Analysis of an Internal Kinetic Model for Free and Bound Tritium

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ABSTRACT

Internal dosimetry models that currently drive regulatory compliance decisions assume that tritium retention kinetic behavior can be modeled by a single exponential function. This is contrary to the results of a number of modeling techniques, which indicate that while elemental tritium (HT) and tritiated water (HTO) are the most commonly released forms of tritium, organically-bound tritium (OBT) doses can be quite significant. In this paper, a unified two-compartment model of the retention kinetics of HTO and OBT is examined for the purpose of investigating the importance of metabolic routes not considered in the ICRP one- and two-exponent models; namely the transfer of tritium from the HTO compartment to the OBT compartment and vice-versa. In particular, the effect of intake ratio is investigated and a detailed analysis of dosimetric implications is performed. For typical combined intakes of HTO and OBT, the number of disintegrations from the two tritium forms can be roughly equal. This result, when combined with the suggested greater biological effectiveness of OBT, indicates effective doses will be greater than those obtained from a single exponential model. The results of this study corroborate previous findings using the two-compartment model for the cases of HTO-only and or OBT-only intakes, and compare well with data taken from studies on animals and human subjects.

INTRODUCTION

Tritium exists in one of three basic chemical forms: 1) in an elemental form (HT; gaseous), 2) in an oxide form or "free-water" tritium (HTO; vapor or liquid), or 3) bound to non-exchangeable sites (i.e., bonding with carbon) in an organic molecule (OBT). The majority of tritium released from nuclear facilities is in the form of either elemental gas or tritiated water. Although the magnitude of elemental tritium releases can equal those of oxide releases, HT is more than four orders of magnitude less hazardous than HTO, in terms of radiation dose (ICRP 1978). Once in the environment, however, HT can be converted through oxidation to HTO; either of these forms of tritium can become bound to organic material (Hill and Johnson 1993). Internal dosimetry models that currently drive regulatory compliance decisions (ICRP 1978) assume that tritiated-water retention follows a single exponential, essentially considering only that which exists in the oxide form, stating that forms with longer retention contribute less than 10% to total dose. Even though some authors also claim this to be the case (Snyder et al. 1968; Lambert and Clifton 1967; Trivedi et al. 1997; Trivedi et al. 2000), various modeling techniques have shown the potential for OBT to be much more significant (Crawford-Brown 1984; Dunford and Johnson 1987; Johnson and Dunford 1985; Etnier et al. 1984; ICRP 1989).

Because of the physical and chemical nature of operations at nuclear facilities, the production of tritium in the presence of organic matter typically does not occur. Organically-bound tritium is at most only a few percent of the total tritium release, occurring as the result of lubricating oils or solvents becoming tritiated. The majority of tritium in the environment that is OBT was originally released in the elemental or oxide form, all of which is available for

elemental exchange with natural organic matter in soils and vegetation (Belot et al. 1988; Bogen and Welford 1976; Murphy 1993).

Tritium can enter the body in any of its chemical or physical forms. Elemental tritium can be inhaled; HTO can be inhaled, ingested, or absorbed through the skin; and OBT is generally ingested as a constituent in foods. Tritium in the form of HTO passes through the body quite rapidly, behaving similarly to other water molecules. However, if tritium enters the body as an organic or becomes organically bound, the nuclide will remain in the body for longer periods.

If estimates of radiation dose are based solely on measurements of HTO in the environment, or if metabolic models consider tritium only in the oxide form (or as a single exponential), there is the chance for underestimating the public impact due to total tritium. This possibility exists since typical analytical methods used to measure HTO do not detect tritium that is organically bound. However, if measurements of tritium are made at the release point, total tritium can be determined, thus lessening the potential effect of not considering OBT in dosimetric calculations.

We have examined a unified two-compartment model of the retention kinetics of HTO and OBT (Crawford-Brown 1984). The assessment is intended to show the importance of metabolic routes not considered in the ICRP one- and two-exponent models, namely the transfer of tritium from the HTO compartment to the OBT compartment, and vice-versa.

