UNCERTAINTY OF THE TRITIUM
DOSE CONVERSION FACTOR

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ABSTRACT

Environmental releases of tritium oxide at a number of Department of Energy nuclear weapons facilities contribute to a significant portion of environmental dose. Several conversion factors are utilized in the estimation of human impact from these releases, e.g., dispersion coefficients, consumption rates, uptake factors, transport factors, dose conversion factors, risk coefficients. A probabilistic determination of the tritium dose conversion factor (DCF) was generated in this work to assess the uncertainty of the internal dosimetry required to estimate dose equivalent given an intake of tritium oxide. The tritium DCF was found to vary by a factor of about 15 with a median value of 2.2×10^{-11} Sv Bq^{-1} when considering orthovoltage x-rays as the standard for estimating the relative biological effectiveness (RBE) of tritium. The median DCF increases by about 50%, however, when gamma radiation is considered as the standard. The current deterministic estimate of the tritium DCF published by the DOE and the EPA is 1.7×10^{-11} Sv Bq^{-1}, 25-50% lower than the median probabilistic values. The tritium oxide DCF model was found to be most sensitive to biological half-life and quality factor and is highly dependent on the standardizing radiation for RBE assessments.

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INTRODUCTION

The U.S. Department of Energy (DOE) has generated special nuclear materials and related by-products for more than 50 years in support of the Nation's nuclear weapons needs. At several of the DOE Sites, a large component of the historical offsite dose is the result of atmospheric and aqueous releases of tritium oxide. Over the last decade, tritiated water vapor released to the environment from operations at the Savannah River Site (SRS), for example, accounted for the majority of the off-site dose via both atmospheric and liquid effluent pathways. Probabilistic environmental dose estimates for the SRS (Hamby 1993) show that the uncertainty in the atmospheric tritium dose potentially spans more than two orders of magnitude. In that estimate of uncertainty, the tritium dose factor was allowed to vary only by including the input variability of body mass and effective half-life. In this work, however, all parameters are varied over their entire range of values to determine the variability, and hence the uncertainty, of the tritium internal dose conversion factor. This uncertainty will shed light on the accuracy of dose estimates from environmental exposures to tritium oxide.

Because the energy of the beta particles emitted from transitioning tritium atoms is relatively small, they do not penetrate the dead-layer of skin and there is no resultant dose from external exposures to tritium. Once internal, however, tritium dose can be significant. Whether through inhalation, skin absorption, or ingestion, internal exposure to tritium oxide results in rapid and complete absorption throughout the body. Typically, dose conversion factors are specific to ingestion or inhalation routes of uptake, but the tritium oxide DCF is applicable to three intake pathways, with skin absorption being related to inhalation intake (ICRP 1978).

METHOD

Monte Carlo technique. The uncertainty of the tritium internal dose conversion factor was determined using a numerical method in which multiple calculations were performed with values that span the entire range of each input variable. In the calculations, parameter values were assigned using a routine that employed Latin hypercube sampling (LHS). This method considers the probability distribution of each parameter to be composed of a given number of non-overlapping intervals of equal probability. For a given parameter, values are selected at random from each interval based on the probability density function in that interval. The Latin hypercube approach is a constrained random sampling technique that results in added precision over conventional random sampling methods (e.g., Monte Carlo) since the entire range of the distribution is sampled in a more systematic manner (Hamby 1993). Thus, fewer iterations are needed in order to provide adequate statistical input for the probabilistic dose estimate. Model input includes the type of distribution (e.g., normal, lognormal, uniform) and parameters describing its range (e.g., mean, standard deviation, minimum, maximum).

Five-thousand dose-factor estimates were calculated using the DCF model and the parameter assignments described below. The Latin hypercube routine selected 50 parameter values from each of 100 intervals within each parameter distribution. All dose-factor input parameters were assumed to be independent. Five-thousand trials using the Latin hypercube method provided frequency distributions of dose with mean standard errors of less than 2%. Statistics of the dose probability distribution were not significantly improved by increasing the number of trials above 5,000.

