ENVIRONMENTAL TRITIUM AND THE DOSE TO MAN

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Abstract

The dose to man from acute or chronic intake of tritiated water has been determined using a three compartment model to describe the retention of tritium in loose water and in bound organic form in the body. The retention times determined from occupational exposure cases are reviewed. The total tissue dose, using representative retention half-times for the three components of 9, 30, and 450 days, is 84 mrad per 1 mCi intake by standard man, 84% of which is due to tritium in body water and 16% to bound tritium in tissue. The record of environmental tritium concentrations in surface waters has been compiled. Maximum levels caused by weapons testing occurred in 1963-64. Subsequent decrease is exponential with a half-time of 3.2 years. By relating environmental tritium levels to daily intake by man and applying the dose model, the dose commitment of 1.5 mrad from fallout tritium was determined.

Introduction

Tritium is produced naturally by cosmic ray interactions in the atmosphere. Superimposed on the natural tritium background are varying amounts of man-made tritium. Nuclear weapons testing activity introduced substantial amounts of tritium into the environment, although the levels of fallout tritium are currently declining. The expanding nuclear power industry is expected to cause increasing amounts of tritium to be released. These continual additions of man-made tritium to the environment are cause for repeated monitoring of environmental levels of tritium and for re-evaluation of the dose consequences.

In this study a detailed tritium dose model is presented which allows one to determine accurately the contribution to tissue dose from tritium in loose water in tissue and from tritium combined in tissue following acute and chronic intakes. The experience from occupational exposure cases is reviewed to indicate the average half-times of the retention components. A record of tritium concentrations in environmental surface water is compiled. By relating these levels to the daily tritium intake by man and applying the dose model, the dose commitment from weapons-produced tritium has been computed.

Tritium Retention in Man

The experience from observations of human cases of accidental tritium exposure is summarized in Table 1. The initial rate of elimination of tritium from the body is exponential with a half-time ranging from 4 to 18 days.[1-8] Identification of additional retention components is limited to the few cases reported with intakes large enough to allow relatively long-term monitoring of tritium excretion. In the 415 day observation of a case, Sanders and Reinig[9] identified three retention components of half-times 6.1, 23, and 344 days. The first component can be associated with retention of free water in the body and the other two components with bound tritium in tissue. It should be noted, that a diuretic was administered in this case from the 3rd to 35th day following the exposure, which undoubtedly increased the turnover rate of body water.

TABLE 1

TRITIUM RETENTION HALF-TIMES IN MAN

Year	Investigator	Cases Studied	Notes	T_1	T_2	T_3
1951	Pinson, Anderson[1,2]	9	range 9.3-13	11.3		
1957	Fallot et al.[3]	20	range 5-11	8.5		
1960	Foy, Schnieden[4]	10	high ambient temp.	7.5		
1962	Richmond et al.[5]	5		9.5		
1963	Wylie et al.[6]	7	range 6-12	8.5		
1965	Butler, Leroy[7]	310	range 4-18	9.5		
1966	Osborne[8]	30	range 6.4-14.4	10.5		
1968	Snyder et al.[10]	1		8.7	34	
1968	Sanders, Reinig[9]	1	diuretic used	6.1	23	344
1969	Minder[13]	1			10-30	139-230
1971	Lambert et al.[12]	1		9.1	36	
1972	Moghissi[14]	3			21-26	280-550
			Reasonable Range:	8.5-11	20-36	200-550
			Assumed Average:	9	30	450

Synder et al.[10] studied another case for 255 days and identified two retention components of half-times 8.7 and 34 days. No particular treatment procedure was applied to this individual following exposure. Further analysis of this data has shown that the data are not inconsistent with a fit by three components of half-times 8.7, 30, and 550 days.[11]

Another case was recently reported by Lambert, Sharpe, and Dawson.[12] Retention half-times of 9.1 and 36 days are very similar to the results reported by Snyder et al. The observation period, 161 days, is not quite long enough to allow a definitive fit with three retention components.

Approximate long-term retention components have been inferred from a few additional studies of occupational exposures.[13,14] The differences in retention times in the cases reported allude to the variability which can be anticipated due to differences in metabolism, age, water intake, ambient temperature and treatment procedures. Reasonable ranges of the retention components have been indicated in Table 1. Approximate average retention half-times of 9, 30, and 450 days have been assumed for the model calculations.

