TRITIUM TRANSFER AND CONCENTRATION IN THE OCEAN

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ABSTRACT

Environmental transport models have been developed for evaluation of radiation doses from tritium released into the atmosphere. Recently, models contain not only inhalation and skin absorption as routes of tritium transfer from the atmosphere to humans, but also the ingestion pathway. Tritium releasing to the environment is contribute to added tritium concentration in the ocean. This paper describes the tritium transfer from the environment to the human body and the possible health effects if it is taken inside the body, the factors which influence the differences of tritium concentrations in the coastal seawater, and the formula of the tritium concentration balance in the ocean surface.

Key words: Tritiated water, HTO, HT

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INTRODUCTION

Tritium ($^3$H) is a radioactive form of hydrogen. Tritium is produced both by natural process, the interaction of cosmic rays with the atmosphere and by man-made process (in nuclear reaction). The half-life of tritium is 12.3 years. This means that the concentration of tritium in the environment is reduced by one-half in every 12 years, disregarding newly generated tritium. When tritium undergoes radioactive decay, it is transformed into non-radioactive helium through the emission of a “beta” particle, or electron from its nucleus. The very low energy radiation emitted by tritium is too weak to cause a radiation hazard outside to the human body. The radiation from tritium can only travel about 5 millimeters in the air and can be stopped completely by a sheet of paper or by ordinary clothing.

Tritium can deliver a radiation dose, if it is taken inside the body. Such an intake could occur by breathing tritiated water vapor in the air, or by eating or drinking tritium-contaminated foods or water. Even though tritium radiation cannot penetrate skin, tritiated water vapor in the air, like regular water vapor in the air, may be absorbed through the skin. Likewise, a person might absorb small amounts of tritiated water through the skin when swimming or wading in contaminated water. A developing fetus could also receive tritium absorbed into its mother’s body through one of these routes.

Tritium in organism is classified into two types: free water tritium (FWT) and organically bound tritium (OBT). FWT exists in organisms as HTO, and metabolized in a similar manner to H$_2$O. OBT is usually found as either tritium directly bound to C-C skeletal materials or as a part of compound such as -COOH,
OH, -SH and –NH. The skeletally bound tritium is not easily exchanged with FWT, while the tritium in other organic compounds quickly equilibrates with FWT. Limited data concerning the non-exchangeable fraction of OBT are available, and 60-90% of organic materials are estimated to be non-exchangeable. In comparison with FWT, OBT generally remains in organisms for longer periods, and tritium is more easily assimilated into OBT fraction of organisms.

Tritium in food is also classified into two types: FWT and OBT. The contribution of OBT in foods, after a radiation dose of released tritium, strongly depends on season that the tritium released. In the environment, tritium is also classified into gas tritium (HT) and metan tritium (CH$_3$T).

While most of the leaked tritium will be in the form of HT and HTO, other tritiated organic compounds like CH$_3$T may be included. Since the bioavailability of HT to plants and animals is lower than HTO, HT will give lower dosage of radioactivity per unit than HTO. However, when HT is deposited on the ground, it is quickly oxidized into HTO, mainly by microbial activity. HTO is incorporated into the human body by both inhalation and by ingestion of contaminated foods.

Natural process of tritium is produced in the environment as the result of the interaction of cosmic rays and the atmosphere gas. After tritium formed in the atmosphere, it changed into the water molecule through oxidation process, and then would reach the terrestrial surface and the ocean surface by the rain.

Since the early 1960’s when a large amount of tritium was discharged into the atmosphere and that the number of atmospheric nuclear test which is a major source in the atmosphere has decreased, tritium levels in recent precipitation have decreased in the environment. From a global point of view, low tritium levels have been measured in places where oceanic climate predominates over continental climate (Sheell et al., 1974).

Momoshima et al. (1986) reported that Japanese coastal seawater has about two times higher tritium levels than eastern Pacific surface water indicated that coastal seawater is apparently by runoff from land in spite of the sampling being carried out the place where river water does not flow into the ocean.

**Atmospheric Tritium to Human**

The transfer pathways of tritium released into the atmosphere, in the form of HTO, are outlined in Figure 1. Atmospheric HTO in puff from sources, is deposited from the atmosphere to the ground where part of it may absorbed by plants via the root system.

