adapted from Luykx and Fraser 1986.
[b] These figures represent only order of magnitude estimates and assume that all tritium used in consumer products is consigned to waste.

* Health Physics Department, Pacific Northwest Laboratory, P.O. Box 999, Richland, WA 99352.

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[†] HTO is used as the chemical notation for tritiated water with other hydrogen isotopes, unless noted otherwise. HTO will represent HTO, DTO, and T_2O.

[‡] OBT is used throughout this paper to represent both exchangeable and nonexchangeable organically bound tritium, except where noted otherwise.

[§] HT is used throughout this paper to denote HT and T_2 hydrogen gas.

creased by a factor of 1.5 to account for skin absorption from tritiated water vapor.

No significant skin absorption of HT in air occurs. Studies by Hutchin and Vaughan (1965), Eakins et al. (1975), and Johnson et al. (1988) have shown that if skin comes into contact with surfaces that have been exposed to high concentrations of HT gas, significant uptake and retention of tritium occurs in the skin, as well as in other organs. The processes by which this occurs are not fully understood.

Distributional effects

Even though tritium can enter the human body by several different routes, the subsequent behavior of the tritium will depend more on its chemical form than its mode of entry. The two main forms of tritium to consider are water and organic molecules. Tritiated water rapidly diffuses across all cell membranes and quickly equilibrates with the body fluids, attaining a uniform concentration throughout the body; however, tritiated organic molecules can be localized in relatively small numbers of cells at relatively high concentrations. The mean range of the tritium beta particle, 0.68 μm, is small compared to the dimensions of cells. Consequently, while the average dose to the tissues may be very low, the dose to cells where the tritium compound is concentrated may be large.

When tritium is homogeneously distributed throughout the body, such as for HTO, the decay of tritium will result in uniform irradiation to the body. As HTO, irradiation of the body from tritium will be similar to that received from external irradiation. If the HTO is present in concentrations that will deliver an absorbed dose of a few gray or more over a short time period, many aspects of the acute radiation syndrome, similar to those seen for external irradiation, may appear (NCRP 1979b).

For radiation protection purposes, inhomogeneous distribution of tritium at the subcellular level as a result of exposure to HTO is generally not considered (Saito and Ishida 1988); however, localization of other tritiated compounds occurs in certain cells or tissues and may cause biological effects that do not resemble those seen with HTO. An inhomogeneous concentration of tritium will generally result in doses to the involved cells that are larger than the average tissue dose, which may be negligible. As a result, the concept of average tissue dose for many tritiated compounds especially (especially those involved in nonexchangeable bonds) would be inappropriate and a poor estimate of the hazard associated from exposure to such compounds, especially if the exposure results in the tritium being incorporated into sensitive regions of the cells, such as DNA.

Several studies have looked at the effects of tritiated thymidine since it is a DNA precursor. There has been concern that DNA precursors containing tritium would lead to selective deposition of tritium in the cell nucleus. A number of studies have shown that the distributional effect of tritium incorporated into DNA results in selective irradiation of only part of the cell (the nucleus) and that the resulting risk of any biological effect to the cell might be greater than tritium incorporated more uniformly throughout the cell (Bond and Feinendegen 1966; Zhuralev and Kalyazina 1979). When tritiated thymidine[1] was injected into humans, 40–60% was found to be metabolized into HTO, with the remainder being incorporated into DNA. The rate of incorporation of tritiated thymidine into DNA will depend on the cell turnover rate; i.e., the higher the cell turnover rate, the higher the rate of DNA synthesis, resulting in a higher rate of tritiated thymidine incorporation into DNA. Inhomogeneous distribution of the tritiated thymidine within the body will result since only those cells undergoing DNA synthesis will assimilate tritiated thymidine.

Transmutational effects

Transmutational effects on molecules can result from the incorporation of tritium. Tritium decay can induce mutations in DNA molecules by two possible mechanisms: 1) the ionization from the emitted beta particle; and/or 2) by transmutation of the ^{3}H to ^{3}He with an associated energy of excitation and recoil (Halpern and Stocklin 1976; NCRP 1979b; Feinendegen et al. 1980; Carsten 1986). While the biological effects of such transmutations in DNA might be easily detected, it requires very accurate microdosimetry to be able to separate the observed effects of ionizations from the beta particle and those seen for transmutation (NCRP 1979b). Since the maximum recoil energy associated with this transmutation is only 3 eV, it is thought to be too small to break most chemical bonds and is generally ignored (NCRP 1979b). Helium atoms which replace the transmutated hydrogen atoms have extremely weak bonding with carbon. This instability leads to dissolution of the carbon-helium bond, yielding free helium atoms and reactive carbon ions. It is the resulting carbon ions which can cause molecular alterations in the DNA molecule, presenting as single-strand breaks (Krasin et al. 1973; Krasin et al. 1976a) or interstrand cross-links (Krasin et al. 1976b), depending on their location within the DNA molecule. The NCRP (1979b) supports the conclusion that energy deposition from beta radiation of most nuclides in nucleic acids in humans significantly outweighs the damaging contribution from transmutation effects.

METABOLIC MODELS OF TRITIUM

As an isotope of hydrogen, tritium can enter a biological system in the form of many different chemical compounds. For most types of tritium exposure described in this paper, two chemical forms of tritium (HTO and OBT) contribute the majority of the radiation dose to the body regardless of whether they were taken directly into the body or enter into *de novo* synthesis of other tritiated compounds (Taylor et al. 1990). With HTO playing a primary role in most metabolic processes associated with tritium, the accuracy of the risk assessment from exposure to tritium will largely be dependent on the accuracy of the tritiated water dosimetry. Fortunately, the dosimetry of HTO is relatively straight-forward and is based on simple, scientifically defendable assumptions about water distribution in the body. Consideration must also be given to the metabolic fate of elemental tritium gas, organically bound tritium, biotransformed tritium in foodstuffs, and the age-dependent effects.

As a precursor to the development of a mathematical model, a conceptual model of the metabolic components and the relationships of tritium transfer between them needs to be developed first. The metabolic components are referred to as compartments, which represent reservoirs of tritium within the body, such as the respiratory tract, the GI tract, or the soft tissues. In a conceptual model, the transfer from one compartment to another is specified by a mathematical relationship that represents this transfer as a function of time. For instance, in cases where tritium is continuously taken into the body and eliminated (i.e., when drinking contaminated water or breathing contaminated air over a considerable time period), tritium will accumulate within the body. If elimination rates are about equal to intake rates, then a state of equilibrium exists. The amounts within the body may increase if intake rates are greater than elimination rates (Quimby et al. 1970). A decrease of activity in any organ can result either by physiological elimination of the compound containing the radionuclide or by physical decay of the radionuclide. If properly developed, the conceptual model can be solved using exact analytical methods or numerical approximations to provide an estimate of concentrations in the defined compartments as a function of time.

The metabolic models discussed in the following sections represent the conceptual interpretation of metabolic processes for a variety of different forms of tritium including HTO, HT gas, HT-contaminated surfaces, OBT in foodstuffs and other organic compounds, metal tritides, tritiated methane, and radioluminous paints.

HTO metabolism

There is a general consensus that the current understanding of the metabolism of HTO, which is the result of many studies of HTO metabolism, is sufficient for radiation protection purposes. The detailed knowledge of HTO metabolism has led to the development of models which result in acceptable estimations of absorbed doses from tritium concentrations seen in the urine (Johnson 1976, 1982).

The greatest majority of tritium in the atmosphere exists in the form of HTO, which can be transferred to humans primarily by inhalation, skin absorption, or ingestion of drinking water or food (ICRP 1979–1982; Belloni et al. 1983). The kinetics of HTO in the body follow that of body water, except for a small portion of metabolized tritium that becomes bound to organic molecules. HTO is excreted in urine, feces, sweat, and breath (NCRP 1979b).

If individuals are at rest at the time of exposure to HTO vapor, the amount of tritium entering the body by inhalation will be approximately equal to the amount entering by skin absorption (Pinson and Langham 1980); however, physical activities during exposure will result in approximately twice as much tritium entering the body by the inhalation pathway (Myers and Johnson 1991). In addition, some endogenous HTO may be formed following metabolic breakdown of various tritiated compounds (e.g., inhaled HT gas) as a result of metabolic combustion of OBT, or following ingestion of foodstuffs or water containing OBT. In normal adults, $\simeq$70% of the total body water is from ingested water and the rest originates from either water in food or from catabolized organic molecules (van Den Hoek et al. 1980).

HTO exposure is generally the most important to consider since HTO quickly reaches equilibrium with hydrogen in the body and imparts a uniform dose to all soft tissues. The ICRP (1979–1982) recommended the assumption be made that internalized HTO is completely and instantaneously absorbed and mixed rapidly with the total body water so that at all times, the concentration in sweat, sputum, urine, blood, insensible perspiration, and exhaled water vapor is the same. Following HTO exposures, a small fraction of tritium will eventually become incorporated into organic molecules. The OBT will have different metabolic rates than those for total body water, therefore, the retention of tritium following an exposure to HTO should be described by more than one component, *viz.*, one component that is characteristic of the retention of body water and one or more components that are characteristic of the retention of hydrogen (or tritium) in organic molecules or OBT (Johnson 1982; Taylor et al. 1990).

In the past, urine concentrations were used to directly obtain an estimate of the whole-body dose based on the argument that the time-integrated tritium concentration (which is proportional to accumulated dose) in any tissue would not exceed that in the body water (or urine) (Sanders and Reinig 1968; Snyder et al. 1968; Osborne 1972; NCRP 1979b); however, this procedure has been reevaluated following the publication of ICRP recommendations regarding the concept of E_c (ICRP 1991). The ICRP Committee 2 recommended that instead of using the total body water, the average soft tissue should be used to estimate the E for HTO exposures and that only retention in total body water be considered in dose estimation (ICRP 1979–1982). In other words, the dose contribution from OBT was to be ignored. This recommendation was based on results of several studies (Sanders and Reinig 1968; Snyder et al. 1968; Moghissi et al. 1972) which indicated that OBT contributed <10% of the E_c in the Reference

[1] Tritiated thymidine is sometimes denoted as ^{3}HTdR.

assumed to go directly to urine, and 50% of the HTO in body fluids was assumed to be excreted by pathways other than urinary excretion. The model predicts that the integrated amount of OBT in skin at the point of contact is ≃0.5 Bq d^{-1} Bq^{-1} of intake, and 40% of the intake is excreted as OBT. For a contaminated area of skin of 20 cm^2, the skin dose would be ≃2 × 10^{-8} Sv. This would result in a dose conversion factor of 5 × 10^{-8} Sv Bq^{-1} of OBT excreted (assuming a contaminated skin area of 20 cm^2), which is sufficiently large that the skin dose may be limiting in certain situations (McElroy et al. 1989). (Note: Two areas of contaminated skin of different sizes can have the same skin dose but different resulting E. The skin section having the largest area will have more associated OBT, thereby contributing to a higher E.)

OBT metabolism

Little is currently known of the metabolism of organically bound tritium in humans. OBT can enter the body directly by ingestion of tritiated foodstuffs, by inhalation of volatile organic vapors or aerosols, or can be formed *in vivo* from tritium that is present in the general body pools after exposure to other tritium-containing compounds (Diabaté and Strack 1993). Once in the body, OBT can be incorporated into a variety of biochemical compounds such as amino acids, sugars, proteins, starches, and other structural materials (Saito and Ishida 1989).

With such a diversified array of biochemical compounds associated with OBT and each having unique metabolic processes associated with it, uniform distribution or retention of OBT compounds cannot be assumed. For instance, tritiated compounds may have a shorter retention time than HTO (Osborne 1966); however, much of the tritium may be in exchangeable sites, and when these compounds are catabolized, HTO will be produced. Tritiated nucleic acid precursors generally have a longer retention than HTO even though much of the incorporated tritium may exist at exchangeable sites or be catabolized (ICRP 1979–1982). Generalities can be made based on how easily tritium exchanges with hydrogen in the hydrogen pool in the body. Organic molecules, in general, have slower turnovers and are retained in the body for longer periods of time than water. Tritium found in nonexchangeable organic binding sites (i.e., tritium bound to carbon atoms) will contribute an increased average soft-tissue dose over that seen for HTO alone, primarily due to the longer retention times in the body (ranging from hours to years) and inhomogeneous distribution among the different organs (Taylor et al. 1990). The increment of this increase in dose will vary depending on the chemical nature and metabolism of the molecule containing the tritium-carbon bond. In contrast, the specific activity of tritium in exchangeable hydrogen binding sites (≃30% of total hydrogen in organic molecules within the body) should be identical to that for hydrogen in body water and, thus, will have metabolism essentially the same as for HTO.

Information gained from human studies indicates that the retention of tritium can generally be characterized by a three-component exponential function having biological half-lives of 10 d, 21–76 d, and 280–550 d. As previously described, the first component represents body water metabolism while the latter two components generally correspond to OBT. The second component may be related to tritium involved in carbon-tritium bonds of organic molecules since it is eliminated with approximately the same half-life as carbon, namely 40 d (ICRP 1979–1982; Taylor 1990). The latter component may represent tritium incorporated into organic molecules having very slow turnover rates.

The fate of OBT after an intake is dependent primarily on the hydrogen metabolism of the human body. Hydrogen accounts for 60% of all atoms in the human body, as described for the Reference Man (ICRP 1975). The possibility of tritium substitution is of major interest because every day 2.1 × 10^{26} hydrogen atoms (5% of all hydrogen atoms in the body) takes part in metabolic processes (ICRP 1975). The net exchange of tritium with hydrogen always occurs toward lower concentrations and is dependent on the type of chemical bond. While OBT has been separated into two general forms, namely exchangeable OBT (i.e., tritium bound to nitrogen, oxygen, phosphorus, or sulfur) and nonexchangeable OBT (i.e., tritium bound to carbon atoms), the proportion of tritium entering into each form is highly variable (Rochalska and Szot 1977; Pietrzak-Flis et al. 1978; Kowalska 1985). It should be noted that the division between exchangeable and nonexchangeable forms of OBT implies a simplification that does not reflect the wide spectrum of organic bonds that are possible.

Tritium in exchangeable OBT is assumed to rapidly exchange with hydrogen found in the body water pool and, thus, will have a metabolism and distribution similar to that of HTO. After equilibration with the body fluids, the exchangeable tritium will be eliminated with a half-life of ≃10 d (ICRP 1979–1982). Nonexchangeable organically bound tritium is firmly bound to carbon atoms and is usually released only during enzyme-mediated reactions (Etnier et al. 1984). The rate of the metabolic breakdown may be fast for certain molecules and very slow for others; the retention times may range from hours to years (Taylor et al. 1990). Analysis of observations of tritium elimination following accidental inhalation or ingestion of tritium has shown that up to three exponential components have been used to describe the retention curves (see Table 2). Besides the HTO biological half-life, two additional exponential components with longer biological half-times were used with ranges of 21–76 d and 280–550 d.

Different types of exposure to tritium will result in different proportions of OBT formed. Following HTO exposures, OBT compartments contribute on the order of 10–20% of the whole-body $H_{c,T}$ that would be seen for HTO alone (Johnson 1982; Etnier et al. 1984; Dunford and Johnson 1987). Following inhalations of HT gas, OBT results from metabolic processes associated with the HTO formed by internal oxidation of HT (Peterman 1985a) and provides a small contribution to the whole-body $H_{c,T}$. With HT inhalations, it is important to note that the dose ratio of HTO to OBT will be similar to that seen for HTO exposures since all OBT will have resulted from HTO metabolism. In contrast, skin contact with HT-contaminated surfaces can result in direct formation of OBT in the skin (Johnson and Dunford 1985; Johnson and Peterman 1987; Johnson et al. 1988; McElroy et al. 1989). In all of these cases, the dose due to OBT distributed throughout the body will be some fraction of the whole-body $H_{c,T}$. However, ICRP 30 recommendations indicate that OBT contributions to dose should be neglected in dose estimation for both HTO exposures and HT exposures since it is expected to be <10% of the total.

OBT from foodstuffs

When dose assessments for tritium exposures are performed, the fact that OBT can be directly incorporated from foodstuffs has generally been ignored since little was known about this exposure pathway for tritium (Travis et al. 1984). However, recent reports using advanced environmental tritium models indicate that the ingestion doses from OBT in the food chain may be 4–23 times higher than doses resulting from inhalation and skin absorption of HTO (Gulden and Raskob 1992). Diabaté and Strack (1993) have provided an in-depth review of the current knowledge of OBT formation and translocation in plants in this issue.

Tritium in foodstuffs as OBT is known to be mostly in a nonexchangeable form (Konig 1990). OBT from foodstuffs in the diet can be used as metabolic precursors that will be directly incorporated into organic metabolic pathways or can undergo degradation to HTO and enter the general body water pool (Etnier et al. 1984; Travis et al. 1984). The relative proportions of exchangeable and nonexchangeable OBT in various components of the human diet vary greatly due to the influence of many different factors such as the history of exposure to tritium during food growth, the differences between plants and animals, and differences in wet and dry food. The nonexchangeable-to-exchangeable OBT ratios in foodstuffs reported in the literature have a wide range of values. For instance, Bogen et al. (1973) have reported that the tritium concentration in nonexchangeable components of food from New York was about twice that of exchangeable components and water components. Belloni et al. (1983) reported even higher ratios in Italian food (i.e., nine times more nonexchangeable tritium). The difficulty in defining a realistic range for the nonexchangeable-to-exchangeable OBT ratio has hampered the development and use of general metabolic modeling of OBT turnover in humans (Ueno and Kawamura 1975).

Etnier et al. (1984) have proposed a four-compartment model (Fig. 5) that is based primarily on hydrogen balance within the human body. This model was adapted from the NCRP model (NCRP 1979a) which was originally based on a three-compartment model developed by Bennett (1973). No physiological significance should be attached to the model compartments since they are meant to represent only differences in carbon-tritium bonding and molecular turnover rates. The four-compartment model allows for input of OBT directly into compartments representing tissues rather than into the body water pool. This feature is desirable but care must be taken to distinguish between the different OBTs. The catabolism of OBTs in foodstuff will be governed by the catabolism of the carbon compounds with which the tritium is bound; and it is unlikely that only four compartments will give a reasonable estimate of retention. For example, when the model alone was used for a hypothetical diet consisting of total intakes amounting to 10 times more nonexchangeable OBT than exchangeable OBT, the resulting dose estimate was reported to be 4.5 times higher than the dose contributed from free body water alone. This ratio value is considerably higher than expected based on animal studies (NCRP 1979b).

A complete characterization of the retention of tritium from ingested foodstuffs would require modeling carbon retention, which was a major task. This is likely not warranted as the nonexchangeable OBT, in even dry food, and can add (at most) ≃6% to the total cell dose (Myers and Johnson 1991). However, when considerations are made for what is known about tri-

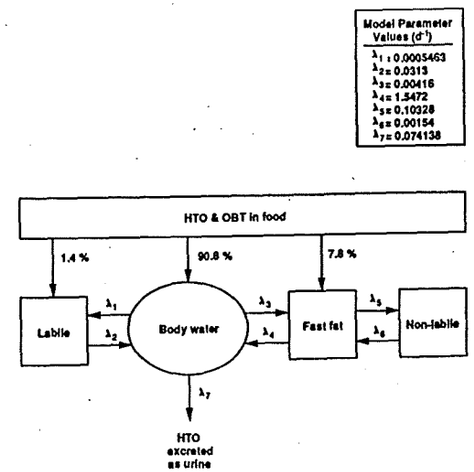

Fig. 5. OBT compartment model (adapted from Etnier et al. 1984).

Table 3. EDE rates for tritiated methane [Sv s^{-1} (Bq m^{-3})$^{-1}$].[a]

Age at intake (y)	Absorbed gas $\dot{H}_L$[b]	Absorbed gas $\dot{H}_A$[c]	Metabolized gas[d]
Adults (20)	3.2×10^{-19}	2.4×10^{-19}	4.5×10^{-17}
Children (10)	6.4×10^{-19}	2.4×10^{-19}	4.7×10^{-17}
Infants (1)	9.0×10^{-19}	2.4×10^{-19}	1.9×10^{-17}

[a] Adapted from Phipps et al. (1990).
[b] $\dot{H}_L$ is the dose rate from gas contained in the lung.
[c] $\dot{H}_A$ is the dose rate from gas absorbed into body fluids but not metabolized.
[d] Dose rate where it is assumed that 1% of inhaled tritiated methane is metabolized.

inated by the dose in the small fraction of methane that is absorbed and metabolized. If the ICRP model for vapors had been used, the doses from the inhaled tritiated methane would have been higher by a factor of 100, which results from assuming 1% of the absorbed material being metabolized. If it is found that methane could present a hazard, several parameters will have to be more carefully evaluated. These would include a more realistic determination of the portion of inhaled methane that is metabolized and incorporated into body fluids, evaluation of the temporal and spatial distribution of absorbed methane that remains in the form of methane, an investigation of the nature and type of the metabolic products following breakdown of methane *in vivo*, and a study of methane's potential to penetrate intact skin.

Radioluminous paints

Tritiated luminous compounds, which have been used for many years, are prepared by adding tritium to basic starting materials for specific plastics, polymerizing these tritiated compounds, dissolving them in organic solvents, mixing them with fluorescent zinc sulfide, and finally evaporating the solution (Seelentag 1973). The end product, in the form of fine crystals that are coated with a thin layer of this plastic material, is used in luminous paints and light sources. A worker may be exposed to tritium from either production or application of the luminous compounds. The potential hazard may result from skin absorption, inhalation, or even ingestion. During all of these procedures, tritium evaporates as a gas (Seelentag 1973) or HTO (Rudran 1988a), and in the first few months the end product may lose up to 50% of the tritium initially present (Seelentag 1973; Rudran 1988a). The chemical form of the tritium by-products, as well as their route of absorption and elimination, will depend on the chemical nature of the luminous compounds. The more biologically active the material, the greater the tritium absorption.

