Determination of the Deposition of Cosmogenic ^{7}Be in Daejeon, Korea during Spring Season

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1. Introduction

Cosmogenic radionuclide, ^{7}Be ($T_{1/2}$=53.3 days) is produced by spallation reactions of light elements (^{14}N, ^{16}O, and ^{12}C) with cosmic radiations in the stratosphere (67%) and troposphere (33%) [1]. Cosmogenic radionuclides are absorbed onto fine particulate matter (PM) as carriers and transported to the earth's surface by downward wind. These particles are cleared out in the lower atmosphere through dry/wet deposition. The amount of ^{7}Be production depends on the intensity of cosmic-ray flux and the number of sunspot. The concentration of ^{7}Be in PM also depends on the residence time, atmosphere distribution (air exchange between troposphere and stratosphere), vertical and horizontal movement of air mass [2]. Several atmospheric models were conducted to identify air mass transportation path and rate of fallouts, but discrepancies were found between models and observations results. In recent years, berylliosis (ex. incurable lung disease) has been increased due to beryllium exposures. Thus, it is necessary to identify the cosmogenic ^{7}Be contribution in atomosphric aerosols. In addition, ^{7}Be has short half life and used as a short term sediment tracer for sediment erosion rates, mobilization, transporatation and storage.

The concentration of PM in the atmosphere is affected by natural pollution including mineral dust which originated from deserts. Cosmogenic radionuclides concentration increases with carrier / PM concentrations. North-East Asian countries are highly affected by Asian dust during winter and spring seasons. South Korea is relatively affected because the country is located on a downstream path of the air mass, and is close to their source. During the spring season, particularly dust storm periods, a high amount of Asian dust is depositing in Korea (0.2 Tg), which is traveling through the middle and lower troposphere regions. A high amount of ^{7}Be is contained in the troposphere due to air mass exchange between stratosphere and troposphere during the spring season, which will be attached to the PMs [2]. In previous studies in Daejeon, ^{7}Be activity in fine particles (PM_{10}) was found ranged 0.05–0.3 Bq/g in the summer to winter period (July - November), and in winter and spring season (December - May) 3–7 times higher. The variation of ^{7}Be activity in seasons due to wind direction and precipitation [3]. Inhalation of high amount of radionuclides is causes the considerable health risks. Thus, It is necessary to determine and monitor the natural radionuclides in PMs.

North-East Asian countries are highly affected by Asian dust, particularly in early spring season, which is transporting from the China region through the lower region of the troposphere. In the present study, an intense dust fallout period (2–4 days) during the spring season (2017–2019) was selected for the measurement of ^{7}Be activity in precipitation. High efficiency gamma ray spectrometry and absolute efficiencies of volume samples were employed for high precision results. Back trajectory models run to determined the approximate residence time of the particles.

2. Experimental

Daejeon, a metropolitan city, has a population of more than 1.5 million and is located in the middle of South Korea. The city is downstream along the major Asian dust transportation pathway. The sampling site is located at 36.3853 N, 127.3683 E and is about 15 m above ground at the top of a building to minimize the local dust contribution. The dust that settled on the polyethylene (PE) film (~ 3.0 m^{2}) was collected using a vacuum cleaner (Oneida cyclone dust collector) with a clean container. Fresh PE film was used during every collection event. The dust samples were collected for three consecutive years 2017–2019 and total of 10 samples were selected for the present study. A period of high dust deposition (dust storm) occurred on May 6–8, 2017, and the deposition rate reached 335 µg/m^{3}. The 3 to 10 g sample was collected in each selected period and sealed in the PE bottle.

The activity of ^{7}Be was measured using a high purity germanium detector with a relative efficiency of 54 % at 10 cm distance and relatively low background shielding setup. A passive gamma ray spectrum of dust particles was counted as shown in Fig. 1.

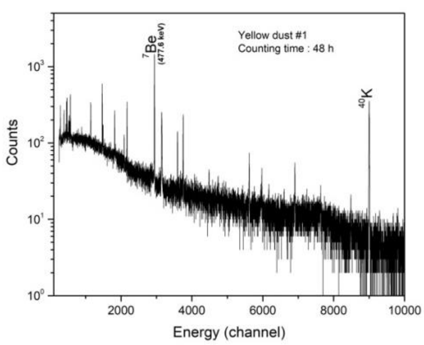

Fig.1. Passive gamma-ray spectrum of PMs

Absolute detection efficiency of the detector was calibrated through EFFTRAN code (Efficiency transfer and coincidence summing corrections for environmental gamma-ray spectrometry) with the help of multi-radionuclide point source certified by Korea Research Institute of Standards and Science, Korea. The intensity of 477.6 keV gamma line used to calculate the ^{7}Be activity in dust samples using the following equation.

