1. Dry deposition and reemission

1.1 Dry deposition of HTO and reemission

During passage of the primary plume, atmospheric HTO vapor diffuses into soil, and HTO condensation occurs at the surface soils (dry deposition). Physical mechanism of the dry deposition of HTO is basically the same that of H_2O , but the deposition (also for reemission) of HTO and H_2O should be treated separately because each molecule follows its individual vapor-pressure deficit (Feinhals and Bunnenberg, 1988; Täschner et al., 1995 and 1997).

Generally the exchange process of HTO between the atmosphere and the soil is expressed in a form of bulk flow (Garland, 1980), as:

 $F_{HTO} = v_{HTO}(\chi_{atm} - \chi_{sa}),$ (1.1.1)

where F_{HTO} is the exchange flux (Bq m⁻² s⁻¹), v_{HTO} is the exchange velocity (m s⁻¹), χ_{atm} and χ_{sa} respectively are the HTO concentration (Bq m⁻³) in the atmosphere at a reference height and in the soil. Flux F_{HTO} expresses HTO deposition flux when $\chi_{atm} > \chi_{sa}$ while re-emission if $\chi_{atm} < \chi_{sa}$. Exchange velocity v_{HTO} depends on both the atmosphere and the soil conditions and is formulated as the inverse of the resistances (Raskob, 1992), as:

 $v_{HTO} = (r_a + r_b + r_s)^{-1},$ (1.1.2)

where r_a is the aerodynamic resistance (m⁻¹ s) characterizing the transfer in the free atmosphere from the reference height to the soil surface, r_b is the boundary layer resistance (m⁻¹ s) describing the mass transfer in the quasi-laminar flow layer adjacent the soil surface and r_s is the soil resistance (m⁻¹ s) regulated by the HTO transport in soil. Resistances r_a and r_b are characterized by the wind speed or the friction velocity (Raskob, 1992; Täschner et al., 1997), whereas the resistance r_s can be calculated from the effective diffusivities of HTO in soil (Garland, 1980; Raskob, 1992; Täschner et al., 1995). In the soil, air HTO concentration is related with the aqueous HTO concentration assuming the equilibrium between the two phases, as:

$$\chi_{sa} = \frac{h_{sat}}{\rho_w} \chi_{sw}, \qquad (1.1.3)$$

where h_{sat} is the saturation of the absolute humidity (kg m⁻³) at the soil temperature, ρ_w is the density (kg m⁻³) of liquid water and χ_{sw} is the HTO concentration (Bq m⁻³) in the soil water. Aqueous HTO transport in soil is normally calculated by a diffusion/advection equation:

$$\theta \frac{\partial \chi_{sw}}{\partial t} = \frac{\partial}{\partial z} D \frac{\partial \chi_{sw}}{\partial z} - q \frac{\partial \chi_{sw}}{\partial z}, \qquad (1.1.4)$$

where θ is the volumetric soil water content, *t* is the time (s), *z* is the vertical coordinate (m) in soil, *D* is the effective diffusivity for the aqueous HTO in soil (m² s⁻¹) and *q* is the advective water flow (m s⁻¹).

To investigate the deposition of the atmospheric HTO to the soil, several field and laboratory

experiments have been conducted, and the results demonstrate that the HTO deposition mainly occurs within the top several millimeters of the soil and steeply decreases with the soil depth (Feinhals and Bunnenberg, 1988; Täschner et al., 1995 and 1997; Atarashi et al., 1998; Yokoyama et al., 1998), indicating that the thickness of the uppermost soil layer should be on the order of millimeters to precisely predict the HTO deposition and also the reemission successively occurs (Täschner et al., 1995). Due to the difference in the hydraulic characteristics, HTO deposition velocity also depends on the textures of the exposed soil (Feinhals and Bunnenberg, 1988).

Soon after the passage of the primary plume, reversal of the HTO gradient across the atmosphere-soil boundary, i.e. χ_{atm} being smaller than χ_{sa} in Eq.(1.1.1), causes the reemission of the deposited HTO to the atmosphere, which enhances the atmospheric HTO level over the subsequent period (secondary plume). Reemission of the HTO is generally the largest at the end of the deposition and becomes lowered as time passes according to the decrease in the aqueous HTO concentration at top soil layers through the HTO evaporation to the atmosphere and the downward HTO diffusion (Garland, 1980; Täschner et al., 1995; Yokoyama et al., 1998 and 2004). Such reduced HTO concentration in the top soil-layers then causes the HTO remission over the subsequent period (Garland, 1980; Täschner et al., 1997; Yokoyama et al., 1998 and 2004).

