RETRIEVAL ESTIMATION OF THE GASEOUS ^{14}C DISCHARGE RATE USING ^{14}C AND ^{3}H CONCENTRATIONS IN PINE TREE RINGS IN THE VICINITY OF WOLSUNG NUCLEAR POWER PLANT

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ABSTRACT. In order to evaluate past levels of ^{3}H and radiocarbon in the environment near the Wolsung Nuclear Power Plant (NPP), Korea, pine trees were sampled at 3 locations within 1 km of the Wolsung site in 2002, and their concentrations of ^{3}H and ^{14}C were analyzed. The α-cellulose was extracted from pine tree rings using the common chlorite method with a subsequent treatment. The concentrations of ^{3}H and ^{14}C were measured by a liquid scintillation (LS) counter. Both organically bound tritium (OBT) and ^{14}C concentrations gradually increased after the commercial operation of the Wolsung NPP, reached a maximum, and then recently decreased. The OBT concentrations of pine tree rings ranged from 34.9 to 393 Bq/L. The OBT concentrations and the gaseous ^{3}H discharge rate were linearly related, and their linear dependence changed with sampling directions and improved by considering the effect of the meteorological conditions, i.e. wind patterns. The ^{14}C concentration of pine tree rings ranged from 0.272 to 1.70 Bq/g C. The concentrations of OBT and ^{14}C in pine tree rings were linearly related, and their linear dependence also changed with sampling directions, which might be explained by the different diffusion mechanism of ^{3}H and ^{14}C with distance from the stack. By using the linear dependence of concentrations of OBT and ^{14}C and the gaseous ^{3}H discharge rate, the gaseous ^{14}C discharge rate in the past could be estimated.

INTRODUCTION

The production of ^{3}H and radiocarbon in the environment originates naturally from cosmic rays and artificially from atmospheric bomb tests and nuclear reactors. Recently, the dominant sources of both radionuclides have been nuclear power reactors (UNSCEAR 1977; NCRP 1978, 1985). The release rates of ^{3}H and ^{14}C from heavy-water reactors are relatively higher than those of light-water reactors.

Since 1983, Wolsung Nuclear Power Plant 1 (Wolsung NPP 1), a CANDU-type pressurized heavy-water reactor (PHWR) with a total gross capacity of 678 MWe, has released small amounts of ^{3}H and ^{14}C during routine operation. Furthermore, Wolsung NPPs 2, 3, and 4—also CANDU-type PHWRs with a total gross capacity of 700 MWe each—have been operating commercially since 1997, 1998, and 1999, respectively. The gaseous ^{3}H discharge rate data of Wolsung NPPs is well documented. However, there are insufficient data on the gaseous ^{14}C discharge rate. For this reason, it is necessary to obtain a retrieval estimation of the gaseous ^{14}C discharge rate from Wolsung NPPs. In this study, the gaseous ^{14}C discharge rate in the past may be estimated using ^{14}C and organically bound tritium (OBT) concentrations of pine tree rings sampled in the vicinity of the Wolsung site.

MATERIALS AND METHODS

Sampling

To measure concentrations of OBT and ^{14}C in pine tree rings, 21-yr-old pine trees were sampled in 2002 at 3 locations within 1 km of the Wolsung NPP (Figure 1). The Wolsung site is located in Naari, Yangnam, Kyungpook, Korea, on the East Sea coast.

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LSC 2005, Advances in Liquid Scintillation Spectrometry
Edited by Stanisław Chałupnik, Franz Schönhofer, John Noakes. Pages 401–406.

