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〈Technical Note〉

Verification of a Dynamic Compartment Model for the Tritium Behavior in the Plants After Short HTO Release Using a BIOMOVS II Scenario

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Abstract

A dynamic compartment model was required for the prediction of radiological consequences of the tritiated vapor released from the nuclear facility after an accident. A computer code, ECOREA-T, was developed by incorporating the unit models for the evaluation of tritium behavior in the environment. Dry deposition of tritiated vapor from the atmosphere to the soil was calculated using a deposition velocity. Transport of tritium from the atmosphere to the plant was calculated using a specific activity model, and the result was compared with the Belot's analytic solution. Root uptake of tritiated water from the soil and formation of OBT from TFWT were considered in the model. The ECOREA-T code was verified by comparing the results from the other computer codes using a scenario developed through BIOMOVS II study. The results showed good agreements.

Key Words : dynamic, compartment, tritium, verification, deposition, OBT

1. Introduction

Many kinds of computer codes are used for the assessment of exposure to the public around the nuclear facilities after accidental releases of radionuclides. Korea Atomic Energy Research Institute (KAERI) is developing a series of computer codes called ECOREA for the prediction of radiological consequences through a food chain after a short-term accidental release of gaseous radionuclides. ECOREA computer codes calculated the radiological consequences using the results of the calculation of FADAS (Following Accident Dose Assessment System) which calculated the atmospheric dispersion of the radionuclides released from the nuclear facilities due to accident. Differences in the behavior of radionuclides in the environment ask the different models. The transfer of H-3 and C-14 is modeled by a specific activity approach because they are readily incorporated into living organisms [1].

Tritium exists mostly in the form of HTO in the atmosphere, moves by advection and diffusion, and a part of tritium is deposited. The dry deposition rates are calculated using two methods. One uses a dry deposition velocity and the other uses exchange velocities [2]. If the exchange velocity method is used, the resuspension of deposited tritium is calculated simultaneously. However, the dry deposition velocity method is used widely because the model is simple and deposition velocities are available.

Plants exchange gases and vapors with the atmosphere through their leaves. During daylight the stomata on the leaves control the exchange rates of gases and vapors. Specific Activity model developed by Belot et al [3] was based on the direct exchange of tritium through the stomata. Murphy developed a atmosphere-plants-soil model for the transport of tritium considering the deposited tritium onto soil [4].

The purposes of this paper were to develop a computer code, ECOREA-T, to simulate the behavior of tritium released from the nuclear facility due to an accident. To this end, a dynamic compartment model consisting of dry deposition model, specific activity model, and OBT (Organically Bound Tritium) conversion model, was developed. Two unit models of the ECOREA-T, dry deposition model and specific activity model, were verified by comparing with the theoretical calculation and an analytic solution, respectively. The verification of ECOREA-T was conducted by assessing the scenario developed through the BIOMOVS II study. The BIOMOVS II study was an international cooperative study for the intercomparison of the computer codes designed to quantify the transfer of radionuclides in the environment. Eight computer codes from 7 countries participated in the tritium working group, but we compared the results with those of 3 representative computer codes.

2. Dynamic Compartment Model

2.1. Air to Soil Transfer

Since majority of tritium exists in the form of HTO in the environment, it moves quickly just like water. The mechanism of deposition onto soil depends on the states of the radionuclides, gas or particles. Whilst the dry deposition of noble gases is negligible, HTO deposits onto soil like vapor [5]. That is, deposition increases when the vapor is below the dew point. Brownian diffusion is more important than gravitational settling and eddy diffusion [6]. There are two models for HTO deposition from atmosphere to soil. In this paper, we used the method based upon deposition velocity. The amount of radionuclides dry deposited onto the soil are obtained as follows [7, 8]:

$$w(x,y)=v_d C(x,y,0) \tag{1}$$

where w is the dry deposition flux of tritium [Bq m^{-2} s^{-1}],
v_d is the dry deposition velocity of tritium [m s^{-1}],
C is the tritium concentration in the atmosphere [Bq m^{-3}].

