

Seasonal variation of ^7Be and ^3H in Korean ambient air and rain

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Abstract The seasonal variation of the ^7Be activities in air was monitored by a continuous sampling with a high volume air dust sampler. And also, ^7Be and ^3H activities in precipitation were determined. The activity level of ^7Be in air was ranged from 1.94 to 47.2 Bq/m³. And ^7Be in the precipitation was separated using cation exchange resin and the monthly average activity level was ranged from 0.29 to 4.77 Bq/L. ^3H was analyzed using electrolytic enrichment method and activity ranged from 0.27 to 2.22 Bq/L.

Keywords Seasonal variation · ^7Be · ^3H · Precipitation · Air

Introduction

Cosmogenic radionuclide such as ^7Be and ^3H produced in the stratosphere and troposphere as a result of cosmic ray spallation of nitrogen and oxygen. ^7Be has a relatively short half-life ($T_{1/2} = 53.3$ days) and emits a gamma-ray of 477.6 keV and ^3H is a pure beta emitter ($E_{\text{max}} = 18.6$ keV) with the half-life of 12.43 years. Shortly after production, ^7Be become attached to aerosols and are transported through the atmosphere and ultimately, via precipitation, to the Earth's surface, where they enter soils, marine sediments and groundwater [1–3]. Tritium enters the hydrological cycle in the form of tritiated water molecules (HTO).

Aerosols ^7Be may subsequently enter the marine as well as the terrestrial environment. It is deposited onto the ground through dry and wet deposition processes and is easily found in soil and plants [4]. Since the deposition rate depends on the local climate conditions, seasonal and inter-annual site-specific data are essential for a realistic estimation of ^7Be in environmental samples. The environmental concentration of ^7Be in the temperate zone is about 3 mBq/m³ in surface air and 700 Bq/m³ in rainwater [5]. ^7Be deposition is strongly dependent on the location of the sample collection, particularly regarding the latitude and the local climate [6–9] and its value in air and land surface may depend on the precipitation. However, Fitzgerald et al. [10] discovered that the activities of ^7Be did not correlate directly with rainfall. Buraeva [11] and Doering [12] reported that the ^7Be content in atmospheric aerosols near the ground in Russia has a seasonal variation of spring-summer maximum and an autumn–winter minimum. However, in East Asia, higher concentrations of ^7Be have been observed in winter [13, 14]. There have been few discussions about the temporal variation of ^7Be activities in air in East Asia, while much data on ^7Be deposition on the earth surface have been reported. The ^3H concentration in precipitation varies with the season and meteorological characteristics at the observation station [15]. The isotopic composition of local precipitation is primarily controlled by regional-scale processes, like the trajectories of water vapor transport over the continents and the average rainout history of the air masses giving rise to precipitation in a particular place [16].

Tritium content of water sample is expressed as tritium unit (TU), where one TU represents one THO molecule in 10¹⁸ H₂O molecules and this means 0.11919 ± 0.00021 Bq/kg of water. Tritium activity measurement in environment is very difficult due to its very low concentration such as 10–20 TU at the Northern Hemisphere and below 10 TU at the Southern

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Hemisphere. To overcome this difficulty, the most of the water samples were enriched using electrolytic enrichment method.

In this study, we observed the seasonal variation of the ^7Be activities in air by a continuous sampling with a high volume air dust sampler and also ^7Be and ^3H activities in precipitation were measured.

Experimental

Sampling

Precipitation and aerosol sample were collected at Daejeon (36.32°N, 127.41°E), Korea. Precipitation was collected for a month using a 20 L plastic bottle fitted with a conical stainless steel funnel. The bottle was wrapped in Al foil to keep the sample out of direct light and paraffin oil was poured into the bottle to avoid evaporation. Aerosol sample was collected using high volume sampler (Thermo GL2000H, USA) with 1.1 m³/min flow rate for 24 h.