FRACTIONATION OF FREE AND BOUND TRITIUM

Evans (1969) showed that, in deer exposed to a tritium environment over the long-term, organically-bound tritium in various organs was significant and an equilibrium between HTO and OBT was attained. With initial and sustained body-water concentrations being identical, he concluded that upper limit calculations of dose for humans exposed chronically to tritium may result in an increase in the amount of tritium resident in the body (i.e., the body burden) by about 40 to 50% over that assumed by the methods of the ICRP (1959; 1978). This estimate assumes, however, that an individual is constantly exposed over a long period with every food/beverage constituent containing tritiated water. Because of exchange mechanisms, it is impossible for the specific activity of organic tritium to greatly exceed the specific activity of body-water tritium (NCRP 1979). Evans bases his calculations on the number of hydrogen atoms in the human body and the assumption that all of those atoms are equally susceptible to being replaced by a tritium atom. This differs from the ICRP (1978) single-compartment dosimetric assumption that tritium is incorporated only in body water.

Laskey et al. (1973) determined that if only tritiated water were consumed, the OBT specific activity in rats would be about 25% of the water specific activity. Additionally, Koranda and Martin (1973) found that kangaroo rats exposed chronically to environmental tritium averaged about 50% greater total tritium than that found only in body-water. And, Thompson and Ballou (1956) concluded that organic tritium in rats exposed for half of a year was about 20-30% of the activity in body water. These studies all tend to show that total tritium, including that

which is organically bound, in the bodies of animals exposed long-term to tritium environments may exceed estimates based solely on water tritium by about 20-50%.

Later work by Crawford-Brown (1984) indicates that if tritium is taken into the body as a single intake in the organic form, body-burden commitments (total radioactive disintegrations or integrated activity) can be nearly 3 times higher than if an equal amount of tritium were taken in as an oxide.

Not only does the organically bound tritium remain in the body for a longer period of time, it also has been shown to be potentially more hazardous than free-water tritium by as much as a factor of 2 (Straume and Carsten 1993; Balonov et al. 1993). The relative biological effectiveness of tritium bound to organic molecules is higher than HTO due to its ability in some cases to become incorporated into the DNA, allowing more direct impact of the beta-particle energy on the DNA molecule and introducing potential chemical damage caused by the presence of ^{3}He at the site of the decayed tritium atom. Tritiated thymadine, for example, is incorporated specifically into the DNA of proliferating cells (NCRP 1979). Thus, the fraction of total tritium in the body that is organically bound is of importance for dosimetric (and microdosimetric) purposes, relative to impacts estimated when modeling tritium solely as HTO.

TRITIUM RETENTION MODEL

ICRP Models for Radiation Protection. Tritium kinetics in the human body have been modeled several ways for estimating internal radiation dose (Hill and Johnson 1993). Typically, tritium is assumed to exit the body in a combination of one, two or three time components (ICRP

1978; ICRP 1989). These components typically represent free-water tritium (HTO), short-term retention of organically-bound tritium (OBT) assumed to follow carbon turnover rates, and longer-term retention of OBT. For radiation protection purposes, however, a simplified one-compartment model is often employed,

$$A_{HTO}(t) = e^{-\left(\frac{0.693}{10d}t\right)},$$

in which tritium is considered to have a retention half-life of 10 days for adults (ICRP 1978).

In a more recent report (ICRP 1989), the ICRP recommends a two-compartment model to describe both HTO and OBT separately, with the only difference between the two models being the coefficients describing the initial fraction of the compound distributed into the short and long retention compartments. The ICRP 56 (1989) models for the amount of tritium activity in the body as a function of time for HTO and OBT intakes, respectively, are:

$$A_{HTO}(t) = 0.97e^{-\left(\frac{0.693}{10d}t\right)} + 0.03e^{-\left(\frac{0.693}{40d}t\right)}, \text{ and}$$

$$A_{OBT}(t) = 0.5e^{-\left(\frac{0.693}{10d}t\right)} + 0.5e^{-\left(\frac{0.693}{40d}t\right)}.$$

These models represent two independent compartments with no allowance for modeling the internal transfer of tritium from a free state to a bound state, and vice-versa. Dose factors

calculated using these models are based on intakes of HTO or OBT separately. A dose factor that considers the ratio of intakes and kinetics of both forms is desirable.