As can be seen in Table 4, tritium absorbed from various types of luminous compounds through the skin or the GI tract of animals is eliminated by two distinct biological half-lives; the first (T_{b1}) was attributed to result primarily from elimination of nonbiotic, unabsorbed tritium, while the second (T_{b2}) had half-lives similar to that documented for HTO. The studies were not carried out long enough to determine if longer-lived components were present. For each type of absorption route, the aliphatic acid and polyester forms of the luminous paint were seen to result in higher absorption levels than for the other forms. Tritium absorbed through the GI tract was eliminated at different rates than that absorbed through the skin (Seelentag 1973; Wawerna 1973).

Rudran (1988a) recently described the retention and elimination characteristics of workers who were exposed to luminous paints by inhalation of insoluble tritiated polystyrene from aerosols of the luminous paints. The high specific activity of the tritiated polystyrene used 7.4 TBq g^{-1} (200 Ci g^{-1}) was thought to cause radiation damage of the compound leading to the production of HT, HTO, and several different, but unknown, organic compounds. The presence of HTO vapor in the workplace also contributed to the workers' exposure through inhalation and skin absorption. The workers were monitored using urine and fecal analysis.

The initial biological half-life of tritium eliminated in the urine was observed to be 7–10 d, indicating elimination of HTO. In addition, delayed excretion of HTO into urine (supposedly from OBT metabolism) and tritium in fecal excretions was seen which had biological half-lives of 35–70 d. This delayed excretion pattern was proposed to result from tritium in the inhaled paint aerosols that was retained for long time periods in the lungs. From this data, a model of the absorption and excretion of HTO was presented (Fig. 6). The annual dose to workers in the luminous paint industry was estimated assuming half-lives of 7.0 for HTO and 100 d for OBT. As can be seen in Table 5, estimations based solely on urinary excretion of HTO will underestimate the dose to the individual worker. The exposures to both OBT and insoluble tritium compounds must be considered when evaluating the exposure to luminous paint workers (Rudran 1988a). The uncertainties in the biological parameters used in the cited study require further investigation.

Age-dependent tritium metabolism

Existing ICRP models for the radionuclide metabolism have been directed toward estimation of dose to the adult male worker (ICRP 1979–1982). The ICRP does not recommended that these biokinetic models be used for calculating equivalent dose for members of the general public (ICRP 1979–1982; ICRP 1989) since differences in growth and other changes may affect not only the metabolism of compounds within the body, but also the biological risk. The problem of age-dependent effects on radionuclide metabolism has recently received considerable attention as evidenced by the occurrence of an international workshop on age-related factors in radionuclide metabolism and dosimetry which was held in Augers, France in 1986 (Gerber et al. 1987), and the issuance of ICRP Publication 56, "Age-Dependent Doses to Members of the Public from Intake of Radionuclides, Part I" in 1989 (ICRP 1989).

Several important factors must be considered when looking at the effects of age on anatomical, physiological, and biokinetic data. The organ mass, shape, and relative position will change with age and will affect $H_{c,T}$ seen in an organ. In general, the dose coefficients [E per unit intake (Sv Bq^{-1})] for the younger members of the public will be greater than those seen for adult workers by factors ranging from 2 to 10 because of the smaller organ and tissue masses (Kaul and Roedler 1987; NRPB 1990).

The biokinetics of the radionuclides will also be affected by differences in age. The tritium elimination is generally fastest in infants and decreases with age (Kaul and Roedler 1987). This generally offsets the increases caused by increases in organ and tissue mass. For tritium, the biokinetics of HTO and OBT must be considered independently. (HT is not considered here since a significant portion of internal dose resulting from inhaled HT comes from HTO resulting from oxidation of HT in the GI tract.)

For simplification, the ICRP has suggested the use of a two-component exponential function to describe the whole-body retention of HTO (ICRP 1989). It is based on the assumption that the biological half-times can be adequately estimated from the daily mass balance and mass of total body stores for water and carbon (i.e., 10 d and 40 d, respectively, for adults), and that OBT formed from HTO contributes 10% of the average

Table 4. Summary of absorption and elimination of tritium from luminous paints via the skin and GI tract.[a]

Compound	Chemical form	Specific activity (GBq g^{-1})	Skin: Percent absorbed	Skin: T_{b1}[b]	Skin: T_{b2}[b]	GI tract: Percent absorbed	GI tract: T_{b1}[b]	GI tract: T_{b2}[b]
MH 200	Aliphatic acid	7.4	8.8	0.5	4.2	21.4	0.6	9.8
CH 250	Polystryene	9.3	0.6	0.4	3.5	4.2	0.4	7.7
RH 250	Silicone rubber	9.3	0.2	0.5	3.2	2.7	0.4	8.8
RH 20	Silicone rubber	0.74	0.1	0.5	4.1	3.0	1.0	7.8
NH 600	Polystyrene	22.0	0.1	0.3	3.2	0.7	0.6	8.3
RC 65	Polyester	2.3	—	—	—	80.0	1.2	14.0
HTO	Control	—	100.0	—	4.1	100.0	—	8.4

[a] Adapted from Wawerna (1973). The experiments done for skin absorption were performed in rats, while cats were used for GI absorption experiments.
[b] T_b = biological half-life for various compartments representing OBT.

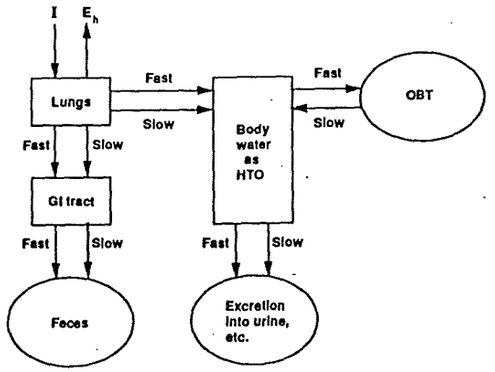

Fig. 6. Metabolic pathways for HTO following inhalation of luminous paints. I = intake by inhalation; E_h = direct exhalation (adapted from Rudran 1988a).

Table 5. Calculated annual doses from inhaled luminous paints.[a]

Target	Dose component	Dose rate (Sv y^{-1})
Whole body	HTO	1.9×10^{-3} to 1.9×10^{-2}
Whole body	OBT	4.2×10^{-3} to 1.2×10^{-2}
Lungs	Insoluble paint dust[b]	2.4×10^{-1} to $1.5 \times 10^{+0}$
Lungs	Insoluble paint dust[c]	8.5×10^{-2} to 1.7×10^{-1}

[a] Adapted from Rudran (1988a).
[b] Calculated using delayed urinary excretion; pathways are given in Fig. 6.
[c] Calculated using data from fecal analysis.

Table 6. Biokinetic data for ingestion of HTO at different ages.[a]

Age	Percent distribution: Body water	Percent distribution: Carbon	Biological half-life (d): Body water	Biological half-life (d): Carbon	Ingestion dose: Coefficient (Sv Bq^{-1})[b]
3 mo	97	3	3.0	8	5.5×10^{-11}
1 y	97	3	3.5	15	4.1×10^{-11}
5 y	97	3	4.6	19	2.6×10^{-11}
10 y	97	3	5.7	26	1.9×10^{-11}
15 y	97	3	7.9	32	1.6×10^{-11}
Adult	97	3	10.0	40	1.6×10^{-11}

[a] Adapted from ICRP (1989).
[b] Effective dose (Sv) per Bq intake.

0.30–0.77 Bq L^{-1} for the Mississippi River) (Kaufmann and Libby 1954). The tritium levels of tap water (drinking water) in Japan, originating from rivers, lakes, or groundwater, now range from 0.3–3.6 Bq L^{-1} (Table 7).

When comparing tritium levels of rice and tap water in Akita City (Table 8) with those of all Japan (Table 7), the Akita levels are within the range of all Japan. Thus, levels observed at Akita could be considered to be similar to those of all Japan.

In Tables 7 and 8, tritium levels are expressed in tissue free water tritium (TFWT) and organically bound tritium (OBT): TFWT represents tritium activity in the "free water portion" of food (or tissue) in which free water is collected by drying a tissue. OBT is estimated in water, which is collected by oxidizing (or burning) the dried tissue. Specific activity ratio (SAR) is a ratio of OBT:TFWT and is assumed to be an indicator for the ability of the biological system to concentrate tritium into organic fractions (e.g., Moghissi et al. 1987). In addition, SAR could sometimes reflect a direct pathway from food OBT to tissue OBT in the system (Etnier et al. 1984; Travis et al. 1984; Takeda et al. 1985), or it could reflect longer retention of OBT fractions and quicker turnover of TFWT fractions in the biological systems (Myers and Johnson 1987).

To examine the "concentration" hypothesis, Moghissi et al. (1987) fed rabbits with alfalfa and water with a SAR of 1 for three generations over 3 y. Rabbits were sacrificed at various intervals. After estimation of TFWT and OBT in six organs, the calculated SAR was found to be 1 in each of six organs. The authors concluded that there is no evidence of tritium concentration in OBT under the "static equilibrium condition."

As shown in Table 8, TFWT in food samples ranged from 0.5–2.5 Bq L^{-1} while their OBT varied from 1.0–2.1 Bq L^{-1}. It is mentioned that tritium in free water of foods was easily influenced by atmospheric HTO (Inoue et al. 1991). This may be one reason for wider variation in TFWT concentration in foods. When pooled portions (daily foods in Table 8) of breakfast, lunch, and supper for 5–6 persons were analyzed, the total TFWT and OBT were similar to TFWT and OBT of three organs in humans. Also, the SAR of 1.2 in the daily foods was the same as three organs of humans. This seems to indicate that tritium in the environment, foods, and humans are nearly in equilibrium in Japan.

The total daily tritium uptake of the Japanese is 4.35 Bq, of which foods contribute 1.71 Bq, water vapor in air contributes 0.40 Bq, and drinking water contributes 2.25 Bq. This shows that tritium intake is 39% from food, 52% from drinking water, and 9% from air (Hisamatsu et al. 1987, 1989b; Okada et al. 1990).

If a total tritium uptake from Japanese food materials is taken as 100%, contribution of cereals constitutes 26% (rice 16%); vegetables 18%; milk and dairy products 14%; fruits 11%; and fish and meats 9% (Hisamatsu et al. 1989b; Okada et al. 1990).

An annual dose equivalent from tritium exposure can be calculated from tritium concentrations in various organs of humans by assuming that tritium in soft tissues is the major contributing factor (ICRP 1978). In an ICRP Reference Man (70 kg of body weight), soft tissue is 63 kg, in which body fluid (water) is 42 kg and other organic matters are 21 kg (ICRP 1975).

In Table 8, TFWT in human organs is 1.6 Bq L^{-1} and it's OBT is 1.8 Bq L^{-1}. Assuming density of soft tissue to be 1 kg L^{-1}, a total tritium content in soft tissues will be 1.6 Bq L^{-1} × 42 kg + 1.8 Bq L^{-1} × 21 kg = 105 Bq.

If 105 Bq of tritium is uniformly distributed in 63 kg of soft tissues, its tritium concentration will be 105 Bq/63 kg = 1.67 Bq kg^{-1}.

The dose equivalent for 1 y from decay of tritium at this constant concentration in equilibrium will be 1.67 Bq kg^{-1} × (28.8 nSv y^{-1}) Bq^{-1} kg^{-1} = 0.0481 μSv y^{-1}. Japanese people, in the present environment, would annually receive 0.048 μSv from tritium decay, which is 0.0034% of their annual natural radiation dose (1.4 mSv) (Abe 1989).

Table 7. Tritium in pine needles, polished rice, and tap water in all Japan.

Samples	TFWT[a] (Bq L^{-1})	OBT[b] (Bq L^{-1})	SAR[c] (average)	References
Pine needles	0.8–3.1	1.4–3.6	0.9–2.1 (1.7)[d]	Takashima (1991)
Polished rice	0.0–4.7	0.2–4.3	0.1–1.6 (0.83)[d]	Inoue and Iwakura (1990)
Tap water	0.3–3.6	—	—	Yamada et al. (1986)

[a] TFWT = tissue free water tritium.
[b] OBT = organically bound tritium.
[c] SAR = specific activity ratio = OBT:TFWT.
[d] Figures in parentheses are average values.

TRITIUM PROBLEMS

Radiation quality and dose rate dependency

Because few data on tritium-induced health injury in humans are available, risk estimation has to be based on animal and cellular experiments with tritium. Such experiments show that acute exposure to tritiated water induced carcinogenesis, fetus malformation, mutagenesis, animal death, or killing of hematopoietic stem cells. The injuries were qualitatively similar to injuries caused by low-LET radiations (e.g., ^{137}Cs or ^{60}Co gamma rays, and x rays). The quantitative difference between tritium beta and a standard low-LET radiation is expressed by relative biological effectiveness (RBE), an inverse ratio of the doses of two kinds of radiation that produce the same biological effect. RBE of tritium beta ranged mostly from 1 to 3 (Sawada and Okada 1990; Straume 1991; Straume and Carsten 1993), and are influenced by various factors (Okada et al. 1986): 1) different RBEs were obtained in the same tritiated-water treated cells, depending upon types of biological endpoints; 2) RBEs for the same endpoint (e.g., killing) were dependent upon cell types; 3) RBEs were sometimes dependent upon dose or dose rate; 4) RBEs for the same endpoint were dependent upon types of tritium compounds (e.g., HTO vs. T-thymidine) (e.g., Ueno et al. 1989); and 5) RBEs are dependent on type of a standard radiation (e.g., x rays, ^{137}Cs gamma rays, and ^{60}Co gamma rays).

RBE values have been a basis for assigning a radiation weighting factor, Wr (ICRP 1991b), which is a correction factor to each of a different quality of radiation, necessary for conversion of an absorbed dose (Gy) to a dose equivalent (Sv) for the purpose of radiation protection. Wr was previously called a radiation quality factor (Q) (ICRP 1978). When the latest ALI of tritium is derived (ICRP 1991c), Wr of 1 (ICRP 1978, 1991b) has been used for the tritium beta. It is added that a joint task group of ICRP and ICRU recommended a Q of 2 for tritium beta from microdosimetric considerations (ICRU 1986) and that radiobiological experiments gave a RBE of 1–3 (e.g., Sawada and Okada 1990). The question of what would be the most appropriate Wr value of tritium beta rays for radiation protection still remains to be decided.

Most radiobiological experiments with tritium have been carried out under acute exposure conditions using tritiated water. One area of study needed now is chronic exposure to tritium by workers in tritium-handling facilities. One of a few studies on this subject was done by Carsten (Carsten et al. 1989), who maintained mice in drinking water at 10×, 33×, 100×, ... of the MPC (1.11 kBq mL^{-1}) of HTO. At 10× MPC, no significant biological effects were found. However, when the HTO concentration was ≥33×, marrow stem cells were slightly reduced. At the higher concentrations of HTO, chromosomal damage in liver and micronuclei formation in red blood cells increased. Carcinogenesis in these mice has not yet been studied. From a literature survey, Ujeno (1983) suggested that RBE would increase with lowering dose rate. Because there are few experimental data available, further studies are definitely needed for assessing health risks from tritium under continuous exposure.

Complex exposures

In tritium-handling facilities and fusion research facilities, tritium in large quantities creates contamination problems on maintenance, clean-up operations, or incidental release situations. Even one of the least

Table 8. Tritium in foods and humans at Akita City, Japan.

Samples	TFWT[a] (Bq L^{-1})	OBT[b] (Bq L^{-1})	Daily uptake of tritium[c] (Bq d^{-1})	SAR[d]	Reference
Uncooked foods[e]					
Rice	2.5	1.8	0.279	0.72	Hisamatsu et al. (1987, 1989b)
Green vegetables	1.4	1.7	0.078	1.19	Hisamatsu et al. (1987, 1989b)
Other vegetables	1.0	1.8	0.229	1.75	Hisamatsu et al. (1987, 1989b)
Fishes/clams	0.49	1.0	0.055	2.11	Hisamatsu et al. (1987, 1989b)
Meats	1.3	1.8	0.094	1.33	Hisamatsu et al. (1987, 1989b)
Eggs	1.7	2.1	0.067	1.20	Hisamatsu et al. (1987, 1989b)
Milk/dairy products	2.0	2.1	0.241	1.04	Hisamatsu et al. (1987, 1989b)
Tap water	1.9	—	2.25[f]	—	Hisamatsu et al. (1987, 1989b)
Daily foods[g]	1.7	1.9	4.2	1.19	Hisamatsu et al. (1987, 1989b)
Human					
Brain	1.6	1.7	—	1.1	Hisamatsu et al. (1989a)
Lung	1.6	1.9	—	1.3	Hisamatsu et al. (1989a)
Liver	1.6	1.8	—	1.1	Hisamatsu et al. (1989a)

[a] TFWT = tissue free water tritium.
[b] OBT = organically bound tritium.
[c] SAR = specific activity ratio = OBT:TFWT.
[d] The table of daily uptake (g) of each food sample from the National Nutrition Survey (Ministry of Health and Welfare 1987) is used to obtain daily tritium uptake (Bq d^{-1}).
[e] Uncooked but edible portion of food materials are used for tritium measurement.
[f] Okada et al. (1990).
[g] Daily foods (from breakfast, lunch, and dinner) consumed by 5–6 persons are collected and analyzed. These exclude beverage taken between meals.

radiotoxic forms, tritium gas (T_2, DT, or HT), could be converted catalitically or enzymatically to HTO with a radiotoxicity 25,000 times higher than elementary tritium (Table 4) (ICRP 1978). Furthermore, tritium gas would be absorbed into various surfaces to form "tritium surface contamination" (TSC) (Carmichael and Haynes 1991), which would be transferred upon human contact (Eakins et al. 1975).

In a fusion research facility (Stencel et al. 1989), radiation protection problems are reported not only from elemental tritium, HTO, and TSC, but also from tritium-contaminated powders (e.g., graphite), and HTO-contaminated vacuum pump oil. In addition, neutrons, radiations from neutron-activated products of materials used for the machines, and bremstrahlung are also problems of radiation protection. Moreover, nonionizing radiations (e.g., magnetic fields, electric fields, and high-frequency waves) of extreme intensities may become another problem (IRPA/INIRC 1985, 1988, 1990). Here, exposures would occur not only from various combinations of tritium forms but also from tritium with other ionizing and/or nonionizing radiations. Although regulatory limits (i.e., ALI, DAC, or effective dose limits) have been assigned only to each specific radiation source (e.g., elemental tritium or tritiated water), organizations need to be prepared for situations of complex exposures.

Oxidation of elemental forms of tritium in humans and its dose contribution

When a human is exposed to pure tritium gas, exposure would occur on the skin from HT in the air, in the lung from HT of inhaled air, on the whole body (soft tissues) from HT dissolved in body fluids, and from HTO in body fluids, which is the oxidation product in the gut from the dissolved HT. When deriving its regulatory limit, the ICRP (1978) considered the following: 1) to neglect the skin exposure because short tracks of tritium beta are not capable of reaching basal cells of the skin; 2) to neglect the HT exposure in body fluids because its contribution is only 1% of the lung dose; and 3) to neglect HTO exposure on soft tissues by assuming its minor contribution. Thus, ICRP (1978) obtained a DAC of 2×10^{10} Bq m^{-3} for elemental tritium on the basis of the lung exposure to HT, alone.

Peterman (1982) set up a modeling of HT gas metabolism and obtained its parameters by fitting the human data (Pinson and Langham 1957). From these calculations, it was found that the whole-body exposure to HTO from gut-oxidized HT is as important as the lung exposure and that DAC is estimated to be 1.08×10^{10} Bq m^{-3}; one-half of what ICRP suggested. The formation of HTO in body fluids from HT inhalation could be detected by the urinary HTO analysis.

The oxidation of HT by intestinal bacteria was first demonstrated in rats by Smith et al. (1953). Their findings of decreased HT fixation in the eviscerated rats or rats treated with antibiotics were further evidence for bacterial involvement in HT oxidation (Smith et al. 1953). Intestinal bacteria with HT oxidation capacity have been studied further (Ichimasa et al. 1989b). In human feces, 63 strains were identified as belonging to families of *Veillonella, Fusobacterium, Escherichia, Bacteroides,* and *Eubacterium.* In rats, 27 strains were identified as being mostly of *Bacteroides* and *Clostridium.* Fecal suspensions of rats, four strains of monkeys, and humans were found to oxidize HT from seven to 20 times more efficiently under anaerobic conditions than under aerobic conditions.

In order to decrease intestinal HT oxidation (Ichimasa et al. 1989c), the administration of Clindamycin (an antibiotic) or Norfloxacin (an antibacterial drug) for 4 d before HT exposure was found to be effective in reducing oxidation by 1/3–1/5 of those of nontreated animals. When these drugs were given to rats after HT exposure, Clindamycin was effective in reducing the biological half-life of tritium to 3/5, but Norfloxacin was not.

It is emphasized that inhaled HTO could be fully incorporated into the human body, while HT would be incorporated only 1/15,000 less efficiently than HTO (Pinson and Langham 1957). Thus, if HTO were present at more than 0.007% of HT as a mixture in air, the dose contribution of the HTO fraction would become greater than that by HT inhalation.

Prevention of HTO intake and enhanced removal of HTO from humans

Biological half-lives of HTO in humans are made up of three components. The first component (T1) is about 9 d, the second (T2) is about 30 d, and the third (T3) is about 450 d (Table 1) (NCRP 1979). The first component would be similar to that estimated from water balance of the Reference Man (ICRP 1978) $T = 0.693 \times$ (body fluid: 42 kg)/(daily water intake: 3 kg) = 9.7 d. Thus, the T1 component would be most likely to be a reflection of water metabolism.