Tritium Dose Model

Acute Intake

The dose to tissue following an acute intake of tritium arises from tritium in body water and from bound tritium in tissue. In computing the dose to tissue, the combined tritium in tissue is often neglected, it being regarded as an insignificant contributor to the total dose. Alternatively, the dose to body water (or a tissue with 100% water) is computed, this dose being a conservative estimate of the total tissue dose.[15] Since the tissue dose is more a measure of the hazard than the water dose, it is useful to determine accurately the dose to tissue.

A three compartment model is necessary to reflect the three retention components that have been identified. Sanders and Reinig[9] suggested the model represented by the diagram in Figure 1. A is the body water compartment, and B and C are bound hydrogen (tritium) compartments. The transfer coefficients represents constant fractional exchange rates of the compartment hydrogen (tritium) contents.

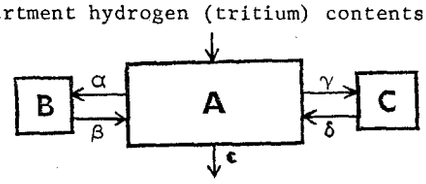

Figure 1. Three compartment model of tritium in the body.

A Markov chain calculation is used to determine the tritium concentrations in the three separate compartments at various times after intake. The fraction of assimilated

tritium in each compartment, f_A, f_B, f_C, are computed for as many iterations as may be required (each iteration representing one day). In analyzing the occupational exposure cases, combinations of exchange and elimination rates were selected so that close agreement was obtained with the measured fractional assimilated tritium in the body water.[9,11] For other calculations the transfer coefficients correspond to assumed retention half-times and compartment sizes.

The cumulative dose to tissue from an acute intake of HTO is computed from this model as follows:

$$\text{Dose (wet tissue)} = .75 \int_0^\infty q_A f_A dt + .25 \int_0^\infty (q_B f_B + q_c f_c) dt$$

The first term is the dose to tissue due to tritium in loose water within tissue. The second term is the contribution to tissue dose from tritium combined in tissue. Wet tissue is assumed to consist of 75% water and 25% tissue solids. A 70 kg man contains an average 42 kg water and an estimated 10 kg active tissue solids (70 kg - 42 kg water - 13 kg fat - 5 kg mineral bone).[16] The specific activities of tritium in the free water and bound compartments following an intake, I (mCi), are q_A = I/42 mCi/kg and $q_{B,C}$ = I/10 mCi/kg times the fractional amount of the intake in these compartments, f_A, f_B and f_C, respectively. The relationship .29 mrad per mCi day/kg is used to convert the units.

A calculation based on average retention half-times of 9, 30, and 450 days results in an estimated total tissue dose of 84 mrad per mCi intake.[11] This dose is due 84% to tritium in water within the tissue and 16% to combined tritium in tissue. Fifty percent of the total tissue dose is delivered within 11 days, and 90% of the dose is delivered within 200 days. The bound hydrogen compartment sizes were inferred to be 120 g for B and 600 g for C, consistent with the total amount of tissue solids (10 kg) consisting of ~7% hydrogen. A maximum of 0.7% of the tritium intake becomes combined, this maximum being reached in about 20 days. The rapidly declining concentration of tritium in the body water allows only relatively small amounts of the intake to become combined. The bound tritium in tissue is, thus, not the major contributor to the total dose to tissue following a single intake of HTO, but its contribution (16%) is not insignificant.

In additional model calculations with the first component half-time T_1 varying from 6 to 12 days with 30 and 450 day combined components, an empirical relationship is obtained.

$$\text{Tissue Dose} = 9\ T_1 + 3 \text{ mrad per mCi intake}$$

The relationship reflects a direct dependence of the amounts of bound tritium on the amount of tritium in the source reservoir, the body water compartment. For the assumed compartment sizes and within reasonable variation of transfer rates from the bound compartments, the second and third component half-times are not required in this relationship. The formula should be generally useful in providing an approximate estimate of the tissue dose following observation of the initial removal rate.

Intake of tritium other than in the form HTO may require special consideration. Bound tritium compounds in food may be more directly assimilated into bound compounds in tissue. Further study will be required to give the significance of the form of the tritium intake.

