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Estimation of Tritium Concentration in Groundwater around the Nuclear Power Plants Using a Dynamic Compartment Model

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Abstract - Every nuclear power plant measured concentrations of tritium in groundwater and surface water around the plants periodically. It was not easy to predict the tritium concentration only with these measurement data in case of various release scenarios. KAERI developed a new approach to find the relationship between the tritium release rate and tritium concentration in the environment. The approach was based upon a dynamic compartment model. In this paper the dynamic compartment model was modified to predict the tritium behavior more accurately. The mechanisms considered for the transfer of tritium between the compartments were evaporation, groundwater flow, infiltration, runoff, and hydrodynamic dispersion. Time dependent source terms of the compartment model were introduced to refine the release scenarios. Also, transfer coefficients between the compartments were obtained using realistic geographical data. In order to illustrate the model various release scenarios were developed, and the change of tritium concentration in groundwater and surface water around the nuclear power plants was estimated.
Key words: tritium, dynamic, compartment model, environment, transfer coefficient

INTRODUCTION

Concentrations of tritium as well as radionuclides of importance in groundwater and surface water around the nuclear power plants have been measured periodically according to MOST Notice 2001-25. Only with these measurement data it was not easy to predict the tritium concentration in the future in case of various release scenarios. Also, recently introduced periodic safety review of the nuclear power plants operating more than ten years asked the estimation of radionuclide concentrations around the NPPs for future 10 years. Even though tritium was the most abundant radionuclide released from the PWR, so far the interest in the behavior of tritium had been focused on the atmospheric dispersion, its concentration in the plants, and ingestion dose rates. Since the ingestion dose rate from tritium was calculated with a specific activity model, the tritium concentration in groundwater or surface water had not been estimated. KAERI (Korea Atomic Energy Research Institute) initiated a new research project to analyze and model the previous tritium measurement data collected around the Kori nuclear power plants (NPPs). Also, the new samples of groundwater and surface water were newly analyzed for the measurement of tritium concentration [1].

Tritium is a radioactive isotope of hydrogen with atomic weight of 3 and decays to ^{3}He emitting

low energy electrons. Its half-life is 12.3 years. The energy of the electron emitted from the decay of tritium is too low to penetrate the human skin. In other words, electrons from the tritium decay can inflict damage on humans only when tritium is present inside the human body.

The sources of tritium are the interactions between gases of the upper atmosphere and cosmic rays, nuclear bomb tests and consumer products as well as nuclear power plants [2]. Tritium is produced by ternary fission of nuclear fuel and from neutron activation of lithium and boron in light water reactors, and the neutron activation of deuterium is the main source exceeding ternary fission in heavy water reactors. The past records showed that around 10 TBq of tritium had been released annually from the Kori NPPs [3].

Even though tritium was the most abundant radionuclide released from the PWR, the research on the behavior of tritium had been restricted to the environment around Pressurized Heavy Water Reactors due to their remarkably great discharge rates. Whereas most of researches on tritium were focused on the behavior in the atmosphere and on the transfer between the atmosphere and the tissue free water [4, 5], the behavior of tritium in the environment including groundwater and surface water was modeled in this study.

The purposes of this paper were to calculate deposition rates of the tritiated compounds and predict the environmental tritium behavior by introducing a dynamic compartment model. The dry and wet deposition rates of tritium were calculated using a computer program, DEPOS. The northern area around the Kori nuclear power plants were represented by 8 compartments in which tritium was assumed to be mixed completely. The several kinds of transfer coefficients that represented the transport of tritium between the compartments were derived. The governing equations for mass transports were solved using a computer program AMBER[6]. Also, the change of tritium concentration was predicted using the compartment model in the case that the source term increased ten times.

2. DEPOSITION OF TRITIUM

Tritium released from the nuclear power plants are transported by bulk of air (advection) and diffusion where tritium or tritiated compound (HTO) moves along a gradient of concentration from high concentration to low concentration. Removal of tritiated compound from the atmosphere takes place by the deposition. The atmospheric removal processes are classified into dry or wet deposition. According to Briggs et al.[7], the amount of radionuclides dry-deposited onto the soil are obtained by multiplying the atmospheric concentration near the soil and dry deposition velocity [8]:

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

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

The tritium concentration in equation (1) was obtained by multiplying x/Q from the Gaussian plume model and the tritium discharge rate from NPPs. Figure 1 showed the discharge rates of tritium and inert gases from the Kori NPPs from the year 1991 to 1999. As shown in Figure 1, tritium discharge rate was around 10 TBq. A computer program, XOQDOQ, was used to get x/Q. Figure 2 showed the x/Q values of 4 directions in 1999. The x/Q values were negligible beyond 8 km from the nuclear power plants. The deposition rate in each sector was obtained by multiplying the concentration of tritium, the area of the sector, and the deposition velocity. The deposition velocity of 4.94×10^{-4} m s^{-1} was used [9].

