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TRITIATED WATER DOSIMETRY

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A two-compartment model of tritium metabolism in human body has been used for dosimetry calculations based on tritium concentrations in urine. In stationary conditions the measured background tritium activity in urine was 4.9 ± 0.5 Bq^{-1} corresponding to a dose of 0.100 ± 0.007 μSvy^{-1}. To exceed the allowed limit for occupational exposure of 50 $mSvy^{-1}$, the maximum concentration of tritium in urine following acute exposure to tritium should be above 60 $MBql^{-1}$.

1. Introduction

Tritium, the radioactive isotope of hydrogen, has chemical properties identical to those of light hydrogen. The health hazard of tritium oxide (HTO) to global population is a consequence of ubiquitous nature of tritium. It follows the same pathways into man as ordinary water (ingestion of liquids and foodstuffs, and absorption through the skin) causing an internal irradiation. When tritium enters the organism in the form of HTO, within a few hours all tissues are permeated and receive a uniform radioactive exposure. So, the critical organ for tritium is the whole body[1].

Until atmospheric testing of nuclear weapons, the dominant source of tritium was cosmic ray interaction with ^{14}N in the stratosphere. Today, the major sources

of environmental tritium are production in the stratosphere, consumer products and the nuclear fuel cycle. In the future, the first generation of fusion power plants is likely to have large inventories of tritium, leading to the possibility of majo chronic and acute releases.

Tritium has a radioactive half-life of 12.3 years and decays to ^{3}He by the emission of a low energy beta particle (maximum energy 18.6 keV and average energy 5.7 keV[2]). The low energy of the beta particle, and the lack of photon emission precludes the use of monitoring *in vivo*. For that reason urinary activity can be used to estimate the activity of body water, i.e. soft tissue.

The committed dose equivalent to an organ or tissue is directly proportional to risk. The annual limit on committed dose equivalent is equal to 0.05 Sv for professionals and 0.001 Sv for the general public. In order to estimate whether the dose exceeds that allowed value, it is necessary to compute the committed dose equivalent integrated over the period of one year. According to the definition, the effective dose equivalent received by the organ or tissue is:

$$H_E = w_T H_T = w_T Q N D_T = w_T Q N \int \dot{D}_T(t)\,\mathrm{d}t$$

$$H_E = w_T Q N \frac{E}{M_T} \int A_T(t)\,\mathrm{d}t. \tag{1}$$

H_E is the dose equivalent,
H_T is the dose equivalent to tissue T,
w_T is the ICRP weighing factor for tissue T,
Q is the quality factor,
N is the product of all the other modifying factors,
D_T is the dose which the organ (tissue) receives,
$D_T(t)$ is the dose rate,
M_T is the mass of target organ T,
$A_T(t)$ is the time-dependent activity in the target organ (tissue) T,
E is the average absorbed energy per radioactive decay.

For tritium, the weighing factor for the soft tissue $w_{st} = 1$. According to the recommendations of the International Commission on Radiological Protection (ICRP), for tritium $Q = N = 1$[3]. The mass of the soft tissue is 60 kg[4]. E, average absorbed energy per radioactive decay is $9.1314 \cdot 10^{-16}$ J, since the average energy of beta particles for tritium is 5700 eV.

In order to calculate the dose, the retention function $A(t)$ which describes activity vs. time in tissue T must be known. Measurements of tritium activity in urine at certain fixed intervals must be carried out to get enough data for fitting an analytical function. Then:

$$A_T(t) = K A_u(t), \tag{2}$$

$A_u(t)$ is the time-dependent activity in the urine and K is the constant of proportionality between the activity in the soft tissue and the activity in body water (urine).

It is equal to the ratio of volume of body water to the body mass. For the ICRP reference man it is approximately equal to:

$$K = \frac{V_{body\ water}}{M_{body}} = \frac{42\,\mathrm{l}}{60\,\mathrm{kg}} = 0.7\,\mathrm{l\,kg^{-1}}, \tag{3}$$

as the body water content in ICRP reference man is 42 l[4].

