< Technical Note >

PREDICTION OF THE TRITIUM CONCENTRATION IN THE SOIL WATER AFTER THE OPERATION OF WOLSONG TRITIUM REMOVAL FACILITY

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The effect of the Wolsong Tritium Removal Facility on the change of tritium concentration in the soil water was assessed by introducing a dynamic compartment model. For the mathematical modeling, the tritium in the environment was thought to come from two different sources. Three global tritium cycling models were compared with the natural background concentration. The dynamic compartment model was used to model the behavior of the tritium from the nuclear power plants at the Wolsong site. The source term for the dynamic compartment model was calculated with the dry and wet deposition rates. The area around the Wolsong nuclear power plants was represented by the compartments. The mechanisms considered in deriving the transfer coefficients between the compartments were evaporation, runoff, infiltration, hydrodynamic dispersion, and groundwater flow. We predicted what the change of the tritium concentration around the Wolsong nuclear power plants would be after future operation of the tritium removal facility to show the applicability of the model. The results showed that the operation of the tritium removal facility would reduce the tritium concentration in topsoil water quickly.

KEYWORDS : Tritium, Environment, Compartment, Model, Deposition, Tritium Removal Facility

1. INTRODUCTION

There are four CANDU (Canadian Deuterium Uranium) pressurized heavy water reactors in operation at the Wolsong site, Republic of Korea. The amounts of tritium in the CANDU reactors increase because the generation rates of tritium are higher than the decay rates. Tritium is mainly produced through neutron capture by the deuterium atom in heavy water, which is used as the moderator and coolant in CANDU reactors [1]. The tritium concentration in the atmosphere and rain water at the Wolsong site was especially monitored, due to the high tritium concentration in the environment. A tritium removal facility (TRF) is being constructed at the Wolsong site to reduce the tritium discharge rates.

We reviewed the record of the discharge rates and tritium concentration at the Wolsong site to model the tritium behavior in the environment. We developed a dynamic compartment model to find the relationship between the tritium release rate and the tritium concentration in the environment [2]. Two different sources of tritium were modeled separately: one from the nuclear power plants and the other from the natural background tritium. The natural background tritium level was generally computed with a global tritium cycling model. Three different models were compared with one another by using the tritium concentration in the soil water. Four different sources were included to estimate the natural background concentration.

The tritium concentration in the atmosphere was calculated with the annual average x/Q values and discharge rates. The calculation was compared with the measurement data at three locations around the site. The wet deposition of the tritium was modeled using the washout ratio approach, since we were interested in the long-term average deposition rates. The wet deposition rates calculated with the washout ratio method were compared using the measurement data at two locations. The dry deposition rates were calculated with the dry deposition velocity.

The topography of the Wolsong site was analyzed using a digital map and the SURFER program to select a representative area for the compartment model. The selected area was represented with four compartments. The tritium source term was calculated from the deposition rates. The transfer coefficients between the compartments were derived for the dynamic compartment model. The annual rates of

evaporation and runoff were calculated using a computer program, Visual HELP, and the groundwater velocity of the site was obtained with the FEMWATER program. A numerical solution for the dynamic compartment model was obtained using the AMBER program. The effect of the TRF operation on the change of the tritium concentration in the soil water was predicted using the dynamic compartment model.

2. NATURAL BACKGROUND TRITIUM

The Korea Institute of Nuclear Safety (KINS), a regulatory body, measures the tritium concentration of South Korea's tap water annually. The recent data showed a concentration of around 0.8 Bq L^{-1} in the nation's tap water [3]. However, an appropriate mathematical model is required to predict the change of the tritium concentration in the future. In general, the global tritium cycling model was used to estimate the natural background concentration. In this paper, we compared three different global tritium cycling models using the tritium concentration in the South Korean tap water. Two NCRP seven compartment models and one model proposed by Killough and Kocher were compared. In NCRP 62 [4], two types of seven-compartment models for the global cycling of the tritium are suggested: one model for the entire planet and the other for the northern hemisphere only. Killough and Kocher [5] modified the NCRP model by separating the tropospheric water volumes on land from those in the ocean to predict the high tritium concentration in the 1960s. We used the transfer coefficients between the compartments given in the UNSCEAR report [6] to calculate the tritium concentration in the compartments.