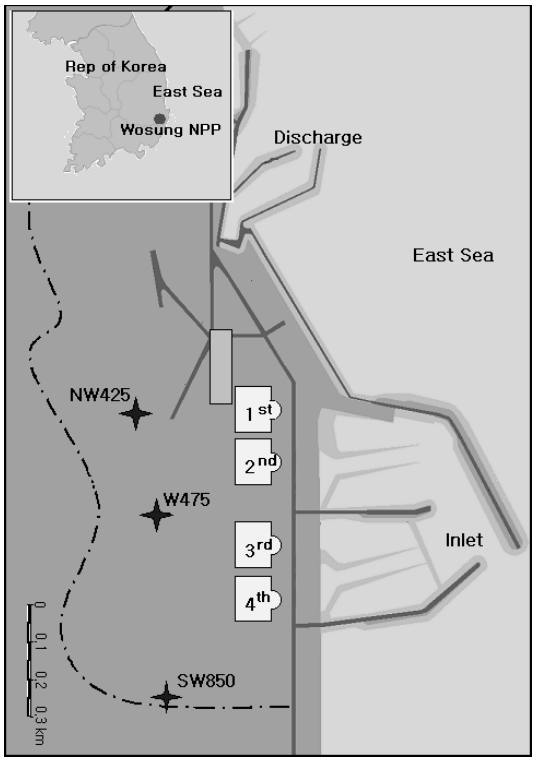

Figure 1 Sampling locations in the Wolsung site

Preparation and Measurement

Each annual growth pine tree ring was separated with a chisel, ground, and dried. Then, the α-cellulose from the tree rings was extracted using the common chlorite method with a subsequent treatment (Green 1963). The freeze-dried α-cellulose was burned in a combustion cup and cooled. Then, water in a combustion vessel was used to measure the OBT concentration. The remaining CO_2 was absorbed in NH_4OH and was precipitated as $CaCO_3$ with $CaCl_2$. The nitric acid was added in the precipitate, then the CO_2 generated was trapped in a liquid scintillator (LS) vial (Fuma et al. 1996; Kim et al. 2000). The activities of ^3H and ^{14}C in the sample were measured for 600 min (30 min × 20 repetitions) by liquid scintillation counting (LSC).

RESULTS AND DISCUSSION

Annual Variations of OBT and ^{14}C

The measured concentrations of OBT and ^{14}C in individual growth rings of pine trees sampled at 3 different locations are shown in Figure 2 and Figure 3. Both OBT and ^{14}C concentrations gradually increased after the commercial operation of Wolsung NPPs, reached a maximum, and then recently decreased. The OBT concentration of pine tree rings ranged from 34.9 to 393 Bq/L. The ^{14}C concentration of pine tree rings ranged from 0.272 to 1.70 Bq/g C.

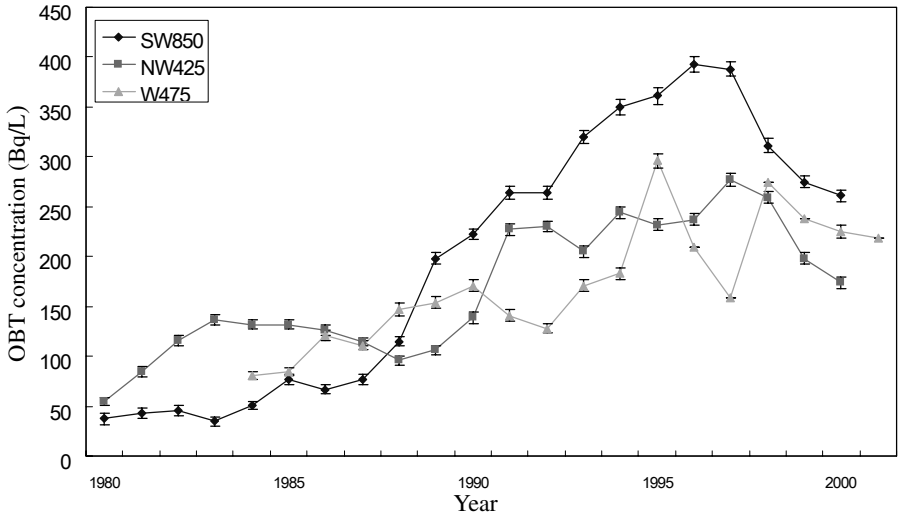

Figure 2 OBT concentrations of pine tree rings in 3 different sampling locations

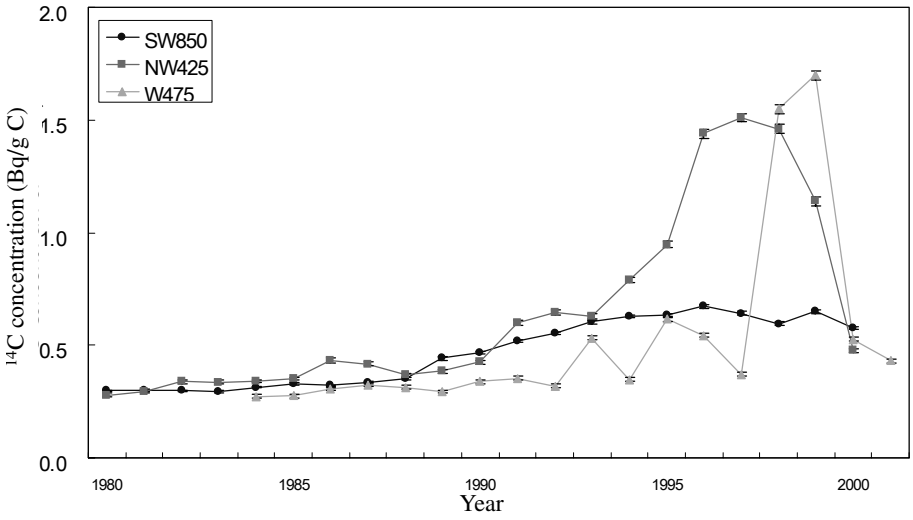

Figure 3 ^{14}C concentrations of pine tree rings in 3 different sampling locations

