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■ 論 文 ■

Washout of Tritiated Water Vapor by Precipitation in the Vicinity of Wolsong Nuclear Power Plant Site

C. K. Kim · S. K. Lee · B. H. Rho · G. J. Park* · W. Kim*
and H. D. Kang*

Korea Institute of Nuclear Safety
*Kyungpook National University

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Abstract - On a basis of the washout model and concentrations of anthropogenic tritium in rainwater around the Wolsong site, the washout coefficients of tritium by rainwater were calculated, and the validity of washout deposition model are estimated. As the result of that, the washout coefficients in 10 sampling stations around Wolsong site were in the range of 2.9×10^{-5} - 16×10^{-5} s^{-1} with the mean value of 7.21×10^{-5} s^{-1}. The validity of the washout deposition model was confirmed by comparing the observed data with the calculated ones.
Key words : tritium, washout coefficient, precipitation, Wolsong site

INTRODUCTION

Tritium, the heaviest and only radioactive isotope of hydrogen, has been a ubiquitous contaminant produced by atomic energy programs. Tritium was distributed worldwide by the nuclear weapons tests that took place in the open atmosphere, particularly during the period 1954 to 1962. Recently, tritium has diminished substantially as a result of the limited nuclear test ban agreement among the major powers, but tritium production by nuclear power reactors is increasing rapidly and will in time become the dominant source. UNSCEAR 2000 report[1] described that the average normalized release of gaseous tritium during the period 1990 to 1994 in HWRs(Heavy Water Reactor) was about three hundred times higher than that in PWRs. The majority of tritium, as a tritiated water vapor form (HTO), is released into the environment from nuclear power plant. HTO is the most commonly encountered form of tritium in the environment, and has a high potential for migration into the terrestrial environment[2]. Once emitted tritium takes the form of tritiated water vapor its properties are essentially the same as those of atmospheric water vapor. Thus tritium becomes a component of the hydrological cycle, and behaves essentially the same as water vapor in the hydrological cycle. The removal of tritium from the atmospheric occurs primarily through the precipitation process, although near the point of release some tritiated water vapor can be diffused to the ground and taken up directly by vegetation. Once deposited, the tritium can infiltrate into the ground water, remain as part of the surface runoff and storage, be assimilated into plants, or transpired and evaporated back into the atmosphere.

Since Wolsong Nuclear Power Plant 1(Wolsong NPP 1), which is a CANDU-type reactor (heavy water reactor), started its operation in 1983, Wolsong 2, 3 and 4 were added to that during the period 1997 to 1999. Tritiated water vapor from Wolsong nuclear

power plant is continuously released to the atmosphere and then scavenged by precipitation. So, the elevated tritium concentrations than the natural level are often observed both in water vapor and in rainwater in the vicinity of the Wolsong site. During rain, the tritiated water vapor(HTO) emitted from a stack of Wolsong nuclear power plant may be exchanged with H_2O in the raindrops, and then the rain will become tritiated. Therefore, rainwater taken downwind of a nuclear plant often contain tritium. Many studies[3-5] on the atmospheric dispersion of HTO vapor and tritium deposition by washout were made in the vicinities of tritium production facilities, reprocessing plants and heavy water reactors. In our previous study[6-7], the tritium levels in the environment around Wolsong nuclear power plant were introduced. The purpose of this work is to estimate the validity of the washout deposition model, comparing the measured value with the calculated one.

EXPERIMENTAL

Site description

Four units of CANDU-type reactor with a total gross capacity of 700 Mwe are under the normal operation in the Wolsong site where is located in Naa-ri, Yangnam-myon, Kyongsangbuk-do in the southeast of the Korean Peninsula bordered by the East Sea (Sea of Japan). The effluents are basically continuous releases, varying with time under normal operation, and discharged from a stack height of about 58 m.

Sampling

A collector of rainwater with a surface area of 0.1 m^2 was placed at a height of 1m above the ground surface. The samples were collected monthly during the period July to December 2000. The sampling stations were located within about 2 km of radius in five directions from Wolsong nuclear power plant as shown in Fig. 1. The determination of tritium was performed with a 100 ml aliquot of each rainwater sample. The atmospheric water vapor at ground level was collected monthly by using an air sampler consisting of a series of molecular sieve adsorption columns[8-9]. Tritium water was recovered from the molecular sieve column in the laboratory by heating at 450°C for 2h.