Unified Two-Compartment Model. In order to discuss our investigation of OBT and HTO retention, three ratios must be defined: 1) an intake ratio - the ratio of specific activities of organically-bound tritium to free-water tritium in media ingested by individuals; 2) an activity ratio - the ratio of activities of OBT to HTO in the two model compartments as a function of time; and 3) an integrated activity ratio - the ratio of integrated activities (cumulative body burden or total disintegrations) of OBT to HTO over a specified integration time. The ratio of integrated activities is of most importance in comparing the potential impact of OBT relative to models that consider only HTO.

The kinetic model analyzed in this paper is taken from the work of Crawford-Brown (1984). It models tritium kinetics in two basic non-physiological representations, one for HTO and one for OBT (Fig. 1). Unlike the models of the ICRP (1989), the compartments are unified by considering a transport route from HTO to OBT and two transport routes from the OBT to HTO compartments that represent its long and short retention time components. Losses from the OBT compartment, representing the exchange of tritium from a bound site to a free site, are modeled assuming that tritium moves back into the free-water pool before it can be eliminated from the body. Direct elimination from the OBT compartment occurs only as the result of radiological decay. This unified model is simple to conceptualize and, as will be seen, provides results similar to the results of more complex models. The model is essentially unchanged from

Crawford-Brown (1984), but a more detailed analysis of intake ratio and dosimetric implications is carried out in this work.

Data from Etnier et al. (1984), indicates that, of the total organically bound hydrogen taken into the body daily, approximately 47% is immediately catabolized into free hydrogen. It also can be estimated from their work that on balance about 93% of hydrogen exchanged daily ends up as free hydrogen while 7% remains bound to organics. Additionally, Hill and Johnson (1993) report that approximately 40-60% of tritiated thymidine is metabolized into HTO when injected into humans. In the unified two-compartment model we have assumed that 50% of organically bound tritium taken into the body is catabolized and that the fractions of tritium initially distributed to the HTO or OBT compartments, F_F and F_B, are dependent on the intake ratio and the catabolized fraction.

The radiological decay constant, λ, is generally of no significance, except for tritium atoms bound at non-exchangeable sites with retention half-times on the order of thousands of days. The values of f_S (0.90) and f_L (0.10) indicate the fractions of tritium being given up from organically-bound sites with relatively short and long half-times, respectively. Age dependency is assumed for individuals less than 21 years, after which all adults are regarded as possessing the same retention characteristics. Age-specific rate constants taken from Crawford-Brown (1984a; 1984b) were used to develop second-order polynomials as a function of age to estimate tritium retention for all ages, infant through age 21 (Table 1).

The system of ordinary differential equations (ODEs) corresponding to the model shown in Fig. 1 is

$$\frac{d\,HTO(t)}{dt} = -(K_4 + \lambda)\,HTO(t) - K_1\,HTO(t) + (f_s K_2 + f_L K_3)\,OBT + F_F Q(t), \text{ and} \tag{1}$$

$$\frac{d\,OBT(t)}{dt} = -\lambda\,OBT(t) + K_1\,HTO(t) - (f_s K_2 + f_L K_3)\,OBT + F_B Q(t), \tag{2}$$

where the source *Q(t)* is defined by

$$Q(t) = \begin{cases} Q_0\delta(t), & \text{single intake at time } t = 0, \\ Q_0, & \text{constant, continuous intake.} \end{cases} \tag{3}$$