Since longer retentions of tritium were found in OBT fractions of animal experiments (Thompson and Ballou 1954; Takeda and Kashida 1979), the longer half-lives of humans might similarly reflect their organically bound (carbon-associated) hydrogens. When a half-life is estimated from the carbon balance of the Reference Man (ICRP 1989), it gives $T = 0.693 \times$ (total carbon content: 16 kg)/(daily carbon uptake: 0.3 kg) = 37 d. This is of similar magnitude to the half-life of the T2 component.

Much longer half-lives were shown in collagen fractions and lipid fractions in fatty tissues and brain in animal experiments (Thompson and Ballou 1954; Takeda and Kashida 1979). Thus, the T3 component of longer half-life would be likely to reflect these fractions.

Under acute HTO exposure, the first component would contribute about 90% of the total dose while the second and the third would be about 10% (ICRP 1978). Myers and Johnson (1987) set up a computer model for more precise dose estimation by introducing corrections for nuclear water content and exchangeable hydrogen and OBT, together with their uncertainties involved.

The T1 component varies from 4–18 d among individuals with a typical value of 10 d (ICRP 1978). Similarly, Japanese data (Akaishi et al. 1984) also showed a wide variation from 5–17 d with an average of 9.4 d. This implies that a radiation dose of an exposed individual would be best calculated from that person's estimated biological half-life.

In the Chernobyl accident, the general public was involved. This shows that the ALI and DAC of tritium should not be limited to the workers, but be extended to the general public of all age groups (ICRP 1989).

Since HTO can easily be taken into the body through respiration, the skin, and ingestion, it needs to be handled with care. For example, HTO (in liquid form) is always vaporizing HTO into the air and use of a "protective hood and other protective devices" would be suggested (Morikawa et al. 1989). Furthermore, it is recommended that the workers who handle large quantities of tritium be routinely monitored. Tritium in body fluids can be monitored either with urine or with exhaled air samples.

In Japan, a vapor collector which traps vapor, from 10 L of exhaled air, into a scintillation vial has been developed. The vials are subjected to liquid scintillation counting with a detection limit of about 2.2 Bq mL^{-1} (Morikawa et al. 1989). It is added that one of the monitoring methods to be developed may be that for estimation of OBT fraction in humans.

When a person is contaminated with tritium, the NCRP (1980) recommends 1) oral intake of fluid (e.g., water, fruit juice, tea, coffee, or beer) at 3–4 L d^{-1}, or 2) instillation with 5% glucose or saline under a doctor's care, together with daily urinary monitoring.

When HTO-exposed rats were treated with daily instillation for 40 d (Ichimasa et al. 1989d), a combination of sodium acetazolamide (a diuretic) and Ringer's solution was the most effective in decreasing the biological half-life to one-third of that of nontreated rats. It is interesting to note that the tritium activities of soft tissues in the treated animals were decreased to 1/10 of the nontreated rats. In mice injected with 811 MBq of tritiated water, 80% of animals without treatment died, while none did if they were given a combined treatment of furosemide (acetazolamide) and 5% glucose (Kunugita et al. 1990). In humans, Lloyd et al. (1986) described the effectiveness of forced diuresis by use of isotonic saline with 5% glucose, 20 mM potassium-chloride and furosemide on a worker contaminated with about 35 GBq tritium.

Uptake of HTO through skin could be prevented by protective coats or gloves, and partially by a hand cream on the skin. Wounds (e.g., incision, abrasion, thermal burn, or acid burn) on the skin of hairless mice were found to accelerate intake of tritiated water or vapor by 2 to 20 times compared to skin without a wound (Sawada 1989).

Biological effects of low levels of tritium

Biological effects of tritium in low levels should be explored in the future from the standpoint of risk assessment. Here, two areas of the recent studies, adaptive response and epidemiological studies, will be described.

Adaptive response

Olivieri et al. (1984) found that when human lymphocytes were cultured with ^{3}H-thymidine and then acutely exposed to x rays, x-ray-induced chromosome aberrations were one-half that of x rays alone without ^{3}H-thymidine pretreatment. This was named "adaptive response" (Wolff 1992).

The adaptive response of lymphocytes was found to be induced not only from pretreatments with ^{3}H-thymidine, but also with x rays (1–3 cGy) (Olivieri et al. 1984; Wiencke et al. 1986), ^{60}Co gamma rays, HTO, ^{14}C-thymidine, and ^{32}P-orthophosphate (Ikushima 1989, Sankaranarayanan et al. 1989). The lowest induction concentration of ^{3}H-thymidine was 0.0185 kBq mL^{-1}, where an average of one disintegration per labeled cell would occur. The highest concentration was 37 kBq mL^{-1}, which produced many aberrations by itself (Wiencke et al. 1986). The induction concentration of HTO was 185 kBq mL^{-1}. In Chinese hamster V79 cells, the range of the induction concentrations of ^{3}H-thymidine ranged from 0.37–1.85 kBq mL^{-1} (Ikushima 1989).

The adaptive responses were expressed in reduction of chromatid aberrations (Olivieri et al. 1984), of induced mutation frequency (Sanderson and Morley 1986) in human lymphocytes, and also, in decrease of micronuclei formation and sister chromatid exchange in Chinese hamster V79 cells (Ikushima 1989).

The response required a time delay for induction after the pretreatment and was expressed for a limited time period (Ikushima 1989; Wolff 1989). The response was abolished by addition of 3-aminobenzoate [an inhibitor of poly(ADP-ribose) polymerase] (Wiencke et al. 1986; Ikushima 1989) and by cycloheximide (a protein synthesis inhibitor) (Youngblom et al. 1989). The cells that received the pretreatment were found to synthesize new proteins detectable in 2-dimensional electrophoresis (Wolff 1989). These findings suggest that the pretreatments with a low dose of tritiated compounds or other inducers would initiate synthesis of one or more repair enzymes as a cellular response to stress. The enzymes, then, would partially repair the chromosomal damage from a subsequent challenging dose of radiations, resulting in reduction of chromosomal damage.

How the adaptive response would contribute to the reduction of health risks of tritium remains to be explored.

Epidemiological studies

Health risks of tritium should be assessed on humans, if possible. Two epidemiological studies available

purification is all that is required. If tritiated organics are to be measured, however, the organics have to be converted to water for analysis.

Once a sample is collected, precautions must be taken to prevent exchange between atmospheric water vapor and the sample. Samples should always be stored in sealed containers. Since HTO will diffuse through plastic, glass or metal containers are recommended for long-term storage. However, plastic containers are acceptable for short-term storage (up to a few months). The bottle cap seals should always be checked.

Water recovery

The methods outlined in NCRP Report No. 47 (NCRP 1976), lyophilization, distillation, azeotropic distillation, and dilution are still the most prevalent methods of recovering tissue-free water from environmental samples. For all of these methods, the sample size required depends on the water content of the sample and the amount of water required for analysis. Assuming the water sample is analyzed by liquid scintillation counting, a 5–10-mL sample is ideal. Larger sample sizes are required if the sample is to be electrolytically enriched (see the section on electrolytic enrichment later in this paper).

Lyophilization. Lyophilization (or freeze drying, as it is more commonly known) consists of freezing the sample and then recovering the water under vacuum. The water is recovered in a cold finger located between the vacuum pump and the sample. The disadvantage of this technique is the time required. Several days may be required to obtain enough water from environmental samples for low-level analysis.

Distillation. Distillation is a common method of recovering water from samples. The sample is simply heated and the distillate is collected on a cold finger. If accurate measurements are required (within 5 to 10%), isotopic fractionation should be considered. The possibility of isotopic separation can be eliminated if the sample is completely distilled, but this often results in contamination of the distillate by volatile pyrolytic products from any organic residue.

Azeotropic distillation. Azeotropic distillation is a convenient method of recovering water from vegetation, soil, urine, and animal tissue. In this technique, the water-containing material is placed in an immiscible hydrocarbon, such as toluene or xylene, and the water is boiled out as a constant boiling mixture of fixed water-hydrocarbon composition. The distillate is collected in a receiver where the phases separate and the hydrocarbon returns to the distillation flask. When taken to completion, there is no isotopic fractionation and the distilled water is free of any constituent that would interfere with liquid scintillation counting. The disadvantage of the technique is that the dried organic residue must be free of hydrocarbon if an organically bound tritium measurement is desired on the same samples.

Dilution. Dilution of a sample with a solvent is a simple way to recover HTO. The solvent can simply be tritium-free water. If left long enough, the water in the sample will come into equilibrium with the added water. This exchange can be accelerated by adding heat. Enough solvent should be added to the sample so that the fraction of HTO remaining in the sample is small compared to the amount of HTO in the solvent. The disadvantage of this method is that the sample is diluted by the solvent. This technique is usually reserved for samples with tritium concentrations significantly above background levels.

Urine

Tritium bioassay is used routinely to assess tritium doses received by workers. Liquid scintillation counting of a worker's raw urine is sensitive enough to detect tritium concentrations of any significance for radiation protection purposes. However, in some applications, such as monitoring environmental transport of tritium in an ecosystem, lower tritium-in-urine concentrations need to be measured. In such applications, the reduction of counting efficiency resulting from chemical and color quenching of the raw urine is limiting. Activated charcoal filters have been used to remove the organic fraction (Eakins et al. 1975; Stencel et al. 1988), greatly reducing the sample quench. The azeotropic distillation method previously mentioned is also suitable for reducing the quench in urine samples.

Organically bound tritium

Once the tissue-free water tritium (TFWT) has been removed from a sample, tritium may still exist in the sample in the form of OBT. The OBT can be recovered from the dried sample by combustion. Moghissi et al. (1975) described a method in which the sample is placed in a 2-L steel Parr[1] bomb, pressurized with oxygen to about 1.5–2 MPa and ignited. The bomb is then evacuated through a cold trap to collect the water. This method has the advantage that it is very rapid, permitting quicker turnaround times than conventional combustion methods, and the water obtained usually does not require further purification. It is limited to about 10 g of dried organic material. Larger samples must be combusted by the conventional technique in a quartz tube with carefully controlled oxygen flow (Hisamatsu et al. 1990a).

The water sample obtained from combustion usually requires purification by refluxing with $KMnO_4$ and/or Na_2O_2, distilled to obtain a pure sample for counting (Hisamatsu et al. 1990a). In a recent interlaboratory comparison of OBT measurements (Hisamatsu et al. 1990b), good agreement was found between all six laboratories participating. However, although the same general procedures were used (combustion followed by purification and distillation), the details were not the same for any of the six laboratories.

[1] Parr bomb, Parr Instrument Company, 211 53rd Street, Moline, IL 61265.

Electrolytic enrichment

One of the limiting parameters of liquid scintillation counting (LSC) and internal gas counting is the amount of sample that can be put into the counting system. When the tritium concentration of a water sample is too low for these methods, isotopic enrichment can be used to concentrate the tritium. Electrolytic enrichment factors of up to 100 are obtainable with two-stage electrolysis. This technique, however, is only suitable if large sample volumes are available (Brown and Grummitt 1956; Östlund and Werner 1962; Östlund et al. 1964; Taylor 1981; Kakiuchi et al. 1991).

MEASUREMENT OF TRITIUM IN WATER

Liquid scintillation counting of discrete samples

The most common procedure for measuring tritium in water is liquid scintillation counting of discrete samples. The water samples to be analyzed are mixed with a liquid scintillation solution (cocktail) in a scintillation vial. Standard scintillation vials have a capacity of 20–24 mL, but smaller vials ("mini vials") are also used if sensitivity is not a problem. Typical water:cocktail ratios between 1:5 and 1:15 are used in routine applications; however, for low-level counting, special cocktails are used that permit ratios of up to 10:10. In a liquid scintillation counter, the prepared sample is placed between two photomultiplier tubes. The beta radiation in the sample interacts with the scintillant, resulting in several light photons being eventually emitted. The summed height of the pulses from the two photomultipliers is proportional to the energy of the beta particle. These counters are run in coincidence mode in order to distinguish photon pulses from photomultiplier noise. Most LSC counting systems are based on commercially available counters using commercially available cocktails.

Typical performance of a regular liquid-scintillation counter is a background of 10–15 cpm with a tritium counting efficiency in water of 35 to 55%, depending on sample composition and sample size. The corresponding detection limit is on the order of 200 Bq L^{-1} for a 1-mL water sample counted for 10 min.

Recent developments in background correction and compensation techniques made possible through improved electronics have greatly improved the performance of commercial liquid scintillation spectrometers (Schönhofer and Henrich 1985; Rozanski et al. 1991b).

Chemiluminescence in a sample prepared for liquid scintillation counting results in single photon events that can be detected by the scintillation counter. Although coincidence counting greatly reduces the possibility of these events being mistaken as tritium events, some random coincidences of these single photon events will occur. Circuits to detect and correct for random coincidences are available for many commercial liquid scintillation counters. Although the details of how instrument manufacturers accomplish chemiluminescence correction vary, they are all based on determining the random coincidence component in the tritium channel and subtracting it from the total signal to obtain the true tritium signal.

Tritium decay in a normal liquid scintillator results in single pulses with a pulse width of a few nanoseconds. When a cosmic-ray particle passes through the sample in the liquid scintillation counter, several smaller "after-pulses" follow the initial pulses resulting from the interaction. Canberra Packard[1] uses a burst counting technique (Roessler et al. 1991) to detect the number of after pulses, referred to as the "pulse index," and only accepts pulses with a low pulse index, effectively discriminating against interferences such as cosmic-ray particles.

The Wallac 1220 Quantulus liquid scintillation counter,[2] designed for very low-level counting, uses a combination of passive and active shielding around the sample detector assembly, in addition to a pulse-shape discriminator and a pulse-height comparator (Kaihola 1991). The passive shielding consists of a copper-lined, 630-kg (considerably more massive than that used in conventional LSCs) lead shield. Between this shield and the sample is the active shield, consisting of a cavity filled with a liquid scintillator and a photomultiplier tube on either side. Single photons are detected directly and multiphoton events are detected with coincidence circuits, both in the active shield and in the sample. In addition, events that cause multiphoton events in the active shield and the sample at the same time can be detected (a third coincidence circuit) and subtracted from the tritium channel.

Automatic optimization, in which the counter determines the optimum window settings (energy windows) for the best figure of merit, is available on some machines. This feature eliminates the time-consuming and tedious task of determining optimum window settings by repeated measurements.

These are just a few representative features available on some modern liquid scintillation counters. More complete information can be obtained from manufacturers of specific products. Although some of these techniques have been known for some time, it is only over the last several years that many of them have become available on commercial instruments.

Background count rates as low as 0.4 cpm with a counting efficiency of 28% have been reported for an 8-mL water sample, corresponding to a detection limit of 0.65 Bq L^{-1} for a 500-min count time (Schönhofer

[1] Canberra Packard, Packard Instrument Company, One State Street, Meriden, CT 06450.

[2] Wallac 1220 Quantulus liquid scintillation counter, Wallace Oy, P.O. Box 10, SF-20 101 Turku, Finland.

creased by a factor of 1.5 to account for skin absorption from tritiated water vapor.

No significant skin absorption of HT in air occurs. Studies by Hutchin and Vaughan (1965), Eakins et al. (1975), and Johnson et al. (1988) have shown that if skin comes into contact with surfaces that have been exposed to high concentrations of HT gas, significant uptake and retention of tritium occurs in the skin, as well as in other organs. The processes by which this occurs are not fully understood.

Distributional effects

Even though tritium can enter the human body by several different routes, the subsequent behavior of the tritium will depend more on its chemical form than its mode of entry. The two main forms of tritium to consider are water and organic molecules. Tritiated water rapidly diffuses across all cell membranes and quickly equilibrates with the body fluids, attaining a uniform concentration throughout the body; however, tritiated organic molecules can be localized in relatively small numbers of cells at relatively high concentrations. The mean range of the tritium beta particle, 0.68 μm, is small compared to the dimensions of cells. Consequently, while the average dose to the tissues may be very low, the dose to cells where the tritium compound is concentrated may be large.

When tritium is homogeneously distributed throughout the body, such as for HTO, the decay of tritium will result in uniform irradiation to the body. As HTO, irradiation of the body from tritium will be similar to that received from external irradiation. If the HTO is present in concentrations that will deliver an absorbed dose of a few gray or more over a short time period, many aspects of the acute radiation syndrome, similar to those seen for external irradiation, may appear (NCRP 1979b).

For radiation protection purposes, inhomogeneous distribution of tritium at the subcellular level as a result of exposure to HTO is generally not considered (Saito and Ishida 1988); however, localization of other tritiated compounds occurs in certain cells or tissues and may cause biological effects that do not resemble those seen with HTO. An inhomogeneous concentration of tritium will generally result in doses to the involved cells that are larger than the average tissue dose, which may be negligible. As a result, the concept of average tissue dose for many tritiated compounds especially (especially those involved in nonexchangeable bonds) would be inappropriate and a poor estimate of the hazard associated from exposure to such compounds, especially if the exposure results in the tritium being incorporated into sensitive regions of the cells, such as DNA.

Several studies have looked at the effects of tritiated thymidine since it is a DNA precursor. There has been concern that DNA precursors containing tritium would lead to selective deposition of tritium in the cell nucleus. A number of studies have shown that the distributional effect of tritium incorporated into DNA results in selective irradiation of only part of the cell (the nucleus) and that the resulting risk of any biological effect to the cell might be greater than tritium incorporated more uniformly throughout the cell (Bond and Feinendegen 1966; Zhuralev and Kalyazina 1979). When tritiated thymidine[1] was injected into humans, 40–60% was found to be metabolized into HTO, with the remainder being incorporated into DNA. The rate of incorporation of tritiated thymidine into DNA will depend on the cell turnover rate; i.e., the higher the cell turnover rate, the higher the rate of DNA synthesis, resulting in a higher rate of tritiated thymidine incorporation into DNA. Inhomogeneous distribution of the tritiated thymidine within the body will result since only those cells undergoing DNA synthesis will assimilate tritiated thymidine.

Transmutational effects

Transmutational effects on molecules can result from the incorporation of tritium. Tritium decay can induce mutations in DNA molecules by two possible mechanisms: 1) the ionization from the emitted beta particle; and/or 2) by transmutation of the ^{3}H to ^{3}He with an associated energy of excitation and recoil (Halpern and Stocklin 1976; NCRP 1979b; Feinendegen et al. 1980; Carsten 1986). While the biological effects of such transmutations in DNA might be easily detected, it requires very accurate microdosimetry to be able to separate the observed effects of ionizations from the beta particle and those seen for transmutation (NCRP 1979b). Since the maximum recoil energy associated with this transmutation is only 3 eV, it is thought to be too small to break most chemical bonds and is generally ignored (NCRP 1979b). Helium atoms which replace the transmutated hydrogen atoms have extremely weak bonding with carbon. This instability leads to dissolution of the carbon-helium bond, yielding free helium atoms and reactive carbon ions. It is the resulting carbon ions which can cause molecular alterations in the DNA molecule, presenting as single-strand breaks (Krasin et al. 1973; Krasin et al. 1976a) or interstrand cross-links (Krasin et al. 1976b), depending on their location within the DNA molecule. The NCRP (1979b) supports the conclusion that energy deposition from beta radiation of most nuclides in nucleic acids in humans significantly outweighs the damaging contribution from transmutation effects.

METABOLIC MODELS OF TRITIUM

As an isotope of hydrogen, tritium can enter a biological system in the form of many different chemical compounds. For most types of tritium exposure described in this paper, two chemical forms of tritium (HTO and OBT) contribute the majority of the radiation dose to the body regardless of whether they were taken directly into the body or enter into *de novo* synthesis of other tritiated compounds (Taylor et al. 1990). With HTO playing a primary role in most metabolic processes associated with tritium, the accuracy of the risk assessment from exposure to tritium will largely be dependent on the accuracy of the tritiated water dosimetry. Fortunately, the dosimetry of HTO is relatively straight-forward and is based on simple, scientifically defendable assumptions about water distribution in the body. Consideration must also be given to the metabolic fate of elemental tritium gas, organically bound tritium, biotransformed tritium in foodstuffs, and the age-dependent effects.

As a precursor to the development of a mathematical model, a conceptual model of the metabolic components and the relationships of tritium transfer between them needs to be developed first. The metabolic components are referred to as compartments, which represent reservoirs of tritium within the body, such as the respiratory tract, the GI tract, or the soft tissues. In a conceptual model, the transfer from one compartment to another is specified by a mathematical relationship that represents this transfer as a function of time. For instance, in cases where tritium is continuously taken into the body and eliminated (i.e., when drinking contaminated water or breathing contaminated air over a considerable time period), tritium will accumulate within the body. If elimination rates are about equal to intake rates, then a state of equilibrium exists. The amounts within the body may increase if intake rates are greater than elimination rates (Quimby et al. 1970). A decrease of activity in any organ can result either by physiological elimination of the compound containing the radionuclide or by physical decay of the radionuclide. If properly developed, the conceptual model can be solved using exact analytical methods or numerical approximations to provide an estimate of concentrations in the defined compartments as a function of time.

The metabolic models discussed in the following sections represent the conceptual interpretation of metabolic processes for a variety of different forms of tritium including HTO, HT gas, HT-contaminated surfaces, OBT in foodstuffs and other organic compounds, metal tritides, tritiated methane, and radioluminous paints.

HTO metabolism

There is a general consensus that the current understanding of the metabolism of HTO, which is the result of many studies of HTO metabolism, is sufficient for radiation protection purposes. The detailed knowledge of HTO metabolism has led to the development of models which result in acceptable estimations of absorbed doses from tritium concentrations seen in the urine (Johnson 1976, 1982).