Sample preparation and tritium measurement

Rainwater samples were purified by single distillation to remove quenching materials and other radioactive materials. Each 20 ml aliquot of the distilled water samples was taken in a 20 ml vial containing 10 ml of a liquid scintillator(Ultima-Gold LLT, Packard Instrument Company). The tritium activity was then measured with a liquid scintillation counter(Quantulus 1220, Wallac Co. Ltd). The counting time was 500 min. The minimum detectable limit was about 0.9 Bq $liter^{-1}$.

RESULTS AND DISCUSSION

Washout coefficient

The washout coefficient was calculated from available data of tritium concentration in atmospheric water vapor and rainwater around Wolsong site during the period July 2000 to June 2001. The washout coefficient (Λ, in s^{-1}) is calculated by washout model (Eq. 1)[10].

$$\Lambda = \omega_0 / (x_0 \cdot H_{eff}) \quad (1)$$

Where H_{eff} (in m) is an effective height calculated from the widely used dispersion formula[10-12], ω_0 (in Bq m^{-2} s^{-1}) is the observed value of the tritium deposition rate, and x_0 (in Bq m^{-3}) is the atmospheric tritium concentration at ground level. The ground level tritium concentration (x_0) was determined from the tritium concentration in atmospheric water vapor. The used tritium concentrations both in rainwater and in water vapor were obtained by subtracting the background level of tritium resulting from cosmic rays and nuclear weapons tests from the data obtained near the

nuclear plants.

The deposition rate is given as

$$\omega_0 = (C \times I_R)/ 3600 \quad (2)$$

where C (in Bq l^{-1}) is the tritium concentration in rainwater, and I_R(in mm h^{-1}) is the mean rainfall intensity during the sampling period. The calculation of tritium washout coefficient in this paper is based on the monthly mean values assuming the continuous tritium release. Using equations 1 and 2 with the meteorological data, the tritium concentration in rainwater and the tritium concentration in atmospheric water vapor, the washout coefficients (Λ, in s^{-1}) at 10 sampling stations were calculated. As shown in Table 1, the washout coefficients around Wolsong site varied widely in the range of 2.9×10^{-5} - 16×10^{-5} s^{-1} with the mean value of $(7.21\pm2.64)\times10^{-5}$ s^{-1}.