2.2. Air to Vegetation Transfer

Plants exchange HTO with the atmosphere through the stomata on the leaves. The transport of HTO is considered by diffusion through the stomatal pores. According to Belot et al. [3], the governing equation for the tritium transfer was given as follows:

$$\mu \frac{dC}{dt} = \frac{1}{r}\left(\chi - \frac{\rho}{\alpha} C\right) \tag{2}$$

where μ is the amount of water per unit area of leaf [g m^{-2}],
χ is the tritium concentration in the atmosphere [Bq m^{-3}],

C is the tritium concentration in tissue-water [Bq g^{-1}],

r is the foliar resistance [s m^{-1}],

ρ is the density of water vapor in saturated air [g m^{-3}],

α is the quotient of isotope ratios T/H in liquid and vapor [= 1.1].

If χ, μ, ρ,and r in equation (2) were constant, an analytical solution could be obtained as follows:

$$\frac{C}{\chi} = \frac{\alpha}{\rho}\left(1-e^{-kt}\right) \tag{3}$$

where $k = \frac{\rho}{\alpha\mu r}$

The solution (3) showed that $\frac{C}{\chi}$ approached $\frac{\alpha}{\rho}$. It meant that steady state tritium concentration in the leaves could be determined by $\frac{\alpha}{\rho}$, which was a function of leaf temperature.

2.3. Conversion of HTO to OBT

HTO is short-lived in plants. Plants convert HTO to organic compounds in which form it lasts longer. HTO is converted to OBT through the following processes [9]. HTO is incorporates into carbohydrates by photosynthesis. Plants synthesize organic compounds such as proteins, oils, alkaloids and vitamins using the energy of respiration. Plant tissue is breaking and rebuilding continuously. HTO and H_2O incorporate together into restored organic compounds during rebuilding. Carbohydrates, proteins, and other organic compounds have labile hydrogen atoms which readily exchange hydrogen and tritium atoms. Once OBT is formed, it is subsequently lost by oxidation. These processes could be modeled as follows [2]:

$$HTO \xrightarrow{k_1} OBT \xrightarrow{k_2} Loss$$

It could be represented as a differential equation:

$$\frac{dC_{OBT}}{dt} = k_1 C_{HTO} - k_2 C_{OBT} \tag{4}$$

where C_{OBT} is OBT concentration in the plant [Bq/g-dry],

C_{HTO} is HTO concentration in the plant [Bq/g-dry],

k_1, k_2 are transfer coefficients [s^{-1}].

2.4. Dynamic Compartment Model

A dynamic compartment model (ECOREA-T) was developed to predict the behavior of tritium by incorporating the unit models mentioned before. Figure 1 showed the relations between the compartments and transfer coefficients used in the ECOREA-T. As shown in figure 1, four compartments including Soil_sink were introduced for the soil layer because the root uptake rates were different with depth of the soil. Plants were divided into 3 compartments, vegetation for leaves, roots, and OBT. An atmosphere compartment (A_sink) was introduced for the

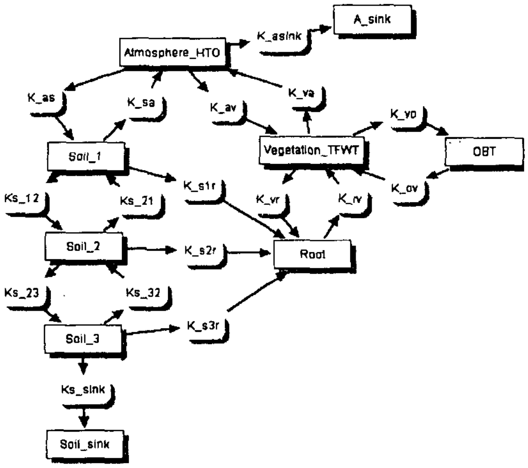

Fig. 1. Compartment and Transfer Coefficient used in ECOREA-T

tritium evaporated into the atmosphere. Mass transfers between the compartments in figure 1 could be represented by nine differential equations. The general form of the equations is as follows:

$$\frac{dA_i^t}{dt} = -\sum_n k_{ij} A_i^t + \sum_m k_{ji} A_i^t - \lambda^t A_i^t + S_i^t \qquad (5)$$

where A_i^t is the amounts of tritium in the compartment-i [Bq],
k_{ij} is the transfer coefficient between compartment-i and compartment-j [yr^{-1}],
λ^t is the tritium decay constant [yr^{-1}],
S_i^t is the source term in the compartment-i [Bq yr^{-1}].

The first term in equation (5) indicated the mass flux coming out of compartment-i into compartment-j, the third term showed the decay, and the final term was the source term to compartment-i. The mass transfer rates between compartments were governed by transfer coefficients. Since the differential equations (5) were coupled each other, it was not easy to solve them analytically. The computer program, AMBER, was used for the numerical solution.

3. Scenario and Transfer Coefficients

BIOMOVS II study developed the scenario for the tritium in the food chain after short exposure. According to the scenario, tritium release starts at 10:00, and the exposure lasts for one hour. The source term is an assumed concentration of HTO in surface air equivalent to 10^{10} Bq m^{-3} sustained for the whole one hour. The exposure occurs sometime in summer 30 days prior to harvesting of each of the crops. The yields of leafy vegetables are 2.0 kg m^{-2} at harvest time in fresh weight. The soil type is loamy sand. The porosity of the soil is 0.4, and initial moisture content is 20%.

BIOMOVS II required the participants to calculate the tritium concentration in three soil layers, leafy vegetables, root vegetables, grains, milk, and beef at the end of each hour for the first 24 hours. However, ECOREA-T calculated the concentrations in soil layers, leafy vegetables, and OBT. We compared results with three representative codes, that is, UFOTRI (FZK, Germany), TRITRAJ (JAERI, Japan), and TRICAROM (Inst. Atomic Physics, Romania). Those three codes were based upon dynamic compartment model that ECOREA used. Each

Table 1. Transfer Coefficients Used in the Verification Study

TRANSFER COEFFICIENTS	FROM	TO	VALUE (h^{-1})
Ks_12	Soil-1	Soil-2	8.75×10^{-3}
Ks_21	Soil-2	Soil-1	5.00×10^{-4}
Ks_23	Soil-2	Soil-3	2.75×10^{-3}
Ks_32	Soil-3	Soil-2	3.42×10^{-4}
K_s1r	Soil-1	Root	1.75×10^{-3}
K_s2r	Soil-2	Root	1.71×10^{-3}
K_s3r	Soil-3	Root	1.17×10^{-3}
K_av	Atmosphere	Vegetation	1.35×10^{-2}
K_va	Vegetation	Atmosphere	0.15
K_vo	Vegetation	OBT	1.04×10^{-4}
K_ov	OBT	Vegetation	1.33×10^{-3}

code had different structures of compartments and used different values of transfer coefficients [2].

Transfer coefficient for the deposition of tritium was derived using the deposition velocity. The deposition rate of tritium was calculated using equation (1). The total amount of tritium below the mixing height, H, was calculated as follows if the vertical concentration gradient was negligible:

$$A = \int_0^H C(z)dz = C \cdot H \qquad (6)$$

Thus, transfer coefficient was calculated from equation (1) and (6) as follows:

$$k_{-as} = \frac{C \cdot v_d}{C \cdot H} = \frac{v_d}{H} \qquad (7)$$

BIOMOVS II did not specify the values, v_d and H in equation (7). Each participant was asked to use the best values of its own in the computer code. ECOREA-T used $v_d = 5 \times 10^{-3}$ m s^{-1} and H = 1,000 m.