Eqs. (1) and (2) are a typical system of linear ODEs with constant coefficients. Techniques to solve these types of ODEs are available in numerous texts (Boyce and DiPrima 1965). For the system of Eq. (3) with a single intake of tritium at time $t = 0$, the solution of these equations is

$$HTO(t) = c_1 \exp(-\omega_1 t) + c_2 \exp(-\omega_2 t), \quad \text{and} \tag{4}$$

$$OBT(t) = c_1 y_1 \exp(-\omega_1 t) + c_2 y_2 \exp(-\omega_2 t), \tag{5}$$

where

$$\omega_{1,2} = \frac{K_1 + K_4 + (f_S K_2 + f_L K_3) \pm \sqrt{X}}{2}, \tag{6}$$

$$y_{1,2} = \frac{K_1 + K_4 - (f_S K_2 + f_L K_3) \pm \sqrt{X}}{2(f_S K_2 + f_L K_3)}, \tag{7}$$

$$\begin{aligned} X = {} & K_1(K_1 + K_4 + 2(f_L K_2 + f_L K_3)) + K_4^2 \\ & -2K_4(f_S K_2 + f_L K_3) + (f_S K_2 + f_L K_3)^2, \end{aligned} \tag{8}$$

$$\text{and} \quad c_1 = \frac{(F_F y_2 - F_B)Q}{y_2 - y_1}, \quad c_2 = \frac{(F_B - y_1 F_F)Q}{y_2 - y_1}. \tag{9}$$

The solution of Eqs. (1) and (2) with a continuous intake of tritium is similar to Eqs. (4) and (5), but has an added term which accounts for the buildup of the source over time and the absence of an initial concentration in either of the two compartments:

$$HTO(t) = d_1 \exp(-\omega_1 t) + d_2 \exp(-\omega_2 t) + z_1, \tag{10}$$

$$OBT(t) = d_1 y_1 \exp(-\omega_1 t) + d_2 y_2 \exp(-\omega_2 t) + z_2, \tag{11}$$

$$z_1 = \frac{Q_0(F_F \lambda + f_S K_2 + f_L K_3)}{\lambda(K_1 + K_4 + \lambda) + (K_4 + \lambda)(f_S K_2 + f_L K_3)}, \tag{12}$$

$$z_2 = \frac{Q_0(K_1 + F_B(K_4 + \lambda))}{\lambda(K_1 + K_4 + \lambda) + (K_4 + \lambda)(f_S K_2 + f_L K_3)}, \tag{13}$$

$$\text{and} \quad d_1 = \frac{(z_2 - z_1 y_2)}{y_2 - y_1}, \quad d_2 = \frac{(z_1 y_1 - z_2)}{y_2 - y_1}. \tag{14}$$

The expressions in Eqs. (4), (5), (10), and (11) are analytic in time and can be easily integrated to yield total disintegrations from a given source. If total disintegrations (HTO^{INT} and OBT^{INT}) are desired at time $t = a$ from the single intake source, the result is

$$HTO^{INT}(t) = \frac{c_1}{\omega_1}\left[1 - \exp(-\omega_1 a)\right] + \frac{c_2}{\omega_2}\left[1 - \exp(-\omega_2 a)\right], \text{ and} \tag{15}$$

$$OBT^{INT}(t) = \frac{c_1 y_1}{\omega_1}\left[1 - \exp(-\omega_1 a)\right] + \frac{c_2 y_2}{\omega_2}\left[1 - \exp(-\omega_2 a)\right], \tag{16}$$

where the variables are defined as before. Likewise, the total disintegrations at time a from the continuous tritium intake are

$$HTO^{INT}(t) = \frac{d_1}{\omega_1}\left[1 - \exp(-\omega_1 a)\right] + \frac{d_2}{\omega_2}\left[1 - \exp(-\omega_2 a)\right] + z_1 a, \text{ and} \tag{17}$$

$$OBT^{INT}(t) = \frac{d_1 y_1}{\omega_1}\left[1 - \exp(-\omega_1 a)\right] + \frac{d_2 y_2}{\omega_2}\left[1 - \exp(-\omega_2 a)\right] + z_2 a. \tag{18}$$

While linear systems of ODE's with constant coefficients can be solved exactly with this technique, regardless of the number of equations, the eigenvalues cannot always be obtained symbolically. As the number of compartments in the model increases, it may be necessary to find the eigenvalues and eigenfunctions numerically.

