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Note

Atmospheric Concentration and Deposition Flux of Cosmogenic Beryllium-7 at Toki, Central Part of Japan

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The atmospheric concentration and deposition flux of cosmogenic ^{7}Be were observed at Toki city, Gifu Prefecture, located in the central region of Japan, from September 2013 to March 2017, to understand the regional characteristics of the atmospheric environment. Atmospheric ^{7}Be concentration ranged from 1.92 to 6.97 mBq m^{-3} and high concentrations were observed in spring and fall. ^{7}Be deposition ranged from 35.2 to 281.6 Bq m^{-2} and it had a single peak in summer depending on monthly precipitation. These results are comparable to the previously reported values. Total deposition velocity of ^{7}Be ranged from 0.25 to 4.92 cm sec^{-1} with a mean value of 1.48 ± 0.88 cm sec^{-1}, and showed a seasonal variation pattern with a single large peak in summer which is similar to deposition pattern. These results suggest that precipitation is an important control factor of total deposition velocity.

Key words: Beryllium-7, Cosmogenic, Atmosphere, Deposition

1. Introduction

Beryllium-7 (^{7}Be, $T_{1/2}$ = 53.3 d) is a short-lived natural radionuclide produced in the upper atmosphere through the spallation reactions of atmospheric molecules of oxygen, nitrogen and carbon by cosmic ray produced neutrons and protons (secondary particles)[1]. Briefly described, 75% of the ^{7}Be production occurs in the stratosphere while 25% is produced in the troposphere, particularly in the upper troposphere[2, 3]. Its production rate depends on the 11-year cycle of solar activity, altitude and the geomagnetic latitude[4]. The produced ^{7}Be rapidly attaches to sub-micron aerosol particles in the upper atmosphere and these are deposited onto the ground surface through the stratosphere[5]. As a result, it has been a useful tracer for atmospheric circulation, tropospheric residence time and scavenging processes of airborne particulates[6, 7]. For example, Uematsu *et al.*[8] reported extensive temporal and areal distribution of atmospheric aerosol concentration in the marine boundary layer using ^{7}Be. Kritzet *et al.*[9] reported about the troposphere-stratosphere exchange inferred from atmospheric ^{7}Be observations. Ishikawa *et al.*[10]

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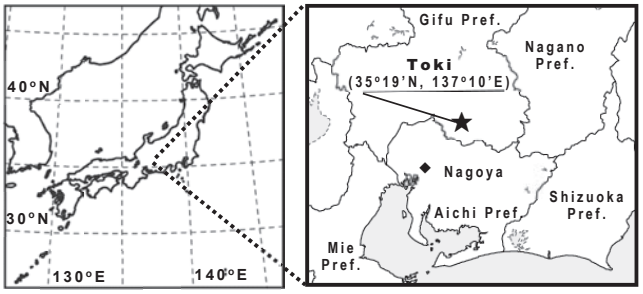

Fig. 1. Location of the sampling site.

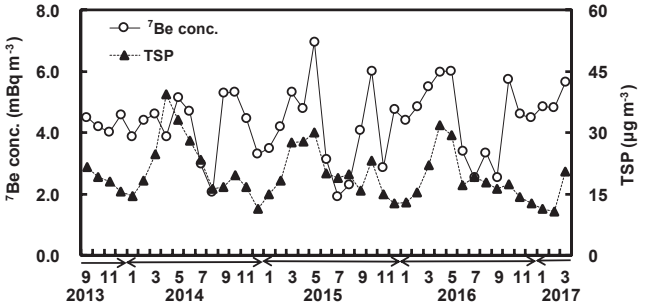

Fig. 3. Atmospheric concentration of ^{7}Be and TSP at Toki.