Four different sources of tritium were considered for the modeling in this study [7]: interactions between gases of the upper atmosphere and cosmic rays, nuclear bomb tests, consumer products, and nuclear power plants [8]. Figure 1 shows the calculation results of the three different models. The NCRP northern hemisphere model result showed the best agreement in 2000. The NCRP northern hemisphere model results and the Killough and Kocher model results agreed well with the tritium concentrations of South Korean tap water in 1960s.

3. TRITIUM FROM THE NUCLEAR POWER PLANTS

3.1 Tritium Concentration in the Atmosphere

The first CANDU reactor began operation at the Wolsong site in 1988. Four CANDU reactors with a power output capacity of 2,780 MWe have been in operation at the Wolsong site since 1998. Since tritium is mainly produced through neutron capture by the deuterium atom in the heavy water used as the moderator and coolant in CANDU reactors, the discharge rates of tritium from the CANDU reactors are larger than those of PWR reactors. Figure 2 shows the tritium discharge rates from the CANDU and PWR (Kori total) reactors [9]. As shown in Figure 2, the amount of tritium discharged from the Wolsong site

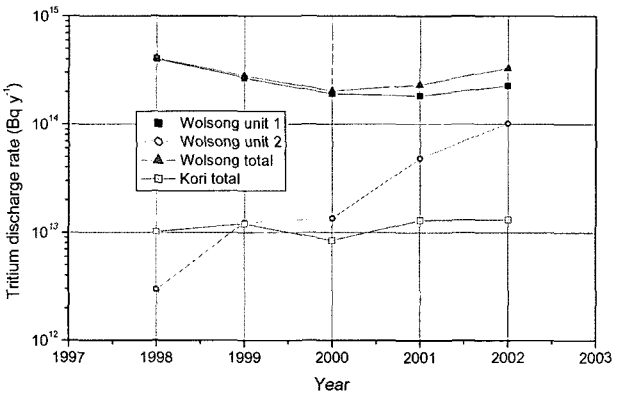

Fig. 2. Tritium Discharge rates from the Wolsong(CANDU) and Kori(PWR) Nuclear Power Plants [9]

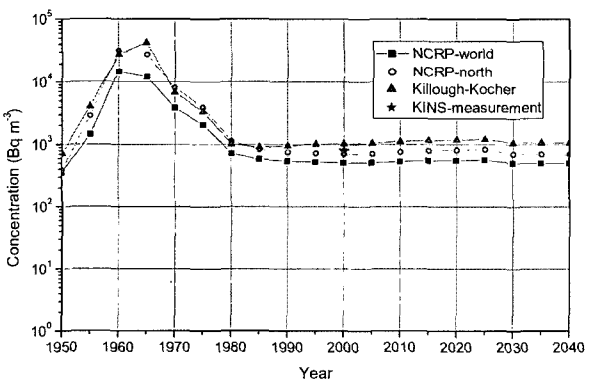

Fig. 1. Comparison of Three Different Global Tritium Cycling Models. NCRP Northern Model Shows the best Agreement with Tritium Concentration Measured in 2000

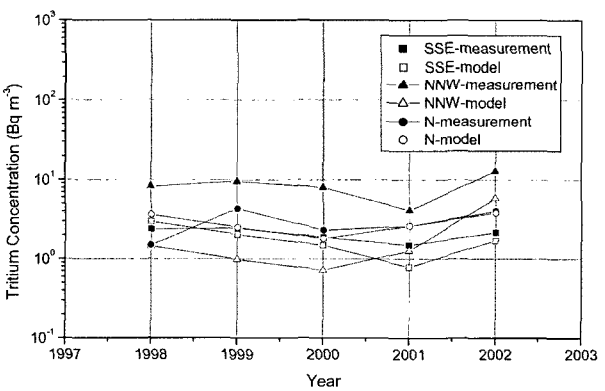

Fig. 3. Comparison of the Result of the Calculation with the Tritium Concentration in the Atmosphere Measured Around the Wolsong Nuclear Power Plants

was much larger than that of the Kori site where four PWR reactors with a power output capacity of 3,140 MWe are in operation. The tritium discharge rate of Wolsong Unit 1 is larger than that of Wolsong Unit 2, due to the short operation time of unit 2.