Metabolic modeling. Tritium in the environment is typically found in one of three forms: oxide, elemental, and organic. The oxide form is more prevalent and is chemically similar to water

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and, as such, is most effective in terms of radiation dose to humans (Hill and Johnson 1993). Elemental tritium is not reactive and its contribution to total dose is limited to that which may be present in the lung at any given time. Many compounds of organically-bound tritium (OBT) are not volatile and thus their probability of being inhaled is very low. Additionally, the majority of OBT taken in through ingestion is likely to be broken down into the oxide form in the gastrointestinal tract. The dosimetry model of the ICRP focuses on tritium oxide and OBT (ICRP 1978). However, for environmentally significant intakes, the committed dose from tritium oxide dominates total effective dose (ICRP 1978).

Being equivalent to water, tritium oxide (HTO) is assumed to be absorbed instantaneously and completely in all soft tissues of the body. The compartmental model structure, therefore, of tritium oxide kinetics is very simple, a one compartment model with a single loss route and a retention time of approximately 10 days (ICRP 1978).

Internal dosimetry. The activity of tritium, A(t), in the body at any given time, t, after an intake of tritium oxide can be described by the simple first order equation,

$$A(t) = A_0 e^{-\lambda_E t}, \tag{1}$$

where A_0 is the initial activity taken into the body and λ_E is the effective decay constant, the sum of the radiological, λ_R, and biological, λ_B, decay constants. Because of the long radiological half-life of tritium compared to its very short biological half-life, the effective time of which tritium remains in the body is completely driven by biological retention. To estimate the internal dose from a unit intake, Eqn. 1 is integrated over 50 years to determine the integrated activity, Ã, of tritium oxide in the body,

$$\tilde{A} = \frac{A_0}{\lambda_B}\left(1 - e^{-\lambda_B t}\right), \tag{2}$$

where t is equal to the integration time. Since 50-years is much larger than the biological half-life, the term in parentheses reduces to unity and the integrated activity is simply the intake activity divided by the biological removal constant.

The internal dose conversion factor can be calculated with knowledge of the integrated activity of tritium oxide and the amount of energy deposited as a result of its decay. Tritium emits a single beta at every nuclear transformation, therefore, the absorbed dose per transformation, S, is simply derived from the mean beta energy of tritium and is applied to all organs as source and/or target and is calculated by,

$$S = \frac{\overline{E}}{M}, \tag{3}$$

where $\overline{E}$ is the average beta energy and M represents the mass of soft tissue of those exposed. The dose conversion factor is then,

$$DCF = \frac{f_1 \cdot QF \cdot \tilde{A} \cdot S \cdot T_B}{\ln(2)}, \tag{4}$$

where T_B is the biological half-life, f_1 is the fractional absorption, and QF is the tritium quality factor.

The intake of tritium oxide can be by either the ingestion or inhalation (and skin absorption) routes. Again, because of the rapid dispersion of HTO in the body, regardless of entry pathway, the

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dose conversion factor is calculated by the same method and, therefore, the same value is used to estimate both inhalation and ingestion dose.

PARAMETER DISTRIBUTIONS

Characteristics of each parameter distribution are given in Table 1. The quality factor for tritium, described below, is assigned one of the distributions presented in Fig. 1 depending on the definition used for the standard radiation in determining radiation quality and bioeffect.

Biological half-life (T_B). Several investigators have studied the biological half-life of tritium in the human body (Pinson and Langham 1957; Wylie et al. 1963; Butler and Leroy 1965; Moghissi et al. 1971). Half-lives associated with the elimination of body water generally range from about 8-12 d and have been shown to be dependent on water intake, age, and mean ambient temperature (Butler and Leroy 1965; Moghissi et al. 1971). The data compiled by Hill and Johnson (1993) show that the weighted average half-life for HTO in the human body is 9.4 d.