Chronic Intake

The model can be used for chronic intake situations by computing the compartment contents on a daily basis. For chronic intake of 1 μCi/ℓ, the equilibrium dose rate to active wet tissue is 95 mrad/yr. The dose to body water is

$$.001 \frac{\text{mCi}}{\text{kg}} \times .29 \frac{\text{rad/d}}{\text{mCi/kg}} \times 365 \frac{\text{d}}{\text{yr}} = 106 \frac{\text{mrad}}{\text{yr}}$$

The dose to tissue containing 75% water is 106 x .75 = 80 mrad/yr, neglecting the dose due to tritium combined in tissue solids. The dose to active tissue is thus 1.2 times the dose due to HTO in tissue.

The dose model assumes that the tritium becomes uniformly combined in actively

metabolizing tissue and that all of the hydrogen of active tissue solids is exchangeable. These assumptions lead to conservative estimates of the dose.

In the model calculations, a water balance of 3.08 ℓ/d has been assumed, based on the 42 kg body water compartment size and transfer coefficients corresponding to the 9, 30, and 450 day retention half-times. For an HTO intake of 1 mCi/d, the equilibrium specific activity is

$$1 \text{ mCi/d} \div \left[3.08 \frac{\text{kg } H_2O}{\text{d}} \times \frac{1 \text{ kg H}}{9 \text{ kg } H_2O}\right] = 2.92 \text{ mCi/kg H}$$

The tritium content of each compartment at equilibrium (the hydrogen content times the specific activity) is 13.6, .35, and 1.75 mCi for A, B, and C, respectively. Radioactive decay allows the long half-time compartment, C, to reach only about 90% of the intake specific activity. The equilibrium dose rate to wet tissue is

$$\text{A} \quad \frac{13.6 \text{ mCi}}{42 \text{ kg } H_2O} \times \frac{.75 \text{ kg } H_2O}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 70 \frac{\text{mrad}}{\text{d}}$$

$$\text{B} \quad \frac{.35 \text{ mCi}}{10 \text{ kg solids}} \times \frac{.25 \text{ kg solids}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 2.5 \frac{\text{mrad}}{\text{d}}$$

$$\text{C} \quad \frac{1.75 \text{ mCi} \times .90}{10 \text{ kg solids}} \times \frac{.25 \text{ kg solids}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 11.4 \frac{\text{mrad}}{\text{d}}$$

$$\text{Total:} \quad 84 \frac{\text{mrad}}{\text{d}}$$

As expected, the dose commitment following a single intake (84 mrad per mCi intake) becomes the equilibrium dose rate for chronic intake (84 mrad/d per 1 mCi/d intake).

Fallout Tritium

Levels in Surface Waters

Tritium is produced by fusion bombs and also by neutrons released during fission bomb explosions and in small amounts by the fission process itself. The amount of bomb-produced tritium is uncertain. Eriksson estimated that 1900 MCi were produced from 1952 through 1962.[17] This compares with natural tritium production of around 2 to 6 MCi/yr. Additional amounts of tritium have been released into the atmosphere by French and Chinese weapons testing conducted since 1964, but the total is a small fraction of that released during the earlier testing period.

The effect of weapons-produced tritium on concentrations of tritium in the environment, in drinking water, food and in man has not been monitored in great detail. Data from the U. S. Geological Survey's program of analyzing river water for tritium content provides the most useful data for ascertaining the tritium levels in environmental waters in the U. S. The data for 1961-68 for 20 streams throughout the U. S. have been published.[18] Preliminary data for 15 rivers for 1969-70 have also been obtained.[19] Tritium data from the earlier testing period, 1952-61, are not nearly as complete. Measurements of Mississippi River water for 1954-57[20,21] and of Rio Grande River water for 1957-58[22] have been included, and comparisons were made with Ottawa River data[23] in arriving at representative average tritium levels.[24]

Figure 2 shows the average U. S. data. The concentration of tritium in environmental waters reflects the weapons testing activity. Sharp increases are indicated following the testing series in 1954, 1956 and 1958. The very active test period in late 1961 and 1962 caused the peak concentrations in U. S. rivers in 1963-64. The declines in concentration during the 1959-60 moratorium and after the 1963 Test Ban Treaty are evident. Recent high yield tests in the Northern Hemisphere by the Chinese may be responsible for the relatively higher tritium levels measured in 1969-70 as compared to the previously declining values.

