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 H₂O, but the deposition (also for reemission) of HTO and H₂O should be treated separately because each molecule follows its individual vapor-pressure deficit (Feinhals and Bünningen, 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:

\[ F_{HTO} = v_{HTO} (\chi_{atm} - \chi_{sa}), \]  

where \( F_{HTO} \) is the exchange flux (Bq m\(^{-2}\) s\(^{-1}\)), \( v_{HTO} \) is the exchange velocity (m s\(^{-1}\)), \( \chi_{atm} \) and \( \chi_{sa} \) 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:

\[ v_{HTO} = (r_a + r_b + r_s)^{-1}, \]

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:

\[ \chi_{sa} = \frac{h_{sat}}{\rho_w} \chi_{sw}, \]

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

\[ \theta \frac{\partial \chi_{sw}}{\partial t} = \frac{\partial}{\partial z} D \frac{\partial \chi_{sw}}{\partial z} - q \frac{\partial \chi_{sw}}{\partial z}, \]

where \( \theta \) 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 (m\(^2\) 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. \( \chi_{atm} \) being smaller than \( \chi_{sa} \) 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 resupply through the upward diffusion of the HTO in the deeper zone, and this maintains 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:

\[
F_{HT} = v_{HT} \chi_{atm,HT},
\]

(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:
\[
\epsilon \frac{\partial \chi_{s,HT}}{\partial t} = \frac{\partial}{\partial z} \left( D_{HT} \frac{\partial \chi_{s,HT}}{\partial z} \right) - e_{HT}, 
\]

where \( \epsilon \) 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 (m\(^{2}\) 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). 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)


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


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.


2. Briefing of a complex model

A complex model SOLVEG-II developed by JAEA computes transport and exchanges of heat, water and CO₂ 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 CO₂ transport and exchanges.
2.1 Atmospheric model

Air HTO concentration $\chi_a$ (Bq m$^{-3}$) in the surface atmosphere is calculated by a diffusion equation:

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

where $t$ (s) is the time, $z$ (m) is the vertical coordinate, $K$ (m$^2$ s$^{-1}$) is the turbulent diffusivity calculated in the heat and water transport process. The sinks or sources $\phi$ (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:

$$\frac{\partial \eta_s \chi_{sw}}{\partial t} = - \frac{\partial E_w \chi_{sw}}{\partial z} + \frac{\partial}{\partial z} D_w \frac{\partial \chi_{sw}}{\partial z} - e_b - e_r, \quad \text{for the aqueous and} \quad (2)$$

$$\frac{\partial (\eta_{sat} - \eta_s) \chi_{sa}}{\partial t} = \frac{\partial}{\partial z} D_s \frac{\partial \chi_{sa}}{\partial z} + e_s, \quad \text{for the gaseous,} \quad (3)$$

where $\chi_{sw}$ and $\chi_{sa}$ are the aqueous and the gaseous HTO concentrations (Bq m$^{-3}$), $\eta_s$ and $\eta_{sat}$ are the soil water content and the porosity, $E_w$ (m$^3$ m$^{-2}$ s$^{-1}$) is the liquid water flux, $D_w$ and $D_s$ (m$^2$ 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 $\chi_{sw}$ 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:

$$\frac{\partial \eta_v \chi_v}{\partial t} = E_{\text{stom}} + E_{\text{root}} - E_{\text{phot}} + E_{\text{res}},$$  \hspace{1cm} (4)

where $\eta_v$ (m\(^3\) m\(^{-2}\)) is the volume of the leaf cellular water existing on an unit leaf area and $\chi_v$ (Bq m\(^{-3}\)) is the TFWT concentration in the leaf cellular water. Terms $E_{\text{stom}}$, $E_{\text{root}}$, $E_{\text{phot}}$ and $E_{\text{res}}$ (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:

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

where $r_a$ (m\(^{-1}\) s) is the leaf boundary-layer resistance, $r_s$ (m\(^{-1}\) s) is the stomata resistance, $\rho_a$ (kg m\(^{-3}\)) is the air density, $\rho$ (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 CO\(_2\) assimilation that is calculated by considering photosynthetic processes (Nagai, 2005).

Flux $E_{\text{root}}$ 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 CO\(_2\) reacts with one-mol of H\(_2\)O through photosynthesis, photosynthetic TFWT assimilation is determined by:

$$E_{\text{phot}} = \chi_v \frac{m}{\rho} fP,$$  \hspace{1cm} (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 H\(_2\)O at the photosynthesis (Diabaté and Strack, 1993), $P$ (mol–CO\(_2\) m\(^{-2}\) s\(^{-1}\)) is the net CO\(_2\) assimilation flux for the unit leaf area.

A respiration reaction decomposes 1/6-mol glucose (C\(_6\)H\(_{12}\)O\(_6\)) and yields one-mol H\(_2\)O. Hence
the TFWT production through the respiration is modeled as:

\[ E_{res} = S_{int} \frac{1}{6} MR, \quad (7) \]

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, \( M = 0.18 \text{ kg mol}^{-1} \) is the molar weight of glucose and \( R \) (mol–CO\(_2\) m\(^{-2}\) s\(^{-1}\)) is the respiration rate for CO\(_2\).

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:

\[ E_P = \frac{1}{6} MP, \quad \text{for input and} \quad (8) \]

\[ E_R = \frac{1}{6} MR, \quad \text{for output,} \quad (9) \]

where the factor 1/6 is the stochastic ratio between CO\(_2\) and C\(_6\)H\(_{12}\)O\(_6\) 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:

\[ Q = \sum S_i w_i \left/ \sum w_i \right. = \frac{S_{int} w_{int} + S_{suc} w_{suc} + S_{str} w_{str}}{w_{int} + w_{suc} + w_{str} + w_{str}}. \quad (10) \]
**Fig. 2.3.1** A carbohydrate compartment model in SOLVEG-II.

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