1.2 Dry deposition for HT and reemission

Because HTO is by four orders of magnitudes more radioecologically-toxic than the tritiated hydrogen (HT), the impact of the release of HT is expected to critically depend on the HT deposition to soil (Täschner et al., 1991), which is caused by the subsurface HT-conversion to HTO by hydrogen-oxidizing microorganisms ubiquitously contained in the surface soils (Conrad and Seiler, 1981; Conrad et al., 1983; Conrad, 1996).

The deposition of the atmospheric HT to the soil is normally evaluated by a resistance approach (Garland and Cox, 1980), as:

 $F_{HT} = v_{HT} \chi_{atm,HT}, \qquad (1.2.1)$

where F_{HT} is the deposition flux for HT (Bq m⁻² s⁻¹), v_{HT} is the deposition velocity (m s⁻¹), $\chi_{atm,HT}$ is the HT concentration (Bq m⁻³) in the atmosphere at a reference height. Similar to the exchange velocity v_{HTO} for HTO expressed by Eq.(1.1.2), deposition velocity v_{HT} for HT is regulated by the turbulent diffusion of HT in the surface atmosphere and by the soil HT transport, but the resistance for the soil transport, corresponding to r_s in Eq.(1.1.2), should include both the limitation through the HT diffusion in the soil and through the microbial HT oxidation there. Soil HT transport is expressed by a diffusion equation (Russel and Ogram 1988) as:

$$\varepsilon \frac{\partial \chi_{s,HT}}{\partial t} = \frac{\partial}{\partial z} \left(D_{HT} \frac{\partial \chi_{s,HT}}{\partial z} \right) - e_{HT}, \qquad (1.2.1)$$

where ε is the air-filled porosity, $\chi_{s,HT}$ is the HT concentration in the soil air (Bq m⁻³), *t* is the time (s), *z* is the coordinate in the soil (m), D_{HT} is the effective diffusivity (m² s⁻¹) for HT in the soil, and herein HT oxidation is termed as a volume sink e_{HT} (Bq m⁻³ s⁻¹). Assuming a first-order reaction for the HT oxidation in soil, i.e. $e_{HT} = k\chi_{s,HT}$, deposition velocity v_{HT} can be formulated with the coefficient *k* and the effective diffusivity D_{HT} (Russel and Ogram, 1988; Raskob, 1992; Täschner et al., 1995). A more sophisticated but practical useful formulations for v_{HT} , as a function of soil conditions, have been proposed by Yamazawa et al. (2011), in which not only the regulation through the soil HT diffusion but also the regulation through the HT oxidation depending on the soil water content and temperature are considered.

To understand the overall HT behavior in the real environments and quantitatively evaluate HT deposition velocity, field HT-release experiments have been conducted from small-scale ones (e.g., Garland and Cox, 1980; Sweet and Murphy, 1981 and 1984; Fallon, 1982a; Förstel, 1986; Diabaté and Honig, 1988; Täschner et al., 1995), to intensive larger-scale ones in France (e.g., Diabaté and Honig, 1988; Täschner et al., 1988) and in Canada (e.g., Belot et al., 1988; Noguchi et al., 1988; Ogram et al., 1988; Spencer et al., 1988; Davis, et al., 1995). These experimental studies demonstrated that the deposition of HT occurs mainly at the top several-centimeter soil and decreases with the depth, and the obtained HT deposition velocities range from 10^{-5} m s⁻¹ to 10^{-3} m s⁻¹ (McFarlane et al., 1978; Garland and Cox, 1980; Sweet and Murphy, 1981; Fallon, 1982a; Förstel, 1986; Diabaté and Honig, 1988; Noguchi et al., 1988; Ogram et al., 1988; Spencer et al., 1988; Amano et al., 1995; Davis et al., 1995). Variations in the HT deposition velocities over a few orders of magnitude seem to be mainly affected by the soil conditions (water content and temperature), regulating either the HT diffusion in the soil or the microbial HT oxidation (Garland and Cox, 1980; Sweet and Murphy, 1981; Fallon, 1982a; Förstel, 1986; Ogram et al., 1988; Täschner et al., 1988). For modeling the HT oxidation by soil alone, some laboratory experimental results are useful. The oxidation rate of HT (defined by the amount of HT converted to HTO by the unit dry soil mass by the unit time (Bq $kg^{-1}s^{-1}$)) depends on the soil temperature and the soil water content, and seems to have slight dependence on the texture (McFarlane et al., 1979; Fallon, 1982b; Ogram et al., 1988; Ichimasa et al., 1988 and 1999; Momoshima et al, 1990; Smith-Downey et al., 2006; Ota et al., 2007 and 2008). Also, the magnitude of the HT oxidation rate becomes smaller as the soil depth increases, for which the depth scale seems to be around several centimeters (Komuro, et al., 2002). Based on these experimental works, the HT oxidation rate is formulated as a function of the soil water content and the soil temperature (Smith-Downey et al., 2006; Ota et al., 2007 and 2008).

