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Dispersion and removal characteristics of tritium originated from nuclear power plants in the atmosphere

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ABSTRACT

The activities of tritium in water-vapor (n = 649) and precipitation (n = 2404) samples were measured from 1998 to 2015 around the Wolsong nuclear power plant (NPP) site where four pressurized heavy water reactors and two pressurized water reactors operated. The activity concentrations of tritium in the water-vapor and precipitation samples were in the ranges of 2.2–2200 Bq/L and 0.3–1090 Bq/L, respectively. The concentrations of tritium in the water-vapor in spring were approximately 7 times higher than those in fall and winter, mainly owing to the wind directions at the power plant location. The annual geometric mean activities of tritium in the water-vapor and precipitation samples varied within 56% and 83% from the average, respectively, depending primarily on the annual discharge amount of tritium to the atmosphere. The activities of tritium in the water-vapor and precipitation samples rapidly decreased away from the power plant. Approximately 0.5–30% of tritium discharged from the NPP site was removed by precipitation to the ground within an area with a radius of 30 km from the NPP site, which linearly depended on the precipitation amount. Our results suggest that the wind direction and precipitation, in addition to the amount of discharge, are important factors that control the tritium concentrations in air near the NPP site.

1. Introduction

Tritium (^{3}H) is a radioactive isotope of hydrogen with a half-life of 12.32 years (Lucas and Unterweger, 2000) and is a low-energy pure-beta emitter with a maximum and average energy of 18.6 keV and 5.7 keV, respectively. Tritium in the environment is naturally produced by cosmic-ray spallation mainly in the upper troposphere and lower stratosphere (Craig and Lal, 1961; Galeriu and Melintescu, 2010; Grosse et al., 1951; Masarik and Beer, 2009; Rozanski et al., 1991). In addition, the level of tritium was enhanced artificially by atmospheric tests of nuclear weapons in the late 1950s and early 1960s. Since the Extended Test Ban Treaty in 1963, the tritium levels in the atmosphere have decreased, approaching the natural tritium level nowadays (Guetat et al., 2011). However, there are additional sources of artificial tritium from the routine operations of nuclear power plants (NPPs) (UNSCEAR, 1988). In general, the tritium production by heavy water reactors (HWRs) is tens of times higher than that by pressurized water reactors (PWRs). These nuclear facilities often become a significant source of tritium in the environment.

Tritium is released in the form of tritium gas (HT) and tritiated water (HTO) from a nuclear power plant (NPP), and most tritium in the environment exists in the HTO and OBT form. The HTO form of tritium is easily incorporated in the global hydrological cycle. Therefore tritium has been used as a tracer in various fields including ocean circulation and ventilation (England and Maier-Reimer, 2001; Fine et al., 1981; Jenkins and Smethie, 1996; Sarmiento, 1983), atmospheric and meteorological studies (Cauquoin et al., 2015; Moon et al., 1992; Rozanski et al., 1991; Yasunari and Yamazaki, 2009), and ground water movement (Santschi et al., 1987; Solomon et al., 1992; Von Buttlar and Wendt, 1958). Tritium that enters the hydrological cycle in the form of HTO can easily enter living organisms; it can enter the human body by ingestion and inhalation. In order to identify and counteract the radiological effect from tritium, many studies have been conducted around nuclear facilities (Fujita et al., 2007; Matsuura et al., 1995). In addition, several studies were performed on the food-chain transfer of tritium (Ciffroy et al., 2006), impact of NPPs on the river environment (Hanslík et al., 2009), verification of the washout model (Köllő et al., 2011; Kim et al., 2003; Piskunov et al., 2012; Tokuyama and Oonishi, 1997), transfer of tritium from water to tissue-free water tritium (TFWT) of organisms in the NPP cooling reservoir (Baeza et al., 2009), conversion of HTO in air to organically bound tritium (OBT) in plants in the vicinity of a nuclear facility (Vichot et al., 2008), and spatial and temporal

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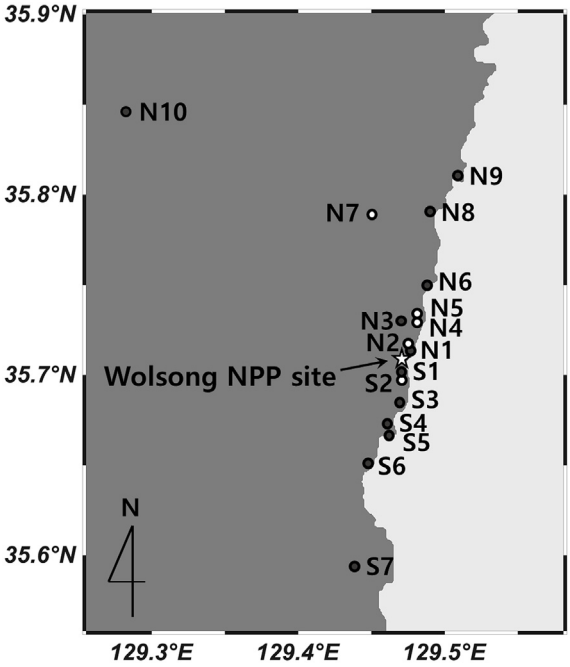

Fig. 1. Map of the sampling stations around the Wolsong NPP site. The star, open circles, and closed circles represent the Wolsong NPP site and sampling stations for both water vapor and precipitation, and precipitation, respectively.

distributions of tritium around nuclear facilities (Chae et al., 2011; Connan et al., 2017; Miljević et al., 2000).

