Prediction of the Tritium Concentration in the Environment around the Wolsong NPPs Using a Dynamic Compartment Model

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Abstract. This paper outlines the current development of the compartment model regarding the tritium behavior around the Wolsong nuclear power plants. For the mathematical modeling the tritium in the environment was deemed to come from two different sources; one from the natural background of the tritium, and the other from the nuclear power plants in operation. The NCRP seven compartment model based upon the global tritium cycling model was used for the calculation of the natural background concentration. Four sources were considered for the modeling of the natural background concentration. The sources of tritium were the interactions between the gases of the upper atmosphere and the cosmic rays, nuclear bomb tests, and consumer products as well as nuclear power plants. For the modeling of the behavior of the tritium from nuclear power plants we developed a dynamic compartment model. The source term for the dynamic compartment model was calculated with the dry and wet deposition rates. The washout ratio was derived using the tritium concentration in the atmosphere and the rain water. The results of the wet deposition calculation showed a good agreement with the measurement. The areas around the Wolsong nuclear power plants were 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 the change of the tritium concentration around the Wolsong nuclear power plants after the operation of the Tritium Removal Facility to show the applicability of the model. The Tritium Removal Facility is being constructed, and its operation will reduce the tritium discharge rate and the tritium concentration in the environment.

1. Introduction

The recently revised Korean Atomic Act asked all nuclear power plants to assess the accumulation trends of the radionuclides in the surrounding environment using measurement data and to predict the change of the concentration in the case of an unexpected release of the nuclides. Only with the measurement data it was not easy to predict the change of the tritium concentration in the future. Korea Atomic Energy Research Institute has developed a dynamic compartment model to find the relationship between the tritium release rate and the tritium concentration in the environment.

There are four CANDU (Canadian Deuterium Uranium) pressurized heavy water reactors in operation at the Wolsong site. The amounts of tritium generated in the CANDU reactors were built up due to their greater generation rates than 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 large discharge rates and high concentration. We reviewed the past record of the discharge rates and concentrations of the tritium at the Wolsong site to model the tritium behavior in the environment.

This paper deals with the application of a dynamic compartment model to predict the tritium behavior in the environment. Two different sources of tritium were modeled separately, that is to say, one from the nuclear power plants and the other from the natural background of the tritium. Natural background tritium was generally computed with a global tritium cycling model. Three different models were compared with each other for the tritium concentration in the soil water for the natural background the tritium concentration. Four different sources were included for the estimation of the natural background concentration.

The tritium concentration in the atmosphere was calculated with the annual average $\chi/Q$ 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 washout ratio was derived using the tritium concentration in the atmosphere and the rainwater. 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 using the dry deposition velocity.

Topography of the site was analyzed with a digital map and the SURFER program to select a representative area for the compartment model. The selected area was represented with four compartments. Tritium source term was calculated from the deposition rates. Transfer coefficients between the compartments were derived for the dynamic compartment model. The future weather conditions were generated using a computer program, Visual HELP, and the hydrology of the site was analyzed with the FEMWATER program. A numerical solution was obtained for the dynamic compartment model using the AMBER program. The effect of the operation of the Tritium Removal Facility on the change of the tritium concentration was predicted using the dynamic compartment model.

2. Natural background Tritium Concentration in Korea

The regulatory body, Korea Institute of Nuclear Safety, measures the tritium concentration of the tap water in the whole country every year. The recent data showed a concentration of around 0.8 Bq/L in tap water [2]. However, an appropriate mathematical model was required to predict the change of the tritium concentration in the future. In general, the global tritium cycling models were used to estimate the natural background concentration. In this paper, we compared three different global tritium cycling models with the tritium concentration in tap water. Two NCRP seven compartment models and one model proposed by Killough and Kocher were compared. NCRP 62 [3] suggested two kinds of 7 compartment model for the global cycling of the tritium. One is for the whole world, and the other is for the northern hemisphere. Killough and Kocher [4] modified the NCRP model by separating the tropospheric water volumes over land from the ocean to predict the high tritium concentration in the 1960s. We used the transfer coefficients between the compartments given in the UNSCEAR report [5].

Four different kinds of sources were considered for the modeling. The sources of tritium [6] were the interactions between gases of the upper atmosphere and the cosmic rays, nuclear bomb tests, and consumer products as well as nuclear power plants. FIG. 1 shows the results of the calculations with the 3 different models. The NCRP northern hemisphere model showed the best agreement with the tritium concentration of tap water in 2000. The NCRP northern hemisphere model and the Killough and Kocher model agreed well for the 1960s.