The tritium concentration in the atmosphere has been measured by KINS, due to the high discharge rates at the Wolsong site. The atmospheric tritium concentration shown in Figure 3 was calculated by multiplying the annual average x/Q values and the discharge rates. The modified x/Q values obtained at the Wolsong nuclear power plants [10] were used in the calculation. Figure 3 shows a comparison of the tritium concentration in the atmosphere at three different locations: the Eastern Gate (denoted by SSE, 0.8 km from the Meteorological Tower), the Waste Storage Building (denoted by NNW, 0.5 km from the Meteorological Tower), and the company houses for the company guards (denoted by N, 1.0 km from the Meteorological Tower). The calculation of the tritium concentration was quite close to the measured value, except for the location denoted by NNW where a waste storage building was located. It seems that an amount of tritium being released from the waste storage building was not included in the calculation, but it became evident as an increased measured concentration.

3.2 Wet and Dry Deposition of Tritium

Tritium released from nuclear power plants is transported by advection and diffusion. Removal of a tritiated compound from the atmosphere takes place by deposition. The atmospheric removal processes are classified into dry or wet deposition. There are two methods by which to calculate the wet deposition. The washout rate approach is used for the short-term release, and the washout ratio approach is used for the long-term release. In accordance with the washout ratio approach [11], the wet deposition rate, $\dot{W}$, was calculated using the following equation:

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

where $\dot{Q}$ is the tritium discharge rate [Bq y^{-1}],
u is the wind velocity [m s^{-1}],
σ_y is the diffusion parameter [m],
ω is the washout ratio [-], and
I is the precipitation rate [m y^{-1}].

In addition, ωI in equation (1) is called the wet deposition velocity. The washout ratio was determined with the following relation:

$$\omega = \frac{R_0}{\chi} \quad (2)$$

where R_0 is the tritium concentration in rain water (Bq m^{-3}), and
χ is the tritium concentration in the atmosphere (Bq m^{-3}).

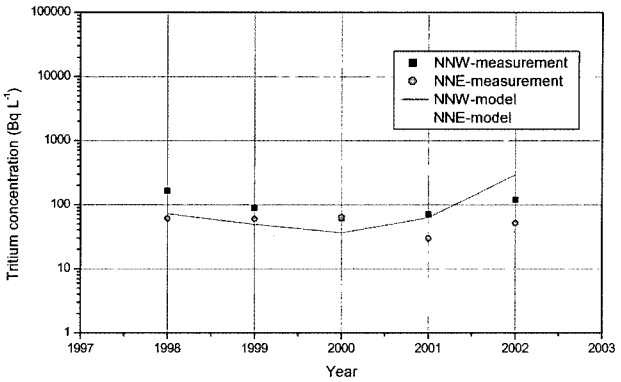

Fig. 4. Comparison of the Modeling and Measurement with the Tritium Concentration in the Rain Water Around the Wolsong Site [15]

The washout ratio around the Wolsong site was obtained with equation (2) [12]. The average value of the washout ratio was 4.85×10^4. Slinn et al. [13] estimated the washout ratio of 10^5 for the HTO. The value obtained in this study was slightly lower than that given by Slinn et al. The wet deposition of the tritium onto the topsoil compartment was calculated using a computer program, DEPOS [14]. Figure 4 shows a comparison of the calculation and measurement results [15] of the tritium concentration at two locations. The results showed a good agreement.

According to Briggs et al. [16], the amount of radionuclides dry-deposited onto the soil are obtained by multiplying the atmospheric concentration near the soil and the dry deposition velocity:

$$w(x,y)=v_d C(x,y,0) \quad (3)$$

where w is the tritium deposition rates (Bq m^{-2} s^{-1});
v_d is the dry deposition velocity of tritium (m s^{-1}); and
C is the tritium concentration in the atmosphere (Bq m^{-3}).

The tritium concentration in equation (3) was obtained by multiplying x/Q from the Gaussian plume model and the tritium discharge rate from the Nuclear Power Plants (NPP). The deposition rate in each sector was obtained by multiplying the concentration of the tritium, the area of the sector, and the deposition velocity. A deposition velocity of 4.94×10^{-4} m s^{-1} was used for soil [17].