For the HT release case, unlike the HTO release case, emission of the deposited HTO to the

atmosphere occurs even during the passage of the HT plume. Dynamics of the HTO emission is basically identical to the ones of the HTO reemission after the dry deposition of HTO (section 1.1), although the profiles of the HTO in the soil differ between the HT and HTO deposition case (Belot et al., 1988; Foerstel et al., 1988; Ogram et al., 1988; Wiener et al., 1988; Täschner et al., 1995).

Reference for the section 1 (Dry deposition and reemission)

Amano, H., Atarashi, M., Noguchi, H., Yokoyama, S., Ichimasa, M., Ichimasa, Y., 1995. Formation of organically bound tritium in plants during the 1994 chronic HT release experiment at Chalk River. Fusion Technol. 28, 803–808.

Atarashi, M., Amano, H., Ichimasa, M., Ichimasa, Y., 1998. Deposition of D_2O from air to plant and soil during an experiment of D_2O vapor release into a vinyl house. Fusion Eng. Des. 42, 133–140.

Belot, Y., Guenot, J., Caput, C., 1988. Emission of tritiated water formed at soil surface by oxidation of HT. Fusion Technol. 14,1231–1234.

Conrad, R., 1996. Soil microorganisms as controllers of atmospheric trace gases (H₂, CO, CH₄, OCS, N₂O, and NO). Microbiol. Rev. 60, 609–640.

Conrad, R., Seiler, W., 1981. Decomposition of atmospheric hyfrogen by soil microorganisms and soil enzymes. Soil Biol. Biochem. 13, 43–49.

Conrad, R., Weiber, M., Seiler, W., 1983. Kinetics and electron transport of soil hydrogenases catalyzing the oxidation of atmospheric hydrogen. Soil Biol. Biochem. 15, 167–173.

Davis, P.A., Galeriu, D.C., Spencer, F.S., Amiro, B.D., 1995. Evolution of HTO concentrations in soil, vegetation and air during an experimental chronic HT release. Fusion Technol. 28, 833–839.

Diabaté, S., Honig, D., 1988. Conversion of molecular tritium to HTO and OBT in plants and soils. Fusion Technol. 1988. 14, 1235–1239.

Fallon, R.D., 1982a. Molecular tritium uptake in southeastern U.S. soils. Soil Biol. Boiochem. 14, 553–556.

Fallon, R. D., 1982b, Influences of pH, temperature, and moisture on gaseous tritium uptake in

surface soils, Appl. Environ. Microbiol. 44, 171–178.

Feinhals, J., Bunnennberg, C., 1988. Laboratory investigations of HTO deposition to soils. Fusion Technol. 14, 1253–1257.

Foerstel, H., Lepa, K., Trierweiler, H., 1988. Re-emission of HTO into the atmosphere after HT/HTO conversion in the soil. Fusion Technol. 14, 1203–1208.

Förstel, H., 1986. Uptake of elementary tritium by the soil. Radiation Protection Dosimetry. 16, 75–81.

Garland, J.A., 1980. The absorption and evaporation of tritiated water vapor by soil and grassland. Water, Air, and Soil Pollut. 13, 317–333.