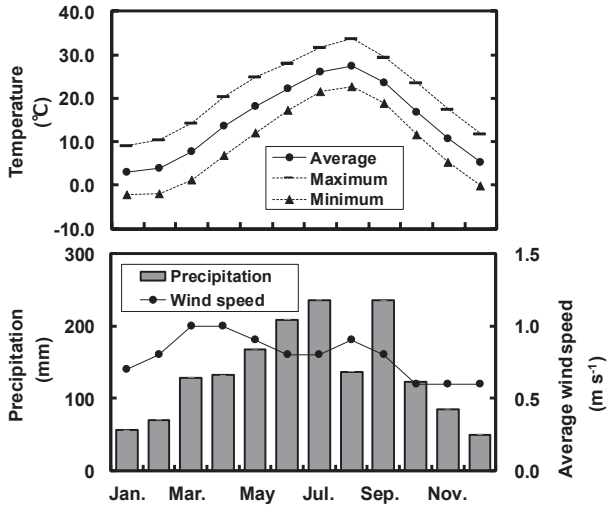

Fig. 2. Average weather conditions for 30 years (1981-2010) at the TajimiAMeDAS site.

investigated the precipitation scavenging process of radionuclides from the atmosphere using ^{7}Be. It is known that local weather condition is the main controlling factor for ^{7}Be scavenging from the atmosphere[11]. Narazaki *et al.*[12] described the seasonal variation of ^{7}Be deposition in Japan and its relation to local meteorological geographical conditions, while Hasegawa *et al.*[13] found the local precipitation mode is an important controlling factor of ^{7}Be deposition flux using an estimated simple simulation model. The total deposition velocity is an index for the removal mechanism of aerosol particles from the atmosphere[14, 15].

In Japan, these surveys have been conducted nationwide. However, the comprehensive observation of atmospheric concentration and deposition flux of ^{7}Be in the central area of Japan has been limited. In this paper, we reported the seasonal variation of the atmospheric concentration and deposition flux of ^{7}Be at Toki, central part of Japan and calculated its total deposition velocity for the understanding of the regional characteristics of atmospheric environment.

2. Experimental

2.1. Overview of study site

The sampling site was on the roof of the National Institute for Fusion Science (NIFS) at Toki City, Gifu Prefecture (35°19'N, 137°10'E). Figure 1 shows the sampling location. Toki City is located approximately 30 km northeast of the Nagoya metropolitan area in the central region of Japan. This area is located in a small-scale basin, approximately 10 km in diameter, and it is surrounded by lower elevation mountains. The average weather conditions for 30 years (1981-2010) are shown in Figure 2. These data were observed at Tajimi AMeDAS (Automated Meteorological Data Acquisition System) site which is located 6 km northwest from the sampling point[16]. The average monthly precipitation was high in summer and low in winter, while the average wind speed was lower than 1.0 m s^{-1} in that time period. Average monthly temperature ranged from 9.2 to 33.7℃. Recently, research on extreme high temperature events relating to local climate change has proceeded in this area[17].

2.2. Sample collection and analysis

Aerosol samples were collected monthly from September 2013 to March 2017 using a high-volume air sampler (HV-1000F, Sibata, Japan) with pre-weighed glass-fiber filters (GB-100R, ADVANTEC, Japan). The collection efficiency was 99.99% for particulate matter with a diameter of 0.3 μm. The airflow rate was approximately 1000 $Lmin^{-1}$. After sampling, the dried filter was re-weighed and the aerosol mass concentration, total suspended particle (TSP), was calculated using the difference in filter weight, and the flow rate. The air filter samples were cut and placed into plastic containers (U8, AS ONE, Japan).

Bulk atmospheric deposition samples which mixed wet and dry depositions were collected monthly from December 2013 to March 2017 using a small polyethylene basin with an open surface area of 0.178 m^2. Deionized water was poured into the sampling basin to ensure the capture of dry deposition. Collected samples were passed through a column (Powdex resin, Ecodyne Co., USA) for

Table 1. Seasonal average values of atmospheric ^{7}Be and TSP concentrations

	Spring (Mar. - May)	Summer (Jun. - Aug.)	Fall (Sep. - Nov.)	Winter (Dec. - Feb.)
^{7}Be concentration (mBq m^{-3}) Average ± S.D.	5.38 ± 0.86	2.93 ± 0.86	4.47 ± 1.05	4.33 ± 0.53
TSP concentration (μg m^{-3}) Average ± S.D.	28.61 ± 5.56	20.06 ± 3.66	17.80 ± 2.70	14.03 ± 2.52