3.3 Dynamic Compartment Model

Most tritium exists in the form of HTO in the environment. Since HTO is very mobile in the environment, its behavior can be described using a compartment model. Figure 5 shows the compartments for the tritium movement modeling. The mass transfer between the compartments was expressed using the following mass conservation equations:

$$\frac{dA_i}{dt} = -\sum_n k_{ij} A_i + \sum_m k_{ji} A_j - \lambda A_i + S_i \qquad (4)$$

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 (y^{-1});
λ is the tritium decay constant (y^{-1}); and
S_i is the source term in the compartment-i (Bq y^{-1}).
The computer program, AMBER [18], was used to numerically solve equation (4).

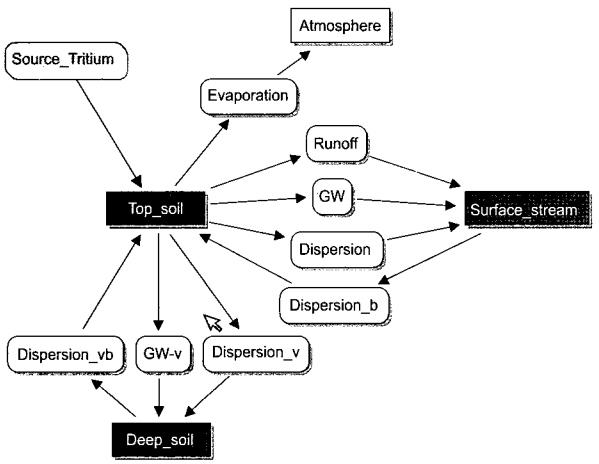

Fig. 5. Compartments and Transfer Coefficients Used for Modeling the Tritium Behavior

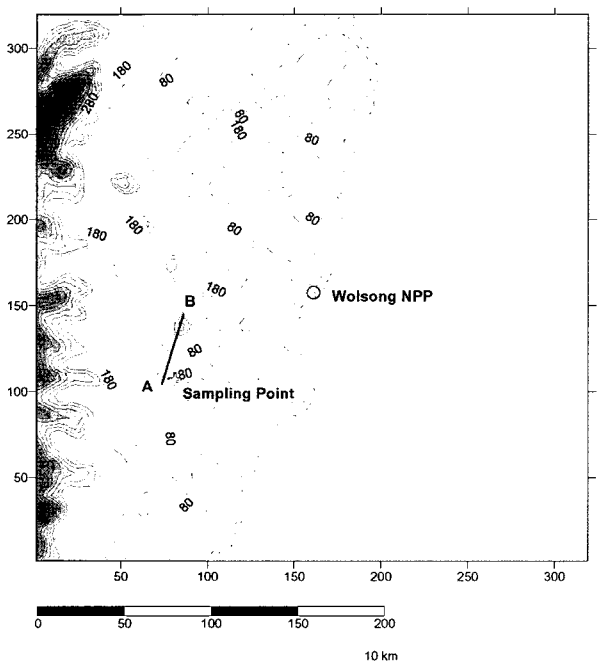

Fig. 6. Contour map of the Wolsong Site. Cross Mark Indicates the Sampling Point

3.4 Source Term and Transfer Coefficients

The geography of the Wolsong site was investigated using a digital map to derive the transfer coefficients. Figure 6 shows the topography of the Wolsong site. The area around the sampling point marked by a cross was selected and represented by four compartments. The sampling point was located at the Kiguri near the Wolsong NPP in the WSW direction. The distance from the nuclear power plants to the sampling point was between 4.8 km to 6.4 km. Even though the surface stream was not the final sink of the tritium, the sea was not included, since we were only interested in the concentration of tritium in the topsoil water.

The source term of the tritium was calculated from the wet and dry deposition rates. The atmospheric concentration was calculated using the discharge rates (given in Figure 2) and the x/Q values at the sector in the WSW direction. Figure 7 shows the time-dependent source term used for the dynamic compartment modeling. Since the data were limited to five years, we assumed that the fifth year deposition rate continued for 10 years.