The unified model was solved analytically and the solution coded into an Excel spreadsheet. Certain input parameters were allowed to vary in order to determine their influence on model outputs. These included: age, specific-activity intake ratio, catabolized fraction, and the f_S and f_L fractions. Cumulative body burdens, integrated over 50 years, were modeled in the HTO and OBT compartments for both single and continuous intakes. From these outputs, integrated activity ratios in the two compartments were determined.

The integrated activity of tritium following a single intake in both free and bound forms, $\tilde{A}$, can be determined by,

$$\tilde{A} = \left\{ \frac{c_1}{\omega_1}(1+y_1)[1-\exp(-\omega_1 a)] \right\} + \left\{ \frac{c_2}{\omega_2}(1+y_2)[1-\exp(-\omega_2 a)] \right\}. \quad (19)$$

For an intake by an adult and assuming an integration time of 50 yrs, Eqn. (1) reduces to,

$$\tilde{A} = 14.91 + 50.42 F_B, \quad (20)$$

where F_B is the fraction of the total tritium intake remaining in the bound form after catabolism in the gut and bloodstream. This fraction is calculated by,

$$F_B = CF \cdot \frac{R_I}{(1+R_I)}, \quad (21)$$

where CF is the catabolized fraction and R_I is the intake ratio (OBT:HTO).

The total integrated activity of tritium in the body following a single intake of both HTO and OBT, weighted by a factor related to the intake ratio, can be calculated from the individual bound- and free-tritium models of the ICRP (1989). For example,

$$\tilde{A} = \left(\frac{1}{1+R_I}\right)\int A_{HTO} + \left(\frac{R_I}{1+R_I}\right)\int A_{OBT} \qquad (22)$$

Equations parallel to those of Eqn. (19) through (22) can also be determined for continuous release scenarios in this same manner.

MODELING RESULTS

Using the two-compartment model of Crawford-Brown (1984a), we have confirmed his age-dependent estimates of the two extremes (intakes of 100% HTO or 100% OBT), and have further shown how combined uptakes of HTO and OBT impact the retention of tritium atoms in the body and, hence, the estimation of integrated activity.

Figures 2a and 2b show that the integrated activity in adults from single intakes can vary by as much as a factor of 2.7 for the case where only HTO is ingested (100% HTO or an intake ratio of 0.0) compared to an instance when only OBT is ingested (100% OBT or an infinite intake ratio). For a model that considers only a single compartment of HTO (ICRP 1978), the integrated activity is equal to the intake activity divided by the biological removal constant (assuming a long integration time). This value of integrated activity for adults is represented by the first (1) heavy dotted line in Fig. 2a and is essentially equal to the unified two-compartment

value for pure HTO intakes since the rate at which HTO converts to OBT in the body is relatively slow (half-time of about 1,000 days). The second (2) and third (3) dotted lines in Fig. 2b represent the integrated activity when using the ICRP 56 (1989) two-exponent model to estimate tritium retention for single HTO and OBT intakes, respectively.

We have found that single intakes of OBT and HTO, regardless of intake ratio, equilibrate to constant activity ratios in the two compartments rather quickly according to the unified model. This occurs after about 125 days in children (Fig. 3a) and about 200 days in adults (Fig. 3b). For continuous intakes, the equilibrium activity ratio is intake-ratio dependent, yet varies insignificantly with age (Figs. 4a and 4b). When examining integrated activity ratios (i.e., dose contribution) at various times after the first intake (Fig. 5), activity ratios from single intakes reach equilibrium faster than do those from continuous intakes. These results are also consistent with Diabate and Strack (1993).

