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Modeling of the Environmental Behavior of Tritium Around the Nuclear Power Plants

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Abstract

The relationship between the tritium release rate from the nuclear power plant and tritium concentration in the environment around the Kori site was modeled. The tritium concentration in the atmosphere was calculated by multiplying the release rates and x/Q values, and the dry deposition rate at each sector according to the direction and the distance was obtained using a dry deposition velocity. The area around Kori site was divided into 6 zones according to the deposition rate. The six zones were divided into 14 compartments for the numerical simulation. Transfer coefficients between the compartments were derived using site characterization data. Source terms were calculated from the dry deposition rates. Tritium concentration in surface soil water and groundwater was calculated based upon a compartment model. The semi-analytical solution of the compartment model was obtained with a computer program, AMBER. The results showed that most of tritium deposited onto the land released into the atmosphere and the sea. Also, the estimated concentration in the top soil agreed well to that measured. Using the model, tritium concentration was predicted in the case that the tritium release rates were doubled.

Key Words : periodic safety review, nuclear power plant, tritium, dry deposition, compartment model

1. Introduction

The PSR (Periodic Safety Review) of the existing nuclear power plants is required every ten years according to the recently revised atomic energy regulations. The relationship between the radionuclide release rates from the plants and the concentration of radionuclides in the environment is to be estimated for the PSR in order to confirm the safe operation of the plants. For this purpose the 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 estimate the environmental tritium behavior by introducing a compartment model. The concentration of HTO in the atmosphere and the deposition rates were calculated. The environments around the nuclear power plants were represented by 14 compartments in which tritium was assumed to be mixed completely. The 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, tritium concentration was estimated using the model in the case that the tritium source terms were doubled due to the nuclear power plants that will be constructed in the near future.

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

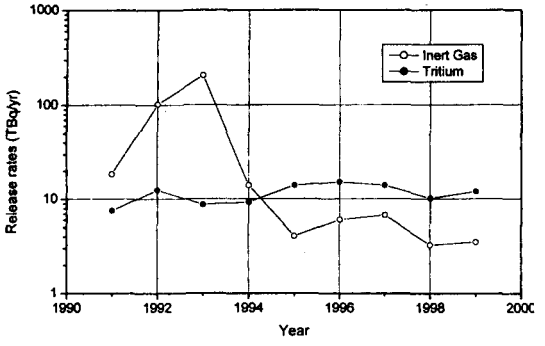

Fig.1. Release Rates of Tritium and Inert Gas from the Kori Nuclear Power Plants. The Release Rates of Tritium Seemed to be Constant for 10 Years

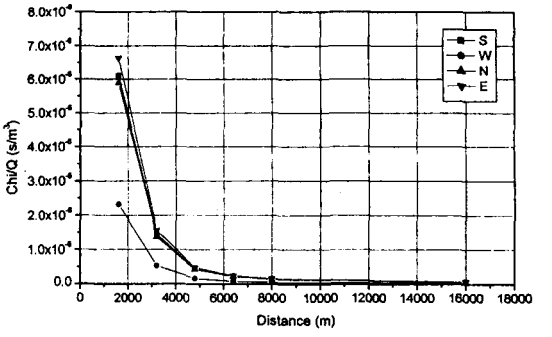

Fig. 2. χ/Q Values in 1999 Showed That They Decreased Remarkably Beyond 8,000m

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 was used [9]. Tritium deposition rates in the land and the sea were computed respectively. The calculation result showed the deposition rate in the sea was 2.13 times as much as that in the land. That is, more than 60% of tritium and other radionuclides released as a gas phase from the Kori-nuclear power plants deposited in the sea due to wind profile in 1999. In this study the tritium deposited onto the soil was assumed to be 50% due to the coverage of land with plants.

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:

$$\frac{dA_i^t}{dt} = -\sum_n k_{ij} A_i^t + \sum_m k_{ji} A_j^t - \lambda^t A_i^t + S_i^t \qquad (2)$$

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 (2) 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 (2) were coupled each other, it was not easy to solve them analytically. The computer program, AMBER, was used in this study.

4. Source Terms and Transfer Coefficients

The source term was required to model the

Table 1. Location of Sectors and the Deposition Rates on the Sectors

ZONE	AREA (m^2)	DIRECTION	DISTANCE (km)	ANNUAL DEPOSITION RATES (Bq)
zone-1	1.81×10^7	NE, NNE, N, NNW	4.8 km	4.20×10^{10}
zone-2	3.22×10^7	NE, NNE, N, NNW	4.8 ~ 8.0 km	8.45×10^9
zone-3	1.36×10^7	NW, WNW, W	4.8 km	1.43×10^{10}
zone-4	2.41×10^7	NW, WNW, W	4.8 ~ 8.0 km	2.87×10^9
zone-5	6.53×10^6	WSW, SW	4.8 km	9.15×10^9
zone-6	1.61×10^7	WSW, SW	4.8 ~ 8.0 km	6.40×10^9