BIOMOVS II did not provide any transfer coefficients for the mass transfer between soils and plants and even in the plants. Most of values should be determined through the experiment, but it was not easy to find the values, either. In this paper we used the transfer coefficients given in computer code, UFOTRI [10]. Most of the codes used the values. The transfer coefficients used in the verification were given in Table 1.

4. Results and Discussion

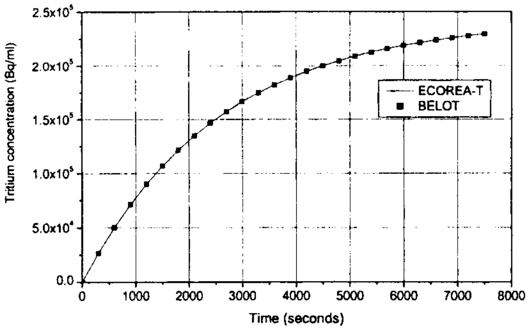

Fig. 2. Comparison of Solutions: ECOREA-T and Belot's Analytic Solution

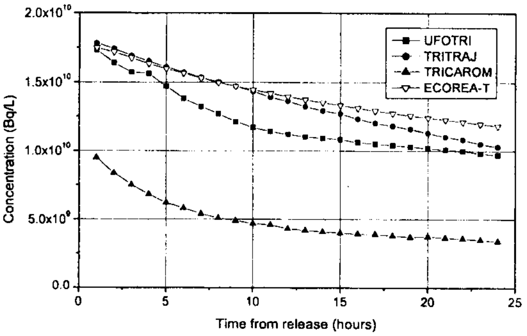

Fig. 3. Tritium Concentration in Soil-1 Layer

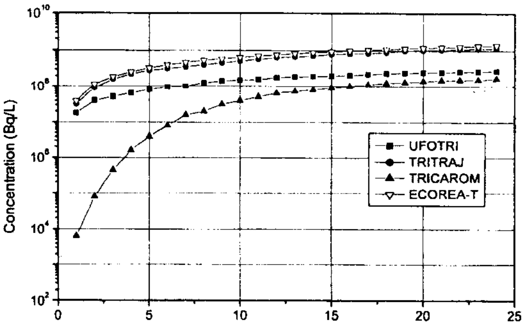

Fig. 4. Tritium Concentration in Soil-2 Layer

In this study the verification of ECOREA-T was carried out using several examples. The numerical solution for the transfer of tritium from atmosphere to the leaves of the plant was directly compared with Belot's analytical solution given in equation (3). For the comparison we used the input values of χ = 6.0 Bq cm^{-3}, $\rho = 2.72 \times 10^{-5}$ g cm^{-3}, r = 4 s cm^{-1}, and $\mu = 1.6 \times 10^{-2}$ g cm^{-2}. The result of the calculation was shown in Figure 2. The results in Figure 2 showed that the numerical solution of ECOREA-T agreed well with Belot's analytical solution for the whole range.