Integrated activity ratios for adults and children plotted as a function of intake ratio (Fig. 6) suggest that once the OBT intake amount is about 10 times greater than the HTO intake amount, further increases of the intake ratio do not influence the integrated activity ratio. Additionally, the integrated activity ratio plateaus at a value between 1.5 and 1.7, indicating that the number of disintegrations of tritium in the form of OBT is 50-70% larger than the number of disintegrations due to HTO. Intake ratios of this magnitude are highly unlikely, however. In a study of NY foods by Bogen and Welford (1976), bound-to-free tritium ratios were as high as 4.5 for bakery products and root vegetables, but ranged from about 1.4 to 2.4 in protein foods. In studies of food samples collected in Japan, Hisamatsu et al. (1987; 1989) measured OBT and

HTO concentrations in various foods and found that their ratio ranged from 0.57 ± 0.12 to 2.9 ± 1.0 with an average of 1.5 ± 2.5. They also determined a composite specific activity intake ratio of 1.2, based on a sampling of breakfast, lunch, and dinner foods for several Japanese persons. The composite intake ratio is more realistic because it considers ratios in various food types as well as the amounts of those foods consumed.

From our analysis, an intake ratio of 1.2 results in an integrated activity ratio of about 0.995 for adults, meaning essentially equal numbers of disintegrations from tritium in both OBT and HTO forms. The integrated activity ratio varies with age (Fig. 7a) from approximately 0.96 to 1.02. As modeled, infants would seem to have the greatest OBT fractional body burden, but all ages essentially have integrated activity ratios of unity. In contrast, integrated activity ratios are much lower (~0.035) for adults when the intake is assumed to contain no organically-bound tritium (Fig. 7b).

The NCRP (1979) states that there are extensive animal studies showing that between 1 and 3 percent of an intake of tritiated water is incorporated into organic constituents of the body; the unified two-compartment model gives similar predictions for adults. A single intake (or even a continuous intake) of tritiated water results in an integrated activity in the OBT compartment of about 3.5% of the amount in the HTO compartment (see Fig. 7b). Additionally, at equilibrium the activity present in the OBT compartment is about 2.6 times the amount in the HTO compartment, given a single intake (see Fig. 3b). During a continuous intake though, the OBT activity reaches only 3.5% of the HTO activity in adults (see Fig. 4b). These findings are consistent with those of Diabate and Strack (1993).

Using a four-compartment model of hydrogen metabolism, Etnier et al. (1984) estimated the ratio of effective dose from total tritium to the dose only from free-water tritium. This ratio, as a function of intake ratio, is compared to our results (Fig. 8) from the unified two-compartment model and to the estimates of Evans (1969) and Bennett (taken from Etnier et al. 1984). Etnier's model is unique in that three of its four compartments represent tissue-solids, enabling different metabolic treatments of carbohydrates and fats. Neither Evans nor Bennett correlated their data with intake ratios of OBT and HTO. Comparing Etnier's and our results with the work of Evans, it appears that the two-compartment model more closely predicts the data from an animal population exposed over the long-term.

ESTIMATE OF THE RELATIVE DOSE INCREASE

From the findings presented, an estimate of potential increase in effective dose can be obtained. In addition to evidence suggesting that the biological effectiveness of OBT is twice that of HTO, Figure 2a indicates that estimates from the unified two-compartment model using an intake ratio of 0.0 are very similar to dosimetric estimates obtained from a simple one-compartment model (ICRP 1978). Additionally, the results from Fig. 7a show that for an intake ratio of 1.2, a value typical of many food items, the ratio of cumulative body burdens from a given intake is approximately 1.0, such that the number of OBT and HTO disintegrations occurring in the body over a 50-yr period are essentially equal. These points suggest, therefore, that it may be appropriate for tritium dose estimates determined using a one-exponential model to be increased following a given total intake, dependent on the intake ratio (OBT:HTO) in foodstuffs.

Assuming a one-compartment kinetic model with a retention time of 10 days, and that all tritium is instantaneously distributed throughout soft tissue upon intake, the ingestion of 1 Bq of tritiated water results in a cumulative body-burden of 14.4 Bq-days over a long integrating time (i.e., 50 yrs). The dose conversion factor then can be calculated using:

$$DCF_{HTO} = \frac{QF_{HTO} \cdot A_{HTO} \cdot \overline{E} \cdot K}{M_{ST}},$$

where QF_{HTO} = quality factor [1 Sv/Gy];

A_{HTO} = total integrated activity [14.4 Bq-d];

$\overline{E}$ = energy imparted per disintegration [5.69 keV];

M_{ST} = mass of soft tissue [63,000 g]; and

K = conversion constant [1.38×10^{-8} g Gy s keV^{-1} d^{-1}].