The greatest majority of tritium in the atmosphere exists in the form of HTO, which can be transferred to humans primarily by inhalation, skin absorption, or ingestion of drinking water or food (ICRP 1979–1982; Belloni et al. 1983). The kinetics of HTO in the body follow that of body water, except for a small portion of metabolized tritium that becomes bound to organic molecules. HTO is excreted in urine, feces, sweat, and breath (NCRP 1979b).

If individuals are at rest at the time of exposure to HTO vapor, the amount of tritium entering the body by inhalation will be approximately equal to the amount entering by skin absorption (Pinson and Langham 1980); however, physical activities during exposure will result in approximately twice as much tritium entering the body by the inhalation pathway (Myers and Johnson 1991). In addition, some endogenous HTO may be formed following metabolic breakdown of various tritiated compounds (e.g., inhaled HT gas) as a result of metabolic combustion of OBT, or following ingestion of foodstuffs or water containing OBT. In normal adults, $\simeq$70% of the total body water is from ingested water and the rest originates from either water in food or from catabolized organic molecules (van Den Hoek et al. 1980).

HTO exposure is generally the most important to consider since HTO quickly reaches equilibrium with hydrogen in the body and imparts a uniform dose to all soft tissues. The ICRP (1979–1982) recommended the assumption be made that internalized HTO is completely and instantaneously absorbed and mixed rapidly with the total body water so that at all times, the concentration in sweat, sputum, urine, blood, insensible perspiration, and exhaled water vapor is the same. Following HTO exposures, a small fraction of tritium will eventually become incorporated into organic molecules. The OBT will have different metabolic rates than those for total body water, therefore, the retention of tritium following an exposure to HTO should be described by more than one component, *viz.*, one component that is characteristic of the retention of body water and one or more components that are characteristic of the retention of hydrogen (or tritium) in organic molecules or OBT (Johnson 1982; Taylor et al. 1990).

In the past, urine concentrations were used to directly obtain an estimate of the whole-body dose based on the argument that the time-integrated tritium concentration (which is proportional to accumulated dose) in any tissue would not exceed that in the body water (or urine) (Sanders and Reinig 1968; Snyder et al. 1968; Osborne 1972; NCRP 1979b); however, this procedure has been reevaluated following the publication of ICRP recommendations regarding the concept of E_c (ICRP 1991). The ICRP Committee 2 recommended that instead of using the total body water, the average soft tissue should be used to estimate the E for HTO exposures and that only retention in total body water be considered in dose estimation (ICRP 1979–1982). In other words, the dose contribution from OBT was to be ignored. This recommendation was based on results of several studies (Sanders and Reinig 1968; Snyder et al. 1968; Moghissi et al. 1972) which indicated that OBT contributed <10% of the E_c in the Reference

[1] Tritiated thymidine is sometimes denoted as ^{3}HTdR.

assumed to go directly to urine, and 50% of the HTO in body fluids was assumed to be excreted by pathways other than urinary excretion. The model predicts that the integrated amount of OBT in skin at the point of contact is ≈0.5 Bq d^{-1} Bq^{-1} of intake, and 40% of the intake is excreted as OBT. For a contaminated area of skin of 20 cm^2, the skin dose would be ≈2 × 10^{-8} Sv. This would result in a dose conversion factor of 5 × 10^{-8} Sv Bq^{-1} of OBT excreted (assuming a contaminated skin area of 20 cm^2), which is sufficiently large that the skin dose may be limiting in certain situations (McElroy et al. 1989). (Note: Two areas of contaminated skin of different sizes can have the same skin dose but different resulting E. The skin section having the largest area will have more associated OBT, thereby contributing to a higher E.)

OBT metabolism

Little is currently known of the metabolism of organically bound tritium in humans. OBT can enter the body directly by ingestion of tritiated foodstuffs, by inhalation of volatile organic vapors or aerosols, or can be formed *in vivo* from tritium that is present in the general body pools after exposure to other tritium-containing compounds (Diabaté and Strack 1993). Once in the body, OBT can be incorporated into a variety of biochemical compounds such as amino acids, sugars, proteins, starches, and other structural materials (Saito and Ishida 1989).

With such a diversified array of biochemical compounds associated with OBT and each having unique metabolic processes associated with it, uniform distribution or retention of OBT compounds cannot be assumed. For instance, tritiated compounds may have a shorter retention time than HTO (Osborne 1966); however, much of the tritium may be in exchangeable sites, and when these compounds are catabolized, HTO will be produced. Tritiated nucleic acid precursors generally have a longer retention than HTO even though much of the incorporated tritium may exist at exchangeable sites or be catabolized (ICRP 1979–1982). Generalities can be made based on how easily tritium exchanges with hydrogen in the hydrogen pool in the body. Organic molecules, in general, have slower turnovers and are retained in the body for longer periods of time than water. Tritium found in nonexchangeable organic binding sites (i.e., tritium bound to carbon atoms) will contribute an increased average soft-tissue dose over that seen for HTO alone, primarily due to the longer retention times in the body (ranging from hours to years) and inhomogeneous distribution among the different organs (Taylor et al. 1990). The increment of this increase in dose will vary depending on the chemical nature and metabolism of the molecule containing the tritium-carbon bond. In contrast, the specific activity of tritium in exchangeable hydrogen binding sites (≈30% of total hydrogen in organic molecules within the body) should be identical to that for hydrogen in body water and, thus, will have metabolism essentially the same as for HTO.

Information gained from human studies indicates that the retention of tritium can generally be characterized by a three-component exponential function having biological half-lives of 10 d, 21–76 d, and 280–550 d. As previously described, the first component represents body water metabolism while the latter two components generally correspond to OBT. The second component may be related to tritium involved in carbon-tritium bonds of organic molecules since it is eliminated with approximately the same half-life as carbon, namely 40 d (ICRP 1979–1982; Taylor 1990). The latter component may represent tritium incorporated into organic molecules having very slow turnover rates.

The fate of OBT after an intake is dependent primarily on the hydrogen metabolism of the human body. Hydrogen accounts for 60% of all atoms in the human body, as described for the Reference Man (ICRP 1975). The possibility of tritium substitution is of major interest because every day 2.1 × 10^{26} hydrogen atoms (5% of all hydrogen atoms in the body) takes part in metabolic processes (ICRP 1975). The net exchange of tritium with hydrogen always occurs toward lower concentrations and is dependent on the type of chemical bond. While OBT has been separated into two general forms, namely exchangeable OBT (i.e., tritium bound to nitrogen, oxygen, phosphorus, or sulfur) and nonexchangeable OBT (i.e., tritium bound to carbon atoms), the proportion of tritium entering into each form is highly variable (Rochalska and Szot 1977; Pietrzak-Flis et al. 1978; Kowalska 1985). It should be noted that the division between exchangeable and nonexchangeable forms of OBT implies a simplification that does not reflect the wide spectrum of organic bonds that are possible.

Tritium in exchangeable OBT is assumed to rapidly exchange with hydrogen found in the body water pool and, thus, will have a metabolism and distribution similar to that of HTO. After equilibration with the body fluids, the exchangeable tritium will be eliminated with a half-life of ≈10 d (ICRP 1979–1982). Nonexchangeable organically bound tritium is firmly bound to carbon atoms and is usually released only during enzyme-mediated reactions (Etnier et al. 1984). The rate of the metabolic breakdown may be fast for certain molecules and very slow for others; the retention times may range from hours to years (Taylor et al. 1990). Analysis of observations of tritium elimination following accidental inhalation or ingestion of tritium has shown that up to three exponential components have been used to describe the retention curves (see Table 2). Besides the HTO biological half-life, two additional exponential components with longer biological half-times were used with ranges of 21–76 d and 280–550 d.

Different types of exposure to tritium will result in different proportions of OBT formed. Following HTO exposures, OBT compartments contribute on the order of 10–20% of the whole-body $H_{c,T}$ that would be seen for HTO alone (Johnson 1982; Etnier et al. 1984; Dunford and Johnson 1987). Following inhalations of HT gas, OBT results from metabolic processes associated with the HTO formed by internal oxidation of HT (Peterman 1985a) and provides a small contribution to the whole-body $H_{c,T}$. With HT inhalations, it is important to note that the dose ratio of HTO to OBT will be similar to that seen for HTO exposures since all OBT will have resulted from HTO metabolism. In contrast, skin contact with HT-contaminated surfaces can result in direct formation of OBT in the skin (Johnson and Dunford 1985; Johnson and Peterman 1987; Johnson et al. 1988; McElroy et al. 1989). In all of these cases, the dose due to OBT distributed throughout the body will be some fraction of the whole-body $H_{c,T}$. However, ICRP 30 recommendations indicate that OBT contributions to dose should be neglected in dose estimation for both HTO exposures and HT exposures since it is expected to be <10% of the total.

OBT from foodstuffs

When dose assessments for tritium exposures are performed, the fact that OBT can be directly incorporated from foodstuffs has generally been ignored since little was known about this exposure pathway for tritium (Travis et al. 1984). However, recent reports using advanced environmental tritium models indicate that the ingestion doses from OBT in the food chain may be 4–23 times higher than doses resulting from inhalation and skin absorption of HTO (Gulden and Raskob 1992). Diabaté and Strack (1993) have provided an in-depth review of the current knowledge of OBT formation and translocation in plants in this issue.

Tritium in foodstuffs as OBT is known to be mostly in a nonexchangeable form (Konig 1990). OBT from foodstuffs in the diet can be used as metabolic precursors that will be directly incorporated into organic metabolic pathways or can undergo degradation to HTO and enter the general body water pool (Etnier et al. 1984; Travis et al. 1984). The relative proportions of exchangeable and nonexchangeable OBT in various components of the human diet vary greatly due to the influence of many different factors such as the history of exposure to tritium during food growth, the differences between plants and animals, and differences in wet and dry food. The nonexchangeable-to-exchangeable OBT ratios in foodstuffs reported in the literature have a wide range of values. For instance, Bogen et al. (1973) have reported that the tritium concentration in nonexchangeable components of food from New York was about twice that of exchangeable components and water components. Belloni et al. (1983) reported even higher ratios in Italian food (i.e., nine times more nonexchangeable tritium). The difficulty in defining a realistic range for the nonexchangeable-to-exchangeable OBT ratio has hampered the development and use of general metabolic modeling of OBT turnover in humans (Ueno and Kawamura 1975).

Etnier et al. (1984) have proposed a four-compartment model (Fig. 5) that is based primarily on hydrogen balance within the human body. This model was adapted from the NCRP model (NCRP 1979a) which was originally based on a three-compartment model developed by Bennett (1973). No physiological significance should be attached to the model compartments since they are meant to represent only differences in carbon-tritium bonding and molecular turnover rates. The four-compartment model allows for input of OBT directly into compartments representing tissues rather than into the body water pool. This feature is desirable but care must be taken to distinguish between the different OBTs. The catabolism of OBTs in foodstuff will be governed by the catabolism of the carbon compounds with which the tritium is bound; and it is unlikely that only four compartments will give a reasonable estimate of retention. For example, when the model alone was used for a hypothetical diet consisting of total intakes amounting to 10 times more nonexchangeable OBT than exchangeable OBT, the resulting dose estimate was reported to be 4.5 times higher than the dose contributed from free body water alone. This ratio value is considerably higher than expected based on animal studies (NCRP 1979b).

A complete characterization of the retention of tritium from ingested foodstuffs would require modeling carbon retention, which was a major task. This is likely not warranted as the nonexchangeable OBT, in even dry food, and can add (at most) ≈6% to the total cell dose (Myers and Johnson 1991). However, when considerations are made for what is known about tri-

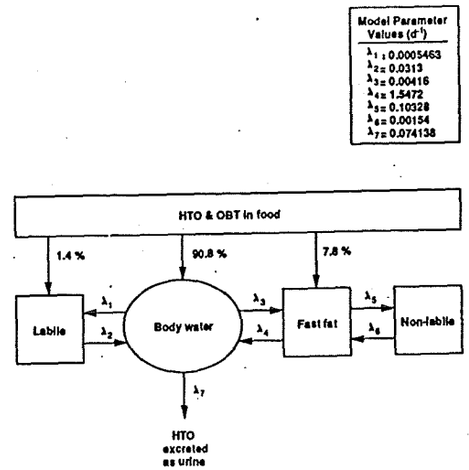

Fig. 5. OBT compartment model (adapted from Etnier et al. 1984).

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Paper

TRITIUM RADIOBIOLOGY AND RELATIVE BIOLOGICAL EFFECTIVENESS

T. Straume* and A. L. Carsten†

Abstract—**During the past decade, a large number of radiobiological studies have become available for tritium—many of them focusing on the relative biological effectiveness of tritium beta rays. These and previous studies indicate that tritium in body water produces the same spectrum of radiogenic effects (e.g., cancer, genetic effects, developmental abnormalities, and reproductive effects) observed following whole-body exposure to penetrating radiations such as gamma rays and x rays. However, tritium beta rays are of greater biological effectiveness than gamma rays and x rays. For example, tritium in the oxide form is about 2 to 3 times more effective at low doses or low dose rates than gamma rays from ^{137}Cs or ^{60}Co. When tritium is bound to organic molecules, relative biological effectiveness values may be somewhat larger than those for tritium in oxide form. Tritium administered to animals or to cells *in vitro* as tritiated amino acids results in relative biological effectiveness values that appear similar to those obtained for tritium in oxide form; however, if administered as tritiated thymidine, the relative biological effectiveness values appear to be about two-fold higher. It is clear from the wealth of tritium data now available that relative biological effectiveness values for tritium beta rays are higher than the quality factor of unity generally used in radiation protection.**
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Key words: tritium; cancer; genetic effects; relative biological effectiveness

INTRODUCTION

THIS paper is a summary of the current radiobiological knowledge of the carcinogenic, genetic, developmental, and reproductive effects associated with exposures to tritium. Key findings from the literature that exemplify the current understanding in these particular areas of radiobiology are presented. Also, relative biological effectiveness (RBE) values are summarized to the extent such information is presented in published papers and reports. The emphasis here is on low-level radiation, which is of greatest concern in radiation protection. Information concerning acute mortality and radiation sickness, which occur only at much higher doses, is not included.

A brief discussion of certain radiobiological concepts (radiosensitive targets, dose-response models, and the quality of radiations) is helpful in the interpretation of the material presented in this paper.

First, in radiobiology, it is generally assumed that for most of the endpoints considered here the radiosensitive target is DNA (among the exceptions may be reproductive effects in the female and central nervous system damage due to *in utero* exposure). The evidence that DNA damage can result in cell death, mutations, and even cancer is substantial. For example, unstable chromosomal abnormalities such as dicentrics are known to lead to reproductive cell death, and stable damage in DNA (e.g., point mutations and translocations) may lead to transmitted genetic effects as well as cancer through the activation of oncogenes (UNSCEAR 1986).

Second, it is also generally assumed (although with exceptions) that the shape of the dose-response curve at low to moderate doses follows a linear-quadratic function of the form, $E = aD + bD^2$, where E is the proportion affected, a and b are linear and quadratic coefficients, and D is the radiation dose (e.g., BEIR 1980). For acute exposures from low-LET radiations, the function may have an additional multiplicative term, $\exp(-a'D - b'D^2)$, that becomes important at high doses. This is illustrated in Fig. 1, where typical curves for low linear energy transfer (LET) and high-LET radiations are shown. Separate curves are shown for single acute exposures and for chronic exposures. The low-LET curves are representative of gamma rays, x rays, and beta rays (including those from tritium), and the curves for high LET are representative of fission neutrons and apparently alpha particles.

As can be seen in Fig. 1, the dose-response curve for acute exposures to low-LET radiation tends to be upward curving, indicating increased effectiveness as dose increases, with bend over at very high doses. The responses for chronic low-LET radiations, on the other hand, appear to be linear at all doses and considerably less steep than those for acute low-LET radiations. The slopes of the chronic curves equal the slopes of the acute curves at points-of-departure. In contrast, the curves for high-LET radiations are much steeper than those for low-LET radiations. Also, several studies using

* Lawrence Livermore National Laboratory, P.O. Box 5507, Livermore, CA 94550; †Brookhaven National Laboratory, Upton, NY 11973.
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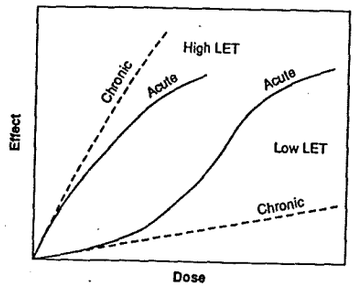

Fig. 1. Curves representing the emerging consensus on dose response for carcinogenesis and mutagenesis endpoints in the range 0 to ~3 Gy absorbed organ dose. The high-LET curves are typical for fission neutrons and low-LET curves are typical for gamma rays, x rays, and tritium beta rays. Curves for tritium beta rays are similar in shape to the curves for gamma rays and x rays but are somewhat steeper, particularly at low doses or low-dose rates. Solid curves are for single acute exposures and dashed curves are for chronic or fractionated exposures.

cells *in vitro* or experimental animals show that fission neutrons given chronically or in fractions appear to produce *greater* (not smaller) effects at high and moderate doses than do acute exposures (e.g., Thomson et al. 1982; Hill et al. 1985). However, at doses less than ~0.1 Gy of fission neutrons, the curves for acute and chronic exposures appear to become identical, at least for *in vivo* exposures (Thomson et al. 1982; Sinclair 1985; Straume 1985). Although not shown in Fig. 1, different dose-response curves exist at all intermediate LETs, ranging from low to high.

The third concept, which is also illustrated in Fig. 1, is that biological effectiveness depends on the "quality" of the radiation. In radiobiology, the RBE provides a quantitative comparison between radiations of differing qualities. RBE is defined as the ratio of doses from two different radiations that produce equal levels of biological damage in the same biological system (i.e., RBE equals the dose of reference radiation divided by the dose of test radiation at an equal level of effect). The reference radiation is generally 250-kVp x rays, ^{137}Cs- or ^{60}Co-gamma rays, and the test radiation is the radiation for which RBEs are to be evaluated—for the present paper, this is beta radiation from tritium, either in water molecules or organically bound. (Note that the reference radiation used *does* make a difference in the RBE estimates and should, therefore, be specified. This has been done for all RBE estimates listed in the present paper.) Because the RBE is a simple ratio, it can be modified by any factor that affects the two curves differently, including both physical (e.g., dose, dose rate, and LET) and biological (e.g., endpoint, gender, and age) factors.

DOSIMETRIC CONSIDERATIONS

Detailed reviews and updates of the dosimetry of tritium are presented in other papers in this issue (Hill and Johnson 1993; Wood et al. 1993). The reader is, therefore, referred to those papers for additional dosimetric information. Here, only an overview of tritium dosimetry is presented that is most relevant to the interpretation of the present paper.

For reproductive and transmitted genetic effects, the relevant doses are those to the germ cells (and their progenitors) of the male and the female. For tritiated water (HTO), which is generally distributed in body water, the dosimetry is relatively straightforward for both genders. For an acute exposure to HTO (e.g., a single inhalation or ingestion), the mean dose to a cell (or DNA) is about 0.7 times the dose to body water, which comprises ~70% of soft tissue, with very little tritium exchanged with tissue-bound hydrogen (1–2%) (NCRP 1979). However, for low-level chronic exposures to HTO, as much as ~35% of the tissue-bound hydrogen has been experimentally observed to be exchanged with tritium that then can become part of biomolecules such as proteins and DNA (NCRP 1979). Because these biomolecules have a slower turnover than body water, the tritium in them is retained longer and, after cessation of HTO exposure, the organically bound tritium (OBT) will become relatively more important as the tritiated water disappears.

Exposures to tritium *in utero* involve special dosimetric considerations. Although fetal cells rapidly divide during certain stages of development, they also differentiate to form organs and tissues that, in certain cases, involve very little or no subsequent cell proliferation. Thus, the incorporation of tritium into biomolecules of long-lived cells (e.g., neurons and oocytes) could result in large integrated doses over the lifetime of the cells. This is illustrated by Commerford et al. (1982), who exposed mice to constant internal levels of HTO from conception to 8 mo of age. They found that, although tritium was incorporated into biomolecules such as DNA and histones, the dose to rapidly dividing cells from such incorporation was small compared with the dose from HTO. However, for long-lived cells, their results indicated that the dose from tritium incorporated into DNA and histones could exceed that from HTO.

In contrast to HTO, tritiated biomolecules confined to the tissue-bound fraction may be retained for much longer time periods than tritium in water. For example, tritiated thymidine (^{3}H-Tdr) is incorporated into nuclear DNA during synthesis preceding cell division and will remain there until the cell divides again, at which time it will, if no new tritiated thymidine is available, be diluted by a factor of 2 by incorporation of unlabeled thymidine in the DNA of the progeny nuclei. The dose from exposure to tritiated thymidine is, therefore, closely related to the duration of exposure and cell-cycle time. For cells that cycle rapidly, both

incorporation and dilution would rapidly occur following a single exposure; thus the likelihood that rapidly dividing cells would incorporate ^{3}H-Tdr from a brief exposure is high, but the dose to DNA would be relatively low. On the other hand, cells that divide slowly would have a correspondingly smaller chance of incorporation, but those few cells that actually become labeled would receive a larger dose.

Isotopic and transmutational effects have also been of some concern in the dosimetry and health-risk assessment of tritium. The isotopes of hydrogen—protium, deuterium, and tritium—differ in mass by factors of 1, 2, and 3 and are the largest differences for any isotope. Although the data concerning isotopic effects indicate lower diffusion rates for tritium than for the lighter isotopes of hydrogen, it appears that the isotopic effects are not biologically significant (Carsten 1979). Transmutation is the conversion of a parent atom by radioactive decay to an atom possessing different chemical and physical properties, for example, the decay of tritium to helium. If the tritium atom is in a biologically important molecule, its decay to helium may result in detectable biological damage that could not be fully accounted for by the beta ray. Such effects were observed when certain nucleosides were labeled in specific positions and incorporated into DNA (Carsten 1979; Myers and Johnson 1991); however, for the usual modes of exposure to tritium through environmental media, it is not expected that a significant transmutation effect would exist (Myers and Johnson 1991).