The data of Butler and Leroy (1965) were used to estimate the biological half-life distribution used in this study; their data set is significantly larger than others (310 subjects). They estimated that tritium was removed from the body with a half-life of 9.4 ± 4.1 d (90% confidence). A log-probability plot of the cumulative probability vs. half-life shows that their data are best approximated by a lognormal distribution. The data have a geometric mean (median) of 8.7 d and a geometric standard deviation of 1.31.

Soft tissue mass (M). Reference Man data from ICRP Report No. 23 (ICRP 1974) were used to determine a range and distribution of the mass of soft tissue in an adult male. Stoudt et al. (1960) reported that the mean height of an adult male is about 1.75 m. Tables of weight as a function of height and age were referenced in ICRP Report No. 23 to determine the distribution of

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body weights averaged for adult males ages 18-79 y. These tables show that the 25th, 50th, and 75th percentiles of weight are 69, 78, and 85 kg, respectively.

A log-probability plot of these weights indicates that a lognormal distribution is appropriate and that the geometric standard deviation is 1.15 (geometric mean is 78 kg). Approximately 90% of body mass is soft tissue (ICRP 1978), therefore, the median soft tissue mass is about 70 kg.

Quality factor (QF). In this study, because of the estimation of a probabilistic dose factor and having the ability to consider a distribution of quality factors, relative biological effectiveness (RBE) is being considered as synonymous with QF. While there are many studies in the literature that report the RBE for tritium (see review by Straume and Carsten 1993), it is difficult to settle on a value for the tritium quality factor since the RBE is dependent on several experimental factors (e.g., the radiation used as the reference or standard, the biological endpoint considered, the chemical form of tritium, the dose rate, the assumed dose-response model). The debate continues as to which standard radiation should be used, which biological endpoint is most significant, or whether chronic or acute dose rates are appropriate. An ICRP and ICRU committee (Lamberton 1963) established to examine the use of RBE values in radiation protection stated that radiation quality should be presented in terms of an average number of ion pairs formed per micron or an average linear energy transfer. The ICRP (1977) also stresses the importance of relating radiation quality to linear energy transfer. For radiation protection and deterministic estimates of impact, the tritium quality factor should certainly be standardized, but when determining a probabilistic dose estimate, the variability in radiation quality can and should be considered. Although the tritium quality factor has varied over the years (ICRP 1991; NCRP 1993; Lamberton 1963; ICRU 1986), a value of 1 (ICRU 1993) currently is used for deterministic dose assessment.

Bond et al. (1978) compared the radiation quality of photons at various energies. From microdosimetric considerations, they found that the radiobiological quality of ^{60}Co gamma photons

is about twice as great as 250 kVp x rays. Ellett and Braby (1972) had earlier determined that the RBE of tritium, when measured microdosimetrically, was 1.5 when using a 250 kVp x-ray standard and 3.8 when using a ^{60}Co standard. More recently, Straume and Carsten (1993) have indicated that HTO is about 2 to 3 times more effective than ^{137}Cs or ^{60}Co. In their review of the literature, Straume and Carsten (1993) report tritium oxide RBEs, obtained by using either of the two standards, ranging from 1 to 1.8 for carcinogenic endpoints, 1.1 to 3.4 for chromosomal aberrations in human lymphocytes, 1 to 3 for genetic endpoints, 1.3 to 2.6 for developmental effects, and 1.4 to 2.9 for reproductive effects.

In this study, we have chosen to estimate the uncertainty of the tritium dose conversion factor using two separate distributions for quality factor, one considering orthovoltage x-rays as the standard and one considering gamma rays as the standard (Fig. 1). This distinction has been made because of the dramatic differences seen in RBE values when using the two different baselines (Ellett and Braby 1972; Bond et al. 1978; Straume and Carsten 1993). The RBE distributions applied to this work follow the data presented by Straume and Carsten (1993) and were determined based on a histogram of the number of reported RBE values for various biological endpoints. The distribution of RBE, hence quality factor, for tritium using orthovoltage x rays (180-250 kVp) is skewed to the right with the majority of values occurring near a value of 1. The RBE distribution using gamma rays as a standard, however, is approximately normally distributed with values lying between 1 and 3.5.

