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RADIOTOXICITY OF TRITIATED WATER AND TRITIATED HYDROGEN

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Tritium in the form of tritiated water is much more radiotoxic than tritiated hydrogen. The effective dose which results from tritiated water as the oxidation product of inhaled tritiated hydrogen gas makes about 55 per cent of the effective dose due to direct irradiation of the lungs by tritiated hydrogen only. The inclusion of the dose due to the production of tritiated water would lower the relative toxicity ratio as well as the value of derived air concentration for tritiated hydrogen. Therefore, to assess the health hazard from tritium exposure, for the two species the relative significance of 1:12000 should be used, instead of the relative radiotoxicity ratio of 1:25000 as given by the International Commission on Radiological Protection.

Key terms
derived air concentration, dose estimate, environmental tritium, tritium health hazard

Tritium (^{3}H or T), a radioactive isotope of hydrogen, decays by beta decay with a half-life of 12.35 years, maximal energy of 18.6 keV, and average energy of 5.6 keV. Owing to low energy and, accordingly, a short range of beta particles originating from tritium decay, tritium does not pose an external radiation hazard. However, as water vapour is readily absorbed by the body, tritium being its constituent poses an internal hazard.

In the environment, tritium is sometimes found around nuclear facilities, particularly close to heavy water reactors. Due to its proposed use as fuel in the first generation of fusion reactors, it is expected to be present in large quantities in those facilities as well as around them, particularly in the gaseous form.

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Generally, the radioactive material released to the environment becomes involved in a complex series of physical, chemical and biological processes. Some of those processes lead to progressive dilution, others to physical or biological reconcentration followed by transfer through various, sometimes interdependent, pathways to man (1). Tritium is a good example of how environmental pathways and a consequent impact of the radionuclide released to the atmosphere depend on its chemical form. Environmental tritium can be found in the form of tritiated water (HTO, T_2O or DTO) or tritiated hydrogen gas (HT, T_2 or DT). Generically, tritiated water is usually referred to as HTO, and tritiated hydrogen as HT.

Tritium in the form of tritiated water is much more radiotoxic (2) and has a considerably shorter tropospheric lifetime than tritiated hydrogen. An estimate of the relative radiotoxicity of tritiated hydrogen to tritiated water can be obtained from the ratio of the respective derived air concentrations (DAC). DAC_{HTO} is $8x10^5$ Bqm^{-3} (2-4) and DAC_{HT} $2x10^{10}$ Bqm^{-3} (2), the ratio being about $2.5x10^4$.

Although *Pinson and Langham* have shown that in humans tritiated hydrogen will oxidize to tritiated water, probably because of the bacterial activity in the gut (5), dose calculations of the International Commission on Radiological Protection (ICRP) take into account the dose due to the tritiated hydrogen gas in the air to the lungs only (2). Once tritiated water has entered the body, uniform mixing with the body water will occur within a few hours (6) i.e. tritiated water will be uniformly distributed through the whole body. The inclusion of the dose due to the production of tritiated water would increase the HTO:HT toxicity ratio, and therefore lower the value of derived air concentration for tritiated hydrogen.

DOSE ESTIMATES

The dose caused by tritiated hydrogen inhalation consists of two segments: one which is due to the presence of tritiated hydrogen gas in the lungs (H_L) and the other which is due to the tritiated water in soft tissues (H_{ST}). Thus, the overall effective dose due to tritiated hydrogen inhalation can be expressed as:

$$E = \sum_T W_T H_T = W_L H_L + W_{ST} H_{ST} \qquad /1/$$

where E is effective dose, H_T are equivalent doses for tissues T and W_T are tissue weighing factors (7, 8) for the lungs and soft tissue; i. e. $W_L = 0.12$ and $W_{ST} = 1$.

The latest recommendation for occupational dose limit is 0.02 Sv per year per person (8). However, as DAC_{HTO} of $8x10^5$ of Bqm^{-3} and DAC_{HT} $2x10^{10}$ Bqm^{-3} taken from references (2-4) are based on the dose limit of 0.05 Sv per year, for consistency sake, the latter value is used in calculations.

To estimate whether E exceeds the occupational dose limit of 0.05 Sv per year, the effective dose integrated through a period τ, where τ=1 year, must be determined:

$$E_T = \int_0^\tau \dot{E}(t)dt = \int_0^\tau (W_L\dot{E}_L + W_{ST}\dot{E}_{ST})dt \qquad /2/$$

$\dot{E}_T$ is effective dose rate for tissue T.