Thus, an intake of 1 Bq of tritium would result in a dose commitment of 1.8×10^{-11} Sv. This is essentially the same calculation carried out in determining the dose conversion factor for tritium by the ICRP in Report No. 30 (1978). An analysis of the uncertainties in this calculation indicate that this estimate varies by as much as a factor of 15 from it's highest to lowest modeled values, with much of the uncertainty attributable to varying estimates of the biological effectiveness of HTO (Hamby 1999).

Using these same methods and taking into account a different total integrated activity, a dose conversion factor can be calculated for organically-bound tritium. The ICRP, in Report No.

56 (1989), estimates that the adult dose factor for the ingestion of HTO is 1.6×10^{-11} Sv/Bq and for the ingestion of OBT the dose factor is 4.0×10^{-11} Sv/Bq, two-and-one-half times larger, accounting only for longer retention of a greater fraction of the tritium intake.

As stated above, however, the relative biological effectiveness of OBT is about twice that of HTO. Additionally, our estimates indicate that for a typical OBT:HTO intake ratio in food stuffs, the integrated activities of OBT and HTO in the body are essentially equal. Thus, by doubling the quality factor, the OBT dose conversion factor would be two times higher than the HTO dose conversion factor, given the same integrated body burden of each form.

Using the unified two-compartment model, one that differentiates the behavior of tritium as OBT and HTO, yet allows for interaction and exchange of the two forms, an intake of 1 Bq (containing 20% more OBT than HTO, i.e., an intake ratio of 1.2) will result in a modeled effective dose of 1.8×10^{-11} Sv for the integrated activity of tritium as HTO, plus 3.6×10^{-11} Sv for the integrated activity of tritium as OBT, for a total of 5.4×10^{-11} Sv, a factor of 3 greater than an HTO-only dosimetric model would estimate.

CONCLUSIONS

The ICRP (1978) states that dose from tritium bound to organic compounds accounts for less than 10% of the total dose when evaluating tritium exposures. It appears, however, that organically-bound tritium may be more significant for dosimetry assessments of environmental tritium than considered in ICRP 30, even if the entire intake is in the form of HTO. If tritium dosimetric models are based solely on the retention characteristics of free tritium, that which is

not labeled to organics at non-exchangeable sites, historical dose estimates may be underestimated by as much as a factor of 3, not accounting for longer retention times nor for differences in the biological effectiveness of organically bound tritium.

For a single exposure, the OBT/HTO intake ratio is unimportant since the integrated activity ratio of the two components is constant and only varies with age. However, for continuous exposures, age is unimportant whereas the intake ratio determines the integrated activity ratio in the two compartments.

The unified model shows that metabolic modeling of tritium is dependent on the ratio of OBT to HTO in foodstuffs being consumed and on the total intake of tritium in the bound and free forms. An assessment of intake ratios for individuals in proximity to nuclear facilities where tritium is released to the atmosphere on a continuing basis is needed for improved modeling of the impact of tritium releases. Research directed toward developing a large database of organic-to-free tritium ratios in foods in an effort to improve predictive models is in order.

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FIGURE CAPTIONS

Fig. 1. Schematic of the unified two-compartment HTO and OBT kinetic model.

Fig. 2. Total integrated activity as a function of intake ratio and age using the unified two-compartment tritium model. Integrated activity following (A) a single intake of 1 Bq and (B) a continuous intake of 1 Bq/d for intake ratios (OBT/HTO) of 0.0, 0.5, 1.0, 2.0, and ∞. The three dashed lines in (A) represent results from (1) the ICRP 30 one-exponent HTO model; (2) the ICRP 56 two-exponent HTO model; and (3) the ICRP 56 two-exponent OBT model.