Table 2. Summary of atmospheric concentration of ^{7}Be

City	Country	Latitude	Longitude	Period	^{7}Be (mBq m^{-3}) Range	Reference
Kiruna	Sweden	67°84′N	20°34′E	Jan. 1975 - Dec. 2000	0.042- 8.159	Kulan et al. (2006)[19]
Grindsjön	Sweden	59°07′N	17°82′E	Aug. 1972 - Dec. 2000	0.602- 9.086	Kulan et al. (2006)[19]
Ljungbyhed	Sweden	56°08′N	13°23′E	Feb. 1975 - Dec. 2000	0.599- 7.938	Kulan et al. (2006)[19]
Prague	Czech Republic	50°05′N	14°25′E	Apr. 1986 - Dec. 2002	1.01 - 9.87	Kulan et al. (2006)[19]
Dijon	France	47°20′N	5°02′E	Sept. 1984 - Feb. 2003	1.08 - 9.254	Kulan et al. (2006)[19]
Milan	Italy	45°28′N	9°06′E	Sep. 1993 - June 1995	0.2 - 6.9	Vecchi and Valli (1997)[7]
Michigan	USA	42°58′N	83°01′W	Oct. 1999 - Feb. 2001	1.5 - 9.8	McNeary and Baskaran (2003)[20]
Rokkasho	Japan	40°57′N	141°21′E	Mar. 2000 - Mar. 2006	1.28 - 6.90	Akata et al. (2008)[15]
Beijing	China	39°54′N	116°24′E	Jul. 2009 - Jul. 2010	1.35 - 15.64	Tan et al. (2013)[21]
Nagano	Japan	36°39′N	138°127E	Aug. 2000 - Mar. 2005	3.3 - 14.0	Muramatsu et al. (2008)[22]
Tatsunokuchi	Japan	36°26′N	136°36′E	Apr. 1993 - Dec. 1994	1.82 - 8.89	Yamamoto et al. (1998)[23]
Tsukuba	Japan	36°03′N	140°07′E	Nov. 1987 - May 1992	1 - 6	Sato et al. (1994)[24]
Osaka	Japan	34°32′N	135°30′E	Jan. 1978 - June 1993	1.8 - 9.6	Matsunami and Megumi (1994)[25]
Kumamoto	Japan	32°48′N	130°43′E	Dec. 2001 - Nov. 2003	1.44 - 5.66	Momoshima et al. (2006)[26]
Okinawa	Japan	26°14′N	127°46′E	Feb. 2004 - Dec. 2010	1.45 - 15.1	Tanahara et al. (2014)[27]

collecting the various radioactive species[18]. The homogenized resin was dried in an electric oven at 80℃ for over 48 hours, followed by packing into plastic containers similarly to the air filter samples.

^{7}Be concentrations in samples were measured by a high-purity Ge spectrometer (HPGe, GX-3018, Canberra, USA) with a multi-channel analyzer for 250,000 sec, using γ-rays of 477.6 keV for the determination. Counting uncertainty was less than 4% expressed as relative standard deviation. Measured values were corrected for radioactive decay to the middle of the sampling period[15].