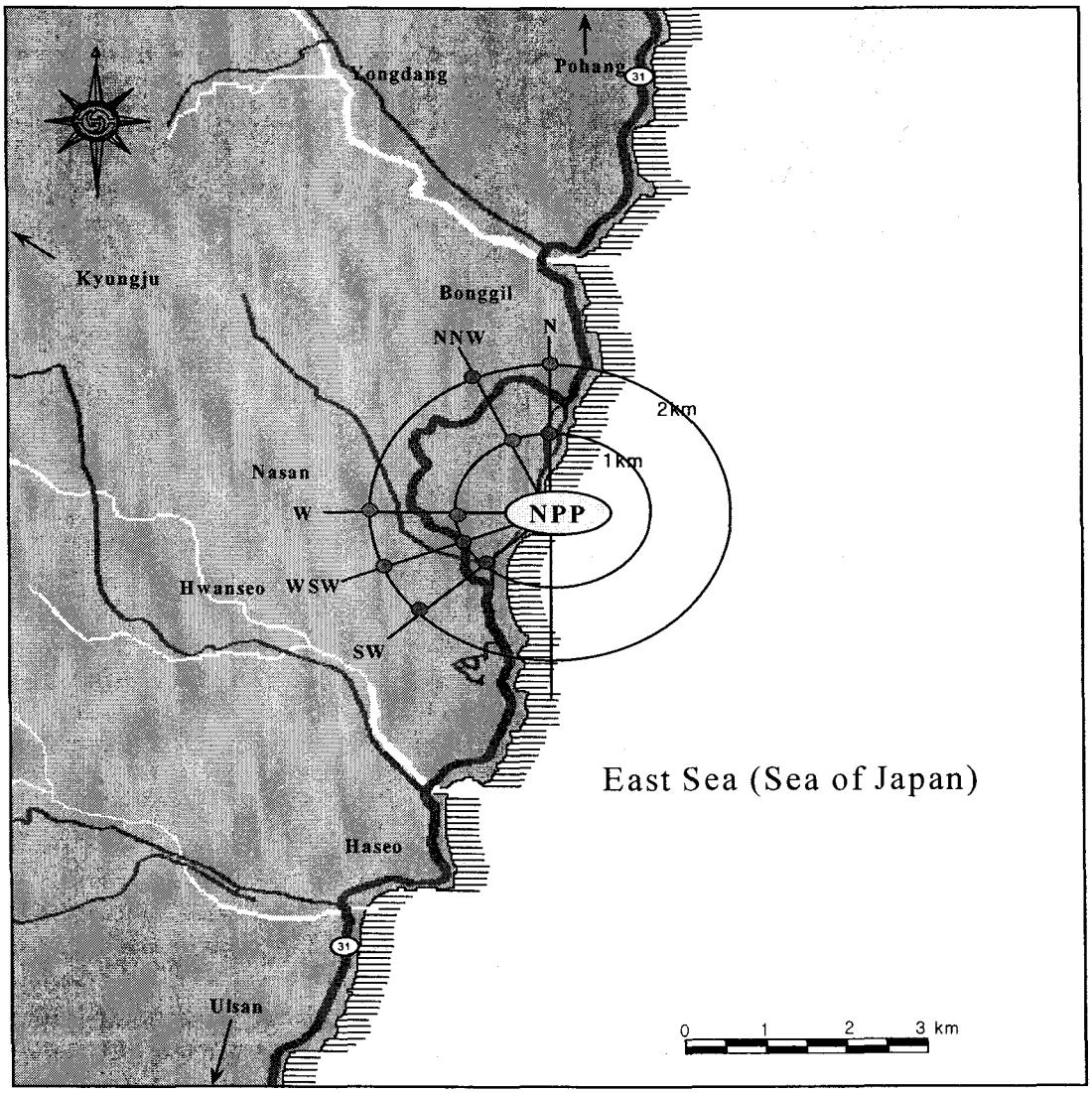

Fig. 1. Sampling stations of atmospheric water vapor and rainwater around Wolsong nuclear power plant

Table 1. The mean value of calculated washout coefficients for the Wolsong site during the period July 2000 to June 2001

Sampling stations	Atmospheric ^3H concentration (x_0; Bq m^{-3})	^3H concentration in rainwater (C; Bq L^{-1})	Effective Height (H_{eff} m)	Rainfall intensity (I_R; mm h^{-1})	Deposition rate (ω_o; Bq $m^{-2}s^{-1}$)	Washout coefficient (Λ; $x10^{-5}s^{-1}$)
N 1km	2.13	81	306	1.79	0.04	6.53
N 2km	0.735	43	335	1.73	0.025	7.53
NNW 1km	0.767	33.1	373	1.85	0.018	6.23
NNW 2km	0.264	9.79	326	1.78	0.006	6.46
W 1km	0.800	34.2	282	1.79	0.016	7.48
W 2km	0.496	15.9	343	1.86	0.008	6.92
WSW 1km	0.755	55.2	413	1.80	0.028	7.72
WSW 2km	0.437	24.9	342	1.74	0.011	7.08
SW 1km	0.937	79.4	733	1.69	0.037	8.31
SW 2km	0.576	22.1	371	1.78	0.012	7.50

Many studies[10-12] reported that meteorological parameters such as rainfall intensity and raindrop size may influence on the deposition by washout. Jacob[11] obtained a washout coefficient of 3.6×10^{-4} s^{-1} from the theoretical considerations by assuming an average rainfall intensity of 4 mm h^{-1}. Inoue et al.[13] reported that their washout model led to a washout coefficient of 4.6×10^{-4} s^{-1} at a rainfall intensity of 2 mm h^{-1}. Tokuyama and Oonishi[14] stated that the tritium washout coefficients in the Tsuruga and Mihama areas varied widely in the range of 1.3×10^{-5} 16×10^{-5} s^{-1} with the mean value of 7.3×10^{-5} s^{-1} at a rainfall intensity of 2 mm h^{-1}. This result is similar to the result obtained from the present work.