Fig. 3. Activity ratio as a function of time and intake ratio. Activity ratios following a single intake of 1 Bq for (A) 5-yr old children and (B) adults. Calculated for intake ratios of 0.0, 0.5, 1.0, 2.0, and ∞.

Fig. 4. Activity ratio as a function of time and intake ratio. Activity ratios for continuous intakes of 1 Bq d^{-1} for (A) 5-yr old children and (B) adults. Calculated for intake ratios of 0.0, 0.5, 1.0, 2.0, and ∞.

Fig. 5. Integrated activity ratio as a function of time after first intake. Modeled for adults and an intake ratio (OBT/HTO) of 1.0. The solid line results from a single intake and the heavy dashed line from a continuous intake.

Fig. 6. Integrated activity ratio as a function of intake ratio for adults (solid line) and 5-yr old children (heavy dashed line).

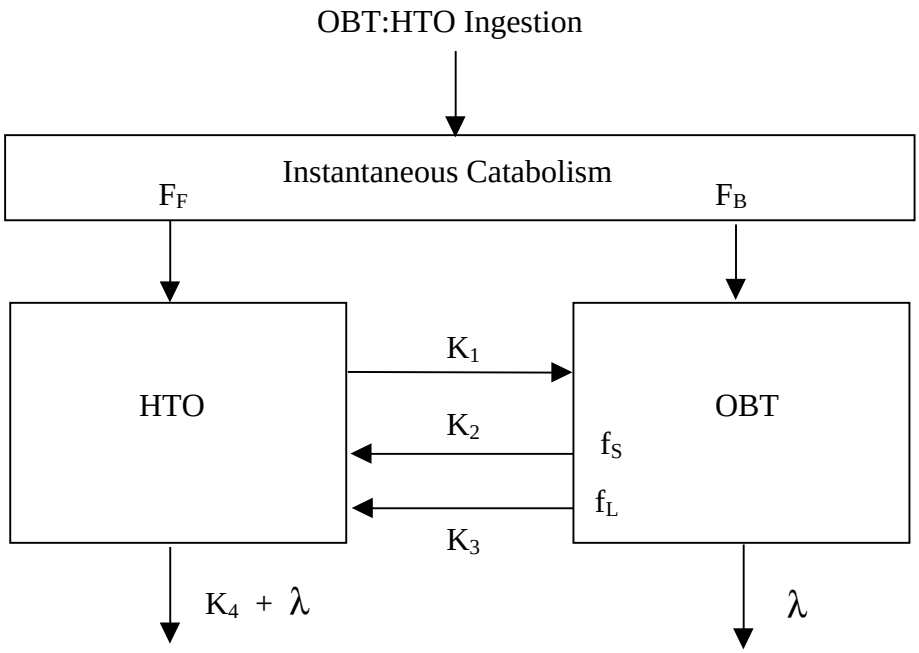
Fig. 7. Integrated activity ratio as a function of age for single and continuous intakes assuming (A) an intake ratio of 1.2 and (B) an intake ratio of 0.0. In (A), the single intake is represented by the solid line and the continuous intake is represented by the heavy dashed line. In (B), the integrated activity ratios for single and continuous intake lie on top of one another, hence only the single line.

Fig. 8. Comparison of the ratio of total integrated activity to HTO integrated activity as a function of intake ratio for four tritium models.

Table 1. Rate constants and parameter values used in the unified two-compartment tritium model.

Parameter	Value
CF	0.5
F_F	$CF \cdot \frac{1}{1+R_I}$
F_B	$CF \cdot \frac{R_I}{1+R_I}$
f_S	0.9
f_L	$1 - f_S$
K_1	$99.7 + 63.6A - 0.981A^2$
K_2	$15.0 + 1.33A - 0.0225A^2$
K_3	$26.3 + 5.38A - 0.0869A^2$
K_4	$4.93 + 0.392A - 0.00704A^2$
λ	0.000158

Note: A = age and R_I = intake ratio (OBT:HTO)


OBT:HTO Ingestion
Instantaneous Catabolism
F_F
F_B
HTO
OBT
K_1
K_2
f_S
f_L
K_3
$K_4 + \lambda$
λ