3. Results and Discussion

3.1. Atmospheric concentration

Monthly atmospheric concentrations of ^{7}Be together with TSP are shown in Figure 3, while the seasonal average values of atmospheric ^{7}Be concentration and TSP concentration are summarized in Table 1. Atmospheric ^{7}Be concentration ranged from 1.92 to 6.97 mBq m^{-3} and the annual average ^{7}Be concentrations in 2014, 2015 and 2016 were 4.17, 4.15 and 4.45 mBq m^{-3}, respectively. The maximum value was detected in May 2015, and the seasonal variation showed double peaks in spring and fall. High atmospheric ^{7}Be concentrations in these seasons are generally observed in mid-latitude areas, since ^{7}Be in the stratosphere is transported to the troposphere by tropopause folding events, which are a vertical transport from the upper troposphere to the stratosphere[4]. Many researchers reported the ^{7}Be concentration range observed in various locations, the summarized list is shown in Table 2, mainly describing the data of Japan[7,15,19-27]. Atmospheric ^{7}Be concentration in various location ranged from 0.042 to 15.64 mBq m^{-3}. Our results are comparable to the other reported values. The TSP concentration ranged from 10.8 to 39.4 μg m^{-3}, and high TSP concentrations were observed in spring season. It is known that northwestern monsoons from the Asian continent blow into Japanese archipelago from winter to spring, and they carry air mass with natural and/or anthropogenic materials from the continent. This seasonal variation of TSP concentration is a general trend in Japan. Average TSP concentrations in 2014, 2015, and 2016 were 21.8, 20.3 and 18.8 μg m^{-3}, respectively, which are comparable to the reported values at the western part of Gifu Prefecture, located 30 km west of the sampling site[28]. There is no correlation between atmospheric ^{7}Be concentration and TSP was observed (correlation coefficient R = 0.33). This result means that the main sources of atmospheric ^{7}Be and particulate matter are

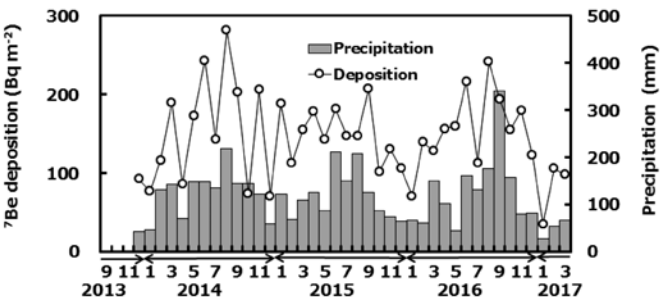

Fig. 4. Monthly ^{7}Be deposition and precipitation at Toki.

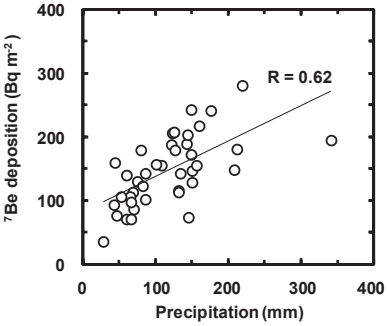

Fig. 5. Monthly ^{7}Be deposition and precipitation at Toki.

different, which are from the upper atmosphere and the troposphere over the Asian continent, respectively.

3.2. Atmospheric deposition

The monthly atmospheric ^{7}Be deposition and monthly precipitation are shown in Figure 4. ^{7}Be deposition ranged from 35.2 to 281.6 Bq m^{-2} and it had a peak in every summer. The maximum value was observed in August 2014. Narazaki *et al.*[12] investigated seasonal variation patterns of ^{7}Be deposition throughout Japan and classified them into four types: winter peak, double peaks (spring and fall), spring peak and non-peak. Our observed pattern was different from the previously reported ones; it was a summer peak pattern. Figure 5 shows the relationship between ^{7}Be deposition and precipitation, which has a relatively good correlation coefficient. As mentioned before, generally the precipitation is high in summer and low in winter in this area (Fig. 2). It seems that the ^{7}Be deposition pattern is dependent on precipitation. The annual ^{7}Be depositions in 2014, 2015 and 2016 were 1864, 1800 and 1879 Bq m^{-2}, respectively. The annual depositions of ^{7}Be throughout Japan from 1989 to 1995 ranged from 290 to 6500 Bq m^{-2} reported by Narazaki and Fujitaka[29], and the data of Gifu Prefecture during 1991 to 1995 ranged from 290 to 1200 Bq m^{-2}. Our results were higher than the reported values of Gifu Prefecture and on the same level with Shizuoka Prefecture which is located at the Pacific Ocean side. The reported values of Gifu Prefecture are being observed in the Seino area which is located at the western part of Gifu Prefecture. The geographical and climatological condition of the Seino area is different from the Tono area including Toki City. This result indicated that ^{7}Be deposition is controlled by the meso-scale local geographical and climatological conditions like atmospheric circulation and/or precipitation[30].