There are two methods to calculate the wet deposition of HTO. The washout rate approach is used to calculate the short-term release, and the washout ratio approach is used for the long-term release. According to washout ratio approach, the wet deposition rate of tritium was calculated using the following equation:

$$W(x,y) = \frac{\omega I Q}{(2\pi)^{1/2} u \sigma_y(x)} \exp\left[-\frac{y^2}{2\sigma_y^2(x)}\right] \tag{2}$$

where ω is washout ratio [-],
I is the precipitation rate [mm yr^{-1}].

ωI in equation (2) was called a wet deposition velocity like a dry deposition velocity. The dry and wet deposition rates of tritium onto each sector and zone were calculated using a computer program, DEPOS, developed by authors.

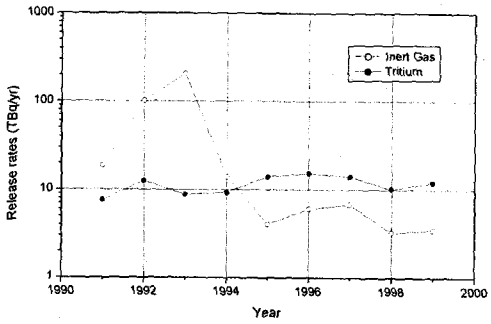

Fig. 1. Release rates of tritium and inert gas from the Kori nuclear power plants. The release rates of tritium seemed to be constant for 10 years.

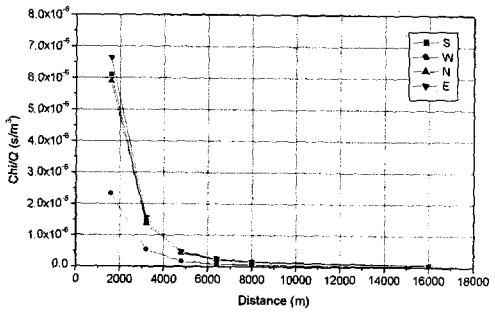

Fig. 2. x/Q values in 1999 showed that they decreased remarkably beyond 8,000 m.

3. COMPARTMENT MODEL

Most of tritium existed in the form of HTO in the environment. HTO deposited onto the soil behaved just like water: some of them were evaporated into the atmosphere, moved along with the surface water or groundwater. Since HTO was very mobile in the environment, their behavior could be described using a compartment model. The mass transfer between the compartments was expressed using the following mass conservation equations:

$$\frac{dA_i'}{dt} = -\sum_n k_{ij} A_i' + \sum_m k_{ji} A_j' - \lambda' A_i' + S_i' \qquad (3)$$

where A_i' 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' is the tritium decay constant [yr^{-1}],

S_i' is the source term in the compartment-i [Bq yr^{-1}].

The first term in equation (3) 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 9 differential equations (3) were coupled each other, it was not easy to solve them analytically. The computer program, AMBER, was used.

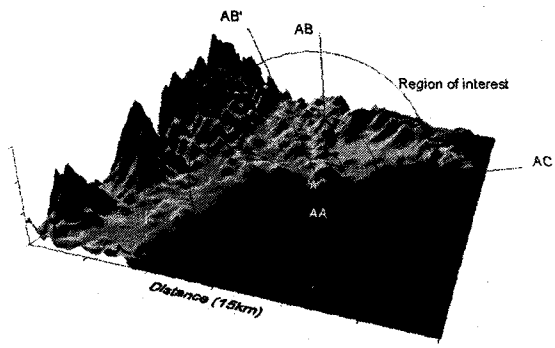

Fig. 3. Geography of the area around Kori site. The star mark indicates the location of nuclear power plants.

