A Study on the Atmospheric Environment using Accelerator Mass Spectrometry: Effect of Neutron Rays on the Atmosphere Generated by Nuclear Power Plants at Different Altitudes

Je-Ho Jang [a], Jae-hyeon Park [a], Seung-won Lee [a], Sae-Hoon Park [a], Sang-Hun Lee [a], Yu-Seok Kim [a*]
[a] *Department of Nuclear & Energy System Engineering, Dongguk University, 123, Dongdae-ro, Gyeongju-si 38066*
[*] *E-mail: unison@donggu.ac.kr*

1. Introduction

At the core of a nuclear power plant, approximately 2.4 high-speed neutrons are generated per fission. These high-speed neutrons are decelerated by the moderator and repeat nuclear fission. During this process, large quantities of neutrons leak from the reactor. Although these neutrons do not adversely affect life or the atmosphere of external life, they react with nitrogen in Earth's atmosphere to produce C-14. C-14 is also produced from various other reactions. C-14 reacts quickly with atmospheric oxygen to become ${}^{14}_{6}CO_2$. Then, ${}^{12}_{6}CO_2$, which is the most abundant carbon isotope in the atmosphere, becomes evenly distributed [1]. Further, the atmosphere surrounding a nuclear power plant in operation has a particularly higher ${}^{14}_{6}CO_2$ concentration than in areas that are sufficiently far from or without nuclear power plants. Carbon dioxide in the atmosphere is absorbed by plants, and C-14 accumulates through growth activities. By using this, if parts of the plants growing in a specific area are collected and analyzed using accelerator mass spectrometry (AMS), the atmospheric effects of neutron rays can be examined.

While investigating data on atmospheric conditions related to C-14, we speculated if the effects of neutron rays on the atmosphere would differ with altitude [2]. Under these conditions, there are two practical variables. At lower altitudes, the concentration of ${}^{12}_{6}CO_2$ is greater because of the use of fossil fuels for human consumption, while at higher altitudes, the amount of ${}^{12}_{6}CO_2$ is varied, as it is affected by cosmic rays. First, due to the use of fossil fuels, the global concentration of C-14 is decreasing. However, some areas show much lower concentrations of C-14 due to high levels of carbon dioxide emissions, such as areas with heavy traffic and industrial complexes. This situation is called the 'regional Suess effect' [3]. Second, regarding cosmic rays, the majority of C-14 ($16{,}400 \sim 18{,}800\ \text{atoms}/s * m^2$) is produced approximately in 9-15 km above the sky and at high-altitude regions (about 9~15 kilometers), so will not significantly impact the quantity of ${}^{12}_{6}CO_2$.

Previous studies collected samples from the air or from the growth rings, branches, and leaves of trees. We chose to sample leaves because atmospheric composition data from recent years could not be acquired. In addition, the collection and pretreatment of leaves are more simple than those of samples from any other source. Further, one power plant was selected from among the four nuclear power plants located in Korea.

Then, areas located at equal distances from the power plant, with no obstructive terrain, were selected, and the samples were collected at different altitudes. After measuring and comparing the concentration of C-14 in the atmosphere using the leaf samples, the extent of the effects on the atmosphere could be determined for each altitude.

2. Methods and results

In this section, selection of the samples and some techniques used to examine the samples are explained. Additionally, the chemical pretreatment and sample graphitization are described.

2.1 Selection of tree species and locations

By referring to the distribution of trees in Gyeong-ju, a tree that exists in all the selected locations in this study was chosen. This is because each tree species has different abilities to absorb and store carbon. For this reason, it is important to find the same species that exists in all the places [4]. The maple tree was selected after exploration and various investigations.

Samples were collected on July 9, 2020 from four locations, based on the Wolsong Nuclear Power Plant (Wolsong NPP) in Gyeong-ju (35°42'21″N, 129°28'14″E). At this time, the operation status of the Wolsong NPP had been checked by the Korea Hydro & Nuclear Power Co., and the entire reactor was in operation. The sampling location near the plant was determined as Mt. Toham. Mt. Toham (35°80'N, 129°34'E) has a maximum altitude of 729 m and is approximately 15.3 km from the Wolsong NPP. The samples were collected at altitudes of 350m (35°47'22″N, 129°20'27″E) and 700 m (35°47'45″N, 129°20'51″E) from Mt. Toham. Yongdong-ri, Yangbuk-myeon (35°50'05″N, 129°26'29″E), was selected as a non-mountainous region, and was used as the ground reference for comparison. The altitude and coordinates were measured using a GPS.

2.2 Examining and storage of samples

The precaution that should be taken during sampling of the leaves is to avoid any contact with carbon-containing substances. After contact, the sample comprised infinitesimal carbon compounds from the carbon-containing substances, which resulted in a large error during the experiment.

Therefore, disposable plastic gloves should be used to collect leaves, which should then be wrapped in aluminum foil to avoid contact with the atmosphere and stored in a cool place.

2.3 Chemical pretreatment

The four collected leaves were approximately 8 cm × 6 cm in size, and each specimen was cut to approximately 1 cm × 1 cm using a knife, scissors, and tweezers. The tools were cleaned with ethanol and methanol after each specimen was subdivided. Afterward, the samples were washed with distilled water and washed again following the acid-alkali-acid (AAA) method [5]. AAA processing comprises three stages. First, 10 mL of 1.0 mol HCl was injected into the washed samples using a separate pipette. Each sample was heated to 80 °C by shaking for 30 min using a shaking water bath. Then, the samples were cleaned with distilled water to realize neutral pH (pH 6 to 7). The acidity was confirmed using litmus paper. Second, 10 mL of 0.5 mol NaOH was injected and heated in a shaking water bath in the same way. Then, neutral pH was achieved using distilled water. Finally, 10 mL of 1.0 mol HCl was injected and heated in a shaking water bath for 15 min. Then, the samples were neutralized in the same way, and placed in a dryer for one day to fully dry.