2. Material and methods

Analysis of HTO (tritiated water) was performed in a low-level tritium laboratory. Azeotropic distillation of urine with toluene was performed in order to extract water.

Samples were counted on Tri-Carb 2060XL liquid scintillator spectrometer coupled to an IBM PC 286 microcomputer. Counting was performed in the low-level mode. The volume of samples was 20 ml (8 ml of water and 12 ml of *Instagel* scintillator). The counting time depended upon the activity of samples, but it was not less than 100 min. Efficiency was determined by internal spiking. Standards were obtained from Amersham and Atomic Energy of Canada Limited. Efficiency was $21.5 \pm 0.1\%$.

The water used for background determination was deep ocean water obtained from the World Health Organization (WHO).

Typical background was 2.18 ± 0.15 cpm.

3. Results and discussion

Mathematical model of HTO metabolism in human body

Tritiated water, whether inhaled, ingested or absorbed through the skin is assumed to be immediately and completely mixed with the total body water[1]. Some of the tritium in body water will exchange with hydrogen in tissue cells. Thus, two main compartments are the tritium in body water (tissue free water tritium — TFWT) and the tritium in the organic material (organically bound tritium — OBT). The OBT is usually considered to have a few sub-compartments (the tritium bound to carbon, oxygen, nitrogen and sulphur). As the main fraction of tritium in the OBT compartment is bound to carbon, for calculating the dose from tritium which has entered the body as tritiated water, the OBT compartment is not further subdivided[5]. The model is presented in Fig. 1.

The main routes of entry into the TFWT compartment are from food, water and penetration through the skin. The main route of entry into the OBT compartment is from the TFWT compartment.

The rate constants k_{12}, k_{21} and k_{1out} are fractions of activity exchanged between the compartments or excreted in unit time.

The unit for the rate constants is d^{-1}. The constants k_{21} and k_{1out} are connected with tritium half-lives t_i in respective compartments by the relation:

$$k_{ij} = \frac{\ln(2)}{t_i} = \frac{1}{t_i'}, \tag{4}$$

t_i' is the mean residence time of tritium in compartment i. As t_1 closely approximates the turnover of body water in compartment 1, it can be calculated from daily intake and the mass of total body water to be about 10 days for the ICRP reference man, the actual range being 4—18 days[5]. t_2 can be calculated from the daily carbon balance and the mass of total body carbon to be approximately 40 days[5].

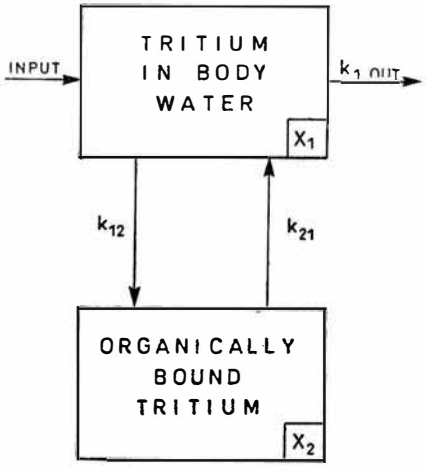

Fig. 1. Model of HTO Metabolism in Human Body.

According to equation (4) k_{1out} and k_{21} can be calculated to be: 0.0693 and 0.0173 d^{-1}, respectively. k_{12} was estimated to be about 25 times lower than k_{21}[6-8] i. e. 0.0007 d^{-1} for the ICRP reference man.

The model is mathematically described by the system of first-oder linear differential equations:

$$\frac{dX_1(t)}{dt} = -(k_{12} + k_{1out})X_1 + k_{21}X_2 + I(t), \tag{5}$$

$$\frac{dX_2(t)}{dt} = k_{12}X_1 - k_{21}X_2, \tag{6}$$

where $I(t)$ is input in Bqd^{-1}.

The functions $X_1(t)$ and $X_2(t)$ are the activities present in compartments 1 and 2 at time t and $I(t)$ is input in Bqd^{-1}.