4. SOURCE TERMS AND TRANSFER COEFFICIENTS

Figure 3 showed three-dimensional geography of Kori site. The northern area (denoted by AC to AB in Figure 3) was selected as the most important based upon the wind profile around the Kori site. The northern area of the site was divided into 3 zones depending on x/Q values, and the deposition rates onto the zones were obtained using a computer program, DEPOS, in order to model the behavior of tritium in the environment. DEPOS calculated the deposition rates using equations (1) and (2) and integrated the deposition rates onto sectors to calculate the deposition onto zones. The deposition rates in 3 zones given in Table 1 were used as source terms.

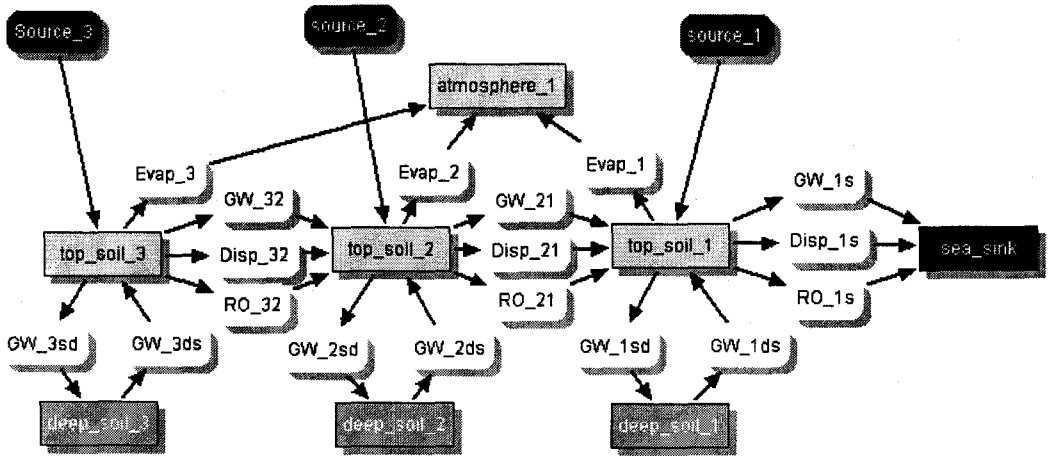

Fig. 4. Compartments and transfer coefficients used in the modeling of tritium movement.

Table 1. Location of zones

ZONE	AREA(m^2)	DIRECTION	DISTANCE
zone-1	1.320×10^{9}	NE, NNE, N, NNW	1.6 km
zone-2	1.477×10^{7}	NE, NNE, N, NNW	1.6 ~ 4.8 km
zone-3	3.020×10^{7}	NE, NNE, N, NNW	4.8 ~ 8.0 km

There were twenty sectors in the northern area. They were grouped into 3 zones. Each zone was divided into 2 compartments. Six compartments were introduced for the soil layer in the northern area. The porosity of the top soil was 0.3, and that of the deep soil 0.1. Total 8 compartments were used to describe the tritium behavior in the environment. Figure 4 showed the compartments used in this study. One atmosphere compartment was introduced for the evaporation of tritium, and one sea compartment as the sink in which tritium in groundwater or runoff water flew.

The transfer of tritium between the compartments was due to evaporation, groundwater flow, infiltration, runoff, and dispersion. The ten year average precipitation rate and evaporation rate were used to derive the transfer coefficients. The annual precipitation rate was 1,390 mm, and annual evaporation rate was 1,150 mm [10]. Runoff was estimated 140 mm which was 10% of the precipitation[11]. According to the water budget, the infiltration of rain was estimated around 100 mm per year. The transfer coefficient of evaporation between the atmosphere and the top soil was calculated by dividing the evaporation rate with pore volume of compartment. It was 0.128 yr^{-1} regardless of the compartments. The transfer coefficients of runoff were given in Table 2.