Table I: Information of samples.

Sample number	Sampling region	Tree species
JH0001	Yongdong-ri	Maple tree
JH0002	Toham mountain (350 m)	Maple tree
JH0003	Toham mountain (700 m)	Maple tree
JH0004	Wolsong nuclear power plant	Maple tree

The completely dried samples were subdivided to 2.15 mg, based on the amount of carbon contained in the samples. To verify the measurement accuracy, five standard samples (Wood, Ox-II, C7, C8, and phthalic acid) were also investigated. Each specimen was burned in an elemental analyzer, and the resulting carbon dioxide was heated with an iron catalyst (r-ferrite) and hydrogen using a reduction system.

2.4. Results and discussion

Table II: Eigen values of standard samples.

Sample	$\delta^{13}C$ (‰)	$F^{14}C$	C^{14}/C^{12} (10^{-12})
Phthalic acid	-25	0	0
IAEA C7	-14.48 (±0.21)	0.4953 (±0.0012)	0.5895
IAEA C8	-18.31 (±0.11)	0.1503 (±0.0017)	0.1755
Ox-II	-17.8	1.3408	1.5850

Table III: Measured values of collected samples.

Sample	$\delta^{13}C$ (‰)	$F^{14}C$	C^{14}/C^{12} (10^{-12})
JH0001	-24.3	1.0507	1.1798
JH0002	-28.6	0.9969	1.1103
JH0003	-28.7	1.0178	1.1330
JH0004	-22.0	3.7640	4.2193

Table IV: Measured values of standard samples.

Sample	$\delta^{13}C$ (‰)	$F^{14}C$	C^{14}/C^{12} (10^{-12})
Phthalic acid	-23.1	0.0081	0.0019
IAEA C7	-6.9	0.4955	0.5816
IAEA C8	-13.0	0.1493	0.1803
Ox-II	-15.4	1.3454	1.5353
Ox-II	-13.2	1.3408	1.5368

The samples that were pretreated were used as the targets and measured using AMS. The AMS facility at Dongguk University was used in this experiment (Ionplus's MICADAS 200keV model). The results are displayed in Tables III and IV. When comparing Tables II and IV, there is no large error for all standard samples. This confirms that the experiment was successful and that the resulting values are relatively reliable.

While the average amount of C-14/C-12 value in the general atmosphere is $1.0 \sim 1.2 \times 10^{-12}$, sample JH0004 obtained from the nuclear power plant indicates that the value of C-14/C-12 in the general atmosphere is very large. In addition, the C-14/C-12 values of JH0002 and JH0003 are slightly smaller; thus, the lower the altitude, the greater the regional Suess effect. However, the C-14/C-12 value of JH0001 was relatively large, which may be due to topographical characteristics. In the case of Mt. Toham, although the distance from the nuclear power plant to the center is the same, various natural features exist between the mountain and the power plant. Meanwhile, in the case of Yeongdong-ri, the terrain is almost flat. Since June 2020, 56,571 climbers have visited Mt. Toham, while Yongdong-ri is inhabited by approximately 100 residents, and is a very small floating population. For this reason, it was concluded that the regional Suess effect was not as effective here, and the C-14/C-12 value of JH0001 was large. Finally, to achieve the objective of this experiment, significant values will only be obtained if the study is conducted

using samples from locations closer to the plant.

3. Conclusions

From this study, we carried out differences in the effects of neutron rays leaked from nuclear power plants if different altitudes.

In order to reduce the error of the result value, the source of contamination was avoided as much as possible during the collection, storage of tree species, and pretreatment process. Based on the results of leaf samples and standard samples, the standard samples were not much different from the actual value, indicating that the experiment had a relatively small error.

When comparing the C-14/C-12 ratio of samples taken at each altitude except Nuclear Power Plant, they were large in the order of Yongdong-ri, 700m and 350m. We could determine the distribution of radioactive carbon in the measurement areas as well as the regional Suess effect. However, the distance from the nuclear power plant was quite large which resulted in a negligible neutron radiation, and the effects of other altitudes were unknown. To achieve a definite result, a study that determines the optimum distance and geography should follow.

REFERENCES

[1] W. Hong, N. B. Kim, and H. J. Woo, Description: Radioactive carbon dating, Analytical Science & Technology, Vol. 3, pp. 281-298, 1990.
[2] C. B. Mun, Accelerator Mass Spectrometry in Environmental Research, Korea Society for Atmospheric Environment, Vol. 16, No. 4, pp. 421-427, 2000.
[3] K. S. Sung, C. H. Eum, W. Hong, J. H. Park, and J. Y. Park, Distribution of CO_2 produced from fossil fuel by accelerator mass spectrometry: in Daejeon, Analytical Science & Technology, Vol. 21, pp. 9-13, 2008.
[4] H. K. Jo and T. W. Ahn, Carbon Storage and Uptake by Deciduous Tree Species for Urban Landscape, Journal of the Korean Institute of Landscape Architecture, Vol. 40, pp. 160-168, 2012.
[5] W. Hong, J. H. Park, H. J. Woo, J. G. Kim, H. U Choei, G. J. Kim, J. Y. Park, and G. H. Lee, A Project for Installation of Accelerator Mass Spectrometry, Korea Institute of Geoscience and Mineral Resources, 2008.