3. Tritium concentration in the atmosphere

FIG. 1. Comparison of three different global tritium cycling models. NCRP northern model showed the best agreement with tritium concentration in 2000.
The first CANDU reactor started to operate at the Wolsong site in 1988. Now four CANDU reactors have been in operation at the Wolsong site since 1998. Since 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, the discharge rates of tritium from the CANDU reactors are larger than those of PWR reactors. FIG. 2 shows the tritium discharge rates from the CANDU and PWR (Kori) reactors. As shown in FIG. 2, the amount of tritium from the Wolsong site was much larger than the Kori site. The tritium discharge rate from the Wolsong unit 2 increased due to its short operation time compared with unit 1.

The tritium concentration in the atmosphere has been measured due to the high discharge rates at the Wolsong site. The tritium concentration was calculated by multiplying the annual average $\chi/Q$ values and the discharge rates given in FIG. 2. The $\chi/Q$ values were calculated with the XOQDOQ computer program. FIG. 3 shows the comparison of the tritium concentration in the atmosphere at three different locations. The calculation of the tritium concentration was quite close to the measured value except the location denoted by NNW where a waste storage building existed. It seems that some amounts of the tritium released from the waste storage building which were not included in the calculation and thus it increased the measured concentration.

![Graph showing tritium discharge rates from Wolsong (CANDU) and Kori (PWR) nuclear power plants.](image)

**FIG. 2. Tritium discharge rates from the Wolsong (CANDU) and Kori (PWR) nuclear power plants.**

![Graph showing comparison of calculation and measurement for tritium concentration around the Wolsong nuclear power plant.](image)

**FIG. 3. Comparison of the calculation and measurement for the tritium concentration around the Wolsong nuclear power plant.**
4. Wet and dry deposition of tritium

Tritium released from nuclear power plants are transported by advection and diffusion. Removal of the tritiated compound from the atmosphere takes place by deposition. The atmospheric removal processes are classified into dry or wet deposition. There are two methods 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. According to the washout ratio approach, the wet deposition rate was calculated using the following equation:

\[ W(x, y) = \frac{\omega I Q}{(2\pi)^{3/2} u \sigma_y(x)} \exp\left[ -\frac{y^2}{2\sigma_y^2(x)} \right] \]  

(1)

where

- \( \omega \) washout ratio (-);
- \( I \) the precipitation rate (m/y).

\( \omega I \) in equation (1) is called the wet deposition velocity. The washout ratio was determined with the following relation:

\[ \omega = \frac{R_0}{\chi} \]  

(2)

where

- \( R_0 \) the tritium concentration in rain water (Bq/m\(^3\));
- \( \chi \) the tritium concentration in the atmosphere (Bq/m\(^3\)).

The washout ratio around the Wolsong site was determined with equation (2). FIG. 4 shows the washout ratio and the tritium concentration measured for recent five years. The average value of the washout ratio was 4.85×10\(^4\). Slinn et al. [7] 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 top-soil compartment was calculated using a computer program, DEPOS, developed by authors [8]. FIG. 5 shows the comparison of the calculation and measurement with the tritium concentration at two locations. The washout ratio of 5×10\(^4\) was used in the calculation. The results show that they agreed well.

According to Briggs et al.[9], the amount of radionuclides dry-deposited onto the soil are obtained by multiplying the atmospheric concentration near the soil and the dry deposition velocity:

\[ w(x, y) = v_d C(x, y, 0) \]  

(3)

where

- \( w \) tritium deposition rates (Bq/m\(^2\) s);
- \( v_d \) dry deposition velocity of tritium (m/s);
- \( C \) the tritium concentration in the atmosphere (Bq/m\(^3\)).

The tritium concentration in equation (3) was obtained by multiplying \( \chi/Q \) from the Gaussian plume model and the tritium discharge rate from the NPPs. The deposition rate in each sector was obtained by multiplying the concentration of the tritium, the area of the sector, and the deposition velocity. The deposition velocity of 4.94 × 10\(^4\) m/s was used for soil.
5. Dynamic compartment model

Most of the 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. FIG. 6 shows the compartments for the modeling of the tritium movement. The mass transfer between the compartments was expressed using the following mass conservation equations:

\[
\frac{dA_i}{dt} = -\sum_{j} k_{ij} A_i + \sum_{j} k_{ji} A_j - \lambda A_i + S_i
\]  

(4)

where

- \(A_i\) the amounts of tritium in the compartment-i (Bq);
- \(k_{ij}\) the transfer coefficient between compartment-i and compartment-j (y\(^{-1}\));
- \(\lambda\) the tritium decay constant (y\(^{-1}\));
- \(S_i\) the source term in the compartment-i (Bq/y).