Stationary conditions

In stationary conditions (constant input):

$$\frac{dX_1(t)}{dt} = \frac{dX_2(t)}{dt} = 0. \tag{7}$$

The solutions are:

$$X_1 = \frac{I}{k_{1\,out}} = 14.4 \times I, \tag{8}$$

$$X_2 = \frac{k_{12}\, I}{k_{21}\, k_{1\,out}} = 0.6 \times I. \tag{9}$$

According to Eq. (1), in stationary conditions, the committed effective dose equivalent in a period of one year is:

$$1\,\mathrm{SvGy^{-1}} \times 5700\mathrm{eV} \times 1.602 \times 10^{-19}\mathrm{JeV^{-1}} \times 0.7\,\mathrm{lkg^{-1}} \times C\,\mathrm{Bql^{-1}}$$

$$\times\ 365\mathrm{dy^{-1}} \times 86400\mathrm{sd^{-1}} = 2.02 \times 10^{-8} \times C\,\mathrm{Svy^{-1}}, \tag{10}$$

where C is the activity of HTO in urine in $\mathrm{Bql^{-1}}$.

In stationary conditions, constant concentration of 5×10^4 $\mathrm{Bql^{-1}}$ urine would result in the allowed committed effective dose equivalent of 1 $\mathrm{mSvy^{-1}}$ for members of the general public. Fig. 2 shows background HTO activity in urine in a 30-day

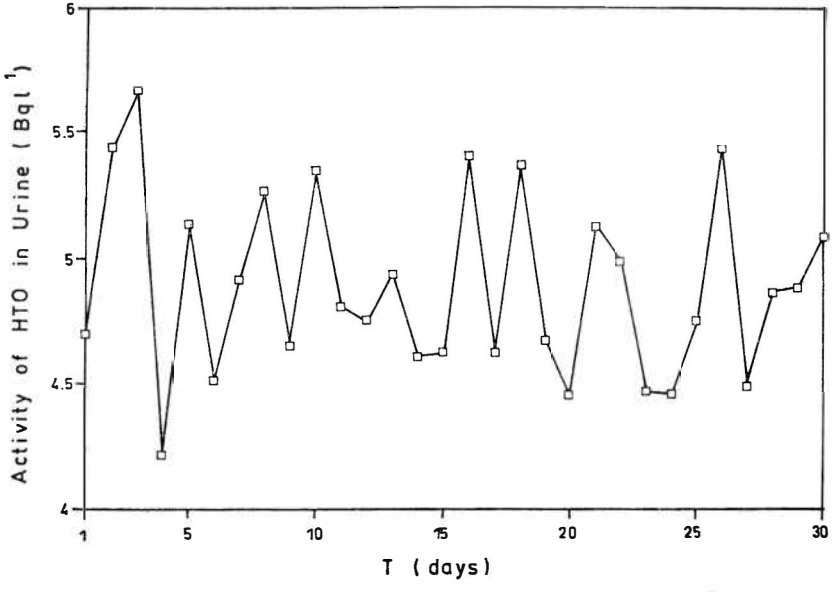

Fig. 2. Background Activity of HTO in Urine in a 30-day Period.

period. The average activity was $4.9 \pm 0\,5$ Bql^{-1}. Using tritium activities measured in various foodstuffs[9], daily intake can be estimated to be up to 15 Bq. Using Eq. (8) total activity in the body water compartment was calculated to be $X_1 = 216$ Bq. Then, the activity in urine is:

$$C = X_1 \text{ Bq} \times \frac{1}{6\theta} \text{kg}^{-1} \times \frac{1}{0.7} \text{kgl}^{-1} \approx 5.1 \text{ Bql}^{-1}, \tag{11}$$

which is in good agreement with the experimentally found value of 4.9 Bql^{-1}. If average activity remained the same (4.9 ± 0.4 Bql^{-1}) throughout the year, the resulting dose would be: 0.100 ± 0.007 μSvy^{-1}.