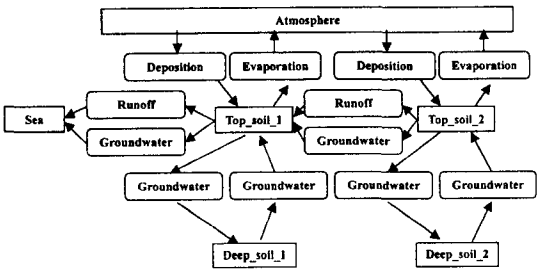

Fig. 3. A Part of Compartments (in zone-1 and zone-2) and Transfer Coefficients Used in the Modeling of Tritium Movement Around the Kori Nuclear Power Plants

behavior of tritium in the environment. The deposition rates in 6 zones given in Table 1 were used as source terms. The source terms for the compartment model were assumed to be constant regardless of time.

The sectors around the Kori NPPs were grouped into 6 zones for the compartment modeling. The soil zones were divided into 2 compartments. 12 compartments were introduced for the soil layer around the Kori-site: 6 compartments were for the top soil and the others for the deep soil. The porosity of top soil was 0.3, and that of deep soil 0.25. Total 14 compartments were used to describe the tritium behavior in the environment. Figure 3 showed the part of the compartments (6 out of 14) used in this study. One compartment for the atmosphere was introduced for the evaporation of tritium, and one sink compartment for the sea in which tritium flew along with groundwater or runoff water.

The transfer of tritium between the compartments was due to evaporation, groundwater flow, infiltration, runoff, and diffusion. The 10 year average precipitation rates and evaporation rates in this area were used to derive the transfer coefficients. The annual precipitation rate was 1,390mm, and annual evaporation rate was 1,150mm [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 100mm 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 13%, and hydraulic conductivity was estimated within the range of 10^{-3}~10^{-5} cm/s. The hydraulic properties of the site were not measured in detail. Groundwater flow was described with the following Darcy' s law [11]:

Table 2. Transfer Coefficients of Runoff and Groundwater Flow

Transfer Coefficient (yr-1)		Value
Runoff	K_{1s}	0.0432
	K_{21}	0.0156
	K_{3s}	0.0431
	K_{43}	0.0156
	K_{5s}	0.0539
	K_{65}	0.0156
Groundwater Flow	K_{1s}	5.68×10^{-3}
	K_{21}	5.32×10^{-3}
	K_{3s}	5.71×10^{-3}
	K_{43}	5.34×10^{-3}
	K_{5s}	7.87×10^{-3}
	K_{65}	5.31×10^{-3}

$$Q = -KA\frac{dh}{dl} \quad (3)$$

where Q is the flow rate of groundwater through the cross section area A[m^2] [m^3/yr],

K is the hydraulic conductivity [m/yr],

$\frac{dh}{dl}$ is the hydraulic gradient [-].

The groundwater flow rate across the vertical cross section between the compartments was obtained from the equation (3) with the hydraulic gradient 0.13, hydraulic conductivity of 10^{-4} cm/s, 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.

Diffusion could lead the movement of radionuclides in the environment. The diffusion was caused by the differences of the concentrations in the compartments. The diffusion was expressed in terms of Fick' s law. The Fick' s law in the porous media was expressed as follows:

$$J = -D_e A\frac{dC}{dx} \quad (4)$$

$$D_e = D_p\varepsilon_p \quad (5)$$

where J is the tritium flux through the cross section area A[m^2] [Bq/yr],

D_e is the effective diffusivity [m^2/yr],

D_p is the pore diffusivity [m^2/yr],

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 (4) with the effective diffusivity of 4.3×10^{-4} m^2/yr. 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.

5. Results and Discussion

The changes of tritium activity were calculated using the computer program AMBER with the constant source terms of 4.20×10^{10} Bq for zone-1 and 8.45×10^{10} Bq for zone-2. The transfer coefficients between the compartments given in Table 2 were used for the calculation of tritium

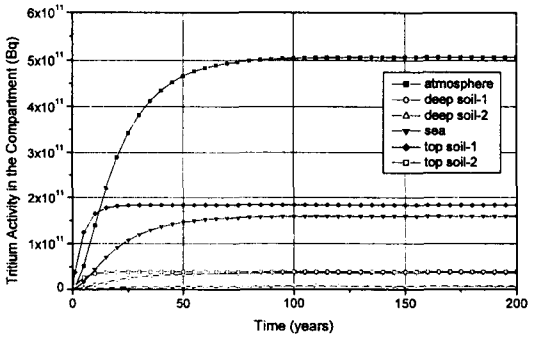

Fig. 4. Calculation Result: Tritium Activity in the Compartments of Zone-1 and Zone-2. Tritium in Top Soil Approached Steady State Quickly