By definition, the effective dose in an organ or tissue T is:

$$E = W_T H_T = W_R W_T D_{T,R} = W_R W_T \int \dot{D}_{T,R}(t)dt = W_R W_T \frac{\varepsilon}{M_T} \int A_T(t)dt \qquad /3/$$

where:
D_T is the absorbed dose to the target organ T, $\dot{D}_T$ is absorbed dose rate, M_T is the mass of the target organ, $A_T(t)$ is the time dependent tritium activity in the target organ, ε is the average energy released per disintegration of a tritium atom, which is 5.7 keV, i.e. 9.13×10^{-16} J.

Combining /2/ and /3/:

$$E_T = 7.89 \times 10^{-11} \int_0^\tau [\frac{W_L}{M_L} A_L(t) + \frac{W_{ST}}{M_{ST}} A_{ST}(t)]dt \qquad /4/$$

The constant in front of the integral has dimension $JBq^{-1}d^{-1}$ as, for convenience sake, integration is given in days (i.e. τ=365 days). $A_L(t)$ and $A_{ST}(t)$ are functions which represent time dependent activities in the lungs and soft tissue. M_L is the mass of the lungs, that is 1 kg, and M_{ST} is the mass of the soft tissue which is 63 kg (9). As the activity in urine is used for calculating tritium activity in soft tissue, equation /4/ can be rewritten as:

$$E_T = 7.89 \times 10^{-11} \int_0^\tau [\frac{W_L}{M_L} A_L(t) + \frac{W_{ST}K}{M_{ST}} A_U(t)]dt \qquad /5/$$

K is the constant of proportionality between the concentration of tritiated water in urine and in the soft tissue and $A_U(t)$ is the time dependent tritiated water activity in urine after tritiated hydrogen exposure. In order to solve equation /5/, K and $A_U(t)$ must be determined.

THE RELATIONSHIP BETWEEN TRITIATED WATER ACTIVITIES IN URINE AND IN SOFT TISSUE

The metabolic considerations of tritium in human body include water balance in the body, and the amounts of hydrogen (tritium) in free water and in the organically bound form in the tissue. For that purpose, usually three-compartment models are used, accounting for body water and for the fast and slow components of the organically bound tritium (10, 11). However, in order to account for the tritiated hydrogen intake, the HT/HTO GENMOD model was used (12), which is

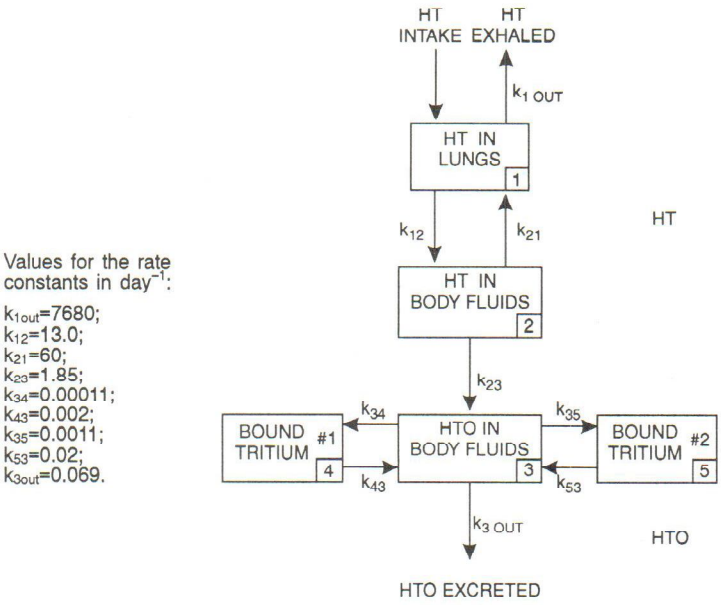

Figure *Model of tritiated hydrogen (HT) and tritiated water (HTO) metabolism*

graphically presented in the Figure. The model is represented by a set of first-order linear differential equations with constant coefficients. For tritiated hydrogen exposures, rate constants given in the Figure are based on the work of *Peterman* and *Peterman and co-workers* (13, 14).

The water balance of 3 l day^{-1} and the mass of body water of 42 l (9), assumed in most tritiated water models, give the mean residence time of approximately 14 days:

$$\frac{1}{k_{3out}} = \frac{T_{1/2}}{\ln(2)} = \frac{42\,l}{3.0\,ld^{-1}}\,14\,d \qquad /6/$$

Therefore, the half-time of body water is 9.7 days or in practical calculations 10 days.

