原著

Seasonal variations in atmospheric deposition of radioactive beryllium and lead associated with atmospheric removal processes at Fukuoka, Japan

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Abstract : Environmental radioactivity related to Be and Pb was surveyed at a site in Fukuoka. To determine the contributions from atmospheric deposition, both precipitation and aerosols were sampled from February 2014 to December 2018. The average annual total deposits of ^{7}Be and ^{210}Pb were 1,509 Bq/m^2 and 256 Bq/m^2, respectively. The depositional velocities of ^{7}Be and ^{210}Pb were strongly correlated. Seasonal variations in radioactive deposition were marked, with higher levels in spring and winter, and lower ones in summer. Suspended particulate matter showed a negative correlation with ^{7}Be and ^{210}Pb between February and April. Analysis of the depositional velocities revealed that in wet seasons, both wet and dry depositional processes are equally important for removing ^{7}Be and ^{210}Pb from the atmosphere. In contrast, in drier seasons, ^{7}Be levels in precipitation are typically higher than ^{210}Pb, suggesting that wet deposition is more effective at removing ^{7}Be, while dry deposition is more effective at removing ^{210}Pb from the atmosphere.

Keyword : *environmental radioactivity, atmospheric deposition, ^{7}Be, ^{210}Pb*

1. Introduction

Radionuclides occurring in our natural environment can be classified into three general categories: primordial radionuclides, cosmogenic radionuclides, and artificial radionuclides. Beryllium-7（^{7}Be）is a cosmogenic radionuclide formed by spallation of oxygen and nitrogen nuclei by energetic cosmic rays. In contrast, lead-210（^{210}Pb）is a decay product of gaseous radon-222, which emanates primarily from continental land masses. ^{7}Be is considered to be a tracer of upper atmospheric air masses, while ^{210}Pb can be an effective tracer of continental surface air. However, concentrations of these radionuclides in the atmosphere are variable because of aerosol transit and residence times in the troposphere, aerosol deposition velocities, and aerosol trapping by ground vegetation[1-2)]. In addition, geographical attributes, including latitude, longitude and altitude affect radionuclide deposition rates. Thus, recognizing patterns of environmental radioactivity is important.

Seasonal variations of these radionuclides in air are widely reported in the literature, especially in precipitation. Precipitation is clearly important because of producing washout effects on atmospheric aerosols[3-9)]. Therefore, we surveyed environmental radioactivity and determined the various contributions of atmospheric removal processes at a site in Fukuoka on Kyusyu Island. This site is also an important one for studying of the effect of continental aerosol transport over Japan.

2. Materials and methods

2.1 Atmospheric deposition

Atmospheric deposits were collected using an open plastic collector（with an area of 2,520 cm^2）, located on a rooftop（at a height of 21.0 m above ground）of a building belonging to Junshin Gakuen University, Fukuoka, Japan（33° 56' N; 130° 42' E; at an altitude of 20.7 m above sea level）（Fig. 1）.

Generally, radionuclides in the atmosphere are removed in two ways; one is by gravitational settling to the ground known as “dry deposition”, and the other is by precipitation scavenging known as “wet deposition”. To collect both wet and dry deposited

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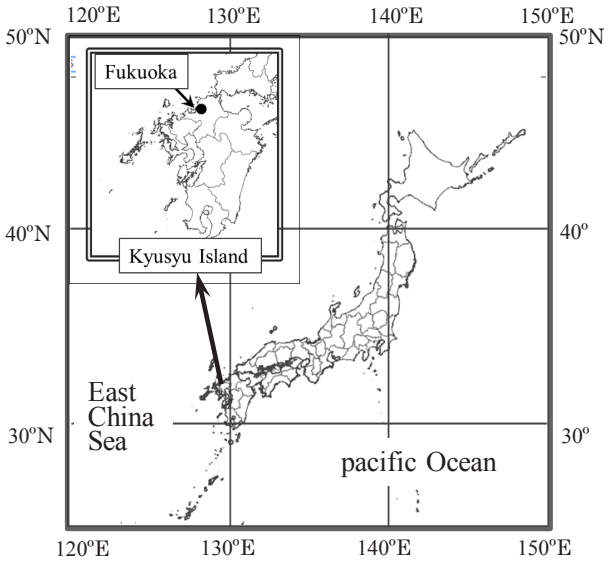