**Fig. 1.** Conceptual model of tritium transfer from atmosphere to humans
Direct deposition of HTO on plan surfaces is also considered to be an important transfer pathway. After the puff has passed, HTO is re-emitted from the soil and plants back into the atmosphere. Tritium is also transferred from the atmosphere to farm animals via inhalation, skin absorption, and ingestion of contaminated foods. The skin absorption pathway is not considered to be as important as the inhalation route. Tritium is taken by humans via inhalation of atmospheric HTO and ingestion of tritium contaminated plant and animal foods.

HT does not remain in plant and animal tissues for long periods, therefore radiation doses from HT are thought to be insignificant unless the receptor is directly immersed in the puff (Okada and Momoshima, 1993). The residence time of HTO in organisms is generally far longer than HT (Murphy, 1993). Therefore, oxidation of HT to HTO is an important factor in the dose evaluation of tritium.

Oxidation of HT in the atmosphere is negligible (Brown et al., 1990). While plants and animals do not oxidize HT at a significant rate, it is well documented that soil may effectively oxidize HT (McFarlane et al., 1978). HT oxidation in the soil is mainly a biotic process, although a weak abiotic activity is known to occur. Sterilization of soil with heat (McFarlane et al., 1978), chemicals (Suscat and Murphy, 1981) or radiation (Momoshima et al., 1992) inactivates the majority of the soils oxidation capability. Deposition velocity and oxidation of HT, from the atmosphere to the soil, depends on the soil’s void content, temperature and location (Dunstall et al. 1985; Forstel 1986, Spencer and Dustal, 1986). Water content is an important factor controlling void content and biotic activity of the soil. The low soil water content increases the number of voids, and allows easier penetration of HT into the soil. Although deposition velocity increases with decreasing water content, it decreases in conditions of extremely low water content because of the reduction in biotic activity. Since oxidation capability depends on the sampling location of the soil, local parameters are important in realistic dose estimations.

Environmental models have been developed to evaluate radiation doses of tritium released into the atmosphere. Raskob developed the UFOTRI code which is the first environmental transfer model to combine inhalation with the long term ingestion pathway. The UFOTRI code consists of a Gaussian dispersion model and a biosphere compartment model. Doses of released tritium depend on many factors such as release height, climatic condition and season. Table 1 shows a few examples of the model’s result for a person receiving maximum exposure; 1 km away from a release point. In these cases, the dose contributions via the ingestion pathway exceed those of the inhalation route, although all foods were assumed to be produced locally (Raskob, 1995). However, the fractional dose contributions of the ingestion depend on the various local conditions. Generally, higher contributions were observed in high total dose cases (Gulden and Raskub, 1992).

Table 1. Estimated dose to maximum exposed person from accidental

<table>
<thead>
<tr>
<th>Released compound</th>
<th>Effective dose (nSv)</th>
<th>Contribution (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HTO</td>
<td>6.4</td>
<td>19</td>
</tr>
<tr>
<td>HT</td>
<td>0.4</td>
<td>0.3</td>
</tr>
</tbody>
</table>

Because the energy of electrons emitted during the decay of tritium nucleus is insufficient to penetrate skin, this report does not address external radiation exposure, but only internal dose routes. The following exposure pathways were considered:

1. Tritiated water vapor entering the body through respiration.
2. Tritiated ingested with water during swimming or wading, home-grown foods, or breast-milk (in the case of infants) and absorbed into the body through the gastro-intestinal tract.

3. Tritium from tritiated water vapor in air taken through the skin.

4. Tritium from tritiated water in surface water taken through the skin during activities that involve dermal contact with the contaminated water, i.e., washing, swimming and wadding in surface water.

5. Tritium transferred from the body water of pregnant women to the developing fetus.

**ATMOSPHERIC TRITIUM TO RICE**

Although rice is an important food source, the transfer dynamics of tritium from the environment to rice are poorly understood. A small greenhouse was constructed outside for the exposure experiments, and HTO vapor was introduced into it for 24 hours. HTO transfer from the atmosphere to rice plant was examined at different intervals after anthesis.

The transfer of HTO from the atmosphere to plant tissue is described by the following formula (International Atomic Energy Agency, 1990).

\[
C_p = a(R_hC_a) + (1-R_h)C_s(1-e^{\lambda t})
\]

With \(C_p\) is HTO concentration in plant tissue, \(C_a\) is HTO concentration in air, \(C_s\) is HTO concentration in soil, \(R_h\) is relative humidity in air, \(a\) is rate constant for transfer from atmosphere to plant tissue, \(\lambda\) is isotope correction and \(t\) is time elapsed from the beginning of exposure. Rate constant \(a\) was obtained from the measurement results of plant samples by non-linear least square fitting method under the assumption of constant \(\lambda\) during exposure.