The relationship between tritium activities in urine and in the soft tissue is:

$$A_{ST}(t) = KA_{U}(t) \qquad /7/$$

where $A_{ST}(t)$ is average soft tissue tritium activity in Bqkg^{-1}, and $A_{U}(t)$ is average urine activity in Bql^{-1}. However, since the mass of 1 l of urine equals approximately 1 kg, $A_{U}(t)$ can also be expressed in Bqkg^{-1}. The proportionality constant K converts tritiated water activity in urine (i.e. body water activity) into tritiated

water activity in the soft tissue. After chronic irradiation this constant increases from the value $M_{BW}/M_{ST} = 42/63 = 0.667$, where $M_{BW} = 42$ kg is mass of body water (9), to a somewhat greater value. K can be calculated using a three-compartment model of tritiated water metabolism (the bottom part of the Figure). K is given by:

$$K = \frac{A_{ST}(t)}{A_{d}(t)} = \frac{(X_3 + X_4 + X_5)/M_{ST}}{X_3/M_{BW}} \qquad /8/$$

X_i is tritium activity in i^{th} compartment. Differential equations which give X_i are:

$$\frac{dX_3(t)}{dt} = -(k_{34} + k_{35} + k_{3out})X_3 + k_{43}X_4 + k_{53}X_5 + I(t) \qquad /9/$$

$$\frac{dX_4(t)}{dt} = k_{34}X_3 - k_{43}X_4 \qquad /10/$$

$$\frac{dX_5(t)}{dt} = k_{35}X_3 - k_{53}X_5 \qquad /11/$$

In equilibrium input $I(t) = I$ where I is constant. With time derivatives $dX_i/dt = 0$, the set of differential equations has the solutions:

$$X_3 = \frac{k_{3out}}{I} \qquad /12/$$

$$X_4 = \frac{k_{34}X_3}{k_{43}} \qquad /13/$$

$$X_5 = \frac{k_{35}X_3}{k_{53}} \qquad /14/$$

Substitution into equation /8/ leads to:

$$K = \frac{M_{BW}}{M_{ST}}\left(1 + \frac{k_{34}}{k_{43}} + \frac{k_{35}}{k_{53}}\right) \qquad /15/$$

From equation /15/ and from the rate constants in the Figure, K is calculated to be 0.74. The K determined in this way is in good agreement with the estimated value of 0.8 reported in literature (15).

TRITIATED HYDROGEN METABOLISM IN HUMANS

The metabolism of tritiated hydrogen gas in humans (Figure) can be mathematically described by a set of first-order linear differential equations:

$$\frac{dX_1(t)}{dt} = -(k_{1out} + k_{12})X_1 + k_{21}X_2 + I(t) \qquad /16/$$

$$\frac{dX_2(t)}{dt} = k_{12}X_1 - (k_{21} + k_{23})X_2 \qquad /17/$$

$$\frac{dX_3(t)}{dt} = k_{23}X_2 - (k_{34} + k_{35} + k_{3out})X_3 + k_{43}X_4 + k_{53}X_5 \qquad /18/$$

$$\frac{dX_4(t)}{dt} = k_{34}X_3 - k_{43}X_4 \qquad /19/$$

$$\frac{dX_5(t)}{dt} = k_{35}X_3 - k_{53}X_5 \qquad /20/$$

The analytical solutions of equations /16/ and /18/ give tritiated hydrogen activity in the lungs and tritiated water activity in the body water. For a long enough exposure to a constant tritiated hydrogen concentration in the air, stationary conditions can be assumed. The solutions are:

$$X_1 = \frac{(k_{21} + k_{23})I}{[(k_{1out} + k_{12})(k_{21} + k_{23}) - k_{12}k_{21}]} \qquad /21/$$

$$X_3 = \frac{k_{12}k_{23}I}{k_{3out}[(k_{1out} + k_{12})(k_{21} + k_{23}) - k_{12}k_{21}]} \qquad /22/$$

For a constant exposure to a unit tritiated hydrogen concentration in the air (i.e. 1 Bqm^{-3}), with the breathing rate of 20 $lmin^{-1}$, the input is 28.8 $Bqday^{-1}$. From /21/ and /22/ the total tritiated hydrogen activity in the lungs is 0.00375 Bq and total tritiated water activity in the body water is $X_3 = 0.02103$ Bq. By substituting these values into /5/ and solving the integral, a numerical value for the effective dose integrated over a period of one year is obtained:

$$E = 1.296 \times 10^{-11} + 6.648 \times 10^{-12} \qquad /23/$$

It is evident that the dose to the soft tissue due to tritiated water as oxidation product of tritiated hydrogen inhalation is approximately 55 per cent of the tritiated hydrogen lung dose.