$$A(Bq/kg) = \frac{C}{\gamma_a m \varepsilon} \qquad (1)$$

A=Activity of analyte (Bq/kg); C=net peak area of 477.6 keV; γ_a=Gamma ray emission probability; m=Mass of the sample (kg); ε= Full energy peak efficiency of detector

3. Results and Discussion

Full energy peak efficiency of cylindrical shape with different weights of few samples (1 g, 1.3 g and 4.42 g) was derived from the EFFTRAN code shown in Fig. 2. Second-order exponential decay fit was used to make a smooth efficiency curve at the energy range from 121 to 1810 keV. A multi nuclide point source contains ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, and ^{60}Co. EFFTRAN code considers the mass attenuation factor and coincidence factors, and that was validated by analyzing details of code and comparing it to other similar software[4]. The absolute efficiency of various dimensions of cylindrical samples is shown in Fig.2.

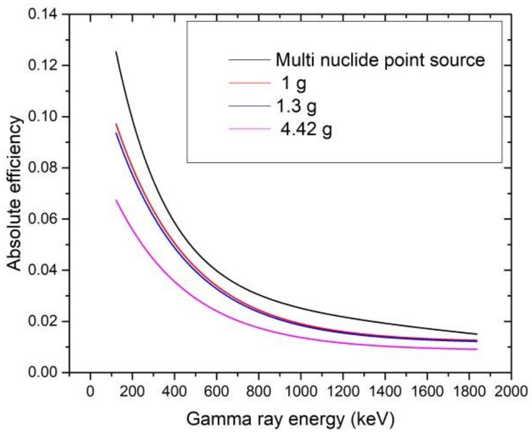

Fig. 2. Absolute efficiency of different volume samples using the EFFTRAN code and reference point source.

The approximate residence time (in the troposphere) of dust particles was determined using the HYSPLIT model [5]. The residence time of the particles is 1 to 7 days and traveled approximately 2000 km from source to the destination. The total activity of ^{7}Be in collected samples was calculated using Equation (1). The details of ^{7}Be activity, residence time and precipitation per day (one sample per year) are presented in Table 1. Standard deviation of triplicate measurement given as uncertainty. A high amount of ^{7}Be was reached to the Earth during the dust storm period (Sample No. 1 is collected during the dust storm period in 2017) along with Asian dust which traveled through the troposphere. The other two samples are relatively low precipitation with high residence time than sample No.1. The activity of ^{7}Be is proportional to residence time and precipitation amount of dust particles. The ^{7}Be activity in dust samples is influenced by latitude and direction of travel path.

Table 1. ^{7}Be activity in total suspended particles of spring season

Sample No.	^{7}Be activity (Bq/g) / day	Residence time (days)	Precipitation (g) / day
1	0.996 ± 0.039	1.5	8.86
2	0.084 ± 0.005	2.6	2.87
3	0.112 ± 0.019	6.5	1.23

Additional factors affect on ^{7}Be activity in the winter and spring seasons [2],

i. A high amount of dust transporting through troposphere due to northwesterly wind
ii. The high rate of air mass exchange between stratospheric and troposphere in late winter and early spring seasons
iii. Local metrological conditions (humidity and temperature).

4. Conclusion

In the present study, ^{7}Be activity in particulate matter was determined using the EFFTRAN code for the absolute efficiency of large volume samples. A high amount of cosmogenic radionuclide is reaching to the Earth's surface in the early spring season due to a high amount of Asian dust (as a carrier) transporting to Korea, which was traveling through the troposphere region. The activity of ^{7}Be is proportional to residence time in the troposphere and amount of precipitation.

REFERENCES

[1] J. Masarik, J. Beer, Simulation of particle fluxes and cosmogenic nuclide production in the Earth's atmosphere, Journal of Geophysical Research, 104, 1999, 12099-12106.
[2] H.W. Feely, R.J. Larsen, C.G. Sanderson, Factors that cause seasonal variations in Beryllium-7 concentrations in surface air. Journal of Environmental Radioactivity 9, 1989, 223-249.
[3] Y. Y. Yoon et. al., Seasonal variation of 7Be and 3H in Korean ambient air and rain, Journal of Radioanalytical and Nuclear Chemistry 307, 2016, 1629-1633.
[4] T. Vidmar, Testing efficiency transfer codes for equivalence, Applied Radiation and Isotopes 68, 2010, 355-359.
[5] L.Y. Lee, et.al., Analyses of airborne ^{7}Be concentrations in Hong Kong using back-trajectories, Atmospheric Environment 38, 2004, 7033–7040.