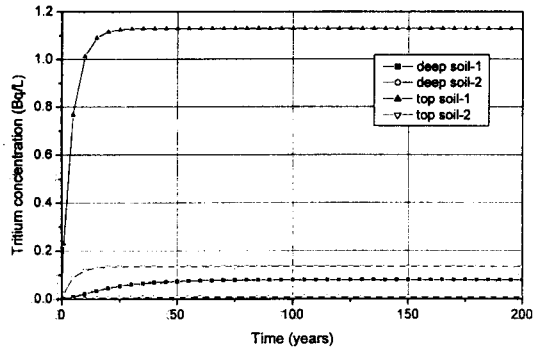

Fig. 5. Calculation Result: Tritium Concentration in the Compartments of Zone-1 and Zone-2

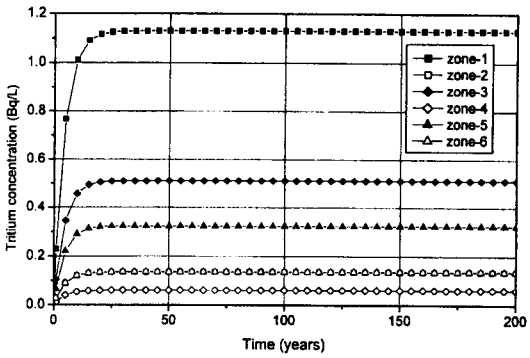

Fig. 6. Comparison of Tritium Concentrations in the Top Soil. Zone-1 Showed the Highest Concentration

concentrations in the compartments. Figure 4 showed the change of total tritium activity in the compartments of zone-1 and zone-2. The behaviors of tritium activity in the other sectors were very similar to those of zone-1 and zone-2. As shown in Figure 4, the tritium activities in the compartments of our interest, top soil-1 and top soil-2, approached steady state values of 1.84×10^{11} Bq and 3.92×10^{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-1 showed high due to large deposition rates.

Figure 5 showed the tritium concentration in zone-1 and zone-2 to compare with the measured data. The results of the AMBER calculation were summarized in Table 3. As shown in the Figure 5 and Table 3, tritium concentration in zone-1 was estimated to around 1.748 Bq/L considering the background tritium concentration. Global tritium cycling model was used for the estimation of background tritium concentration. The background tritium concentration in Korea was mostly originated from the interactions between the upper atmosphere gases and cosmic rays according to our previous result [12]. The

Table 3. Comparison of AMBER Calculation with Measured Data

Zone	Location	Tritium concentration measured (Bq/L)	Background tritium concentration estimated (A) (Bq/L)	Tritium concentration from NPPs (B) (Bq/L)	Tritium Concentration estimated (A+B) (Bq/L)
zone-1	Hyoam Kangwol Kogyeongsa	2.58 1.88 1.69	0.618	1.13	1.748
zone-2		-	0..618	0.135	0.753

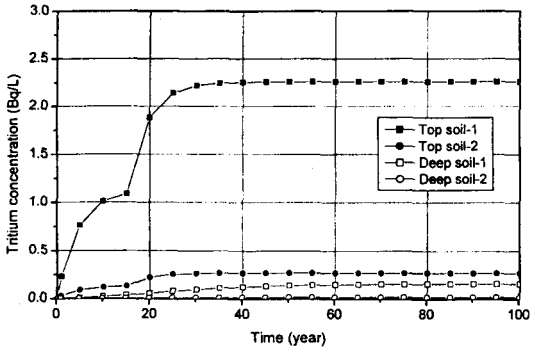

Fig. 7. Changes of Tritium Concentration in the Case that the Tritium Release Rates were Doubled

estimation of tritium concentration was not far away from the measured concentration in zone-1. The more precise calculation would be obtained if the compartments are refined and input parameters are supplied with the in situ data. Figure 6 showed the tritium concentrations in the 6 top soil compartments. As expected, the sectors with odd number showed higher concentrations than those with even number because they were located nearer.

The tritium concentrations were calculated in the case that the tritium release rates were doubled due to the addition of new nuclear power plants at the same site. That is to say, the source terms were doubled when the tritium concentration approached steady states. For the calculation it was assumed that the meteorological and hydrological states of the site were not changed. Figure 7 showed the change of tritium concen-trations. The result showed that the highest tritium concentration around the nuclear power plants was still low even though new reactors were added.

6. Conclusions

Tritium behavior in the environment including groundwater and surface water was studied based upon a compartment model to find 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 calculated with a XOQDOQ computer program. Deposition rates showed that more than 60 percents of tritium released from the NPPs fell down to the sea.

The environmental behavior of tritium was studied using a compartment model. The area around the Kori site was divided into 14 compartments accounting for the deposition rates and the distance from the NPPs. 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 deposition rates. Five different kinds of mechanisms were reviewed for the tritium movement in the environment. Among them transfer coefficient of diffusion was neglected due to too low value and that of evaporation was of the most importance.

Tritium concentrations in surface soil water and groundwater were calculated based upon the compartment model. The semi-analytical solution of the compartment model was obtained with the computer program, AMBER. 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 estimated tritium concentration in the surface soil water agreed well to that measured. Also, tritium concentration was predicted for the scenario in which the release rates were doubled. The result showed that the tritium concentration increased a little.

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