3.3. Total deposition velocity

The total deposition velocity (Vd) of ^{7}Be is determined by following equation:

$$Vd = \frac{Ft}{Ca} \quad (1)$$

where Ft is the monthly ^{7}Be deposition flux and Ca is the atmospheric concentration of ^{7}Be. The time series of total deposition velocity of ^{7}Be is shown in Figure 6. The total deposition velocity of ^{7}Be ranged from 0.25 to 4.92 cm sec^{-1} with a mean value ± S.D. of 1.48 ± 0.88 cm sec^{-1}, and it had a seasonal variation pattern with a single peak, high in summer and low in the other three seasons. Crecelius[14] reported that the total deposition velocity of ^{7}Be at Quillayute, Washington (USA) was 1.0 cm sec^{-1}. McNeary and Baskaran[20] observed that the total deposition velocity of ^{7}Be at Michigan varies between 0.2 and 6.1 cm sec^{-1}. Our results are comparable to these previously reported values. Akata *et al.*[15] reported that total deposition velocity of ^{7}Be at Rokkasho, northern Japan had clear seasonal variation, which is large in winter and summer, and small in spring and fall. As mentioned before, the factors control the atmospheric concentration and deposition flux of ^{7}Be are the local geographical and meteorological conditions. Relationship between ^{7}Be deposition and precipitation rate has a good correlation coefficient (Fig. 7) which is same the relationship between ^{7}Be deposition and precipitation rate. This result indicates that precipitation is an important control factor of the total deposition velocity in central region of Japan.

Conclusion

We investigated the atmospheric concentration and deposition flux of ^{7}Be at Toki, the central region of Japan from September 2013 to March 2017 to understand the regional atmospheric environment. The atmospheric ^{7}Be concentration ranged from 1.92 to 6.97 mBq m^{-3} and it had double peaks in spring and fall. On the other hand, ^{7}Be deposition ranged from 35.2 to 281.6 Bq m^{-2} and it had a single peak in summer. Total deposition velocity of ^{7}Be ranged from 0.25 to 4.92 cm sec^{-1} with a mean value of 1.48 ± 0.88 cm sec^{-1}, and it had a seasonal variation pattern

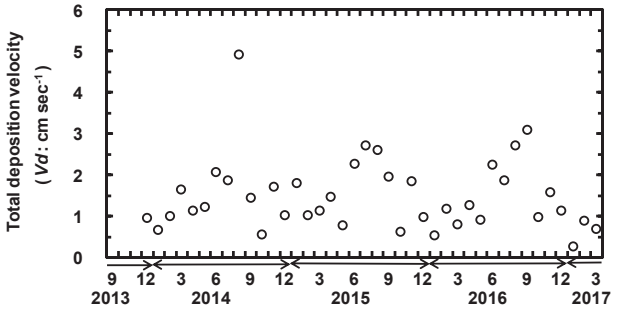

Fig. 6. Seasonal variation of total deposition velocity of ^{7}Be at Toki.

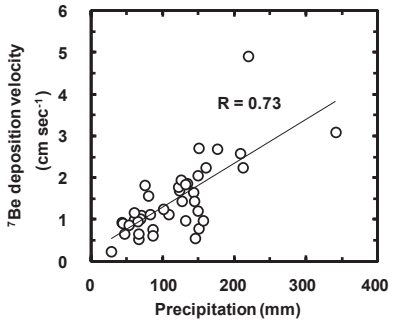

Fig. 7. Relationship between precipitation and total deposition velocity of ^{7}Be at Toki.

with single peak which was the same as the one for deposition and precipitation. These results suggest that the precipitation is the most important control factor of total deposition velocity of ^{7}Be in this area.

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Conflict of interest disclosure

The authors declare that they have no conflicts of interest.

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