Reagents and instruments

For the analysis of ^7Be in precipitation, filtered precipitation was acidified by HCl about pH 2 and passed through cation exchange resin (Dowex 50X 8). ^7Be was separated using 150 mL 4 N HCl solution and activity was counted by ultra low level HPGe detector (relative efficiency; 100 %, FWHM; 2.4 keV at 1.3 MeV, Canberra, USA). And also, aerosol sample was collected glass fiber filter and total air particulate samples were sampled onto 1.0 μm glass fiber filters of 210 mm \times 297 mm size (Gelman Co., USA) continuously with a high volume sampler. The ^7Be activity of the glass fiber filters collected sample was determined by gamma-spectrometry with HPGe detector. The counting rates of the sample were corrected for the relative decay rates of the gamma radiation versus total decay processes and the blank filter subtraction from the gamma-spectra. The detection limit at a 95 % confidence level was 0.01 Bq at a counting time of 200,000 s

Precipitation was filtered with 0.45 μm nuclear pore filter and transferred to a polyethylene bottle. 1 L sample was distilled to remove impurities and 1 w% GR grade Na_2O_2 (97 %, Sigma-Aldrich) was added as an electrolyte. After electrolytic enrichment process which was discussed our previous work [17], the distilled 10 mL water sample was mixed with a 10 mL cocktail (Ultima Gold LLT, Perkin Elmer Co, USA) which was a mixture of scintillator, solvent and surfactant. The tritium measurement was performed with a Quantulus 1220 (Wallac, Perkin Elmer) low level liquid scintillation counter. Tritium counting efficiency was estimated using the NIST standard water sample (SRM 4926E, tritiated water). With a 10 mL sample and 600 min of counting time, the detection limit was determined 2.17 Bq/L, which corresponds to about 18 TU. Therefore, most of the water samples could not be analyzed directly without enrichment. The tritium free background water was acquired from a 500 m deep groundwater well located in the Korea Atomic Energy Research Institute. The age of the background water sample was determined to be about 2500 year by the ^{14}C counting method. The counting vial was a 20 mL polyethylene vial with its inner side coated with Teflon.

Results and discussion

The ^7Be results from Daejeon Korea monthly air filter and rainwater samples ranges from 1.68 to 47.2 Bq/m³ and precipitation ranges from 0.29 to 4.77 Bq/L. These results are shown in Table 1, Fig. 1 and Table 2, Fig. 2. The results showed ^7Be activity of the aerosol was increased winter and this result was correlated with some other research conducted in East Asia [7, 13, 14]. The wind direction of winter season in Korea was subject to cold and dry continental winds, which originated from the northern part of Asian continent and Siberia. This cold air mass with a high content of ^7Be from Siberia gives rise to the ambient ^7Be levels of South Korea. Lee [13] has also found the high ^7Be activity at Hong Kong in winter. This is due to the

Table 1 ^7Be activity of air dust samples

Sampling	Aerosol amount ($\mu\text{g}/\text{m}^3$)	^7Be activity (Bq/m^3)	^7Be activity (Bq/mg)
2014-06-30	34.25	1.94 ± 0.72	56.6 ± 7.5
2014-07-16	21.68	2.48 ± 0.64	114.4 ± 10.7
2014-07-30	23.69	1.68 ± 0.77	70.9 ± 8.4
2014-08-15	36.63	10.87 ± 0.30	296.8 ± 17.2
2014-08-28	19.02	4.55 ± 0.47	239.2 ± 5.4
2014-09-10	30.61	17.30 ± 0.24	565.2 ± 23.8
2014-09-28	55.75	29.69 ± 0.18	532.6 ± 23.1
2014-10-30	53.77	47.18 ± 0.15	877.4 ± 29.6
2014-11-27	50.29	39.48 ± 0.16	785.1 ± 28.0