Fig. 1. Sampling location in Fukuoka. (33° 56' N; 130° 42' E; at an altitude of 20.7 m above sea level)

material, distilled water covering the bottom of the plastic collector was added in advance to prevent resuspension of any dry deposited material from the collector. Samples were collected from February 2014 to December 2018. At the time of collection, nitric acid was added to the samples at a rate of milliliters per liter to preserve them and prevent any loss of radionuclides by absorption to the container walls. After collection, the samples were transferred to beakers and reduced in volume by heating them on hot plates. The residues of these samples were transferred to plastic column-shaped containers (ϕ 56 × 68 mm; Umano Co., Ltd. Japan) to undergo natural drying. Our method for collecting environmental samples complies with the Ministry of Education[10]. As the number of days in a month is different, the monthly data obtained with the device (Bq/kg) were converted to a daily amount of deposition per unit area (Bq/m^2·day).

2.2 Supplementary data on samples

Weather information was obtained from the Japan Meteorological Agency[11]. Atmospheric information on the concentration for aerosols was obtained from the Environmental Conservation Section of the Environmental Bureau of Fukuoka[12], conducted in Oohashi, Minami-ku, Fukuoka City. The kind of collected data were suspended particulate matter (SPM) and fine particulate matter (PM2.5). Monitoring information on spatial dose rates of radioactivity was obtained from the Nuclear Regulation Authority[13], conducted at Fukuoka Prefectural Government, Hakata-ku, Fukuoka City. As the measurements value was updated every 10 minutes in terms of μSv per hour, the converted value detected into a time average value was used in this study.

2.3 Detection of radioactivity and statistical analysis

Radioactivity of the deposits was determined using a gamma-spectrometer, equipped with a low-background

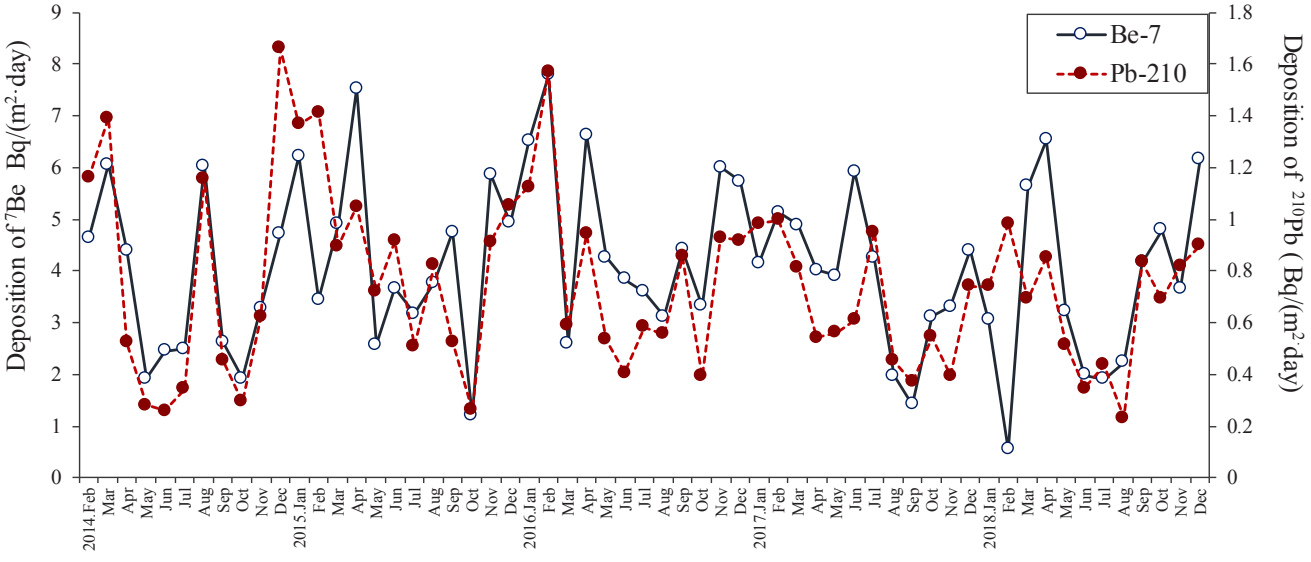

Fig. 2. Variations in Be-7 (○) and Pb-210 (●) concentrations in atmospheric deposition from 2014 to 2018.