Although various studies have been performed on the distributions of tritium in the atmosphere around nuclear facilities, the factors controlling the long-term spatio-temporal changes in tritium near nuclear facilities are poorly understood. Therefore, in this study, we evaluated (1) the dispersion characteristics and (2) removal proportions to the ground by precipitations for tritium released from the NPPs based on extensive monitoring data of tritium in water vapor and precipitation obtained in the vicinity of the Wolsong NPP site. This information may provide important information on the modelling of tritium behavior and radiation protection around NPPs in normal operation as well as emergency situations.

2. Materials and methods

2.1. Study area

The Wolsong NPP site is located on the southeastern coast of the Korean Peninsula. The site is surrounded by small mountains on the west side and borders on the East Sea (Japan Sea) on the east side. In the Wolsong NPP site, the Wolsong NPP Unit 1 began operation in April 1983, and the Wolsong NPP Units 2, 3 and 4, Shinwolsong NPP Units 1 and 2 have operated commercially since July 1997, July 1998, October 1999, July 2012, and July 2015 respectively. Among the six NPPs, the Wolsong NPP Units 1, 2, 3, and 4 are HWRs (CANDU-type reactors), while the Shinwolsong NPP Units 1 and 2 are PWRs. The annual tritium discharge amounts from the Wolsong NPP site ranged 125–403 TBq during 2001–2015. Although the amount of discharged tritium has been reduced by the operation of a tritium removal facility (TRF) since 2007, a considerable amount of tritium over 100 TBq per year has still been released into the environment.

Several studies have been performed around the Wolsong NPP site. When only the Wolsong NPP Unit 1 was operating, the TFWT and OBT activity concentrations were found to be affected by the released amount of tritium (Kim et al., 1998, 2000). The annual effective dose was estimated to be approximately 1.3 μSv/y in the vicinity of the site (within 1.6 km from the NPP) for environmental samples collected in 1992–1993 (Kim and Han, 1999). This dose level was significantly lower than the limit (1 mSv y^{-1}) of the effective dose to the general public recommended by the International Commission on Radiological Protection (ICRP) (ICRP, 2007). The distribution of tritium around this area has been reported (Chae et al., 2011; Kim et al., 1998).

2.2. Data acquisition

Basic data for tritium concentrations in water vapor and precipitation are available in the annual reports published by Korea Institute of Nuclear Safety (KINS) (KINS, 1998–2015). Atmospheric water-vapor tritium data are available for five sampling sites (N2, N4, N5, N7, and S2) from 1998 to 2015 (Fig. 1). The results for the monthly sample collection periods at each site are shown in Tables 1 and 2. Precipitation tritium data are available for 17 sampling sites (N1–N10, S1–S7), located within 0.6–22 km from the Wolsong NPP site (Fig. 1). Meteorological data are available from Korea Hydro &Nuclear Power (KHNP), which manages the meteorological station located in the Wolsong NPP site.

2.3. Analytical methods

We briefly describe the analytical method used for the monitoring. Atmospheric water samples were collected using an active air sampler, which includes a series of molecular sieve adsorption columns (ÖStlund and Mason, 1974). The sampler was installed approximately 1 m above the ground with a flow rate of 0.52–1 L/min. Precipitation samples were collected using a rainwater sampler with a surface area of 0.1 m^2. The sampler was positioned at a height of 1 m above the ground. HTO was extracted from the molecular sieve column (45 cm × 5 cm i.d.) by heating at 450 °C for 2 h in the laboratory. After distillation, 10 mL of the water sample was added to a 20-mL Perfluoroalkoxy (PFA) vial, together with 10 ml of Ultima gold LLT cocktail (Packard, USA); the mixture was then sufficiently mixed. The prepared sample was measured 10 times for 50 min by using a liquid scintillation counter (1220 Quantulus, PerkinElmer). The rainwater samples were distilled, and the method used for the tritium analysis in a water vapor sample was employed.

Table 1
Concentrations of tritium in the water vapor around the Wolsong NPP.

Station	Location		Range (Bq/L)	GM[a] (Bq/L)	SD[a] (Bq/L)	Period
	Cardinal direction	Distance (km)				
N2	N	0.9	68–2200	431	517	2003.4–2008.12
N4	NNE	1.8	37–1810	262	358	1998.1–2003.3
N5	NNE	2.2	14–798	120	142	1998.1–2015.12
N7	NNW	8.8	2.2–67	12	12	2008.5–2015.12
S2	SSW	1.7	18–645	75	77	1998.1–2015-12

[a] Geometric mean (GM), standard deviation (SD).