Relationship of OBT Concentration to the Gaseous ^{3}H Discharge Rate

The relationship of OBT concentration of pine tree rings and the gaseous ^{3}H discharge rate at the SW850 location is linearly related, with a correlation coefficient of $r = 0.76$ (as shown in Figure 4). The relationship of OBT concentration and the gaseous ^{3}H discharge rate at NW425 and W475 locations are also linearly related, but their correlation coefficients are $r = 0.74$ and $r = 0.44$, respectively. This means that the linear dependence of OBT concentration on the ^{3}H discharge rate changed with the sampling directions and may be improved by considering the meteorological conditions, i.e. wind patterns.

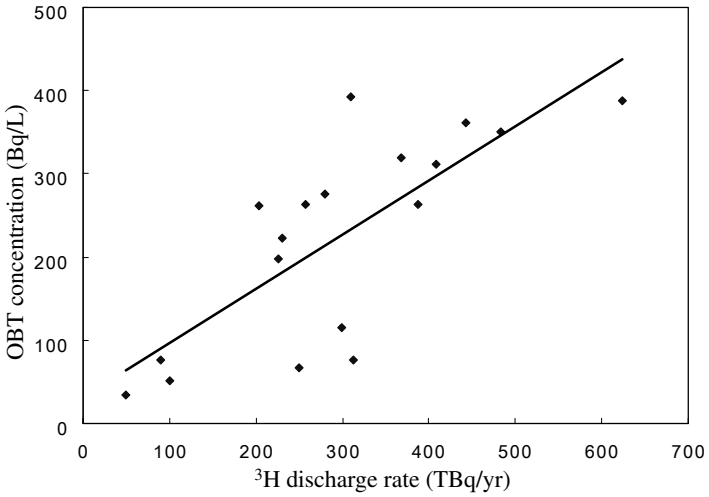

Figure 4 ^{3}H discharge rate and OBT concentration in pine tree rings sampled at the SW850 location.

Relationship of OBT and ^{14}C Concentrations

The relationship between OBT and ^{14}C concentrations in pine tree rings sampled at the SW850 location is linearly related, with a correlation coefficient of $r = 0.98$ (as shown in Figure 5). The relationships of concentration of OBT and ^{14}C sampled at the NW425 and W475 locations are also linearly related, but their correlation coefficients are $r = 0.84$ and $r = 0.66$, respectively. This means that the linear dependence of OBT and ^{14}C concentrations also changes with the sampling directions, which might be explained by the different diffusion mechanism of ^{3}H and ^{14}C with distance from the stack.

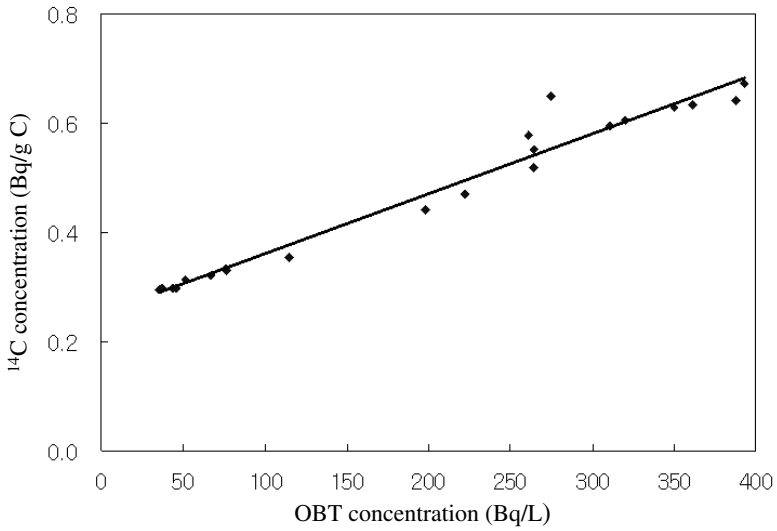

Figure 5 Correlation of OBT and ^{14}C concentrations in pine tree rings sampled at the SW850 location.

Retrieval Estimation of the Gaseous ^{14}C Discharge Rate

By using the linear dependence of OBT concentration on the gaseous ^{3}H discharge rate shown in Figure 4, and that of OBT and ^{14}C concentrations of pine tree rings shown in Figure 5, the gaseous ^{14}C discharge rate in the past could be estimated.