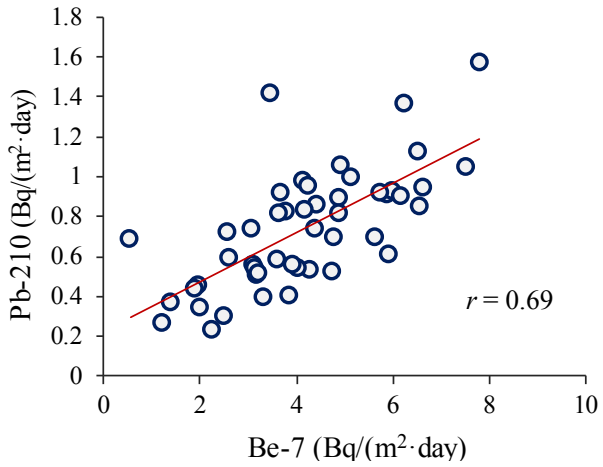

Fig. 3. Correlation between the concentrations of Be-7 and Pb-210 in atmospheric deposition from 2014 to 2018.

type Ge detector (GMX, EG&G Ortec Ltd., Oak Ridge, TN, USA) and shielded with massive lead blocks (15 cm in thickness) and 4-mm-thick oxygen-free copper lining. The spectrum was stored by a 4,096-multichannel analyzer for more than 24 h before being analyzed using Gamma Studio software (V1.22, Seiko Instruments, Inc., Tokyo, Japan).

Correlations were determined using Pearson's product-moment correlation coefficients. A p value of < 0.05 was considered statistically significant. Statistical analyses were performed with Statcel (V3.0, Microsoft Corp., Albuquerque, MN, USA). To determine concentrations of radioactivity, we calculated an average daily concentration to avoid issues related to unequal calendar months.

3. Results and discussion

3.1 Variations of ^{7}Be and ^{210}Pb in deposits

Monthly variations in ^{7}Be and ^{210}Pb in deposits are shown in Fig 2. The average annual total deposits of ^{7}Be and ^{210}Pb in Fukuoka were 1,509 Bq/m^2 and 256 Bq/m^2; these values ranged from 1,250 to 1,757 Bq/m^2, and from 215 to 289 Bq/m^2, respectively. The amounts of ^{7}Be obtained in this study were within the range of annual amounts reported in Japan. Momoshima *et al.*[14] reported a range from 1,486 to 1,693 Bq/m^2 in Kumamoto from 2001 to 2003, Megumi *et al.*[15] reported a range from 967 to 1,752 Bq/m^2 in Osaka from 1983 to 1997, while Narazaki *et al.*[16] reported a value of 1,239 Bq/m^2 in Hokkaido and 1,922 Bq/m^2 in Fukushima in 1995.

^{7}Be is produced in the upper troposphere and stratosphere; but transfer of ^{7}Be to the Earth's surface is accomplished by stratospheric air mass intrusions into the troposphere, followed by gravitational settling and precipitation scavenging processes[17]. In contrast, ^{210}Pb is a decay product of gaseous radon-222, which emanates primarily from the land surface. Despite their different origins, both radionuclides show equivalent geochemical behavior