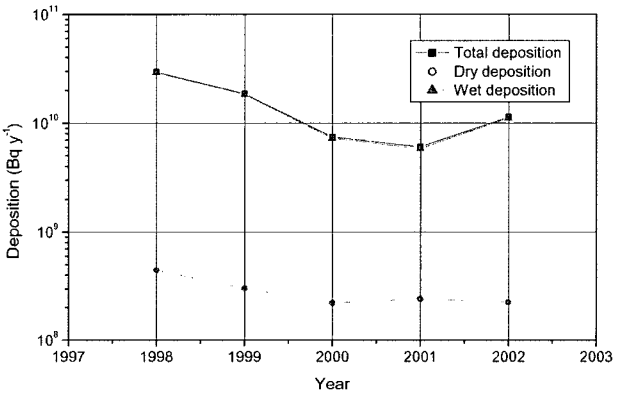

Fig. 7. Tritium Source Term Used for the Dynamic Compartment Modeling

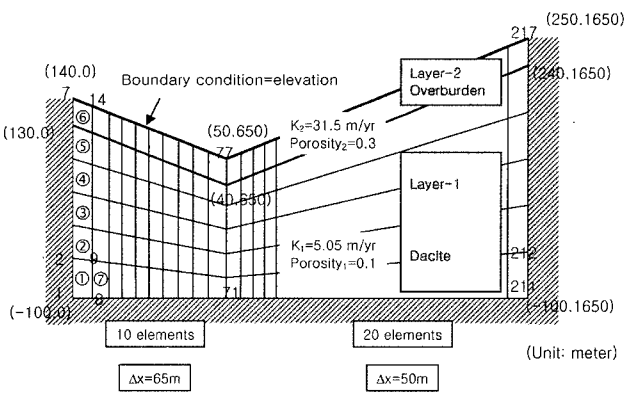

Fig. 8. Elements and Nodes Used for the Modeling of Groundwater with FEMWATER

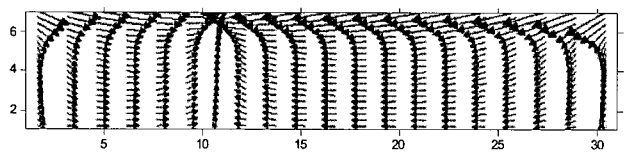

Fig. 9. Groundwater Velocity Vectors Calculated Using FEMWATER. The Numbers in the x- and y-axes Mean the Element Number in Each Direction

The transfer of the tritium between the compartments was mainly due to evaporation, groundwater flow, infiltration, runoff, and dispersion. The annual rates of evaporation and runoff were calculated using a computer program, Visual HELP [19]. The transfer coefficients of the evaporation and runoff between the compartments were calculated by dividing the evaporation and runoff rates by the pore volume of the compartment, respectively.

Tritium deposited onto the soil moved with the groundwater flow. According to the FSAR of the Wolsong nuclear power plants, an overburden was located on the Dacite at the site. Hydraulic conductivities of the overburden and the Dacite were estimated to be 31.5 and 5.05 m y^{-1}, respectively. Groundwater flow was described by Darcy's law. The finite element computer program, FEMWATER, was used to calculate the steady state groundwater flow. Figure 8 shows the vertical cross section of the area denoted by line AB in Figure 7. A total of 180 elements were used to calculate the groundwater flow. Figure 9 shows the groundwater velocity vectors obtained using the FEMWATER program. The numbers in the x- and y-axes indicated the element number. The average groundwater velocity for the overburden was around 4.25 m y^{-1}.

Hydrodynamic dispersion can lead to the movement of the radionuclides in the environment. The coefficient of the hydrodynamic dispersion can be expressed in terms of two components: molecular diffusion and kinematic dispersion. The molecular diffusion was so small that it was not considered. The kinematic dispersion was calculated using the following equation:

$$D_k = \alpha_l \cdot v \tag{5}$$

where α_l is the dispersivity (m);
v is the groundwater flow velocity (m y^{-1}).
A conservative dispersivity value of 1 m was used in the calculation.