The ratio of the average annual OBT concentration of pine tree rings (C_{OBT}) and the average annual gaseous ^{3}H discharge rate from 1983 to 2000 (D_{H3}) is given by:

$$R_{OBT} = \frac{C_{OBT}}{D_{H3}} \tag{1}$$

The ratio of average annual ^{14}C concentration of pine tree rings (C_{C14}) and the average annual gaseous ^{14}C discharge rate (D_{C14}), which is available from 1998 to 2000, is given by:

$$R_{C14} = \frac{C_{C14}}{D_{C14}} \tag{2}$$

Therefore, the gaseous ^{14}C discharge can be calculated by:

$$D_{C14} = \frac{R_{OBT}}{R_{C14}} \times \frac{C_{C14}}{C_{OBT}} \times D_{H3} \tag{3}$$

Figure 6 shows the results of retrieval estimation of the gaseous ^{14}C discharge rate by using Equation 3. The estimated gaseous ^{14}C discharge rate ranged from 0.26 to 3.22 TBq/yr.

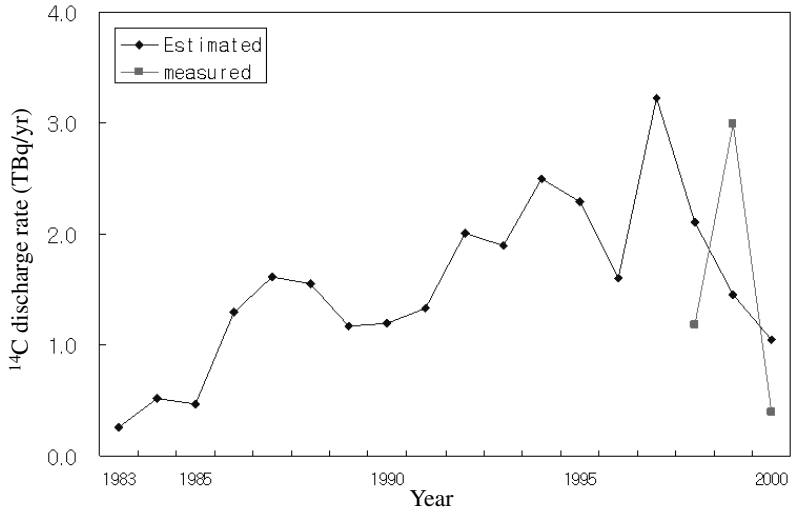

Figure 6 The estimated gaseous ^{14}C discharge rate of the Wolsung NPP

CONCLUSION

The OBT and ^{14}C concentrations of pine tree rings in the vicinity of the Wolsung NPP were measured. Both OBT and ^{14}C concentrations gradually increased after the commercial operation of the Wolsung NPP, reached a maximum, and then recently have decreased. The OBT concentration of

pine tree rings ranged from 34.9 to 393 Bq/L, while the ^{14}C concentration of pine tree rings ranged from 0.272 to 1.70 Bq/g C. The relationship of OBT concentration to the ^{3}H discharge rate depends on the sampling direction and the wind patterns. The relationship of OBT and ^{14}C concentrations also depends on sampling directions, which might be explained by the different diffusion mechanism of ^{3}H and ^{14}C with distance from the stack. By using the linear correlation of OBT and ^{14}C concentrations and the gaseous ^{3}H discharge rate, the gaseous ^{14}C discharge rates in the past were estimated, and ranged from 0.26 to 3.22 TBq/yr.

REFERENCES

Fuma S, Inoue Y. 1996. Measurement of ^{14}C in environmental samples by using CO_2 absorbent. *Radioisotopes* 45:1–7.

Green JW. 1963. Methods in carbohydrate chemistry. In: Whistler RL, editor. *Cellulose*. New York: Academic Press. p 9–20.

Kim CK, Lee SK, Rho BH, Lee YG. 2000. Environmental distribution and behavior of ^{3}H and ^{14}C around Wolsung nuclear power plants. *Health Physics* 78(6): 693–9.

National Council on Radiation Protection and Measurements [NRCP]. 1978. A handbook of radioactivity measurements procedures. Bethesda, Maryland, USA: NCRP Report No. 58.

National Council on Radiation Protection and Measurements [NRCP]. 1985. Carbon-14 in the environment. Bethesda, Maryland, USA: NCRP Report No. 81.

United Nations Scientific Committee on the Effects of Atomic Radiation [UNSCEAR]. 1977. Sources and effects of ionizing radiation. New York: United Nations, E77, IX 1. URL: http://www.unscear.org/unscear/en/publications/1977.html.