CANCER

Radiobiology

Following sublethal whole-body ionizing radiation exposure, a primary concern is the resulting carcinogenic effects in the exposed individual. As previously noted, the sensitive target for most cellular effects, ranging from alterations in mitotic rate to cell death, is the cellular DNA. Among the most important radiation effects on DNA for development of the cancerous state are the activation of an oncogene or the turning off of a suppressor gene that has been controlling the oncogene. Either of these effects, together with possible reduction in immunological competence, may lead to the transformation of a cell and its later clonal expression as a malignancy.

Since the principal exposure to tritium is in the form of HTO, which distributes itself as does other body water, tritium exposures can, for most purposes, be considered as uniform whole-body exposures without the concern for target organs as noted for isotopes such as iodine in the thyroid or strontium in the bone. The situation for OBTs and specific DNA precursors is considered in subsequent sections, and by others in this issue (Diabate and Strack 1993). As might be expected, the spectrum of malignancies observed following HTO exposure is similar to that induced by other low-LET radiations (e.g., x rays and gamma rays) that deliver relatively uniform whole-body dose.

Investigations on the induction of cancer by tritium exposure may be broadly divided into two classes: first, as with other low-level radiation exposure, the evaluation of the incidence of leukemia and other malignancies as a late effect in the exposed individual; second, studies aimed at investigating the relationship of single or multigenerational tritium exposures and development of malignancies in each subsequent generation.

In addition to the overt appearance of the disease, studies of radiation-induced leukemogenesis have included detailed examination of toxic effects on the hematopoietic stem cells, which are readily accessible for study from mammalian bone marrow.

The toxic effects of acute and chronic exposure to HTO were investigated in mice using the spleen-colony technique (Carsten et al. 1982). In the mouse model, reduction in the number of hematopoietic pluripotent stem cells was observed after continuous ingestion of water containing 37 and 111 kBq mL^{-1} HTO. This reduction in stem cells was not accompanied by a reduction in the total number of bone-marrow cells. Tritiated thymidine cytocide measurements on the marrow indicated a higher-than-normal percentage of the surviving stem cells to be in active mitosis leading one to hypothesize that when damaged, but normally resting, stem cells were forced into mitosis to maintain the total number of marrow cells; this could create a greater opportunity for the expression of unrepaired genetic damage leading to leukemia.

As part of these same studies, effects on the DNA due to continuous tritium ingestion were evaluated by counting sister-chromatid exchanges (SCE) in the bone marrow cells of mice maintained on tritiated water (Irushima et al. 1984). In these studies, it was found that maintenance of mice on 111 kBq mL^{-1} HTO resulted in a statistically significant (1% level) elevation of SCE frequency at periods from 81–261 d of HTO ingestion. The frequency of induced SCEs increased linearly with the ingestion time. These results paralleled similar findings for the induction of liver-chromosome aberrations in the same animals after 100, 330, and 500 d of HTO ingestion (111 kBq mL^{-1}) (Brooks et al. 1976).

The ability to induce myeloid leukemias and characteristic cytogenetic changes by the prolonged exposure to tritiated water was also established in rats (Nikolaevskaya et al. 1990). These studies indicated differences in quantitative and structural damage of the chromosomes in rats developing acute myeloid leukemia and chronic myeloid leukemia.

In other recent studies, Myers and Johnson (1991, 1993) and Yokoro et al. (1989) demonstrated that low-level chronic ingestion of tritiated water induces leukemia and other malignancies. Details of these and related results are discussed in the RBE section.

In addition to carcinogenic effects in the exposed

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(696)

Paper

TRITIUM TRANSPORT AND CYCLING IN THE ENVIRONMENT

Charles E. Murphy, Jr.*

Abstract—**The transport and cycling of tritium in the environment can be understood in terms of the role of hydrogen in the environment. Physical and chemical isotopic effects, while present in some transport mechanisms and chemical reactions, are not important factors in environmental tritium dynamics. In addition, because of the role of hydrogen in metabolism and its ubiquitousness in the environment, organisms have not evolved mechanisms to accumulate or concentrate hydrogen or its isotopes in food chains. Therefore, biomagnification of tritium is not a factor in food chain transfer. The lack of significant isotopic effects or biomagnification means that tritium transport and cycling in the environment can be predicted on the basis of the transport processes, hydrogen content, and chemical transformation of hydrogen and its compounds in the environment.**
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Key words: tritium; environmental transport; hydrogen; radiation dose

INTRODUCTION

THE EMPHASIS of our interest in environmental tritium is how tritium transport and cycling leads to exposure and radiation dose to humans. To accomplish this task it is necessary to know the tritium concentration of components of the environment that can lead to human exposure. Examples are the concentration of tritium in air, in water used for drinking or bathing, and in foods. All of the components of the human environment are interconnected with each other and with other environmental components which do not normally come into contact with humans. It is necessary to account for all significant transport and cycling of tritium among these components in order to determine human exposure. The sources of information available for understanding environmental transport and cycling are the results of tritium experiments on individual components of the environment, experience from designed or inadvertent releases of tritium to the environment from nuclear facilities and nuclear weapons tests, and extrapolation of tritium behavior from the behavior of hydrogen and its compounds in the environment.

As an isotope of hydrogen, much of the behavior of tritium in the environment is similar to that of protium and deuterium, the other isotopes of hydrogen. Hydrogen is the most abundant element on earth in terms of number of atoms. It forms an enormous number of compounds, including its association with carbon in almost all organic compounds. In terms of abundance and mobility, the most important compound of hydrogen is water. The transport of tritiated water, HTO, is extremely important in determining the transport of tritium in the environment and the subsequent exposure to humans.

While the relative mass difference of tritium with respect to protium, the most abundant natural hydrogen isotope, is large, the isotopic effects of tritium transport are so small as to be difficult to detect in environmental tritium transport measurements. The small difference in the physical characteristics of tritiated and non-tritiated compounds can be explained by the small contribution of hydrogen to the mass of most hydrogen-containing compounds. The vapor pressure of tritiated water is around 90% of that of natural water (Jacobs 1968). The molecular diffusion coefficient is approximately 40% less for molecular tritium, HT, than molecular hydrogen but only 5% less for tritiated water. Similar ratios are the rule for other physical isotopic effects of tritiated compounds.

The mass difference in the isotopes of hydrogen leads to isotopic differences between tritium and protium in chemical reactions that can be much greater than in physical processes (Jacobs 1968). These effects are most pronounced in the kinetics of hydrogen reactions (Weston 1973). Reactions with tritium are slower than those with protium. However, the net hydrogen isotope effect on isotopic abundances in organisms appears to be very small (Bruner 1973). In most cases, an isotopic effect cannot be detected against other sources of environmental variability. In a very carefully controlled experiment, Garland and Ameen (1979) found that grain plants grown in a tritiated water environment were at 80% of tritium equilibrium with the tritium to hydrogen ratio in the water.

The observation that the ratio of tritium to hydrogen in organisms is similar to that in their environment also suggests that there is no biological sequestering of

* Westinghouse Savannah River Company, Aiken, South Carolina 29802.
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tritium against an energy gradient (biomagnification). Unlike many essential nutrient elements, hydrogen is so ubiquitous in the environment that there is no reason for organisms to evolve mechanisms for hydrogen or hydrogen isotope enrichment.

In summary, the small magnitude of either physical or chemical isotopic effects and the apparent absence of biomagnification means that tritium transport and cycling in the environment can be inferred from the cycling of hydrogen and its compounds. The most important of these compounds is water, followed by organic compounds and molecular hydrogen.

The relationship between tritium transport and the tritium concentration of the components leading to exposure of humans can be described in the framework of the mass balance of tritium transported between specified components through the action of identified processes. In the language of systems modeling, the components are the compartments and the processes are the paths between compartments. The compartments can be individual organisms or assemblages which can be characterized by an average behavior and tritium concentration. Examples of compartments are a single leaf, all the vegetation at a geographical location, the rooting depth of the soil, or all the fish of one species in a stream.

The processes transporting tritium can be physical, chemical, or biological in nature. Physical processes can usually be defined as bulk transport or flow (advection), where tritium moves with the bulk of the compound with which it is associated, or diffusional transport or flow in which the tritium or a tritiated compound moves along a gradient of concentration from high concentration to low concentration. Chemical processes involved in tritium transport are largely associated with the change in the chemical species involved in transport. As discussed previously, because there is no evolutionary advantage to concentrating hydrogen, active chemical or biological pumps moving tritium against a concentration gradient are very rare and of little practical importance in environmental tritium transport.

Once the transport processes and the capacity of the compartments have been identified, the behavior of the system can be described in terms of the magnitude of the transport by the processes moving tritium between compartments and the capacity of the compartments to store tritium. This type of system can also be characterized by rate of tritiated material flowing through the compartments, the steady-state concentration in the compartments, and the half-life of tritium concentration following a step change in input (Anspaugh et al. 1973). In some instances where the system has been exposed to a constant level of tritium, the steady-state concentration is a sufficient characterization for estimating human exposure.

In the next section, an example of tritium transport will be developed to show how the results of tritium transport in the environment can be explained in terms of the principles of hydrogen transport with small isotopic effects and no biological enrichment of tritium. The example will demonstrate how seemingly anomalous results, which show deviations from equilibrium of hydrogen isotope ratios between compartments, can be explained without isotopic effects or biomagnification. This example will be followed by a section which describes some of the characteristics of broadly defined environmental systems with the objective of identifying the processes which are important in tritium transport.

EXAMPLE: TRITIATED WATER TRANSPORT AND CYCLING IN VEGETATION

The vegetation tritiated water concentration is of interest because of the potential for human uptake by consumption of vegetable foods. Vegetation also serves as the primary path for incorporation of tritium into organic compounds which can move up the food chain. In addition, the leaves of many types of vegetation have proved to be excellent sampling devices which can be monitored to determine the path of tritium releases. The vegetation system, illustrated in Fig. 1, has three compartments: the atmosphere, the plant leaves, and the soil. Because bulk flow dominates transport through the conducting system of the plant roots and stem, this part of the plant can be treated as a time delay in the path between the soil and the leaves. The flow of tritiated water is described in terms of the concentrations in the compartments and the processes transporting tritiated water between compartments. Tritiated water from the soil moves into the leaf by bulk flow through the water-conducting vessels. This water comes into contact with the atmosphere at the thin film in the walls of the mesophyll cells lining the substomal cavity. Transport from inside of the leaf to the atmosphere is by vapor diffusion through small pores (stoma) in the water-impervious cuticle covering the leaf. The flow toward the leaf is proportional to the tritiated water vapor concentration of the air (the product of the tritiated water vapor concentration, C_a, and the air humidity, ρ_{wa}) and the diffusion resistances in the path through the atmosphere, stoma, and substomal cavity (r). The diffusion from the leaf to the air is proportional to the tritiated water vapor concentration (C_v) in the water film in the mesophyll cell walls, approximately the saturation vapor concentration at the leaf temperature (ρ_{wv}), and the same resistance to vapor flow as in diffusion in the opposite direction. The change in leaf tritiated water concentration with time (t) is also controlled by the relationship between leaf water volume (V_v) and leaf area (A_v).

Water flow from the soil into the plant takes place by absorption through the roots and is transported along a water potential gradient through the plant vascular system into the leaves. Tritium is transported by bulk flow with the water. The uptake is driven by evaporation within the plant leaves. Evaporation is essentially the same process as described for tritiated water exchange between the atmosphere and the wet surfaces of the leaf cells. In this case the water vapor concentration gradient between the atmosphere and the wet leaf surfaces ($\rho_{wv}-\rho_{wa}$) is the driving force and the vapor diffusion resistance in the leaf is essentially the same as for tritiated water vapor.

In most cases, the tritium concentration in the soil water is the result of washout of tritium from the atmosphere. Because of the relatively low rate of diffusion between the atmosphere and the soil, dry deposition of tritiated water or tritiated hydrogen to the soil only becomes significant when the atmospheric concentrations of these compounds are very high compared to the concentration in rainfall. Washout is proportional to the tritiated water vapor concentration and the diffusion resistances associated with the air and surface of the individual rain drops. The relationship between the atmospheric tritiated water concentration and the concentration of tritium in rainwater is usually based on an empirically derived washout factor (β) which incorporates the effects of diffusion into the individual raindrops. Other factors affecting the tritiated water concentration of the soil (C_s) are the soil water content (Θ), the infiltrated precipitation (R_i), the rooting depth of the vegetation (D), and the drainage from the rooting depth. Drainage is approximated here as proportional to the water content of the rooted soil layer. The constant of proportionality (α) is a function of the soil water conductivity.

The system of equations which describe this system are as follows:

$$V_v \frac{dC_v}{dt} = A_v\left(\frac{C_a\rho_{wa}}{1.05r} - \frac{0.9\rho_{wv}C_v}{1.05r} + \frac{\rho_{wv}-\rho_{wa}}{r}C_s\right) \quad (1)$$

$$D\frac{d}{dt}(\theta C_s) = \beta C_a R_i - \alpha\theta C_s - \frac{\rho_{wv}-\rho_{wa}}{r}C_s \quad (2)$$

$$D\frac{d\theta}{dt} = R_i - \frac{\rho_{wv}-\rho_{wa}}{r} - \alpha\theta \quad (3)$$

For the soil-plant-atmosphere tritium transport, the average leaf tritium concentration for seasonal or annual periods can be estimated by the steady-state concentration under average air tritium concentration and climatic conditions. The steady-state concentration can be quantitatively described by setting the left hand sides of the eqns (1), (2) and (3) to zero (Raney and Vaadia 1965; Belot et al. 1979; Murphy 1984). Notice that the steady-state value of tritiated water concentration does not depend on the volume of any part of the system, but only on the relative magnitudes of the inputs and outputs. This set of equations can be simplified to show the relationship between the air concentration and the leaf concentration of tritiated water.

$$\frac{C_v}{C_a} = \frac{0.95\rho_{wa}}{0.9\rho_{wv}} + \frac{1}{0.9}\left(1 - \frac{\rho_{wa}}{\rho_{wv}}\right)\beta \quad (4)$$

The results of solving this equation are shown in Fig. 2. The concentration of tritiated water in the leaf is somewhat lower than the atmospheric concentration. The ratio leaf to atmospheric concentration depends on the ratio of the atmospheric water vapor concentration to the vapor concentration at the wet leaf cell surfaces. If leaf temperature is equal to atmospheric temperature, this vapor concentration ratio is identical to the relative humidity of the air. The water in the leaf is a mixture of the water from atmospheric vapor and water from the soil. While the tritium in the soil water is from the atmospheric water vapor, it is diluted by the rainfall which is assumed to be from another location with low tritium content. These calculations show that the concentrations of tritiated water in the leaf and the atmosphere are not in equilibrium. The cause of the difference is the mixing of water from different sources, in this example atmospheric water vapor and

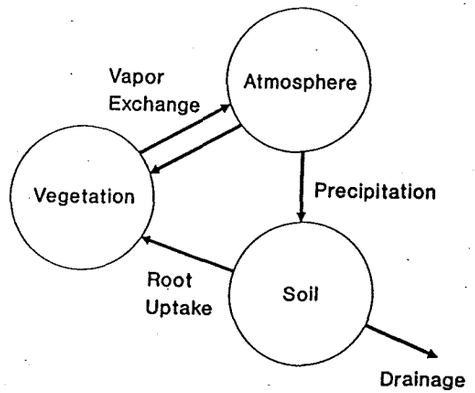

Fig. 1. Conceptual model of the soil-plant-atmosphere tritiated water transport system.

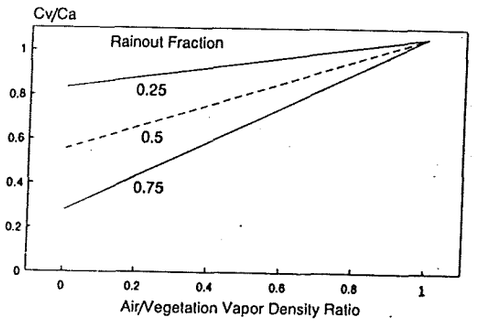

Fig. 2. The results of a steady state analysis of the ratio of tritiated water concentration in vegetation versus the concentration in air moisture.

rainfall. The leaf concentration is bounded by the concentration of the sources.

This analysis can be extended to explain a situation observed first at the Savannah River Site where it was found that, in some years, the annual average concentration of tritiated water in vegetation water was greater than that in atmospheric moisture. The additional source of tritium was from oxidation of atmospheric tritiated molecular hydrogen in soil (Murphy and Pendergast 1979). This can be accounted for in eqn (2) by adding a term for molecular tritium oxidation in the soil of the form $\gamma\ v_d\ C_T$. The rate of tritium oxidation is assumed to be proportional to the atmospheric concentration, C_T. The constant of proportionality is called a deposition velocity, v_d. The constant γ converts molecules of tritiated hydrogen to equivalent molecules of tritiated water. The steady state equation becomes:

$$\frac{C_v}{C_a} = \frac{0.95\rho_{wa}}{0.9\rho_{wv}} + \frac{1}{0.9}\left(1 - \frac{\rho_{wa}}{\rho_{wv}}\right)\left(\beta + \frac{v_d C_T}{R_i C_a}\right) \quad (5)$$

The results are shown in Fig. 3. In addition to the dependence on the ratio of atmospheric and leaf water vapor concentrations, the ratio of leaf to atmospheric tritium concentration depends on the ratio of the depositions of tritiated hydrogen and tritiated rainfall to the soil. At the Savannah River Site the ratio of tritium flux densities is greater than unity, explaining the anomalous high leaf tritiated water concentrations, because the specific activity (tritium to hydrogen ratio) of tritiated hydrogen is much greater than that of the tritiated water in rainfall. In this case, as in most other environmental cases, there was no evidence of biological enrichment of tritiated water in the leaves or in any part of the soil-plant-atmosphere system.

Eqns (1), (2), and (3) can also be used to describe the time-dependent behavior of tritium in a system. Fig. 4 shows the result of simulating an experiment in which a small plot of herbaceous vegetation was covered with a chamber and fumigated with tritiated water for a short period of time (similar to the experiment of Kline and Stewart 1974). After fumigation, the time course of tritiated water in the leaves of the vegetation was measured. The behavior of the system is characterized by the approach of the leaf water to a steady-state concentration with the chamber air. After the source of atmospheric tritium is removed, the leaf concentration decreases as the tritiated water is evaporated and replaced with water from the soil. However, the soil in the chamber has also absorbed tritiated water from the fumigation period. There is a lag between the time when the soil first contains tritium and when the tritium shows up in the leaf because the plant must be purged of the pre-exposure water that remains in its roots and stem. After the soil tritiated water reaches the leaves the leaf water approaches a steady-state with the soil water as the source of tritium. In this transient case there are periods of time when the leaf tritiated water concentration is not at a steady-state with any part of

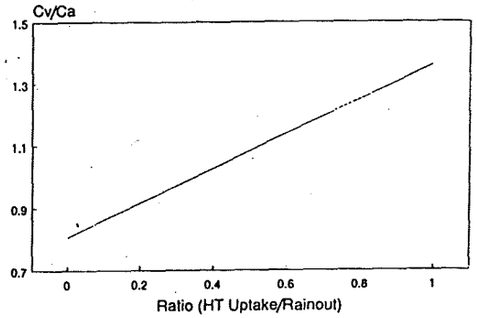

Fig. 3. The results of a steady state analysis of the ratio of tritiated water concentration in vegetation versus the concentration in air moisture when oxidation of HT gas by soil microorganisms is included.

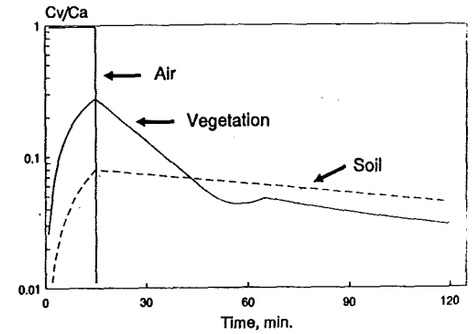

Fig. 4. The results of a time dependent simulation of tritiated water exposure of a soil-plant-atmosphere system during and after exposure to atmospheric HTO vapor.

the soil-plant-atmospheric system. In fact, there is a period when the leaf concentration is lower than the tritiated water concentration of either the concentration of the atmosphere during fumigation or the concentration of the soil after fumigation. This example illustrates the problems of interpreting measurements made of a system which is changing with time. This is a particular problem when sampling is done at different times and/or is characteristic of different averaging times.

The analysis of this simple environmental tritium system illustrates some of the problems in interpreting tritium field measurements. Measurements made without knowledge of all the sources of tritium, or made at different times in systems that are not at steady state can lead to the appearance of anomalies that have been interpreted as isotopic effects or tritium biomagnification. In fact, no firm evidence exists of either of these effects being significant in environmental tritium transport and cycling.

TRITIUM TRANSPORT AND CYCLING IN ENVIRONMENTAL SYSTEMS

Tritium in the atmosphere

It is not the purpose of this paper to review the large volume of literature which describes the transport of materials in the atmosphere (see Pasquill and Smith 1983 or Slade 1968). This section will deal with those characteristic of atmospheric transport which are especially important to understanding tritium transport and cycling in the environment. The most important gaseous chemical species of tritium are tritiated water vapor, tritiated molecular hydrogen, and tritiated methane, listed approximately in the order of importance. In addition to the dilution of the tritiated gas by the turbulent mixing, the transfer from the atmosphere to other environmental systems is affected by the dilution by non-tritiated forms of the same compound, by chemical processes that change the chemical species, and by processes that remove tritium from the atmosphere.