Tritium moved with groundwater. The FSAR (Final Safety Analysis Report) of Kori NPPs was reviewed to derive transfer coefficients of groundwater flow. According to the FSAR, the groundwater flow around the Kori site directed from the land to the seashore. Hydraulic gradient was

Table 2. Transfer coefficients of runoff and groundwater flow

Transfer Coefficient (yr^{-1})		Value
Runoff	K_{32}	0.0156
	K_{21}	0.0426
	K_{1s}	0.491
Groundwater Flow	K_{32}	5.68×10^{-3}
	K_{21}	6.97×10^{-3}
	K_{1s}	2.34×10^{-2}
Dispersion	K_{32}	1.78×10^{-6}
	K_{21}	2.90×10^{-6}
	K_{1s}	1.46×10^{-5}

13%, and hydraulic conductivity was estimated within the range of 10^{-3} ~ 10^{-5} cm s^{-1}. The hydraulic properties of the site were not measured in detail. Groundwater flow was described with the following Darcy's law [11]:

$$Q = -KA\frac{dh}{dl} \tag{4}$$

where Q is the flow rate of groundwater through the cross section area A [m^3 yr^{-1}],
K is the hydraulic conductivity [m yr^{-1}],
$\frac{dh}{dl}$ is the hydraulic gradient [-].

The groundwater flow rate across the vertical cross section between the compartments was obtained from the equation (4) with the hydraulic gradient 0.13, hydraulic conductivity of 10^{-4} cm s^{-1}, the cross section area. The transfer coefficient was derived by dividing the groundwater flow rate with the volume of groundwater in the compartment. The transfer coefficients of groundwater were given in Table 2.

Hydrodynamic dispersion could lead the movement of radionuclides in the environment. The coefficient of hydrodynamic dispersion can be expressed in terms of two components, molecular diffusion and kinematic dispersion.

The molecular diffusion was caused by the differences of the concentrations in the compartments. The diffusion was expressed in terms of Fick's law. Fick's law in the porous media was expressed as follows:

$$J = -D_e A\frac{dC}{dx} \tag{5}$$

$$D_e = D_p \varepsilon_p \tag{6}$$

where J is the tritium flux through the cross section area A[m^2] [Bq yr^{-1}],
D_e is the effective diffusivity [m^2 yr^{-1}],
D_p is the pore diffusivity [m^2 yr^{-1}],
ε_p is the porosity [-],
C is the concentration of tritium in the compartment [Bq m^{-3}].

The amount of tritium transferred between the compartments was calculated using equation (5) with the effective diffusivity of 4.3×10^{-4} m^2 yr^{-1}. The transfer coefficient was in the range of 10^{-10} yr^{-1}. It was so small that the mass transfer of tritium due to diffusion was not considered in the calculation.

The kinematic dispersion can be calculated by the following equation:

$$D_k = \alpha_l \cdot v \tag{7}$$

where α_l is the dispersivity [m],
v groundwater flow velocity [m y^{-1}].

The dispersivity of 1 m was used conservatively in the calculation. The transfer coefficients due to hydrodynamic dispersion were given in Table 2.

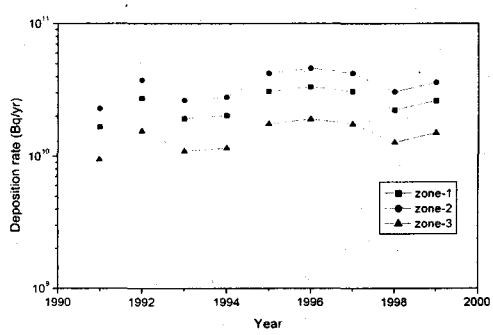

Fig. 5. Time dependent source term in each zone.

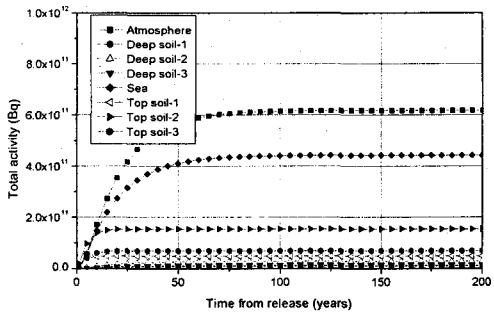

Fig. 6. Total activity of tritium in the compartments calculated using a dynamic compartment model.

5. RESULTS AND DISCUSSION

The changes of tritium activity were calculated with the time dependent source terms shown in Figure 5. The deposition rates were similar to the

release rates shown in Figure 1 because the x/Q values used in the calculation were same every year. The deposition rates in zone-2 were the greatest because of the large area. The transfer coefficients between the compartments given in Table 2 were used for the calculation of tritium concentrations. Figure 6 showed the change of total tritium activity in the compartments of zone-1, zone-2, and zone-3. As shown in Figure 6, the tritium activities in the compartments of our interest, top soil-1, top soil-2, and top soil-3, approached steady state values of 4.82×10^{10} Bq, 1.51×10^{11}, and 6.74×10^{10} Bq within around 10 years after tritium discharge. However, tritium activities in the deep soil, atmosphere, and sea approached steady state more slowly. The amount of tritium in the atmosphere was large since the transfer coefficient of 0.128 yr^{-1} for the evaporation was much higher than the other transfer coefficients. Also, a great amount of tritium flew into the sea. The tritium activity in the top-soil of zone-2 showed the highest due to large deposition rates.