The computer program, AMBER [10], was used for the numerical solution.
6. **Source term and transfer coefficients**

The geography of the Wolsong site was investigated using a digital map to derive the transfer coefficients. FIG. 7 shows the topography of the Wolsong site. The area around the sampling point marked by a cross was selected and represented by four compartments. Even though the surface stream was not a final sink of the tritium, the sea was not included since we were only interested in the concentration of the top-soil water.

The source term of the tritium was calculated from the amount of deposition onto soil. FIG. 8 shows the 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.

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. The transfer coefficients of the evaporation and runoff between the compartments were calculated by dividing the evaporation and runoff rates with 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 31.5 and 5.05 m/y, respectively. Groundwater flow was described by Darcy’s law. The finite element computer program, FEMWATER, was used to calculate the steady state groundwater flow. FIG. 9 shows the vertical cross section of the area denoted by line AB in FIG. 8. Total 180 elements were used to calculate the groundwater flow. FIG. 10 shows the groundwater velocity vector obtained using the FEMWATER program. The average groundwater velocity for the overburden was around 4.25 m/y.

Hydrodynamic dispersion could 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 by the following equation:
\[ D_k = \alpha_t \cdot v \]  

(5)

where

\[ \alpha_t \] the dispersivity (m);
\[ v \] groundwater flow velocity (m/y).

The dispersivity of 1 m was used conservatively in the calculation.

**FIG. 7.** Contour map of the Wolsong site. Cross mark indicates the region of interest.

**FIG. 8.** Source term used for the dynamic compartment modeling.
7. Results and discussion

The changes of the tritium activity were calculated with the time-dependent source term given in FIG. 8. FIG. 11 shows the tritium concentration in the top-soil and deep soil. Most of the tritium deposited onto the top-soil returned to the atmosphere due to a large transfer coefficient of the evaporation and was not accumulated in the top-soil. As shown in FIG. 11, the tritium concentration in the top-soil was estimated to around 4~5 Bq/L including natural background tritium concentration of 0.71 Bq/L in the top soil, which was estimated with the NCRP northern hemisphere model. The calculation showed a little lower tritium concentration 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.

The Tritium Removal Facility (TRF) is being constructed at the Wolsong site to reduce the discharge rates of the tritium. It is scheduled to start operation from 2006. The discharge rates are expected to be reduced by one sixth to one tenth after the operation of the Tritium Removal Facility. One scenario regarding the operation of the TRF was introduced to show the usefulness of the dynamic compartment model developed in this study. We calculated the effect of environmental change of the tritium concentration in the case that the discharge rates were reduced by one tenth of the normal operation of the nuclear power plants since 2006. FIG. 12 shows the results of the calculation. The results show that the tritium concentration in the top-soil starts to decrease from 2006, but the concentration in the deep soil was changed a little due to the low transfer coefficients.
8. Conclusions

The dynamic compartment model was applied to predict the tritium behavior in the environment around the Wolsong nuclear power plants. Due to the high tritium discharge rates of CANDU reactors the tritium concentration was high at the site. Tritium in the environment was thought to come from both the natural background and the nuclear power plants. 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 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 $\chi/Q$ values. The calculation showed a good agreement with the measurement data. The washout ratio of $4.85 \times 10^4$ 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 tritium movement in the environment. The evaporation and runoff were predicted using a computer program Visual HELP. Groundwater flow was analyzed with a finite element computer program, FEMWATER. Transfer coefficients between the compartments were calculated using the pore water volume and the surface area. Among them the transfer coefficient for evaporation was the largest.

Tritium concentration in the top soil 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 top-soil. Tritium concentration in the top-soil was estimated at around 4 ~ 5 Bq/L including the natural background tritium concentration of 0.71 Bq/L in the top soil. It was very close to the measurement of 6.7 Bq/L. One scenario regarding the operation of the Tritium Removal Facility was analyzed to show the usefulness of the dynamic compartment model developed in this study. The change of the tritium concentration was predicted in the case that the discharge rates of the tritium were reduced to one tenth from 2006. The result showed that the tritium concentration in the top-soil started to decrease from 2006, but the concentration in the deep soil was changed a little due to the low transfer coefficients.

Acknowledgements

This study was performed under the long-term nuclear research and development program sponsored by the Korea Ministry of Science and Technology.

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