Since the tritiated compounds and their untritiated counterparts are transported through the environment at approximately the same rate, there will be competition for the same exchange or reaction sites. Given the same tritiated water concentration per unit volume of air, more tritium will be exchanged with an organism at low humidity than at high humidity. In the same way, if a compound is the product of two different chemical species, the product of the species with a higher tritium:hydrogen ratio will have a greater specific activity than the product of a compound with a lower tritium:hydrogen ratio. In the atmosphere away from nuclear facilities, the specific activities of tritiated water, tritiated hydrogen, and tritiated methane were found to be 16, 10^6, and 4×10^4 TU (approximately 1.7×10^4, 1.1×10^9,and 4.3×10^7 Bq/g-H), respectively (Okai and Takashima 1989). This suggests that water from oxidation of tritiated hydrogen would have a much higher specific activity than water condensed from the atmosphere. Of course, once water is produced by hydrogen oxidation it would be expected to be quickly diluted by the great quantity of water in the atmosphere.

Removal of tritiated compounds from the atmosphere can take place by transfer to the surface or by chemical transformation into another compound. The latter process does not actually remove tritium from the atmosphere, it only transforms it into another compound. The only transformation that can accomplish actual removal is radioactive decay. However, chemical transformations are important because they can change the rate of exchange between the atmosphere and the surface.

The most well studied atmospheric chemical transformation involving tritium is the oxidation of tritiated hydrogen gas to tritiated water. The radiotoxicity of tritiated water has been calculated to be 20,000 times greater than that of tritiated molecular hydrogen (ICRP 1979). The results of a number of studies indicated that the oxidation of molecular hydrogen gas in air takes place very slowly, with a half-life on the order of 100 or more y (Eakins and Hutchinson 1973; Easterly et al. 1985). However, Mason and Ostlund (1979) estimate that the half-life of molecular tritium in the atmosphere is only 6.5 y. The difference in these two estimates is resolved by recognizing that the half-life of molecular tritium in the atmosphere is controlled by surface uptake and not oxidation in the atmosphere.

Atmospheric removal processes are classified as dry or wet deposition, based on whether the process does or does not involve precipitation. Deposition of tritium in precipitation can be the result of two processes. Rainout is the process of incorporation of tritium in the rain drops as they form in a cloud. Washout involves the incorporation of tritium into precipitation falling through a plume (Chamberlain and Eagleton 1964). The maximum tritium concentration in a raindrop or other form of precipitation is determined by the equilibrium concentration of the gas in solution. Once saturation is reached, no more tritium will be removed by the precipitation.

Monitoring data suggests that the wet deposition of tritiated water is much greater than that of tritiated hydrogen (Murphy et al. 1991). This is confirmed by the calculations of Long (1978) which show that the lower solubility of molecular tritium vs. tritiated water should lead to less wet deposition.

The rate of dry deposition of tritiated gases is influenced by diffusion from the bulk of the atmosphere to the surface and by processes which are characteristics of the surface. In most cases, the deposition rate is controlled by the surface processes. Removal of tritiated compounds takes place in the vegetation and the soil of terrestrial systems and at the water surface of aquatic systems. As in wet deposition, the dry deposition rate of tritiated water is generally much greater than that of tritiated hydrogen.Ultimately, most of the removal of tritium from the troposphere is the result of deposition. (Radioactive decay is also a factor in the stratosphere where the residence time of tritium is greater than in the troposphere.) It is possible to compare the integrated rates of removal of tritiated water and tritiated molecular hydrogen by all processes by comparing their tropospheric half-lives.

NCRP Report Number 62 (NCRP 1979) summarizes the results of a number of experiments which estimate the half-life of tritiated water in the atmosphere to between 21 and 41 d. This is equivalent to a deposition velocity (the amount of deposition per unit area divided by the concentration in atmosphere) of between 0.4 and 0.8 cm/s if the height of the tropopause is taken to be in the range of 12 to 15 km.

The deposition velocity of tritiated hydrogen can be calculated in the same way. Mason and Ostlund (1979) have estimated that the half-life for HT is 4.8 y.

Again, using the height of the mixed layer as being between 12 and 15 km, the deposition velocity is between 0.04 and 0.05 cm/s.Bardolle (1985) presented data which suggested that the oxidation of tritiated hydrogen in the atmosphere away from the surface might be more rapid than previous studies had indicated. His results led to two experimental releases of tritiated hydrogen gas to determine if the results could be duplicated. One experiment was held in France (Djerassi and Gulden 1988) and the other in Canada (Brown et al. 1988). The results confirmed the earlier work which showed very low oxidation rates under natural conditions (Paillard et al. 1988, Brown et al. 1990).

It should be noted that deposition of tritiated compounds to the surface does not indicate permanent removal from the atmosphere. Tritiated water that is absorbed by vegetation or soil as a plume passes over a location will re-enter the atmosphere after the plume has passed. When the atmospheric concentration is integrated over time periods greater than a few days, only that part of the tritiated water that is removed by percolation into soil below the root zone, or that part which is incorporated in organic matter, is removed from the atmosphere.

Deposited tritiated hydrogen is oxidized at the surface (largely in the soil) and, in this sense, the deposited molecular tritium is removed from the atmosphere (Ehhalt 1973, Murphy et al. 1977, McFarlane et al. 1978). However, the tritiated water produced by oxidation becomes part of the soil water and in this form re-enters the atmosphere (Kirchmann et al. 1986a; Belot et al. 1988). Depositional processes are important because of the influence on atmospheric concentrations and because deposition is the primary path for tritium to enter terrestrial systems after an atmospheric tritium release.

Tritium in the terrestrial environment

In this discussion, the terrestrial environment is defined as the layer at the land surface of the earth that contains the majority of the living organisms. This includes the atmosphere within plant canopies, the soil down to the rooting depth of the vegetation, and the vegetation, animals, and other organisms living in this layer. The characteristics of importance to tritium transport for each part of the terrestrial system will be discussed separately, followed by a discussion of the integrated behavior of terrestrial systems.

Soil tritium. In soils, entrance and transport of tritium takes place by gaseous diffusion or by liquid water movement under the influence of gravity and capillarity. The diffusion of tritiated compounds into the soil is impeded by vegetation and any vegetative litter on the soil surface. For this reason the transport of tritiated water vapor to the soil at sites having a vegetative cover is usually much lower than the transport to bare soil (Sweet et al. 1983; Kirchmann et al. 1986a). Gaseous diffusion in the soil is a function of the gas-filled pores. Course, sandy soils have a more open structure than clay soils to gas diffusion. Diffusion is lower at high soil water contents because gaseous diffusion is limited by the water filling the soil pores. Garland (1979) measured tritiated water deposition velocities between 0.09 and 0.91 cm/s in bare soil.

Tritiated hydrogen also diffuses into the soil, where it is dissolved in the soil water. Once tritiated hydrogen has penetrated into the soil, soil microorganisms, producing the enzyme hydrogenase, are able to break the bonds in the diatomic hydrogen molecule (Schlegel 1966) and the molecular tritium is oxidized to tritiated water. At the low tritiated hydrogen concentration normally found in the environment, the oxidation reaction is very efficient. However, because of the comparatively slow diffusion into the soil and the low solubility of hydrogen in the soil water, the conversion rate of tritiated hydrogen from an atmospheric source to tritiated soil water is not rapid. Measurements have ranged from 0.001 to 0.1 cm/s in a variety of natural soils (Sweet and Murphy 1984; Dunstall et al. 1985; Foerstel et al. 1988; Taechner et al. 1988).

Once tritium has entered the soil through water vapor diffusion, conversion of tritiated molecular hydrogen, or direct application of tritiated water, the transport of tritiated water in the soil will involve the same processes and follow the same paths as other water. Tritiated water is carried by bulk flow with the liquid water in the soil which moves in response to gravity and capillary tension. This causes tritiated water entering the surface of the soil to move downward in a layer as it is displaced by the water entering the soil from above. Even though tritiated water is carried by bulk flow, there is some mixing of water in the soil because the water is dispersed among the different pores in the soil when it is transported. Some water ends up having a longer flow path than other water. Thus some water moves ahead and some lags behind and is mixed with water at either edge of the layer containing tritiated water. Finally, the tritiated water will move below the rooting depth of the vegetation and interaction with the surface will stop.

Tritiated water can also move by vapor transport. Under constant temperature conditions, vapor transport will take place along the same tension gradient as liquid water because the equilibrium vapor pressure in the pores is a function of the tension the water is held under. The vapor pressure is also a function of soil temperature and the vapor pressure gradient will tend to move water from regions of high temperature to regions of low temperature. Vapor transport is by diffusion through the air-filled pores of the soil. Since vapor transport in soil is by diffusion, tritiated water will move in response to the difference in tritiated water concentration between two locations in the soil, independent of the movement of water vapor. This means that tritiated water can diffuse from a region of high tritiated water concentration to a region of low tritiated water concentration even when there is no net transport of water vapor. A thorough discussion of soil water movement can be found in texts such as Hillel (1971) or Childs (1968).

Absorption of soil water by plant roots supplies the majority of water for terrestrial vegetation. The extraction of water is through the action of capillary tension formed in the plants by removal of water from the leaves by evaporation (Kramer 1969). Water moves out of the soil into the plant roots along this tension gradient. Extraction of water from the soil is also a bulk flow process and the water moves into the plant at the same tritium concentration as the tritiated water concentration in the soil pores near the roots. Rooting depths vary greatly between different types of plants, and between the same type of plant grown in different soils and climates. In many soils of the Southeastern U.S., plant roots are impeded by heavy clay subsoils. In arid regions with highly permeable soils, plant roots have been found in a mining excavation at a depth of over 100 meters (Phillips 1963). However, most of the roots of both agricultural and forest species are found within one meter of the surface.

The soil contains many organic compounds. Most of the organic tritium in soil is derived from plants growing in the soil. However, small amounts of tritiated organic matter are produced by autotrophic soil microorganisms that use tritiated hydrogen as an energy source. The effect of soil organic tritium transformations on tritium transport is probably very small compared to transport by water. In practice, the most important transformation of tritiated compounds taking place in soils is the oxidation of tritiated hydrogen to tritiated water (Watts and Murphy 1978; Brown et al. 1990).

Vegetation tritium. The initial process in the chain of processes which leads to deposition of tritiated gases is turbulent diffusion in the fluid boundary layer as eddies of air carry the tritiated compounds from the atmosphere to the surface of the vegetation and the soil. After the tritium has reached the surface of the vegetation, it must cross the fluid boundary layer of the surface elements. The waxy surfaces and bark of vegetation are relatively inert to tritiated compounds, such as hydrogen, water vapor, or methane. The only significant sinks for these compounds are inside the vegetation. The route into the vegetation is through the stoma on the leaf surface. The size of a stoma changes in response to the environment. Plant leaves have evolved to limit water loss while allowing transport of CO_2 into the leaf for photosynthesis. Under conditions of adequate moisture, the stomata will be open during the day and closed at night. If water stress develops in the leaf, the stomata will close to conserve water. The rate of diffusion of any of the tritiated gases will be controlled by the size of the stomata. After diffusion through the stomata, the gas will go into solution in the cell sap which wets the surfaces of the cells in substomatal cavities.

The diffusion of tritiated water vapor in air is in the direction of the gradient from high concentration to low concentration. It is possible for the gradient of water vapor to be in the opposite direction from the gradient of tritiated water vapor, resulting in water evaporating from the leaves while tritiated water vapor is entering the leaves. As the tritiated water concentration of the leaf water increases, the leaf will come to a steady state with the atmosphere and the net uptake of tritiated water will stop. Leaves can come into a steady state with the atmosphere in 0.25 to 2 hours, depending on the uptake rate of tritiated water as controlled by the leaf stomata. The comparatively large initial deposition velocity for tritiated water to vegetated surfaces will decrease to nearly zero as the leaves of the vegetation approach a steady state (Mason et al. 1973).Tritiated methane and tritiated hydrogen also go into solution in the leaf cell sap (Cline 1953; Mason et al. 1973). Since the solubility of both of these gases is low, the cell sap will tend to saturate at low internal concentrations. The fact that tritiated water is detected in vegetation soon after exposure to either of these gases suggests that there are physiological processes which promote absorption and conversion. However, the rates of uptake are small compared to either the deposition rate of tritiated water to vegetation or the conversion rate of tritiated hydrogen to tritiated water in soil (Mason et al. 1973; McFarlane 1978; Sweet and Murphy 1984; Spencer and Dunstall 1986; Murphy 1989).

Tritiated water can also reach vegetation through uptake of soil water. Tritium transport of water from soil is a bulk flow process and the concentration entering the vegetation will be a weighted average of the tritium concentration of the soil layers from which water is being withdrawn. The relative rate of withdrawal from different depths in the soil is influenced by the capillary tension holding water in the soil, the root density, and the depth. Most plant roots are within a few feet of the soil surface. Under conditions of equal soil moisture tension at all depths, most of the water will come from the surface layers of the soil. However, as water is removed from the surface layers, the majority of water will come from successively deeper in the soil.

The water content of whole terrestrial plants varies from less than a gram to thousands of kilograms. The mass of water, relative to the rate of water flow, in forests or other large vegetation types is great enough that the residence time for tritium in this type of vegetation is only a matter of days (Jordan et al. 1970; Kline et al. 1970; Luvall and Murphy 1982). Since the transport of tritiated water is by bulk flow, the effect of storage in the vegetation is a time delay between the soil and the leaves. In some cases water flow through plant organs is not uniform. Some organs, such as fruits, have slower water turnover than the root-stem-leaf

conducting system, therefore leading to longer tritiated water turnover times (Dinner et al. 1980)

One would expect that both the atmospheric and soil water in the vicinity of a source of atmospheric tritium would contain tritium. In this case, the average tritiated water content should be between the concentration of the soil and air water. The average ratio of tritiated water in vegetation to tritiated water in the air of 0.77, calculated from environmental monitoring data collected at Savannah River Site (SRS) during the period 1977 to 1982, agrees well with the value of 0.82 calculated for steady state concentrations (Murphy 1984).

There are a number of mechanisms which incorporate tritium into organic matter (Moses and Calvin 1959; Peng 1973). Exchange of loosely bound hydrogen radicals such as hydroxyls accounts for most of the incorporation during short exposure to tritiated water. Most of the tritium incorporated by exchange is reversibly lost when the concentration of tritiated water in the environment decreases. The primary process for incorporating tritium into the long-residence-time hydrogen sites of plants is photosynthesis in the green leaves (Moses and Calvin 1959; Guenot and Belot 1984). The amount of tritium incorporated into organic matter is small compared to the amount of tritium moving through the plant, from the soil to the atmosphere, as tritiated water. The fraction of hydrogen converted to organic matter varies from 0.06% to 0.3% for rapidly growing crops (Kramer 1969). Therefore, incorporation into organic matter has little effect on the amount of tritiated water moving through the environment. However, tritiated organics need to be considered when dealing with tritium concentrations in organisms feeding on the vegetation. Furthermore, the longest retention times in terrestrial systems are likely to be associated with organic compounds (Stewart et al. 1972; Sanders 1976; Brown 1979).

Animal tritium. Tritium concentration in animals is similar to that in vegetation in the sense that the concentration is the result of tritium transport to and from the animal through a number of paths (Robertson 1973; Yousef 1973). In terms of the amount of water transported, the major path for entrance of tritiated water in animals is through drinking water. A major second source is the water in food eaten by the animal. The water in food is from the moisture in the food and from the water produced by digestion and catabolism (burning for energy) of the food. Tritiated water is also exchanged with body water through breathing and skin exchange. Tritiated water leaves the animal by excretion in urine and feces.

The major path by which water leaves the animal is through excretion. The relationship between the volume of water in the animal and the rate of excretion is the major determining factor for the turnover time of tritium in the body once the animal has been contaminated. Water turnover rates have been found to be closely related to body weight for a wide variety of animals (Yousef 1973). An exception to this trend is the dry-land kangaroo rats which have low water losses compared to their body weight. The water turnover time for most species is less than 10 d. This suggests that most animal species, as with vegetation, will respond to the day to day changes of tritium in their environment. However, unlike vegetation, animals are mobile and will contain water that is averaged over the area in which they range.

The fate of tritium in organic material which is transported to animals through the food chain depends on the chemical form of the compounds containing the tritium. The fraction of exchangeable sites and the metabolism of digestion and assimilation varies among compounds. Carbohydrates have comparatively large numbers of exchangeable sites and are digested to basic units before assimilation. Therefore, tritium in carbohydrates can be lost during ingestion and assimilation. Proteins have fewer easily exchangeable sites and are only broken down to amino acids before assimilation. Lipids are either completely metabolized as energy or assimilated with few exchanges. This is reflected in the studies of van den Hoek et al. (1985) which show that when milk is produced from tritiated feed, fats retain better than 90% of the tritium when assimilated into the consuming animal. In contrast, proteins retain around 50% and carbohydrates around 25%. Since the hydrogen content of animal organic matter is in the range of 40% of the hydrogen content of body water, the tritium content of the organic matter is potentially a significant portion of the total tritium content of the whole animal (Evans 1969).

Residence times for tritium in the organic fraction of animals have not been widely studied. Studies of humans (Sanders and Reinig 1969), marine animals (Harrison et al. 1973), and milk cows (van den Hoek et al. 1979) exposed to tritiated water indicate that there is one or more "slow" turnover components, usually attributed to organic tritium, that are in the order of 100 d to 1 y.

Integrated tritium transport and cycling in terrestrial systems. It is a general property of systems made up of a number of connected components that the residence time of the system will be determined by the longest residence time among the components. Sasser et al. (1973) presents data from a series of terrestrial systems exposed through irrigation with tritiated water. The results (Fig. 5) show that the residence time of the systems, as measured by the residence time in the vegetation water, is determined by the residence time in the soil. In these systems, after initial exposure, the other system components were supplied tritiated water from the soil water. The short residence time in the grassland compared to the desert is a result of the greater amount of rainfall moving tritium out of the rooting zone of soil in the grassland. The residence of the tritiated water in the tropical rain forest is controlled

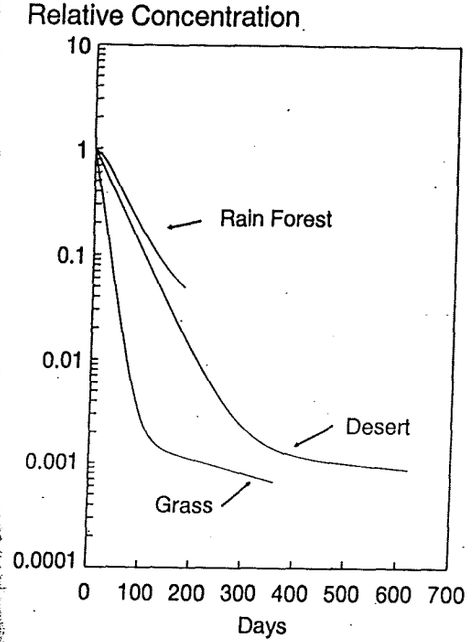

Fig. 5. A comparison of the response of three terrestrial systems to tritiated water additions to the soil. The concentration of the vegetation is controlled by the response time of the soil (redrawn from Sasser et al. 1973).

by a soil layer of low permeability which retards drainage of water from the soil. Miettinen (1979) has presented a summary of soil tritiated water residence times measured in the IAEA coordinated program. The half residence times vary from a few hours to 14 d for the shortest component of tritium loss. Longer components can be as great as several years.

A somewhat different result was found after exposure of forests to tritiated water vapor following an atmospheric release. The short duration release resulted in tritium entering the vegetation but not the soil (Sweet et al. 1983). The residence time of all measurable tritium in the system was less than one day. On the other hand, after a release of tritiated molecular hydrogen most of the tritium remaining in the system was in the form of tritiated water, oxidized from tritiated hydrogen entering the soil. The residence time in the vegetation after the tritiated hydrogen release was found to be on the order of 10 d, which was the residence time in the soil (Murphy et al. 1977).

Average tritium concentrations in system components resulting from chronic releases to terrestrial systems appear to agree with steady state estimates (Murphy 1984). The vegetation has a tritium concentration between the soil water and atmospheric tritium concentration. The vegetation organic concentrations will be close to the vegetation water which is the source of water used in photosynthesis (Guenot and Belot 1984). The animals living in this system will have a tritium concentration in their body water that is a balance between intake by drinking water and vapor absorption, with the drinking water being most important. The concentration of tritium in the organic material of the animals will reflect the concentration in the food and the body water. Monitoring data taken at SRS suggest that the average tritium concentration in farm animals and plants grown near the facility is very similar to the average value of measured tritiated water vapor concentrations in the air in the same locality (Murphy et al. 1991).

A number of studies have reported concentrations of tritium in the organic fraction of vegetation or vegetable food that exceed the concentration of the water in the same vegetation (Stewart et al. 1972; Murphy and Pendergast 1979; Belloni et al. 1984; and Belot et al. 1986). In some cases, these studies have been used to suggest that tritium will accumulate in vegetation or be actively concentrated. However, this has not been observed in carefully controlled experiments where plants grown under equilibrium with tritiated water have slightly lower concentration than the water (Garland and Ameen 1979). In addition, the tritium concentration of the organic fraction of plants grown in areas far removed from tritium sources are nearly in equilibrium with the water in their environment (Hisamatsu et al. 1989). The latter results suggest that the elevated tritium levels are either from the slower turnover of organic matter than water following higher tritium exposure in the past or by uptake of tritium from another source which is at high concentrations (Murphy and Pendergast 1979), rather than from biomagnification.