Fig. 1 Variation of ⁷Be content in the air dust during 2014

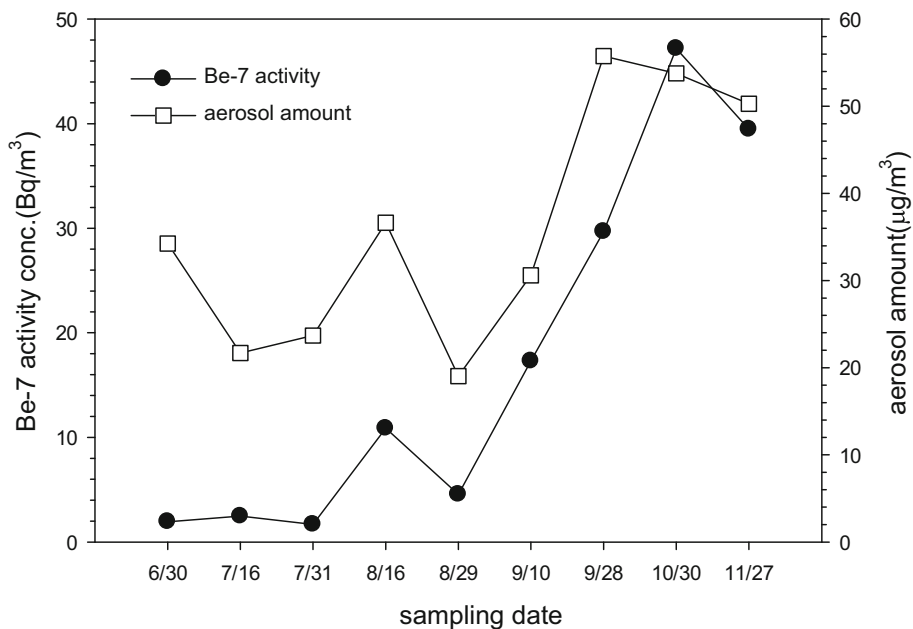


Table 2 ⁷Be activity of rain samples

Sampling	Precipitation amount (L)	Be-7 activity (Bq/L)	Monthly precipitation (mm)
2014-06	14.12	0.51 ± 0.04	143.7
2014-07	14.2	0.55 ± 0.04	177.2
2014-08	14.6	0.46 ± 0.05	240.9
2014-09	13.1	0.29 ± 0.06	118.0
2014-10	17.22	0.99 ± 0.03	169.4
2014-11	4.42	4.77 ± 0.02	40.7