Garland, J.A., Cox, L.C., 1980. The absorption of tritium gas by English soils, plants and the sea. Water, Air, and Soil Pollut. 14, 103–114.

Ichimasa, M., Ichimasa, M., Azuma, Y., Komuro, M., Fujita, K., Akita, Y., 1988. Oxidation of molecular tritium by surface soils, J. Radiat. Res., 29, 144–151.

Ichimasa, M., Suzuki, M., Obayashi, H., Sakuma, Y., Ichimasa, Y., 1999. In vitro determination of oxidation of atmospheric tririum gas in vegetation and soil in Ibaraki and Gifu, Japan, J. Radiat. Res. 40, 243–251.

Komuro, M., Ichimasa, Y., Ichimasa, M., 2002. HT oxidation in soils in Ibaraki and isolation, identification of HT oxidizing soil bacteria, Fusion Sci. Technol. 41, 422–426.

McFarlane, J.C., Rogers, R.D., Bradley Jr., D.V., 1978. Environmental tritium oxidation in surface soil. Environ. Sci. Technol., 12, 590–592.

McFarlane, J.C., Rogers, R.D., Bradley Jr., D.V., 1979. Tritium oxidation in surface soils. A survey of soils near five nuclear fuel reprocessing plants. Environ. Sci. Technol., 13, 607–608.

Momoshima, N., Nagasato, Y., Takashima, Y., 1990. Kinetic studies on oxidation of molecular tritium by soils. Appl. Radiat. Isot. 41, 655–660.

Noguchi, H., Matsui, T., Murata, M., 1988. Tritium behavior observed in the Canadian HT rlease study. Fusion Technol. 14, 1187–1192.

Ogram, G.L., Spencer, F.S., Brown, R.M., 1988. Field studies of HT behavior in the environment: 2. The interaction with soil. Fusion Technol. 14, 1170–1175.

Ota, M., Yamazawa, H., Moriizumi, J., Iida, T., 2007. Measurement and modeling of oxidation rate of hydrogen isotopic gases by soil. J. Environ. Radioact. 97, 103–115.

Ota, M., Yamazawa, H., Moriizumi, J., Iida, T., 2008. Measurement and modeling of the oxidation rate of hydrogen isotopic gases by soil. J. Nucl. Sci. Technol. Suppl. 6, 185–190.

Raskob, W., 1992. Modeling of tritium behavior in the environment. Fusion Technol. 21, 636–644.

Russel, S.B., Ogram, G.L., 1988. Modeling elemental tritium deposition, conversion and reemission using Ontario Hydro's tritium dispersion code. Fusion Technol. 14, 1193–1198.

Smith-Downey N.V., Randerson, J.T., Eiler, J.M., 2006. Temperature and moisture dependence of soil H₂ uptake measured in the laboratory. Geophys. Res. Letters. 33, L14813.

Spencer, F.S., Ogram, G.L., Brown, R.M., 1988. Field studies of HT behavior in the environment: 3. Tritium deposition and dynamics in vegetation. Fusion Technol. 14, 1176–1181.

Sweet, C.W., Murphy, C.E., 1981. Oxidation of molecular tritium by intact soils. Environ. Sci. Technol. 12, 1485–1487.

Sweet, C.W., Murphy, C.E., 1984. Tritium deposition in pine trees and soil from atmospheric releases of molecular tritium. Environ. Sci. Technol., 18, 358–361.

Täschner, M., Wiener, B., Bunnenberg, C., 1988. HT dispersion and deposition in soil after experimental releases of tritiated hydrogen. Fusion Technol. 14, 1264–1269.

Täschner, M., Bunnenberg, C., Gulden, W., 1991. Maximum permissible amounts of accidentally released tritium derived from an environmental experiment to meet dose limits for public exposure.

Fusion Technol. 20, 58-64.

Täschner, M., Bunnenberg, C., Camus, H., Belot, Y., 1995. Investigations and modeling of tritium reemission from soil. Fusion Technol. 28, 976–981.

Täschner, M., Bunnenberg, C., Raskob, W., 1997. Measurements and modeling of tritium reemission rates after HTO deposition at sunrise and sunset. J. Environ. Radioact. 36, 219–235.

Wiener, B., Täschner, M., Bunnennberg, C., 1988. HTO reemisson from soil after HT deposition and dose consequences of HT releases. Fusion Technol. 14, 1247–1252.