Tritium in the aquatic environment

In this review the aquatic environment includes streams, ponds, and lakes. The behavior of tritium in these bodies of water depends on the relationship between the processes exchanging water in the system. Streams are dominated by the flow of water. Most of the water is from precipitation on the watershed of the stream. Water moving through a watershed can reach the stream by a number of surface or subsurface paths. Some of these paths have transit times that are on the order of several radioactive half-lives of tritium. Therefore, stream water may have an average tritiated water content that is lower than that of the precipitation falling on the watershed. Tritiated water is transported with the mass of water in which it is contained. The concentration of tritiated water added directly to a stream is determined by the processes of volumetric dilution, advection, dispersion, and turbulent diffusion

in the stream channel (Thibodeaux 1979). Measurements made at two locations on the Savannah River following an accidental spill of heavy water contaminated with tritiated water showed the peak tritium concentration had decreased by 43% in 87 miles of river transit (Buckner and Hayes 1975).

In pond systems, the volume of water entering the pond directly by rainfall and leaving by evaporation may be greater than input and output from other sources. Most pond systems are well mixed through the action of wind on the pond surface and can be characterized by an average tritiated water concentration. The tritium concentration in ponds can be greatly affected by vapor transport of tritiated water from the surface. In many cases, the residence time of tritiated water in a pond is determined by vapor exchange (Horton et al. 1971).

Tritiated water transport and cycling in lake systems includes all the processes identified for streams and ponds. River-lake systems are much like lakes except the long transit times in a section of these lakes allows for more mixing. Lakes with small water throughflow compared to their volume are more like ponds. Some lakes undergo a characteristic seasonal cycle of stratification. Stratification of lakes has the effect of decreasing the transport between layers of water within the lake. This may exclude part of the lake from tritium released into the lake. It may also restrict tritium transport out of the lake by some processes (Thibodeaux 1979). Because of the wide variation in the characteristics of lakes, residence times can vary from a few days to years.

Aquatic organisms. Since aquatic systems are almost totally immersed in water and generally have high water exchange rates, the tritiated water exchange between the environment and aquatic organisms is very rapid. The turnover time for tritiated water in the tissues of aquatic organisms is of the order of a few minutes to a few hours (Harrison et al. 1973; Morgan et al. 1973; and Blaylock et al. 1986). Therefore, the tissue water concentration is the same as that of the surrounding environment for all conditions other than releases lasting less than a few hours. The exception is aquatic macrophytes which are often only partially immersed and characteristically have lower tritium concentrations than other aquatic vegetation (Blaylock and Frank 1979).

The sources of organic tritium in aquatic systems are the incorporation of tritium from tritiated water by photosynthesis and the uptake of detritus of either terrestrial or aquatic origin. Algae and other aquatic plants are the source of photosynthetically fixed tritium. As in terrestrial plants, under carefully controlled laboratory conditions the tritium to hydrogen ratio in the organic material of aquatic plants grown in tritiated water is near 0.8 (Kirchmann et al. 1979). However, algae can incorporate organic material from the environment as well as make it by photosynthesis. In this case, the tritium in the organic material of the algae will be an average of the two sources. If the tritiated organics have higher concentrations of tritium than the water, the organic material of the algae will also have higher tritium concentrations than the water. This has been shown to be the reason for the relatively high tritium concentration of microorganisms found in small effluent streams receiving waste water containing reactor water purification resins (Kirchmann et al. 1986b).

The transfer of organic tritium from photosynthetic or detritus inputs through the food chain depends on the nature of the organic material consumed (Bruner 1973 and Blaylock et al. 1986). Aquatic consumer organisms raised in tritiated water but fed untritiated food have been found to have organic tritium to hydrogen ratios that are from 10 to 50% of the ratios in the water. When consumers are grown in tritium-free water but fed tritiated food, the tritium to hydrogen ratio of the consumer approaches 50% of that in the food. Few experiments have been run long enough, or controlled well enough, to ensure that all the organisms in a particular aquatic system are at a steady state with the environment; however, under conditions which most closely approach this ideal situation, the tritium to hydrogen ratios in the organisms of these systems generally range from 80 to 130% of the ratio in the environment (Murphy et al. 1991). None of these systems, either laboratory or natural, show any sign of biomagnification of tritium in the food chains.

Integrated tritium transport and cycling in aquatic systems. The residence time of the majority of tritiated water introduced into a body of water is determined by the turnover of water in the stream, pond, or lake. Only a small fraction of the tritium in an aquatic system will be found in organisms. However, there is the potential for some of this tritium to reach humans through the food chain. The tritiated water content of organisms in the body of water will closely follow the concentration in the water, while the organic matter concentration lags behind the concentration in the water (Adams et al. 1979).

Organic matter in sediments may have even lower turnover rates. Bogen and Welford (1976) suggested that the high tritium concentration measured in Hudson River sediments could be explained by the relatively high tritium concentrations in the environment when the organic matter in the sediments was synthesized, immediately following the introduction of weapons testing tritium into the environment, and the long turnover time of tritium associated with the organic residue in the sediments.

Given enough time, an aquatic system will reach a steady state and the concentration of tritium in all components of the aquatic system will be similar to the tritium concentration of the water. The exceptions are systems which have high inputs of tritium in organic matter. In these systems the steady state tritium concentration in organisms is intermediate to the concentrations in the organic matter and the water (Kirchmann et al. 1986b). In no case is there evidence of biomagnification of tritium in the food chain.

Tritium in the groundwater environment

In this section, the groundwater system is defined as the ground, or earth, beginning below the rooting depth of the vegetation and extending down to bedrock. Groundwater hydrology is a well developed field and no attempt will be made to review the literature on that subject here. The following short overview of the principles that effect tritiated water transport are summarized from de Wiest (1965)

In the absence of layers which restrict water movement, the general direction of flow below the rooting zone will be downward from the upper boundary or surface. When a retarding layer is encountered, the water will saturate the pore space in the soil or rock above the retarding layer. The upper surface of this saturated zone will be connected to the atmosphere by the unsaturated pore space above it and the upper surface of the saturated zone will be at atmospheric pressure. This unconfined, saturated surface is called the water table. Water in the pores above the water table is held in the pore space by capillary force and is under tension with respect to the pressure at the surface of the water table. Water in the water table is under hydraulic pressure from the weight of the water above it.

If the impeding layer producing the water table is inclined from the horizontal and/or the impeding layer reaches the surface (e.g., through the down cutting of a stream) the water table will also be inclined, leading to differences in the hydraulic head (the sum of the pressure and elevation geopotential) in the direction of the inclination. The water in the water table will flow, both vertically and horizontally, from areas of high hydraulic head to areas of low hydraulic head.

In many areas, the region below the water table will be made up of a succession of layers of material that conduct water (aquifers) or retard water flow (aquitards). Aquifers can be saturated or unsaturated with water but are often saturated. Some of the water in an aquifer may come from leakage through an aquitard. In many cases, the water enters the aquifer at some point where there are no confining layers between it and the surface. In this case, the geologic layer containing the aquifer is part of the water table and water enters directly by flow from the surface. These locations are often referred to as the source areas for the aquifer. Aquifers can also be fed by source areas under rivers or lakes.

Almost all of the tritium entering the groundwater system is in the form of tritiated water. Tritiated water in the groundwater system is transported by bulk flow from areas of higher hydraulic head to areas of lower hydraulic head. Tritiated water is diluted by other groundwater through the processes of hydrodynamic dispersion, a combination of mechanical mixing and molecular diffusion which causes a spreading of the tritium in the groundwater over a larger area. Most of the tritiated water in the saturated zone is associated with the bulk water in the pore space and moves as a nonreactive, nonabsorbed constituent within the groundwater system.

Under some conditions, tritium in the groundwater can become associated with water of hydration on mineral surfaces. The water of hydration has a number of components varying in their mobility in the groundwater. At ambient temperatures, little tritium is associated with water of hydration. Incorporation at higher temperatures is associated with underground detonation of nuclear explosives which can produce tightly bound tritium (Stewart 1973). The tightly held component of the water of hydration has little significance to tritium movement in most groundwater systems. However, Koranda (1965) found relatively high tritium concentrations in bound water of hydration in the soils of islands in the Pacific Proving Grounds where the tritium had become fixed under the high temperatures produced by the thermonuclear detonations.

The velocity of groundwater moving through aquifers varies through a large range, normally from 1.5 m/y to 1.5 m/d. In highly permeable aquifers, the velocity can be as high as several hundred m/d. With this wide range of permeabilities, which are often exponentially distributed in a given groundwater system, it is difficult to forecast the dispersion or residence time of tritium in a particular groundwater system without knowledge of the characteristics of that system. Groundwater tritium normally contributes to human exposure only if the water is used for drinking or irrigation, or if the tritium bearing aquifer outcrops into a surface water body.

Tritium in the ocean

The ocean is, by a large margin, the major global reservoir of hydrogen and, as such, has an enormous capacity for dilution of tritium. Given time, most of the undecayed tritium released to the environment will finally reside in the ocean. Tritium transport into the ocean from continental sources is through river water discharges, advection of continental air over the ocean, and deposition and discharge of groundwater directly into the ocean (Weiss et al. 1979).

River discharge takes place first through the flow out of the river estuary onto the continental shelf. Estuarine transport is partially controlled by the efflux rate of the river and partly by the tidal and coastal water currents in an area. The characteristic turnover time of water in most estuarine systems is along the order of days or weeks. The mixing of water between the continental shelf and coastal bays and inlets is driven by the local system of currents. In many cases, the mixing is not continuous but driven by events such as storms. The characteristic mixing in most coastal

areas seems to be on the order of months (Hayes 1979; Hetherington and Robson 1979).

Most of what is known about the transport of tritium in the ocean is from the study of the transport of tritium injected into the stratosphere by the nuclear test detonations in the early 1960's. Initial efforts to reconcile measurements of tritium entering the ocean and tropospheric air concentrations indicated that the deposition over the ocean must be greater than that over the continents. This was supported by the fact that the concentration of tritium in the atmosphere over the ocean was less than that over land, suggesting that it was being removed at a higher rate over the ocean. At first this was justified on the basis of a higher rainfall rate over the ocean. However later calculations suggest that a large portion of the tritiated water deposition into the ocean is by dry deposition (Weiss et al. 1979). The large wet and dry deposition rates for tritiated water are the reason for the relatively short residence time of tritiated water in the atmosphere when released at the surface of the earth.

Tritium is initially deposited into the surface "mixed" layer of the ocean. This layer is about 100 meters thick and mixing to this depth is relatively rapid. While the mixed layer has a very large dilution volume for tritium, an even greater volume exists below the mixed layer. Studies of the transport of tritium in the ocean have shown that mixing of the surface layer with the deep ocean takes place at a few locations where cold surface water sinks into the depths (Michel 1976; Ostlund and Fine 1979). At these locations, tritium transport is fairly rapid; however, the average tritium transport from the surface layer to the deep ocean is comparatively slow. The turnover time of the deep ocean is on the order of 100 y or more. This means that tritium reaching the deep ocean is likely to decay to extremely low levels before the water reappears on the surface.

TRITIUM EXPOSURE FROM RELEASES TO THE ENVIRONMENT

When tritium is released into the environment, the exposure to humans from any part of the system will depend on the magnitude of the transport into that part of the system and the residence time of the tritium after initial introduction. The processes that transport tritium in the environment and the storage capacities and residence times of components of the environment have been identified and described. The mode of release also has a strong influence on the exposure. From the previous discussion it is possible to identify those transport and cycling processes which are important under different release conditions.

For a short duration release of tritiated water into the atmosphere, most of the exposure will be from the environment close to the release site. The majority of the tritium will remain in the atmosphere as the tritium is carried away from the site by the wind. Therefore, the most important component in determining exposure is the atmosphere condition near the time of the release. The conditions include the amount of air humidity and the wind speed, and the mixing processes in the atmosphere that will dilute the release. Only the leaf water of terrestrial plants will approach the concentration of tritium in the air but the leaves will quickly lose tritiated water after passage of the release plume, decreasing the opportunity for exposure to humans by this path. Animals will absorb tritiated water from the air through inhalation and skin absorption but the ratio of intake to volume of water in an animal is smaller than in a plant leaf and the concentration of tritium will be lower, although the residence time will be somewhat longer.

In a release of tritiated hydrogen to the atmosphere, an even greater fraction of the tritium will remain in the atmosphere. However, tritiated molecular hydrogen has a low solubility and the uptake by organisms, including humans, is very low. The most important component of human exposure comes from oxidation of molecular tritium in the soil. The residence time is determined by the residence time of tritiated water in the soil. In terms of the total tritium entering the soil, the amount of tritium oxidized in the soil to tritiated water is very small. The only reason that this cannot be totally ignored is that the tritium dose to humans following atmospheric releases of tritiated hydrogen is almost totally from oxidation in the soil and the readmission of tritiated water into the atmosphere. However, since this process is much less efficient than direct absorption of atmospheric tritiated water vapor by man, the dose is always considerably smaller than from a similar sized release of tritiated water.

The exposure from a release of tritiated water to the soil is determined by the transport of tritium from the soil to the other parts of the environment. Tritiated water will be transported by bulk flow in roots and stems and by bulk flow and diffusion in leaves resulting in the concentration of tritiated water in the vegetation being somewhat lower than that in the soil. Animals will take up tritium in the vegetable or animal food they eat and from the water vapor that enters the atmosphere from the soil. The residence time of tritium in this case will be determined by the residence time in the soil. Human exposure is possible from water vapor re-admitted from the soil, especially through vegetation and consumption of food grown in the contaminated soil.

The exposure of aquatic systems following short duration releases is most affected by the physical residence time and dilution through mixing in the stream, lake, or pond. The water in aquatic organisms has a short residence time and the concentration tends to follow the concentration in the water. Only a small fraction of the tritium enters into the organic matter in these systems; however, the residence time in organics, particularly sediments, can by quite long, along the order of tens of years. Exposure is possible by immersion and drinking water. Exposure through the food chain is less important because of the rapid turnover of tritiated water in aquatic organisms and the low potential for incorporation of tritium in organic matter during a short duration release.

Groundwater and ocean systems are not affected greatly by short duration releases. In the case of groundwater, little tritium from short duration atmospheric or surface water releases will reach the groundwater. Only a release directly into the groundwater would be of concern. However, once groundwater has been contaminated, it can control the tritium half-life in streams which are fed by the groundwater system. Estuaries are, to some extent, extensions of the stream systems which feed them and act like the streams with the additional dilution due to mixing with coastal waters. The quantity of water in the open ocean is too large to be affected by any reasonable magnitude, short duration release.

When the environment approaches a steady state following long duration "chronic" releases of tritium, the concentration in different components of the environment is determined by the balance of concentration and flow from other components of the environment. Concentrations are characterized by the long term average, and exposure is this average integrated over the exposure time of interest (usually the lifetime of the individual).

In the vicinity of chronic atmospheric releases of tritiated water, the concentration in the soil will be similar to that in rainfall, which is usually lower than that in the air water vapor because the source of water in rainfall is from a location away from the site. The concentration in vegetation will be intermediate to the concentration in the air and the soil. Vegetation organic tritium concentrations will be slightly lower than those of the water in the vegetation. The concentration in the water of animals will be most strongly influenced by drinking water, which may or may not be rainwater. However, the concentration in animal water is also influenced by the air concentration and the concentration in the water of food. The tritium concentration in the organic matter of animals is influenced by the concentration in the food the animal eats and the water in the animal. The net effect of all of these transfers is that the concentration of tritium in organisms (including humans) chronically exposed to a tritiated environment is somewhat less, but very near, the average concentration in the environment.

The same type of balance determines the steady state tritium concentration in aquatic systems. However, the transport of water between the components of these systems is so rapid that almost all components have identical concentrations of water. Organic tritium transport in aquatic systems is somewhat different than most terrestrial systems because of the potential role of detritus (dead organic matter) originating from outside the system plays in some aquatic systems. In this case, the concentration of tritium in the organic matter of the aquatic organisms may be supplemented or diluted, depending on the ratio of tritium to the other isotopes of hydrogen in the detritus.

CONCLUSIONS

From this review we conclude that tritium transport and cycling in the environment can be explained without postulating large isotopic effects or biomagnification of tritium through food chains. However, care must be taken to identify all of the paths which lead to exposure to humans. Studies which show higher concentrations of tritium in a single component of the biota than expected from identified paths, have been traced to some other source of tritium which is itself at a high concentration or to differences in retention times after transient exposures. An example of this effect in a terrestrial system is the elevated levels of tritium in vegetation traced to high soil water concentration from oxidation of relatively high concentrations of tritiated hydrogen (Murphy and Pendergast 1979). An example in an aquatic system is the high organic tritium concentrations in organisms exposed to relatively high tritium concentrations of reactor water purification resins (Kirchmann et al. 1986b).

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Paper

ORGANICALLY BOUND TRITIUM

Silvia Diabaté and Siegfried Strack*

Abstract—**Tritium released into the environment may be incorporated into organic matter. Organically bound tritium in that case will show retention times in organisms that are considerably longer than those of tritiated water which has significant consequences on dose estimates. This article reviews the most important processes of organically bound tritium production and transport through food networks. Metabolic reactions in plant and animal organisms with tritiated water as a reaction partner are of great importance in this respect. The most important production process, in quantitative terms, is photosynthesis in green plants. The translocation of organically bound tritium from the leaves to edible parts of crop plants should be considered in models of organically bound tritium behavior. Organically bound tritium enters the human body on several pathways, either from the primary producers (vegetable food) or at a higher trophic level (animal food). Animal experiments have shown that the dose due to ingestion of organically bound tritium can be up to twice as high as a comparable intake of tritiated water in gaseous or liquid form. In the environment, organically bound tritium in plants and animals is often found to have higher specific tritium concentrations than tissue water. This is not due to some tritium enrichment effects but to the fact that no equilibrium conditions are reached under natural conditions.**
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Key words: tritium; metabolism; environmental transport; environmental assessment

INTRODUCTION

TRITIUM released into the environment as tritiated water in a gaseous or liquid form (HTO), as molecular tritium (HT), or as volatile organic substances results in labeling of the organic material in plants and animals. Of these, HTO is most relevant to the incorporation of tritium into living organisms and to forming organically bound tritium (OBT). OBT exhibits longer residence times in organisms than tritiated water. Quantitatively, the most important process of OBT production after a tritium release into the environment is photosynthesis in green leaves with HTO as a precursor. Tritiated assimilates synthesized in the leaves may be transported to humans over several trophic levels.

* Kernforschungszentrum Karlsruhe GmbH, Hauptabteilung Sicherheit/Biophysik und Ökologie, Postfach 3640, D-7500 Karlsruhe 1, Germany.
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In the past few years. the significance of OBT with respect to the dose has been recognized. The ICRP has recommended to consider, in estimating the doses to members of the public (ICRP 1990), the contribution made by the generation of OBT following the intake of tritiated water and after direct ingestion of OBT. New, age-dependent dose factors have been introduced to cover the ingestion of tritium as OBT. In advanced models, such as UFOTRI (the KfK code assessing the radiological consequences of accidental tritium releases), this recommendation has already been considered. Parametric studies with UFOTRI have shown that ingestion doses can be 4–23 times higher than doses due to inhalation and skin absorption, depending upon the weather conditions during and subsequent to a release of HTO into the atmosphere (Gulden and Raskob 1991).

This paper summarizes the available information about OBT generation and conversion processes, explains transfer processes and possibilities of modeling, and assesses the significance of OBT in dose estimates. A choice of relevant reviews and some pertinent papers are cited for further reading.

DEFINITION OF OBT

Tritium is bound organically either in exchange reactions or in enzymatically catalyzed reactions in which it replaces hydrogen. Tritiated organic matter is classified as a function of the fractions of the exchangeable and nonexchangeable bound tritium it contains.

Exchangeable tritium

Tritium-hydrogen exchange reactions occur in organic molecules with chemical groups such as

$$=NH;\ -OH;\ -SH;\ \text{or}\ -\overset{\overset{O}{\|}}{C}-CH_2-.$$

The first three hydrogen bonds can easily be exchanged by dissociation and the α-keto group can be transformed into an α-enol group from which hydrogen can dissociate. Since most organic compounds contain some exchangeable hydrogen, contamination with HTO will result in uniform labeling of the exchangeable hydrogen fraction in accessible positions within a few seconds; otherwise, hydrogen atoms at internal sites of coiled macromolecules are exchanged very slowly because of their inaccessibility to cellular water.

After an HTO exposure, the bulk of exchangeable tritium decreases as a function of the tritium concentration in cellular water. Tritium on internal sites and in the hydration water of biological macromolecules will be exchanged more slowly. The hydration water of biological macromolecules represents an integral part of the macromolecular structures and plays a crucial role in maintaining the structural and conformational integrity of all biological structures. Bonding to organic molecules is relatively strong because of electrostatic interactions (Mathur-De Vré and Binet 1984).

Nonexchangeable tritium

Tritium is incorporated into stable bonds to carbon atoms only via enzymatically catalyzed reactions. Since C-H bonds are stable, except in the presence of strong acids, strong bases, or catalysts, this fraction is called "nonexchangeable" OBT. Tritium from HTO may enter an organic molecule by enzymatic reduction processes, most of which involve a reduced coenzyme, and may be brought into exchangeable or nonexchangeable positions. Tritium in an exchangeable position can be relocated subsequently by further enzymatic reactions into a nonexchangeable position or vice versa. Initial incorporation of tritium by physical exchange and subsequent enzymatic reactions can also result in labeling of a nonexchangeable hydrogen position. Furthermore, HTO may be incorporated as a complete molecule, e.g., in the reaction producing malic acid from fumaric acid.

While tritiated organic compounds are transported within organisms or ecosystems, exchangeable tritium is removed, leaving the fraction bound to carbon in place. Therefore, dose estimates should only refer to nonexchangeable OBT. In this paper, the term OBT is used in this sense.