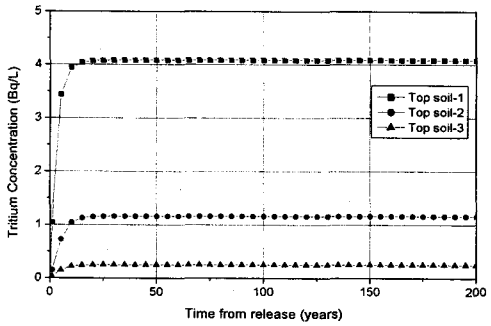

Fig. 7. Tritium concentration in the top soil of each zone.

Figure 7 shows the tritium concentration in zone-1, zone-2, and zone-3. As shown in the Figure 7, tritium concentration in zone-1 was estimated to around 4.08 Bq L^{-1}. Natural background tritium concentration of 0.618 Bq L^{-1} in the top soil, which was estimated using a global tritium cycling model[12]. Those values showed a little higher tritium concentration in the top soil. It was believed that the washout ratio used in wet deposition rates was not measured. If the value was adjusted, we could get more realistic tritium concentration. Since the validation of the model is ongoing, we expect to find the best parameter values later. Also, the more accurate calculation would be obtained if the compartments are refined and input parameters are supplied with the in situ data.

The change of tritium concentration was predicted in the case that the source term increased ten times in tenth year of the release for one year. This accident scenario was introduced to show how the environment of our interest reacted against the sudden increase of tritium release. For the calculation it was assumed that the meteorological and hydrological states of the site were not changed. Figure 8 showed the result of the calculation. The result showed that the sudden increase of tritium concentration in the top soils return to initial steady state concentration within 10 years after the accident if the release rates returned to the original value.

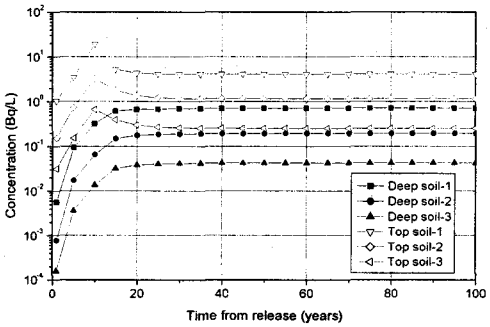

Fig. 8. Prediction of the change of tritium concentration in the environment in the case of accident scenario.

6. CONCLUSION

A dynamic compartment model was developed to predict the tritium behavior in the environment including groundwater and surface water. The model showed the relationship between the tritium release rate from the nuclear power plant and the tritium concentration around the Kori site. The atmospheric tritium concentration in each sector according to the direction and the distance was obtained using the release rates and x/Q values. The deposition rates onto zones were calculated using a computer program, DEPOS.

The northern area of the Kori site was selected as the most important based upon the wind profile around the Kori site. The northern area of the site was divided into 3 zones for the modeling. The transfer coefficients between the compartments were derived from the site characterization data around Kori site, and the source terms were derived from

the dry and wet deposition rates. Five different kinds of mechanisms were reviewed for the tritium movement in the environment. Among them, the transfer coefficient of evaporation was the most important.

Tritium concentrations in surface soil water and groundwater were calculated using the compartment model. The results showed that most of the tritium deposited onto the land released into the atmosphere and the sea and was not accumulated in the surroundings. The tritium activity in top soil approached steady state within around 10 years, but the activities in the deep soil, atmosphere, and sea approached more slowly. The tritium activity in the top soil of zone-2 showed the highest.

The change of tritium concentration was predicted in the case that the source term increased ten times in tenth year of the release for one year. This accident scenario was introduced to show how the environment of our interest reacted against the sudden increase of tritium release. The result showed that the sudden increase of tritium concentration in the top soils return to the initial steady state concentration within 10 years after the accident if the release rates returned to the original value.

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