Single intake

It is assumed that the C_0 activity was taken into the compartment of the body water at time $t = 0$. After initial input $I(t)$ is equal to 0. The model is represented by a set of equations:

$$\frac{\mathrm{d}X_1(t)}{\mathrm{d}t} = -(k_{12} + k_{1out})\, X_1 + k_{21}\, X_2, \tag{12}$$

$$\frac{\mathrm{d}X_2(t)}{\mathrm{d}t} = k_{12}\, X_1 - k_{21} X_2. \tag{13}$$

The initial conditions are: $X_1(0) = C_0$ and $X_2(0) = 0$. C_0 is initial concentration of HTO in the first compartment, in Bqkg^{-1}. Solutions to the system can be found using the Laplace transformations. The solutions are:

$$X_1(t) = \frac{C_0 (k_{21} + s_1)}{s_1 - s_2} \mathrm{e}^{s_1 t} + \frac{C_0 (k_{21} + s_2)}{s_2 - s_1} \mathrm{e}^{s_2 t}, \tag{14}$$

$$X_2(t) = \frac{C_0\, k_{12}}{s_1 - s_2} (\mathrm{e}^{s_1 t} - \mathrm{e}^{s_2 t}), \tag{15}$$

where:

$$s_1 = -\frac{1}{2} [k_{12} + k_{1out} + k_{21} + \sqrt{(k_{12} + k_{1out} - k_{21})^2 + 4k_{21}k_{12}}]. \tag{16}$$

and

$$s_2 = -\frac{1}{2} [k_{12} + k_{1out} + k_{21} - \sqrt{(k_{12} + k_{1out} - k_{21})^2 + 4k_{21}k_{12}}]. \tag{17}$$

The committed effective dose equivalent is calculated by substitution of Eq. (14) into (1) and integrating over the period T:

$$H_E = w_T\, QN \frac{E}{M_T} \int_0^T X_1(t)\, \mathrm{d}t \tag{18}$$

with solution:

$$H_E = w_T\, QNC_0\, \frac{E}{M_T}\left[\frac{k_{21}+s_1}{s_1(s_1-s_2)}(e^{s_1 T}-1)+\frac{k_{21}+s_2}{s_2(s_2-s_1)}(e^{s_2 T}-1)\right]. \quad (19)$$

The committed effective dose equivalent to whole body received in a peroid of one year after a single intake of tritium is then:

$$H_E = 7.97 \times 10^{-10}\, C_0\, \mathrm{Svy}^{-1}, \quad (20)$$

where C_0 is initial concentration of tritium in urine in Bql^{-1}.

Concluding remarks

The tritium concentrations determined in urine of workers in the Nuclear Power Plant Krško, Slovenia, working in containment during 1990 refuelling ranged from 3×10^3 to 10^4 Bql^{-1} leading to doses from 2.4×10^{-6} to 8×10^{-6} Svy^{-1}.

To exceed the allowed dose for occupational exposure of 50 mSvy^{-1}, the maximum concentration of tritium in urine following acute exposure (single intake) of tritium should be above 60 MBql^{-1}.

Above this level, therapeutic measures should be considered, exact time of tritium intake must be determined and working conditions must be checked in order to verify whether other workers have been exposed. If possible, individual retention function must be determined.

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DOZIMETRIJA TRICIRANE VODE

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Na osnovi poznatih koncentracija tricija u urinu izveden je, pomoću dvokompartmentnog modela metabolizma tricirane vode, izraz za dozu. U stacionarnim uvjetima mjerene aktivnosti tricija u urinu čovjeka koji nije bio izložen triciju su bile 4.9 ± 0.5 Bql^{-1} što odgovara dozi od 0.100 ± 0.007 μSvy^{-1}. Da se prijeđe 50 $mSvy^{-1}$, što je dozvoljena godišnja doza za profesionalno izložene osobe, maksimalna koncentracija tricirane vode u urinu (nakon akutne izloženosti triciju) bi trebala biti iznad 60 $MBql^{-1}$.