DERIVED AIR CONCENTRATION FOR TRITIATED HYDROGEN

The derived air concentration is defined as the concentration in the air in a working environment that would result in an intake of one ALI (annual limit on intake) by a worker breathing 1.2 m^3h^{-1} (20 $lmin^{-1}$) for 2000 hours (2) (a working year). Exposure to elemental tritium in air during any year is limited by considerations of stochastic effects in the lungs (2):

$$W_L\dot{H}_L\int C(t)dt \leq H_{WB} \qquad /24/$$

where:
$\dot{H}_L$ (in $Svm^3Bq^{-1}h^{-1}$) is the equivalent dose rate to the lungs from exposure to a unit concentration of tritium in the air (1 Bqm^{-3}).
C(t) (in Bqm^{-3}) is the concentration of elemental tritium in the air at any time t.
H_{WB} is the upper limit of equivalent dose for stochastic effects, because of uniform irradiation of the whole body, which is 0.02 Sv per year for workers, according to ICRP recommendations (8). The limits of integration pertain to a working year (2000 hours). ICRP therefore recommends a DAC_{HT} value for tritiated hydrogen which is 1/2000 of the greatest value of the integral from relation /24/:

$$\int C(t)dt = 2000\ DAC_{HT} \qquad /25/$$

From /3/, the equation for equivalent dose H_L is:

$$H_L = W_R \frac{\varepsilon}{M_L} \int V_L C(t)dt \qquad /26/$$

since in stationary conditions $A_L(t) = V_L \times C(t)$ where $V_L = 0.0043\ m^3$ is vital capacity of the lungs and M_L=1 kg is mass of the lungs (9). From equation /26/ the equivalent dose rate can be calculated to be $1.414 \times 10^{-14}\ Svm^3Bq^{-1}h^{-1}$. This value must be multiplied by factor (1+0.55) to account for the effect of tritiated water irradiation. Substitution into /24/with respect to /25/ leads to:

$$0.12 \times 2000 \times DAC_{HT} \times 1.414 \times 10^{-14} \times (1 + 0.55) = 0.05 \qquad /27/$$

(for consistency sake, as explained before, the value of 0.05 Sv is implemented for H_{WB}).

The DAC_{HT} value is then $\approx 9.5 \times 10^9\ Bqm^{-3}$.

The radiotoxicity of a given radionuclide is inversely proportional to its derived air concentration. The radiotoxicity ratio for tritiated water and tritiated hydrogen (R_{HTO}:R_{HT}), representing also relative hazard, is then:

$$\frac{R_{HTO}}{R_{HT}} = \frac{DAC_{HT}}{DAC_{HTO}} = \frac{9.5 \times 10^9}{8 \times 10^5} \approx 12000 \qquad /28/$$

which means that in human body tritiated water is approximately 12000 times more radiotoxic than the same activity of tritiated hydrogen.

CONCLUSION

Tritium is present in many different chemical and physical forms, the two most important ones being tritiated water and tritiated hydrogen. In the air tritiated water is much more hazardous than tritiated hydrogen because it is readily absorbed into the body both by breathing and through the skin. The International Commission on Radiological Protection assuming that the only hazard posed by tritiated hydrogen is the direct dose to the lungs, claims that the relative radiotoxicity between the two species is 1:25000. The equivalent dose which results from tritiated water as the oxidation product of inhaled tritiated hydrogen is about 55 per cent of the equivalent dose due to direct irradiation of the lungs by tritiated hydrogen inhalation. Therefore, the derived air concentration for tritiated hydrogen must be lowered leading to the relative radiotoxicity ratio R_{HTO}:R_{HT} of about 1:12000.

In addition, according to the ICRP 60 recommendations (8) for dose limit for workers, the derived activity concentration for tritiated water (DAC_{HTO}) should be $(2/5)x810^9=3.2x10^5$ Bqm^{-3}. Consequently, DAC_{HT} value from equation /27/ ($9.5x10^9$ Bqm^{-3}) should be lowered by the same factor to the value $3.8x10^9$ Bqm^{-3}.

Considering the conversion of tritiated hydrogen to tritiated water after release of tritiated hydrogen to the atmosphere (16), it is essential to keep tritiated hydrogen concentrations under control to prevent possible future contamination with tritiated water. To assess the health hazard from tritium exposure, a monitor that can discriminate between the two species, at least one part in 12000, is necessary.

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Sažetak

RADIOTOKSIČNOST TRICIRANE VODE I TRICIRANOG VODIKA

Tricij u obliku tricirane vode mnogo je toksičniji od triciranog vodika. Efektivna doza zračenja od tricirane vode kao oksidacijskog produkta udahnutog plina triciranog vodika pridonosi oko 55% efektivnoj dozi od direktnog ozračivanja pluća triciranim vodikom. Uključenje doze zbog nastajanja tricirane vode smanjilo bi relativni omjer radiotoksičnosti tricirane vode i triciranog vodika kao i vrijednost izvedene koncentracije u zraku za tricirani vodik. Stoga, da bi se procijenio ukupni rizik od ozračivanja tricijem, trebala bi se rabiti relativna značajnost od 1:12000 između ova dva kemijska oblika, umjesto relativnog omjera radiotoksičnosti od 1:25000 koji daje Međunarodna komisija za radiološku zaštitu.

Ključne riječi:
izvedena koncentracija u zraku, procjena doze, tricij u okolišu, zdravstveni učinci tricija

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