The average U. S. river tritium concentrations declined with a half-time of 3.2 years from 1963 until 1969 and 5.0 years during 1969 and 1970. Tap water analyzed at the Health and Safety Laboratory in New York City reflecting the lower Hudson Valley watershed area, showed somewhat lower tritium concentrations than the U. S. average in 1970. More recent tap water samples would indicate that the U. S. average beyond 1970 resumes the 3.2 year half-time decline. The absence of high yield atmospheric tests in the Northern Hemisphere in the past two years would also support this assumption.

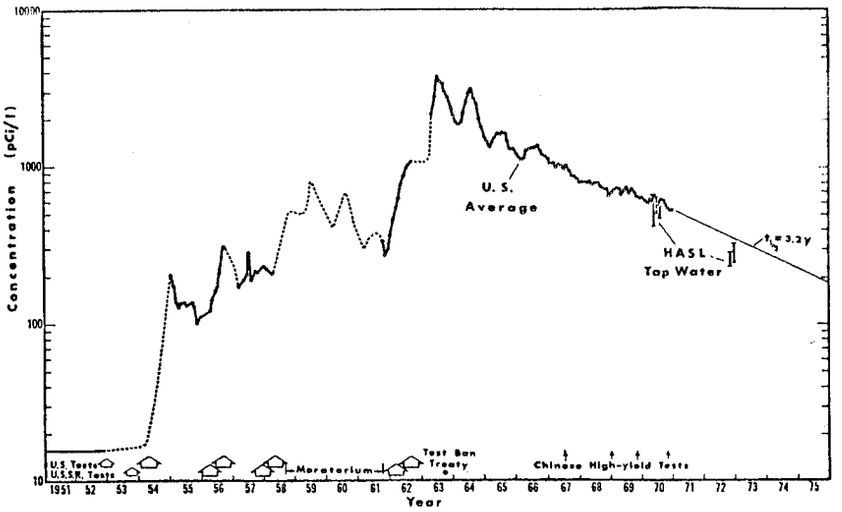

Figure 2. Environmental tritium in surface waters.

Tritium Intake

If a relationship can be found between the variations in tritium levels in environmental waters and the variations in the amounts of tritium intake by man or the levels of tritium in the body, the dose to man can be determined. The data of Bogen[25] show the relationship for New York in 1970 between the tritium levels of drinking water (530 pCi/ℓ), loose water in food (860 pCi/ℓ), oxidation water of food (2050 pCi/ℓ), and water vapor in air (930 pCi/ℓ). The drinking water intake by man can be assumed to be 1.4 ℓ/d. Intake by inhalation and transpiration of water vapor is computed to be .13 ℓ/d, based on average air temperature and humidity. The food water intake, based on analysis of food items of a standard consumption diet, was found to be 1.26 ℓ/d loose water and .29 ℓ/d oxidation water.[25] The effective concentration of the total tritium intake is thus:

$$(1.4 \times 530 + .13 \times 930 + 1.26 \times 860 + .29 \times 2050) \div 3.08 = 825 \text{ pCi}/\ell$$

These data indicate that, currently, one should assume somewhat higher concentrations of tritium intake than indicated by the tritium concentration in drinking water. During the earlier fallout period, when environmental levels of tritium were increasing, the tritium concentrations in water were probably higher than in food. Since specific data are lacking, it is probably most reasonable to assume that the tritium levels in surface waters reflect directly the tritium intake by man.

The Dose Commitment

The average natural concentration of tritium in environmental waters (16 pCi/ℓ) results in a dose to man of 1.5 μrad/yr, based on the three compartment dose model. The dose commitment to man from weapons-produced tritium is the dose due to the increases in tritium intake concentrations above the natural level. Extrapolation of the increased levels beyond 1972 is necessary to account for the exposure which is yet to be experienced. Assuming a 3.2 year half-time for weapons-produced tritium in the environment beyond 1970, a return to the natural background tritium level occurs in 1987.

The tritium intake is assumed to be 3.08 ℓ/d times the effective tritium intake concentration (the average tritium concentration in surface waters). The dose model is used to determine the bound and loose tritium compartment contents and the dose for the entire period that the tritium levels are above the natural background. The dose commitment thus determined is 1.5 mrad. Details of the calculation are shown in Table 2. The highest annual dose due to weapons-produced tritium was .21 mrad in 1963 and 1964. The calculation assumes no further atmospheric weapons testing and takes no account of future tritium releases from nuclear facilities.