PROCESSES OF OBT FORMATION IN THE ENVIRONMENT

Plants and animals in the terrestrial and aquatic environment are the main contributors to processes of OBT formation. Although upper soil layers can contain significant amounts of OBT, most of this OBT is not produced in soil but originates from the metabolism of plants and animals as outlined in the following sections.

OBT formation in plants

Organisms containing chlorophyll, such as algae, mosses, or higher plants, are able to synthesize organic substances from inorganic carbon dioxide and water under the impact of light. Early exposure experiments with tritiated water and $^{14}CO_2$ showed photosynthesis to be the most important process in the photo-induced assimilation of nonexchangeable tritium and ^{14}C. Also in the dark, tritium may be incorporated at low rates, but ^{14}C uptake is much reduced. In chlorella algae (Moses and Calvin 1959), a three-times higher incorporation of tritium into nonexchangeable positions was observed in the light than in the dark. In soybean leaves (Thompson and Nelson 1971), the increase in assimilation in the light over assimilation in the dark was found to be 10-fold for tritium and 100-fold for ^{14}C. The distribution of tritium among the organic compounds was similar to that of ^{14}C in both experimental series. In experiments with wine grape and potato leaves, Guenot and Belot (1984) also found that the amounts of tritium and ^{14}C fixed in the light were nearly proportional.

The complex process of photosynthesis can be divided into light reactions and dark reactions. In light reactions, reduction equivalents ($NADPH_2$), energy equivalents (ATP), and molecular oxygen are produced by the photolysis of water. $NADPH_2$ and ATP are used in the dark reaction (Calvin cycle) to reduce phosphoglyceric acid, the first product of CO_2 fixation, to phosphoglyceric aldehyde. This is the primary reaction under light conditions in which tritium from HTO may be incorporated into a nonexchangeable bond to carbon. Tritium is also incorporated by subsequent highly reversible reactions, such as isomerization reactions.

Sugar monophosphates and phosphoglyceric acid are early products of the reduction processes, found after HTO exposure under light conditions for only a few seconds. Other tritiated substances, which also appear rapidly (aspartic acid, glutamic acid, and malic acid), can be attributed to the addition of water in the fumaric-acid-to-malic-acid reaction of the tricarboxylic acid cycle. Glutamic acid and aspartic acid also incorporate tritium by reductive amination to their respective keto acids (Moses and Calvin 1959).

Nonphotosynthetic assimilation in the dark is more important for tritium than for ^{14}C. After HTO exposure in the dark, the same compounds appear as after exposure in the light, but the relative quantities incorporated into amino acids, namely aspartic acid and alanine, are higher. Tritiated substances synthesized in the dark mainly associate with the tricarboxylic acid cycle and with pyruvate (alanine, malic acid, aspartic acid, and glutamic acid). Proportionally less tritium appears in sugar phosphates. Light and dark labeling with tritium differ in the absence of a *de novo* biosynthesis of organic compounds in the dark due to CO_2 fixation. Labeling with tritium in the dark implies a metabolic turnover of the substances built up by photosynthetic processes in the light. The rate of tritium incorporation into nonexchangeable positions depends on the turnover rate or metabolic activity of the molecular species to which tritium is bound.

All organic matter in organisms containing chlorophyll, higher plants included, originates from the products of photosynthesis. Consequently, tritium will also be incorporated into such complex molecules as polysaccharides, proteins, lipids, and nucleic acids.

Most of the plant mass is made up of polysaccharides (e.g., cellulose or hemicellulose) which occur in the cell walls of plants and serve as structural materials. Other polysaccharides are used as storage material, (e.g., starch in tubers or grains). The mean hydrogen content

The retention of OBT in various organs of rats after the administration of HTO was studied by Takeda and Kasida (1979). They observed high retention levels in those tissues in which the initial tritium concentration was relatively low, such as fatty tissue, brain, and muscle. This suggests that the excretion of OBT is determined largely by the metabolic turnover of those tissue constituents that are considered to be slow in their turnover.

In a long-term experiment, van den Hoek (1988) studied the half-lives of a number of organs in miniature goats which received tritium from their conception. Tritiated hay was given throughout the pregnancy period of some 150 d and also during the lactation period of ~80 d. At this time, the OBT administration was discontinued and the decrease of OBT levels in the organs of the young animals analyzed. As the tritium concentration is reduced not only because of metabolic processes but also because of new growth, a mathematical procedure was developed that yielded growth correction curves for individual organs. The resulting half-lives represent the decrease by metabolic processes only (Table 6).

For most organs showing a single component decrease of the OBT level, half-lives varied between 62 and 318 d. A very high value of 2,712 d was found for tendinous tissue, indicating a very slow metabolic turnover; however, the radiological significance of these tissues is low. Two different half-lives were obtained for other organs ranging from 5–78 d for the first compartment and from 57–328 d for the second compartment.

The decrease of OBT in the body is easily observed in analyses of OBT in secreta, such as milk. Tritium deposited as fat or protein in certain tissues of the body may be mobilized later and reutilized for the synthesis of milk components. The half-lives of OBT in milk components (Table 7) determined after HTO administration to a lactating cow are composed of two compartments. The slow components represent only 4% of the total tritium in milk under conditions of continuous HTO uptake and <0.1% after a single dose (van den Hoek et al. 1983).

Since only tritium atoms located within the cell nucleus can contribute materially to radiation damage, Commerford et al. (1982) studied the half-lives of tritium in the components of cell nuclei of mice after a transient exposure to tritiated water. The tritium concentration in DNA of rapidly proliferating tissues, such as bone marrow and testes, declined rather fast at half-lives of 6.9 d for the first component (90%) and 231 d for the second component (10%). In liver, some 70% of the original tritium present in DNA decreased with a half-life of 182 d and in brain about 90% decreased with a half-life of 257 d. The metabolic turnover of nuclear components, particularly DNA, is very slow in relation to the turnover of water; therefore, only small amounts of tritium are incorporated into these components after a transient exposure to tritiated water. Nevertheless, their contribution to the radiation dose may be significant in cells dividing at the time of exposure and may persist for long periods of time.

Table 6. Half-lives of OBT in organs and tissues of minigoats receiving OBT from conception on (van den Hoek 1988).

	Biological half-lives (d)	
	First component	Second component
Heart muscle	80	
Skeletal muscle	128	
Involuntary muscle of urinary bladder	318	
Subcutaneous and omental fat	45	
Small intestine	62	
Spleen	71	
Adrenal	92	
Uterus	105	
Abomasum	111	
Large intestine	117	
Testicle	132	
Pancreas	208	
Skin	242	
Tendinous tissue	2,712	
Thymus	5	57
Lymph gland	7	85
Liver	13	96
Kidney	14	124
Lung	23	224
Rumen	42	161
Gallbladder	78	239
Brain	29	328

Table 7. Half-lives of tritium in milk components after chronic administration of HTO to a lactating cow (van den Hoek et al. 1983).

	Biological half-lives (d)	
Milk component	First compartment	Second compartment
Milk water	3.6	43.7
Lactose	3.6	43.7
Milk fat	4.1	224.8
Casein	4.3	24.3

Effect of OBT on dose. As previously shown, the rate of tritium incorporation into the organic pool of the body depends on the duration of exposure and on the chemical nature of the compound in which tritium is taken up. The following findings about the contribution of OBT to the dose were condensed from comprehensive review articles (Commerford 1984; Myers and Johnson 1991).

After a single exposure to tritiated water, much less tritium will be incorporated into the nonexchangeable organic compartment than into the water compartment, but the relative contribution to the radiation dose is nearly the same because of the longer half-life of OBT. The dose may even exceed that from tritiated water when tritium is incorporated into proliferating cells which synthesize DNA, for instance in embryos (Commerford et al. 1982).

Although chronic exposure to tritiated water results in higher tritium incorporation into the OBT compartment, the dose increase due to OBT amounts to only a few percent because nearly all the tritium in the cell nuclei is present as HTO in this case.

When tritium is ingested as tritiated food, the OBT compartment becomes relatively more important because more tritium is incorporated. In this case, the dose may be higher by a factor of about 2 for single and chronic exposures.

OBT generation in aquatic systems

Nearly all the tritium released into the environment reaches the hydrosphere unless it is lost by physical decay. HTO is taken up by aquatic organisms very quickly because they are totally immersed in water. For the same reason, the half-lives of tritium are short, for example 0.5 min for planktonic algae (Blaylock et al. 1986) and 0.5 h for fish (Rodgers 1986).

Algae are the primary producers of organic matter in aquatic systems. Tritium from HTO may be incorporated into organic matter by photosynthetic reduction processes as previously described. Under steady-state conditions, the specific activity of OBT is lower by a factor of 0.5–0.8 than that of the ambient water, due to isotopic effects (Weinberger and Porter 1954; Kanazawa et al. 1972; Strack 1978).

However, in algae grown in effluents of nuclear facilities, much higher OBT concentrations were found than in ambient water (Kirchmann et al. 1977b). This is because algae are additionally able to incorporate dissolved organic compounds from the external medium. Such tritiated biomolecules, which originate from plant or animal secreta or from dead and autolyzed organisms, occur in effluents containing tritium. These compounds may be incorporated selectively into algae as well as into mussels and fish and accumulate to significant amounts as shown in Table 8. In experiments with algae, in which synthetically tritiated biomolecules were used as precursors, most compounds were taken up within 30 min. In many cases, the intracellular concentration of the compound to be taken up is lower than that in the medium, but some compounds are concentrated in algae to quite high levels (Strack et al. 1983).

Table 8. Uptake of various tritiated organic compounds by Dunaliella bioculata and Acetabularia mediterranea (according to Strack et al. 1983).

	Specific activity ratios (intracellular/extracellular)	
	Dunaliella bioculata	Acetabularia mediterranea
Thymidine-methyl-^{3}H	0.8	1.8
Adenine-2-^{3}H	122.7	4.6
Uridine-5-^{3}H	2.0	0.1
L-leucine-4-^{3}H	11.4	0.3
Glycine-2-^{3}H	1.1	0.1
L-arginine-3.4-^{3}H	0.6	5.1
L-aspartic acid-2.3-^{3}H	1.0	2.3
L-phenylalanine-2.3-^{3}H	0.5	2.6
D-glucose-1-^{3}H	0.4	5.7
D-glucose-6-^{3}H	0.9	2.7

Since algae incorporate tritium from organic material in the environment as well as in photosynthetic reactions with HTO, the OBT concentration in algae will be an average composed of the two sources.

Fish, as secondary or tertiary consumers in aquatic food webs, may obtain tritium from the ambient water as well as from their food of plant or animal origin. The processes of incorporation are similar to those described for terrestrial animals. The nonexchangeable organic compartment into which tritium may be incorporated amounts to some 13% of the total hydrogen in fish. Tritium incorporation into this compartment is more effective when tritium is available as tritiated food rather than tritiated water, and is a maximum when both sources are tritiated, as has been shown experimentally in rainbow trout (Rodgers 1986). This could explain the instances reported of higher specific activities in OBT relative to ambient water observed in fish living in their natural habitats (Kirchmann et al. 1979).

BEHAVIOR OF OBT IN ECOLOGICAL SYSTEMS

To reflect the distribution and movement of OBT in terrestrial ecosystems, the soil, vegetation, and animals must be considered as subsystems. In this approach, humans are the final consumer in the human food chain.

The soil

The deposition and dispersion of tritium as HTO or HT in soil has been studied in great depth; however, little is known about the fate of organic matter labeled with tritium once it has entered the soil. The soil is penetrated by roots of terrestrial plants and populated by innumerable genera and species of animals and microorganisms. The bulk of organic matter in the soil is supplied permanently or periodically by plants or plant residues. It is degraded, altered, metabolized, and finally mineralized by heterotrophic organisms. Organic material in upper soil layers typically consists of 70% of substances decomposed relatively easily, such as simple carbohydrates, hemicelluloses, and cellulose. Lignin (~20%), proteins (often associated with tannin or lignin), resins, oils, waxes, lipids, etc. are more resistant to microbial digestion and contribute more to the generation of humus, a complex mixture of substances such as humin, humic acid, and fulvic acid.

Where tritium is bound in nonexchangeable positions of the original organic plant material, this humic fraction may represent a long-term tritium reservoir in the soil, depending on the rate of decomposition processes, which is higher in a temperate climate than in an arctic climate.

Measurements in soil of OBT originating from fallout were conducted by Bogen and Welford (1976) in the 1960s. From 1963–1970, those authors observed an exponential decrease of tritium concentration in one soil sampling site in Oklahoma. From these data they estimated a half-life of 3.5 y for OBT, which corresponds to a mean residence time of 5 y.

In many steps of breakdown and condensation, most of the original hydrogen is lost and substituted by hydrogen from the ambient water. The direct uptake of organic compounds by plants is a possible pathway, as is known from studies of pesticides; however, it seems to be insignificant because autotrophic organisms synthesize their biomass from mineral substances. No uptake of tritiated glucose by plant roots has been demonstrated (Momoshima et al. 1991; Papke and Förstel 1991).

In the decomposition processes, on the other hand, nonexchangeable bound tritium will be converted to HTO which will be introduced into the soil water compartment and may become available for plant uptake through the roots. The storage capacity for tritium in soil may be slightly enhanced by organic binding; however, this process cannot be responsible for the enhanced specific activity ratios observed in vegetation, as will be discussed in the next section.

The vegetation

Terrestrial plants are supplied with water by the transpiration stream from the soil to the leaves. Tritium in the tissue water of foliar organs also originates from HTO in the atmospheric moisture taken up by gas exchange through stomata in the leaves, a process described in detail in the past few years (e.g., Belot 1986).

Aqueous tritium from both sources can enter the organic fraction of the plant biomass by photolytic splitting of HTO and reduction of CO_2 in the chloroplasts. The assimilates produced by photosynthesis are used at the beginning of a vegetation period for synthesis of the structural material of the leaf organs. After leaf formation has been terminated, the newly formed assimilates can be stored temporarily in the chloroplasts and then used for the growth of roots and stems. Later in the vegetation period, usually after flowering, most of the assimilation capacity of the plants growing in zones of temperate climates is used in the evolution of fruits and tubers. Seasonal differences in these processes of evolution and translocation must be considered in assessing OBT concentrations in plants for dose assessments under nonequilibrium conditions. The grass consumption by animals, of course, can be modeled in less complicated ways.

As previously mentioned, the majority of the hydrogen is associated with the tissue water fraction; from stoichiometric calculations, the long-term OBT compartment is known to be ~3% of the fresh plant material. This organic compartment usually will not reach an equilibrium with the tritium content in the water fraction under dynamic conditions. Murphy (1990) reported observations that <0.3% of the tritium deposited in plants was incorporated in the organic fraction. If the total deposition onto plants is taken to be <10% of the HTO released from a stack (within 25 km from the release point), only 0.03% of the tritium initially released is seen to enter the OBT compartment. This small fraction of OBT, relative to tritium turnover in aqueous compartments, should be remembered when considering the residence times of tritium in vegetation systems.

Most measurements of tritium in environmental samples show enhanced tritium concentrations in the OBT fraction, compared to the tritium concentration in the tissue-free water (TFWT) fraction, thus resulting in a ratio between these two concentrations greater than unity. Examples from all over the world are summarized in a review by Brown (1988).

In 1979, Bogen et al. (1979) reported ratios in the range of 4 for vegetation samples. Belot (1986) measured OBT concentrations in vegetation from different sampling sites in France by a special method allowing any contamination due to a rapid exchange with water vapor in the laboratory to be discounted. The OBT concentrations of these samples exposed only to fallout tritium in most cases exceed the TFWT concentrations, which were below their detection limit of 6 Bq L^{-1}. Simultaneous measurements of tritium in soil proved that the activity of soil organic matter does not exceed that of plant material.

The authors therefore concluded that tritium previously incorporated into the soil over the period of maximum fallout cannot be responsible for the enhanced concentration ratios. The possible explanation they gave for those elevated OBT concentrations is the direct incorporation (without involvement of HTO) of gaseous tritiated hydrogen or methane which can be detected in the global atmosphere at higher specific activity levels than HTO. However, the deposition rates determined experimentally of atmospheric HT-to-OBT in plants were on the order of 10^{-9} to 10^{-10} m s^{-1} (Sweet and Murphy 1984; Murphy 1989; Strack et al. 1991). Observing rates of uptake that low is not a convincing explanation of ratios greater than unity in environmental samples.

In contrast to the rather inert HT and tritiated methane gases, tritiated formaldehyde seems to be taken up at a rate exceeding even that of HTO (Belot 1992). More work is needed to elucidate the significance of such pathways of tritium into plant organic matter. Belot concluded that an irreversible process of OBT generation normally would result in a considerable accumulation of OBT in vegetation. An effect of tritium accumulation in the organic fraction should be most evident in aerial plant organs with life spans longer than one vegetation period, such as pine needles. In Japan, extensive measurements of tritium concentrations in pine needles were conducted by Takashima et al. (1989). In 21 samples collected at different locations situated far enough from nuclear facilities to be unaffected by atmospheric tritium releases, the OBT:TFWT ratios observed ranged from 0.9–2.1, with an average of 1.5. These ratios are quite similar to those determined by Hisamatsu et al. (1987) in other plant samples, such as fruit and vegetables grown in a single vegetation period in Akita, Japan (e.g., 1.5 and 1.2 for fruits and green vegetables in 1985). In 1986, Hisamatsu et al. (1989a) observed ratios of 2.4 and 1.5 in the same groups of vegetable samples collected during June to July, and of 1.3 and 1.0 in samples collected during October to November, respectively.

A remarkable exception in all the diet samples observed by Hisamatsu et al. (1987, 1989a) are the concentration ratios in polished rice of 0.57 ± 0.12 in 1985, and 0.67 ± 0.11 and 1.3 ± 0.3 in 1986. Also Inoue and Iwakura (1990) determined tritium concentrations in Japanese rice and found a concentration ratio below unity: 0.83 ± 0.32. The authors gave an interesting explanation of their findings, saying that only rice plants show activity ratios below unity in most cases. The special rice cultivation technique seems to give rise to a ratio which is low compared to those of all other terrestrial plants. For most of its growth period, the rice plant is covered by irrigation water, and tritium in the tissue water of the plant and the rice grain is almost in equilibrium with HTO in the irrigation water. The influences of fluctuations in tritium concentrations in atmospheric humidity seem to be negligible when compared with the growth conditions of other terrestrial plants. Therefore, the authors compare their results with the findings of Garland and Ameen (1979), who observed activity ratios of 0.73 in barley and 0.64 in maize grown 30 d under equilibrium conditions in a controlled environment. In these cases, the ratios reflect only a discrimination due to an isotopic effect during photosynthetic assimilation of HTO.

The studies by Inoue and Iwakura (1990) suggest, as one reason for enhanced specific activity ratios in terrestrial plants, the existence of nonequilibrium conditions during growth. Even in regions far away from known tritium emitters, changing wind directions (e.g., from the ocean or from the continent) can give rise to temporarily different tritium concentrations in the atmospheric humidity. As the tritium concentration in tissue-free water of plants is affected by the tritium concentration in soil water as well as in atmospheric moisture, depending on diurnal trends of relative humidity in the atmosphere, different specific tritium concentrations in both sources can lead to variable TFWT concentrations in plants. A longer residence time of OBT, compared to HTO, in leaves may lead to specific activity ratios greater than unity, as is well documented in studies of tritium concentrations in plants grown under dynamic conditions in the vicinity of nuclear installations (Strack 1989).

The effect on OBT:TFWT ratios of translocating tritiated organic compounds into fruits and tubers under nonequilibrium conditions has been demonstrated by Spencer (1984) in well-controlled chamber experiments with tomato plants. He found ratios of up to 1.92 in the green fruits of tomato plants exposed to atmospheric HTO. The results show that the fruits have little exchange with the atmospheric moisture, hence they maintain a low TFWT value; however, the fruits incorporate photosynthetic products labeled with tritium in nonexchangeable positions and translocated from the foliage. This observation is more evidence proving that enhanced specific activity ratios in environmental plant samples are not caused by tritium accumulation or enrichment in the vegetation.

Isotopic effects during sample preparation may also contribute to the specific activity ratios greater than unity observed in plant samples. Baumgärtner and Kim (1990) suggested that tritium enrichment in the organic fraction could be due to the sample preparation, in particular to freeze-drying. Freeze-drying is the usual way of removing tissue water from samples. Because of the low temperature in the sample during water removal, isotope fractionation can cause tritium to be concentrated in the residual moisture of the sample. This water can equilibrate with the exchangeable hydrogen atoms in the organic matter and can result in a higher OBT value, unless the exchangeable OBT is removed.

Animals and humans

Processes of OBT uptake and turnover in animals and humans have been previously described. Living organisms have no site where hydrogen, and hence also tritium, could accumulate in contrast to iodine, for example, which accumulates in the thyroid. Furthermore, experiments with various animals and analyses of animals living in environments of enhanced tritium levels do not indicate tritium enrichment in any compartment against a concentration gradient. Under well-controlled equilibrium conditions, the mean specific activity ratio, OBT to TFWT, in the organs of rabbits showed a value of 0.98. In the experiment (Moghissi et al. 1987), the rabbits were exposed to constant tritium concentrations in drinking water and food for three generations.

Evans (1969) analyzed various tissues in deer chronically exposed to tritium on the Savannah River Plant site. Variations in atmospheric releases caused fluctuating tritium concentrations in the environment of the animals, thus representing typical nonequilibrium conditions. The results show that, in most tissues, the tritium concentrations in the body water equal those in organic matter, reflecting more or less the tritium concentrations in the environment. The most probable specific activity ratios in the animals were found to be between 0.85 and 1.0.

Only a few publications exist about OBT concentrations in human tissues. In a recent publication by Hisamatsu et al. (1989b), tritium concentrations in Japanese human tissue samples were reported and compared with earlier data published by Bogen and Welford (1976) and Belloni et al. (1984). While the American