Fraction of tritium absorbed (f_1). It is generally accepted that all tritium oxide entering the body, by whichever route, will be completely absorbed in soft-tissue. The value for f_1 is, therefore, taken to be equal to unity when calculating the deterministic dose factor for tritium oxide. In this assessment, we have chosen to allow the absorption fraction to vary between 0.9 and 1.0, with a uniform probability for all values. This method provides the least amount of input bias, yet injects the greatest variability, hence maximizing its influence on model output. If the use of this

uniform distribution contributes very little to the output variability, as determined by a sensitivity analysis, the input distribution is not in need of refinement. However, on the contrary, if the parameter were determined to be important to the model output variability, refinement would be imperative.

Average energy per disintegration ($\bar{\text{E}}$). The input distribution describing the average energy of the beta particles emitted by the disintegration of tritium to ^{3}He, is also introduced with the least amount of bias, for the reasons given above. In reality, there is no variability in the average beta energy of tritium. The only uncertainty lies in how well that average value is known. The value of $\bar{\text{E}}$ used here, reported as 5.69 keV, was taken from the National Nuclear Data Center's (NNDC) computer database[*]. Because the value is reported to three significant figures by the NNDC, this parameter will be varied by ±1% and will be assigned a triangular distribution with a mode of 5.69 keV. To determine the significance of the assigned ±1% variability, the impact of $\bar{\text{E}}$ will also be assessed while allowing the parameter to vary by ±10%.

RESULTS

Uncertainty. Variability in the tritium dose conversion factor (DCF) was estimated using two different distributions representing the tritium quality factor (QF). As stated above, the QF distributions represent RBE data based on, in one case, a standard radiation of orthovoltage x-rays and, in the other case, a standard radiation of ^{60}Co or ^{137}Cs gamma rays. The calculated DCF distributions are given in Fig. 2. Log- and linear-probability plots of the frequency distributions, indicate that both functions are best described as lognormal. The DCF distribution using a QF derived from an x-ray standard has a geometric mean (GM) of 2.2×10^{-11} Sv Bq^{-1} and a geometric standard deviation (GSD) of 1.6. Using gamma radiation as the standard for deriving the QF results in a DCF distribution with a GM of 3.0×10^{-11} Sv Bq^{-1} and a GSD of 1.5. The dose conversion factors reported for tritium by the DOE and EPA are 1.7×10^{-11} and 1.73×10^{-11} Sv Bq^{-1},

respectively (USDOE 1988; USEPA 1988). The DOE and EPA dose factors were derived using a quality factor of one (1) for tritium and are approximately 25-50% lower than the median values calculated here. The deterministic values recommended by the DOE and EPA fall at the 24th percentile of the x-ray standard DCF and at the 8th percentile of the gamma-standard DCF (Fig. 2).

To assess the overall influence of quality factor, a DCF distribution was generated while holding QF constant at a value of unity (Fig. 3). As compared to the distributions in Fig. 2, the probabilistic estimate of dose conversion factor, again fitting a log-normal distribution, shifts dramatically to the left being characterized by a geometric mean of 1.3×10^{-11} Sv Bq^{-1} and a geometric standard deviation of 1.4. In this case, the DOE/EPA recommended DCF falls at the 77th percentile.

Sensitivity analysis. The sensitivity of the tritium-DCF model to its input parameters was determined using a rank correlation method (Hamby 1994; Hamby 1995). The results of this analysis (Tables 2 and 3) show that, of the five inputs for the calculation using an x-ray-standard quality factor, the model was most sensitive to the QF itself, followed by the biological half-life of tritium, and the mass of soft-tissue. For the calculation of DCF using a gamma-standard QF, the model was most sensitive to biological half-life, followed closely by the quality factor and then the soft-tissue mass. As expected, the variance associated with the average energy per disintegration and the fraction of tritium absorbed does not contribute significantly to the total variance of either DCF distribution. When the range of possible values of average beta energy was increased from $\pm$ 1% to $\pm$ 10%, there was no significant change in the DCF output (GM <1% and range essentially unchanged).