The ratios of specific activities (μCi ^{3}H per kg hydrogen) of the organic bound and

TABLE 2

TRITIUM TISSUE DOSE AND BOUND-LOOSE RATIO IN THE BODY

Year	Tissue Dose (μrad) Fallout	Total	Bound-Loose Ratio	Year	Tissue Dose (μrad) Fallout	Total	Bound-Loose Ratio	Year	Tissue Dose (μrad) Fallout	Total	Bound-Loose Ratio
1952	0.0	1.5	.92	1965	144.8	146.3	1.21	1978	9.5	11.0	1.32
1953	0.1	1.6	.90	1966	122.0	123.5	1.17	1979	7.4	8.9	1.32
1954	3.8	5.3	.29	1967	97.0	98.5	1.15	1980	5.7	7.2	1.32
1955	10.5	12.0	.73	1968	76.4	77.9	1.28	1981	4.3	5.8	1.32
1956	16.4	17.9	.63	1969	68.4	69.9	1.21	1982	3.2	4.7	1.32
1957	18.4	19.9	.75	1970	58.1	59.6	1.21	1983	2.3	3.8	1.32
1958	30.3	31.8	.57	1971	47.4	48.9	1.26	1984	1.6	3.1	1.32
1959	53.1	54.6	.84	1972	38.0	39.5	1.28	1985	1.0	2.5	1.32
1960	47.2	48.7	1.12	1973	30.4	31.9	1.30	1986	0.5	2.0	1.32
1961	32.3	33.8	.95	1974	24.3	25.8	1.31	1987	0.2	1.7	1.21
1962	76.6	78.1	.61	1975	19.3	20.8	1.32	1988	0.1	1.6	1.08
1963	210.3	212.0	.73	1976	15.3	16.8	1.32	1989	0.1	1.6	1.01
1964	210.9	212.4	1.10	1977	12.1	13.6	1.32	1990	0.0	1.5	.97
								Dose Commitment:		1.5 mrad	

loose water tritium in the body, determined from this computation, are included in Table 2. The ratio would be 1.0 under natural conditions, except that radioactive decay does not allow complete equilibrium in the bound compartment with the slowest turnover time. As the concentrations of tritium intake increase, labeling of the bound components lag with respect to the specific activity of the loose water compartment. The bound-loose ratio is then less than one. During times of decreasing tritium intake, the bound tritium label is retained longer than the activity in body water, and the bound-loose tritium ratio becomes greater than one. The ratio is currently about 1.3 in man, according to this computation, which is not unlike the ratios actually being measured in human tissue.[26]

Other Estimates of the Dose Commitment

Indirect estimates of the dose commitment can be made by comparing the total amounts of weapons-produced tritium with the natural production rate, as is done by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR).[27] The uncertainties involved, however, affect the accuracy of these estimates.

Most of the weapons-produced tritium, an estimated 1900 MCi,[17] was introduced into the Northern Hemisphere. The stratospheric residence time of tritium (~1 yr) does not allow inter-hemispheric mixing to be an important factor. Ten percent may be a reasonable approximation for stratospheric tritium carried to the Southern Hemisphere.[28] The dose commitment for the Northern Hemisphere is consistent with the above direct computation if the natural production rate is taken as 3.4 MCi/yr (1.7 MCi/yr in each hemisphere). The natural background tritium dose to man is .0015 mrad/yr.

$$.0015 \text{ mrad/yr} \times \frac{1900 \text{ MCi} \times .90}{1.7 \text{ MCi/yr}} = 1.5 \text{ mrad}$$

UNSCEAR computed a range of possible dose commitments for the Northern Hemisphere, based on various assumptions of the natural tritium background and inter-hemispheric mixing of stratospheric weapons tritium. A conservative estimate for the Northern Hemisphere fallout tritium dose commitment of 4 mrad was obtained.[27]

Tritium released from nuclear power facilities are as yet insignificant, compared to the amounts of weapons-produced tritium. It will be desirable, however, to continue to measure the environmental tritium levels and to ascertain the contribution from nuclear power activities. With use of the dose model presented here, the dose commitment from future activities involving release of tritium to the environment can be determined.

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