BIOMOVES II scenario was simulated with the

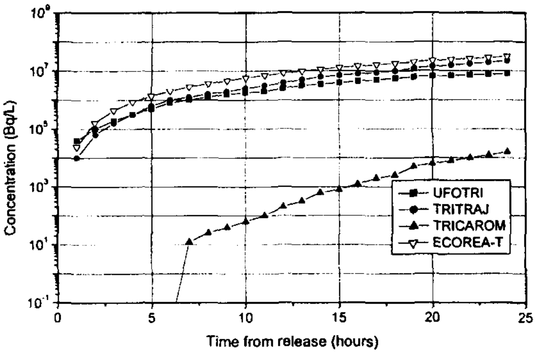

Fig. 5. Tritium Concentration in Soil-3 Layer

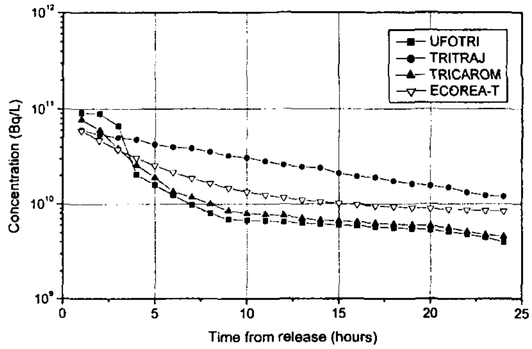

Fig. 6. Concentration of TFWT in the Plants

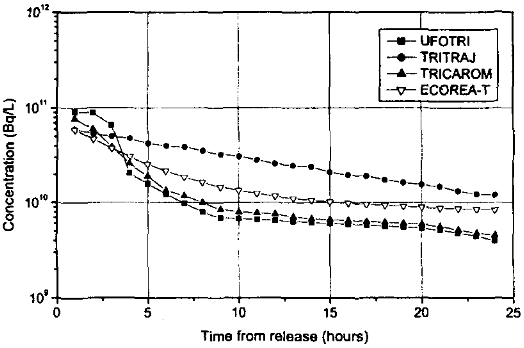

Fig. 7. OBT Concentration in the Plants

input parameters given in Table 1 using the ECOREA-T code. The results of the simulation were shown in Figure 3 to Figure 7. Figure 3 showed the change of tritium concentration in soil-1 layer. According to ECOREA-T calculation, the initial tritium concentration in pore water in soil-1 layer due to the deposition was 1.76×10^{10} Bq/L for one hour after the release. Theoretically, the tritium concentration of 1.8×10^{10} Bq/L was calculated with atmospheric concentration (1.0×10^{10} Bq/m^3), deposition velocity (5×10^{-3} m/s), volume of pore water ($1\times0.05\times0.2$ m^3), and deposition time (3,600 seconds). These two values showed that the solutions were in good agreement. Among other computer codes, TRICAROM showed different results because it used the deposition velocity of 0.0026 m/s. Figure 4 and 5 showed that all the tritium concentration in soil-2 and soil-3 layer except that of TRICAROM agreed well for 24 hours.

Figure 6 and 7 showed the concentration of HTO and OBT in the plants. Four computer codes showed the similar patterns in the concentration of HTO and OBT in the plants unlikely in the soil layers. It was considered that most of the tritium in the plants was not from the soil layers but from the atmosphere.

5. Conclusions

We developed a computer code to assess the radiological consequences of tritium released from the nuclear facilities after an accident. The computer code, ECOREA-T, was based upon the dynamic compartment model. The dry deposition of tritiated water from the atmosphere to the soil was calculated with the transfer coefficient derived from a dry deposition velocity. Transport of tritium from the atmosphere to the plant was calculated using a specific activity model, and the result was compared with the Belot's analytic solution. Root uptake of tritiated water from the soil and formation of OBT from TFWT were considered in the model. The unit models were incorporated into the compartment model, ECOREA-T. The solution of the dynamic compartment model was obtained using AMBER program.

The transport processes involved in the development of the ECOREA-T were so various and complex that it was required to verify it. Two unit models used in the ECOREA-T, dry deposition model and specific activity model, were verified by comparing the results with the theoretical one and analytical solution, respectively. The results showed that the deposition onto soil calculated from ECOREA-T was close to the theoretical calculation value. And both the ECOREA-T and Belot's analytical solution showed the same tritium transfer between atmosphere and the leaves.

Also, the ECOREA-T code was verified by comparing the results from the other computer codes using a scenario developed through BIOMOVS II study. Eight computer codes participated in the intercomparison of tritium working group of BIOMOVS II. However, we compared the ECOREA-T with three representative codes for convenience's sake. Tritium concentrations in three soil layers showed a good agreement except TRICAROM due to different value of deposition velocity. HTO and OBT concentration in the plants showed good agreements.

Acknowledgement

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