Yamazawa, H., Ota, M., Moriizumi, J., 2011. Realistic and practical modeling of tritium deposition to bare soil. Fusion Sci. and Technol. (in press)

Yokoyama, S., Noguchi, H., Ichimasa, M., Ichimasa, Y., Fukutani, S., 1998. Deposition of heavy water on soil and reemission to the atmosphere. Fusion Eng. Des. 42, 141–148.

Yokoyama, S., Noguchi, H., Ichimasa, Y., Ichimasa, M., 2004. Re-emission of heavy water vapor from soil to the atmosphere. J. Environ. Radioact. 71, 201–213.

2. Briefing of a complex model

A complex model SOLVEG-II developed by JAEA computes transport and exchanges of heat, water and CO_2 in a multi-layered atmosphere-vegetation-soil system (Nagai, 2005), and dynamically calculates HTO transfers and OBT formation shown in Fig. 2.1.1 (Yamazawa, 2001; Ota and Nagai, 2011). Here, only the model components calculating HTO transfers and OBT formation are described, although these processes are closely related to other processes and use outputs from the components calculating heat, water and CO_2 transport and exchanges.



Figure 2.1.1 Land surface tritium transfer (black arrows) and exchanges (white arrows) considered in the SOLVEG-II.

2.1 Atmospheric model

Air HTO concentration χ_a (Bq m⁻³) in the surface atmosphere is calculated by a diffusion equation:

$$\frac{\partial \chi_a}{\partial t} = \frac{\partial}{\partial z} K \frac{\partial \chi_a}{\partial z} + \varphi, \qquad (1)$$

where t (s) is the time, z (m) is the vertical coordinate, K (m² s⁻¹) is the turbulent diffusivity calculated in the heat and water transport process. The sinks or sources φ (Bq m⁻³ s⁻¹) for HTO includes HTO exchanges between the canopy air and leaf cellular water through stomata, between the canopy air and leaf surface water, and between the canopy air and rain drops.

2.2 Soil model

Soil model covers diffusion and advection for the aqueous and the gaseous HTO (Yamazawa, 2001), as:

$$\frac{\partial \eta_s \chi_{sw}}{\partial t} = -\frac{\partial E_w \chi_{sw}}{\partial z} + \frac{\partial}{\partial z} D_w \frac{\partial \chi_{sw}}{\partial z} - e_b - e_r, \text{ for the aqueous and}$$
(2)
$$\frac{\partial \{(\eta_{sat} - \eta_s) \chi_{sa}\}}{\partial t} = \frac{\partial}{\partial z} D_a \frac{\partial \chi_{sa}}{\partial z} + e_b, \text{ for the gaseous,}$$
(3)

where χ_{sw} and χ_{sa} are the aqueous and the gaseous HTO concentrations (Bq m⁻³), η_s and η_{sat} are the soil water content and the porosity, E_w (m³ m⁻² s⁻¹) is the liquid water flux, D_w and D_a (m² s⁻¹) are the effective diffusivities for the gaseous and the aqueous HTO. Assuming that the root uptake of soil

water is fully driven by the transpiration, the root-uptake term e_r (Bq m⁻³ s⁻¹) is calculated from χ_{sw} and the aboveground transpiration flux (Ota and Nagai, 2011). Equations (2) and (3) are linked by the HTO evaporation e_b (Bq m⁻³ s⁻¹) (or condensation if negative) formulated by a resistance approach. Soil model is connected to the atmospheric model through the gaseous HTO exchange and the aqueous HTO flux by rain.

2.3 Vegetation model

Vegetation model (Ota and Nagai, 2011) computes tissue free water tritium (TFWT) budget in leaves, as:

$$\frac{\partial \eta_{\nu} \chi_{\nu}}{\partial t} = E_{stom} + E_{root} - E_{phot} + E_{res}, \qquad (4)$$

where η_{ν} (m³ m⁻²) is the volume of the leaf cellular water existing on an unit leaf area and χ_{ν} (Bq m⁻³) is the TFWT concentration in the leaf cellular water. Terms E_{stom} , E_{root} , E_{phot} and E_{res} (Bq m⁻² s⁻¹) respectively express TFWT flux caused by exchange between the canopy air and the leaf cellular water, loading through the root uptake, photosynthetic assimilation and production through respiration.