**TRITIUM CONCENTRATION IN THE OCEAN**

The tritium concentrations of lake and river water show some scatter compared with that of coastal seawater and the variation can be attributed to differences in some geographical and hydrographical situations/residence time of water, tritium concentration of supplied water, a turnover rate of coastal seawater etc.

The tritium balance in the ocean is expressed by the following equation (Altison and Holmes, 1979).

\[
\frac{d(V_1T_1)}{dt} = \sum I_1T_1 - EBT_1s + X(T_a-BT_1s) - T_1\sum O_j - \lambda V_1T_1
\]

with
\(V_1\) : volume of ocean water sample, with tritium concentration \(T_1\)
\(T_{1s}\) : tritium concentration at seawater surface sample
\(I_1\) : rate of inflow from source I with tritium concentration \(T_1\)
\(E\) : rate of evaporation
\(O_j\) : rate of outflow to sink j
\(T_a\) : tritium concentration of atmospheric water vapour
\(X\) : exchange rate
\(B\) : HTO-H2O fractionation factor
\(\lambda\) : decay constant for tritium.

The formula is simplified by ignoring minor contribution terms such as the radioactive decay and the exchange of tritium between the atmosphere and the ocean surface.

**HAZARDOUS TRITIUM TO HUMAN HEALTH**

The fact that tritium emits very low energy radiation, is diluted throughout the body, and is eliminated fairly quickly from the body that makes it as one of the least hazardous radioactive materials. Tritium is a potential health risk only if it is taken inside the body.
The only studies that show radiation effects on human health are studies of individuals exposed to high dose levels (e.g., from the atomic bombing of Hiroshima and Nagasaki) – well above those associated with background radiation, which are orders of magnitude higher than tritium from the environment. It is assumed that low-dose radiation does affect health. The health risk estimates in the risk assessment are extrapolated from effects observed only at high dose levels.

There is evidence from experiments with animals and cell cultures exposed to very high levels of radiation from HTO results in mutations and cell disruption that can lead to health effects associated with radiation, including cancer. Both leukemia and non-leukemia soft-tissue carcinomas are associated with high levels of HTO exposure. Based on experimental evidence, this risk assessment assumes that the likelihood of an individual suffering a cancer as a result of exposure to tritium depends upon the magnitude of the dose of tritium radiation and the time period over which the dose is received.

Risk estimates for low doses of low linear-energy transfer radiation, such as those for tritium releases, are based on linear extrapolation from selected populations exposed to relatively high dose – that is, greater than 100 mSv for very short time periods (seconds or minutes). When dose levels are much lower and permit natural repair of radiation damage, they could result in a much smaller biological effect per unit dose. Dose levels for uncontrolled areas are several orders of magnitude below these small doses and are estimated to be greater than 0.005 mSv.

Health risk estimates for tritium are therefore based on the large number of experiments with animals and cell cultures. These experiments show that exposure to tritiated water results in mutations and cell disruptions can lead to the health effects possible for ionizing radiation-cancer, heritable genetic effects and reproductive and developmental effects. The health risks are associated with exposure to tritium through inhalation, ingestion of HTO or OBT, or absorption of HTO through the skin. The health effects of ionizing radiation are proportional to the energy carried by the radiation and delivered to living cells (www.lb.gov/ehs/epg/tritium.htm, 2003).

Based on the genetic effects and noninheritable developmental effects identified, it is possible that tritium exposure has this potential. As with cancers, it is assumed that the risk of birth defects from exposure to tritium is proportional to the relative magnitude of the dose and time period over which that dose is received.

CONCLUDING REMARKS

1. Tritium is transferred from the atmosphere to humans through inhalation, skin absorption, ingestion of contaminated foods and pregnant women to the developing fetus.
2. Natural tritium has contribution for tritium concentration in the ocean.
3. The tritium concentration of seawater is depend on geographical and hydro-graphical factor.
4. Exposure of tritium can lead to the health effects such as ionizing radiation-cancer, heritable genetic effects and reproductive and developmental effects.

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