CONCLUSIONS AND DISCUSSION

The probabilistic estimates of both tritium dose conversion factors span a range that is a factor of about 15 from the highest to lowest calculated values. Additionally, the minimum and maximum values of the two distributions, whether originating from either using an x-ray or a gamma RBE standard, are different by less than 10%. The skewness of the distributions (see Fig. 2) is their major difference.

The DCF distribution is essentially controlled by equal influence from the quality factor and the biological half-life of tritium oxide in the body. While the input distributions of both variables are somewhat uncertain, the biological half-life of HTO is more easily characterized than the tritium quality factor. The DOE/EPA recommended DCF is comparatively low relative to both DCF distributions calculated here. When the probabilistic DCF is calculated holding QF at a constant value of one (1), the DOE/EPA value appears to be appropriately conservative (Fig. 3). The DCF distributions generated with a variable QF, however, have a definitive impact from, and tend to mimic the general shape of, the quality factor distributions, thus showing the QF to be of paramount importance when determining the tritium DCF. Therefore, in order to reduce uncertainties related to the tritium DCF, our focus should be directed toward determining the relative quality of tritium beta particles and standardizing methods for such. As suggested by Bond et al. (1978) twenty years ago, we certainly need a more narrow definition of the RBE standard radiation.

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FIGURE CAPTIONS

Fig. 1.　　Tritium quality factor (from RBE) distribution using either orthovoltage x-rays or gamma photons as the RBE standard radiation (taken from Straume and Carsten 1993).

Fig. 2.　　Probability distribution of the tritium dose conversion factor as a function of quality factor input.

Fig. 3.　　Probability distribution of the tritium dose conversion factor using a fixed value of 1 for quality factor.

FOOTNOTES

[*]The NNDC database can be found at http://www.nndc.bnl.gov

Table 1. Input parameter distribution characteristics for the tritium DCF model.

Parameter	Symbol	Distribution Type[a]	Median	Variability[b]
Biological Half-Life (d)	T_B	LN	8.7	1.31
Soft-Tissue Mass (kg)	M	LN	70	1.15
Quality Factor (x ray standard)	QF_x	custom[c]	-	1; 3.5
Quality Factor (gamma standard)	QF_γ	custom[c]	-	1; 3.5
Absorption Fraction	f_1	U	-	0.9; 1.0
Avg. Disintegration Energy (keV)	$\overline{E}$	T	5.69	5.63; 5.75

[a]LN = lognormal; T = triangular; U = uniform
[b]Variability for the lognormal distributions is described by their geometric standard deviation; values characterizing variability for the custom, triangular, and uniform distributions are the endpoints of the range.
[c]see Figure 3.

Table 2. Sensitivity analysis results using a QF derived from an x-ray standard.

Parameter	Symbol	Rank Correlation
Quality Factor (x ray standard)	QF_x	0.76
Biological Half-Life	T_B	0.54
Soft-Tissue Mass	M	-0.27
Avg. Disintegration Energy	$\overline{E}$	-0.06
Absorption Fraction	f_1	0.05

Table 3. Sensitivity analysis results using a QF derived from a gamma standard.

Parameter	Symbol	Rank Correlation
Biological Half-Life	T_B	0.66
Quality Factor (gamma standard)	QF_γ	0.60
Soft-Tissue Mass	M	-0.34
Absorption Fraction	f_1	0.08
Avg. Disintegration Energy	$\bar{E}$	-0.03

Intake, A_O

f_1

BODY
A(t)

λ_E

Excretion and Decay

$^{3}_{1}$H (12.33 y)

1/2+   0.0

Q = 5.69 keV

100% β^{-}_{1}

1/2+   0.0

$^{3}_{2}$He (stable)