The exchange flux is calculated by:

$$E_{stom} = \frac{1}{r_a + r_s} \left\{ \chi_a - \frac{\rho_a}{\rho} q_s(T) \chi_v \right\}, \qquad (5)$$

where r_a (m⁻¹ s) is the leaf boundary-layer resistance, r_s (m⁻¹ s) is the stomata resistance, ρ_a (kg m⁻³) is the air density, ρ (kg m⁻³) is the liquid water density, and $q_s(T)$ is the saturation of the specific humidity (kg kg⁻¹) at the leaf temperature T (K). Resistance r_a is assumed to depend on the aerodynamic characteristics of the leaves and wind velocity, and r_s is calculated using a relationship between r_s and net CO₂ assimilation that is calculated by considering photosynthetic processes (Nagai, 2005).

Flux E_{root} for the root uptake is calculated from the root uptake term e_r in Eq (2) by relating the belowground root distribution and the aboveground leaf transpiration.

Given that one-mol of CO₂ reacts with one-mol of H₂O through photosynthesis, photosynthetic TFWT assimilation is determined by:

$$E_{phot} = \chi_v \frac{m}{\rho} f P , \qquad (6)$$

where m = 0.018 kg mol⁻¹ is the molar weight of water, f = 0.78 is the isotopic discrimination factor between HTO and H₂O at the photosynthesis (Diabaté and Strack, 1993), P (mol–CO₂ m⁻² s⁻¹) is the net CO₂ assimilation flux for the unit leaf area.

A respiration reaction decomposes 1/6-mol glucose (C₆H₁₂O₆) and yields one-mol H₂O. Hence

the TFWT production through the respiration is modeled as:

$$E_{res} = S_{\rm int} \frac{1}{6} MR \,, \tag{7}$$

where S_{int} (Bq kg⁻¹) is the OBT contained in the unit mass of the dry matter in an intermediate carbohydrate pool, described later, M = 0.18 kg mol⁻¹ is the molar weight of glucose and R (mol-CO₂ m⁻² s⁻¹) is the respiration rate for CO₂.

Leaf OBT dynamics are calculated with a carbohydrate compartment model having four pools, shown by Fig. 2.3.1 (Ota and Nagai, 2011). The input- and output-carbohydrate flux (kg m⁻² s⁻¹) at the intermediate pool, where the photosynthates firstly enter, are calculated by the photosynthesis rate *P* and the respiration rate *R* as:

$$E_P = \frac{1}{6}MP$$
, for input and (8)
 $E_R = \frac{1}{6}MR$, for output, (9)

where the factor 1/6 is the stochastic ratio between CO₂ and C₆H₁₂O₆ at the photosynthesis and the respiration reaction. Daytime carbohydrates flows are determined by E_P and nighttime flows by E_R (see Fig. 2.3.1), and the amount of carbohydrates w_i (kg m⁻²) in each pool are calculated. With the OBT input by E_{phot} and output by E_{res} (Eqs. (6)–(7)) at the intermediate pool, OBT exchanges between the four pools are calculated and the total OBT amount Q (Bq kg⁻¹) contained in the unit mass of the dry matter is determined by summarizing the OBT content S_i (Bq kg⁻¹) and carbohydrates amount in the four pools, as:

$$Q = \frac{\sum S_{i} w_{i}}{\sum} w_{i} = \frac{S_{int} w_{int} + S_{suc} w_{suc} + S_{sta} w_{sta} + S_{str} w_{str}}{w_{int} + w_{suc} + w_{sta} + w_{str}}.$$
 (10)





References for the section 2 (Briefing of a complex model)

Diabaté, S., Strack, S., 1993. Organically bound tritium. Health Phys. 65, 698-712.

Nagai, H., 2005. Incorporation of CO_2 exchange processes into a multiplayer atmosphere-soil-vegetation model. J. Appl. Meteorol. 44, 1574–1592.

Ota, M., Nagai, H., 2011. Development and validation of a sophisticated atmosphere-vegetation-soil HTO transport and OBT formation model. J. Environ. Radioact. 102, 813–823.

Yamazawa, H., 2001. A one-dimensional dynamical soil-atmosphere tritiated water transport model. Environ. Model. Softw. 16, 739–751.