4. RESULTS AND DISCUSSION

The changes of the tritium activity were calculated with the time-dependent source term given in Figure 7. Figure 10 shows the tritium concentration in the topsoil and the deep soil at the location of the sampling point. Most of the tritium deposited onto the topsoil returned to the atmosphere, due to the large transfer coefficient of evaporation, and did not accumulate in the topsoil. As shown in Figure 10, the tritium concentration in the topsoil was estimated to be around 4 ~ 5 Bq L^{-1} including a natural background tritium concentration of 0.71 Bq L^{-1}, which was estimated with the NCRP northern hemisphere model. The calculation showed a slightly lower tritium concentration as compared with the measurement. It was expected that a more accurate calculation would be obtained if the compartments were refined and the input parameters were supplied by in situ data.

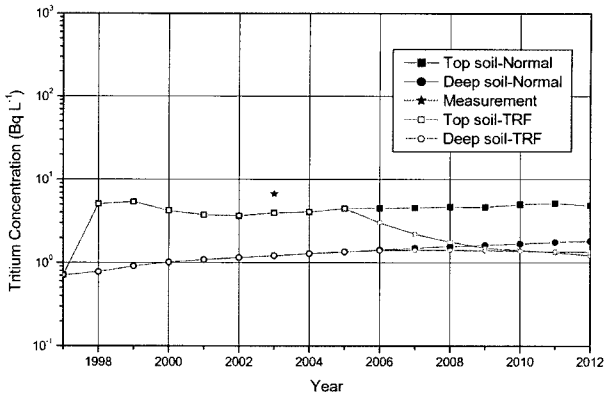

Fig. 10. The Comparison of Model and Measurement with Tritium Concentration in the Top-soil and Deep Soil Compartments and the Prediction of the Change of Tritium Concentration in Soil Water After the Operation of Tritium Removal Facility

A tritium removal facility is being built at the Wolsong site, Republic of Korea, to reduce the discharge rates of tritium. It is scheduled to start operation in 2006. The discharge rates are expected to be reduced by one sixth to one tenth after the operation of the tritium removal facility begins. One scenario regarding the operation of the TRF was introduced to show the usefulness of the dynamic compartment model that was developed in this study. We calculated the change of environmental tritium concentration for a case in which the discharge rates were reduced by one tenth of those of the year 2005. Figure 10 shows the results of the calculation. The results showed that the tritium concentration in the topsoil begins to decrease in 2006, but the concentration in the deep soil changes little, due to low transfer coefficients.

5. CONCLUSIONS

A dynamic compartment model was applied to predict the change of the tritium concentration in the environment around the Wolsong nuclear power plants for future operation of the Wolsong Tritium Removal Facility. Three global tritium cycling models were compared to estimate the natural background tritium concentration. The NCRP northern hemisphere model showed the best agreement

with the measurement in 2000. The tritium concentration in the surface soil water was around 0.7 Bq L^{-1} in 2000, and the interactions between the atmosphere and the cosmic rays were the most important sources of the natural background concentration.

The source term for the dynamic compartment model was derived from the wet and dry deposition rates. The atmospheric tritium concentration was obtained using the discharge rates and the annual average x/Q values. The calculation showed a good agreement with the measurement data. The washout ratio of 4.85×10^4 was derived using the tritium concentration in the atmosphere and in the rain water. The calculation of the tritium concentration with the washout ratio approach showed a good agreement with the measurement data at the two locations.

One area at the Wolsong site was selected as a result of the topographical analysis of the site and was represented by four compartments. Five different kinds of mechanisms were reviewed for the modeling of tritium movement in the environment. Tritium concentration in the topsoil and deep soil water was calculated using the compartment model. The results showed that most of the tritium deposited onto the land was released into the atmosphere and was not accumulated in the topsoil. The tritium concentration in the topsoil was estimated to be around 4~5 Bq L^{-1} including the natural background tritium concentration of 0.71 Bq L^{-1} in the topsoil. It was close to the measured amount of 6.7 Bq L^{-1}.

One scenario regarding the operation of the tritium removal facility was analyzed to show the usefulness of the dynamic compartment model that was developed in this study. The change of the tritium concentration was predicted for a case in which the tritium discharge rates were reduced to one tenth beginning in 2006. The result showed that the tritium concentration in the topsoil started to decrease in 2006, but the concentration in the deep soil changed little, due to low transfer coefficients.

Acknowledgements

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