Fig. 2 ⁷Be content variation in precipitation

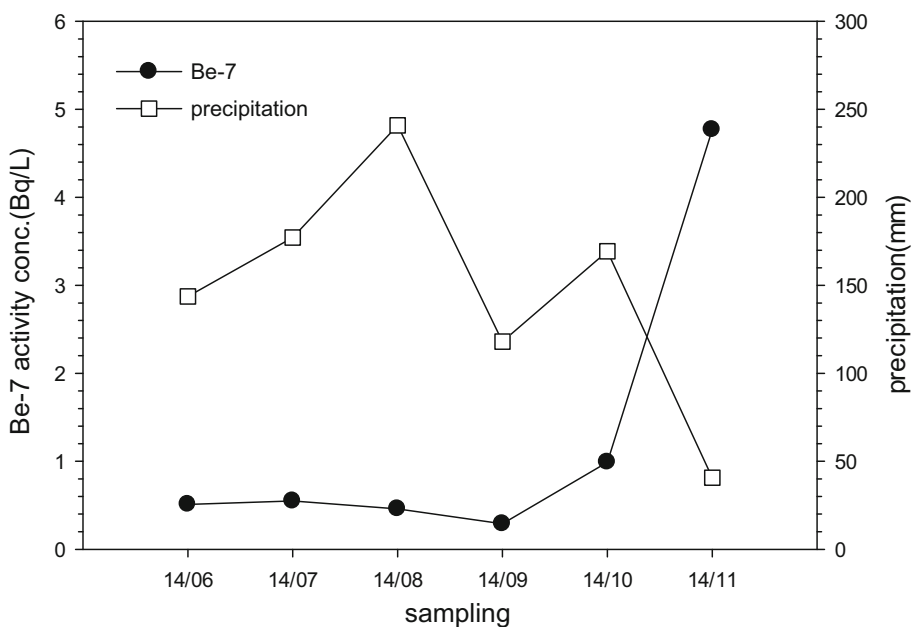
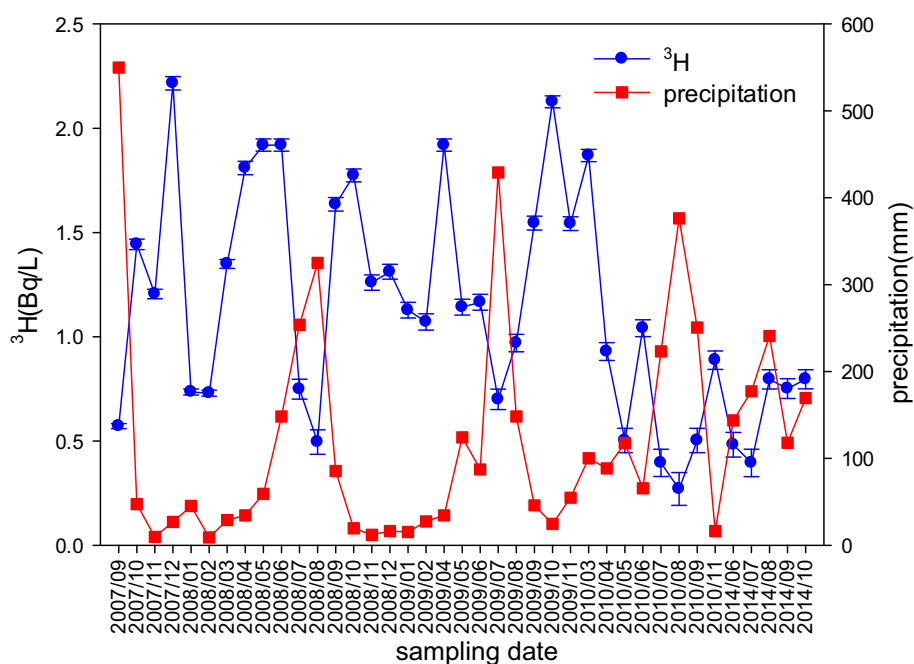


Fig. 3 ^3H content variation in precipitation



Siberian air mass, which travels over South Korea and then moves across southeastern China to Hong Kong.

Measured ^7Be activity in precipitation varies up to ten times on a monthly time collection. This variation is largely a consequence of washout effect. The ^7Be activity in precipitation was also high in winter season. The precipitation in Korea was generally high at summer and most of the rain was precipitated this time. Therefore, ^7Be content was diluted by high precipitation at summer time. Mitchell [18] and Bulloch [19] were also found ^7Be activity variation in precipitation and high activity was observed at low rainfall season.

The tritium concentration monitored from Sep-2009 to Oct-2014 at Daejeon ranged from 0.27 to 2.22 Bq/L and Fig. 3 shows the spatial distribution of the tritium values. The maximum value of the tritium content was in March and the minimum value in the rainy season (Jul, Aug) due to the dilution effect of heavy rain. Vapor produced from Pacific ocean at summer season diluted ^3H concentration with high precipitation. Tritium is uniformly produced by cosmic ray in the stratosphere but ^3H concentration is varied with season and latitude when crack was happened at the spring in the tropopause between the stratosphere and troposphere [20, 21].

Conclusion

This paper attempts to provide a comprehensive survey of the cosmogenic radionuclides composition in the aerosol and precipitation. ^7Be concentration in the aerosol was varied with season due to wind direction and precipitation.

The mean monthly activity level of ^7Be varied from 1.94 to 47.2 Bq/m³ and the precipitation was ranged from 0.29 to 4.77 Bq/L. These results showed a seasonal variation and the ^7Be activity levels were high in the relatively cold season and low in summer due to high precipitation. These results suggest that the ^7Be concentration of East Asia is affected more by the flow pattern of the arctic cold air mass than the vertical convection of air. ^3H concentration ranged from 0.27 to 2.22 Bq/L and also showed a seasonal variation with precipitation pattern.

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