The validity of the model

To verity the usefulness of the washout model, the tritium concentrations in rainwater calculated with the mean value of washout coefficient, 7.21×10^{-5} s^{-1} deposition rates were compared with those observed at 1km of N and SW directions, and 2 km of W direction from Wolsong nuclear power plant during July to December 2001. The comparison results are given in Table 2. The ratios of the calculated values to the observed ones ranged from 0.42 to 1.98 with a mean value of 0.99±0.47. As shown in Fig. 2, hence, it is confirmed that the calculated tritium concentrations in rainwater are accord with the observed ones, even if there are a few exceptions.

Table 2. Observed and calculated monthly tritium concentrations in rainwater during July to December 2001

Sampling stations	Sampling period	Effective Height (m)	Rainfall intensity (mm h^{-1})	Observed ^3H concentration in rainwater (A) (Bq L^{-1})	Calculated ^3H concentration in rainwater (B) (Bq L^{-1})	A/B ratio
N 1KM	'01. 7.1~7.31	68.9	2.52	64.9±1.6	90.8	0.71
	8.1~8.31	71.0	1.71	21.0±1.3	29.3	0.72
	9.1~9.30	74.9	1.41	1.77±0.66	3.79	0.47
	10.1~10.31	160	2.62	9.48±0.88	8.26	1.15
	11.1~11.30	665	1.04	53.2±1.6	42.3	1.26
	12.1~12.29	950	2.65	12.8±1.0	10.7	1.20
SW 1KM	7.1~7.31	189	2.51	32.5±1.1	24.4	1.33
	8.1~8.31	274	1.74	40.7±1.4	45.4	0.90
	9.1~9.30291	291	1.38	88.5±1.8	129	0.69
	10.1~10.31	234	2.44	106±2	82.6	1.28
	11.1~11.30	871	1.07	123±2	62.3	1.97
	12.1~12.29	1383	2.59	33.4±1.3	40.2	0.83
W 2KM	7.1~7.31	120	2.56	15.6±0.8	13.1	1.19
	8.1~8.31	52.8	2.02	2.64±1.04	6.34	0.42
	9.1~9.30	156	1.48	17.0±1.0	23.9	0.71
	10.1~10.31	451	2.83	28.0±1.2	47.6	0.59
	11.1~11.30	117	1.16	15.0±1.0	7.59	1.98
	12.1~12.29	191	2.19	2.78±0.84	5.56	0.50

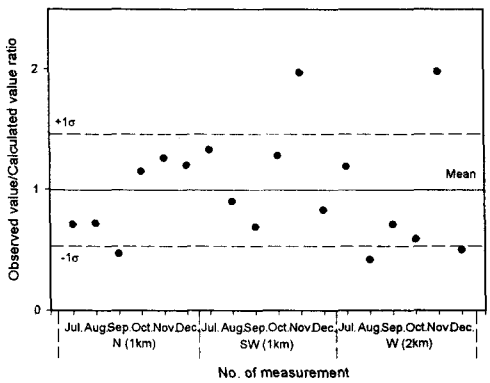

Fig. 2. Variation of ratios of observed values versus calculated ones by washout model

CONCLUSIONS

On a basis of the meteorological data, the tritium concentration in rainwater and the tritium concentration in atmospheric water vapor, which were monthly measured for one year, the washout coefficients in the areas around Wolsong nuclear power plant were calculated. The washout coefficients in 8 sampling stations around Wolsong site varied widely in the range of 2.9×10^{-5} - 16×10^{-5} s^{-1} with the mean value of $(7.21\pm2.64)\times10^{-5}$ s^{-1}. Under the conditions that HTO vapor is continuously released at a constant rate and that the estimated point is located near a

reactor site, the washout model is reasonable and the obtained washout coefficient is useful for estimating the deposition of tritiated water vapor by washout, although the validity of application of this washout model to an intermittent release or an accidental release should be more studied.

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