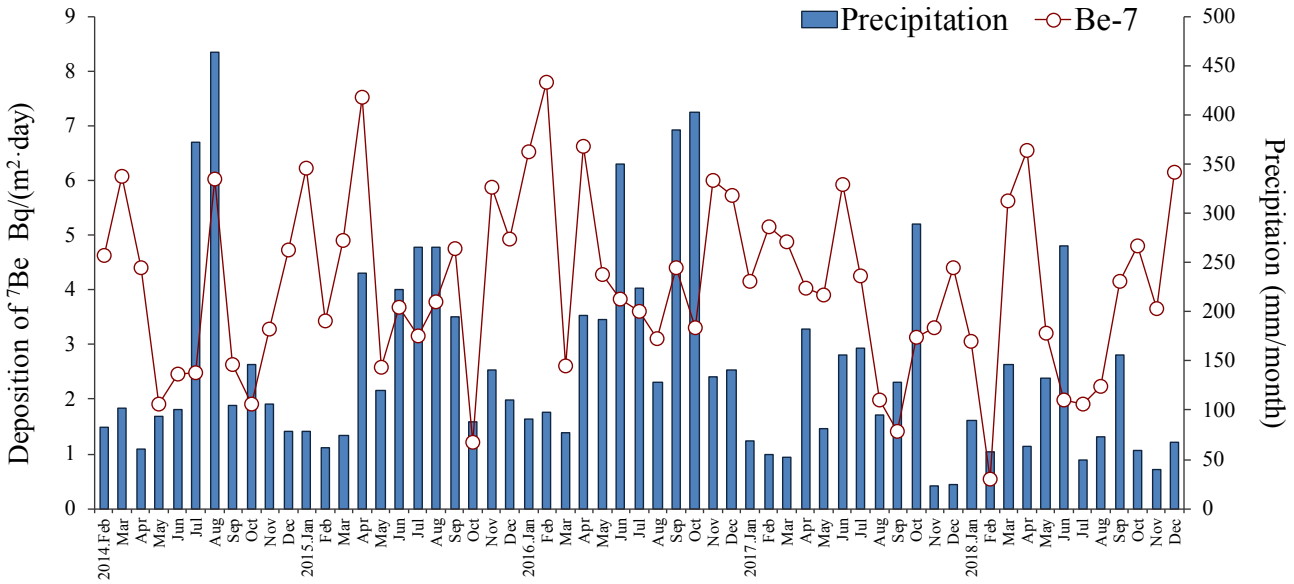

Fig. 4-1. Monthly variations in Be-7 concentrations in atmospheric deposition and precipitation samples. The monthly concentration of Be-7 (line graph) and monthly total precipitation (bar graph) were monitored from 2014 to 2018.

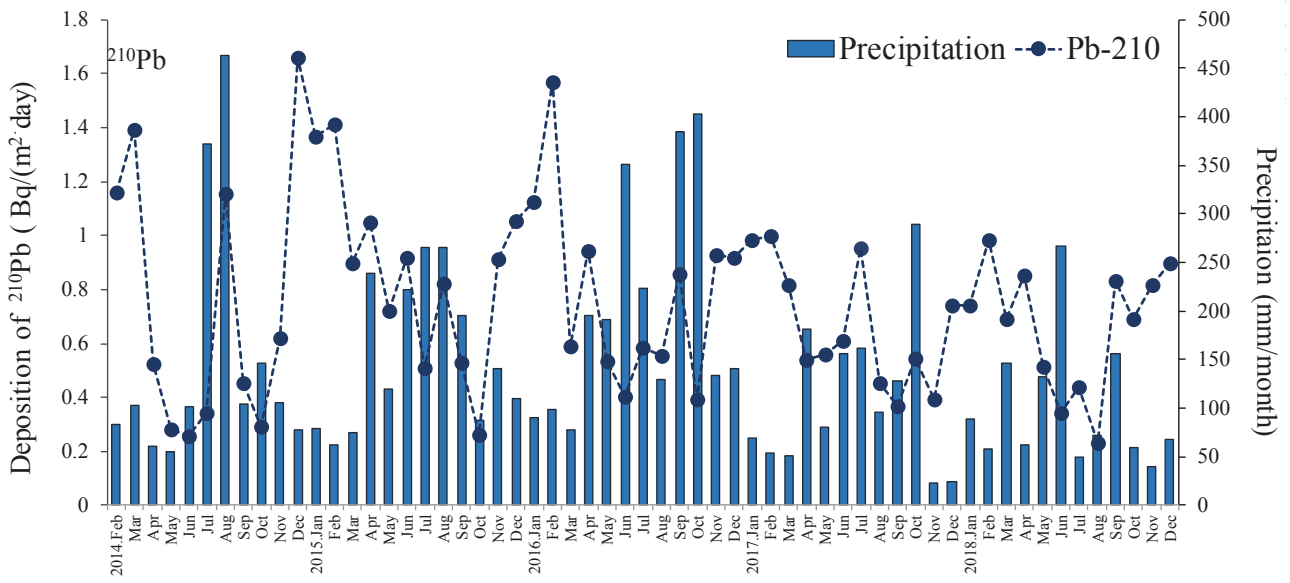

Fig. 4-2. Monthly variations in Pb-210 concentrations in atmospheric deposition and precipitation samples. The monthly concentration of Pb-210 (line graph) and monthly total precipitation (bar graph) were monitored from 2014 to 2018.

in tropospheric air masses. We measured the variations of ^{7}Be and ^{210}Pb concentrations in deposits (Fig. 3). Concentrations of ^{7}Be and ^{210}Pb were highly correlated, according to the Pearson's correlation coefficient ($r = 0.69, p < 0.01$).

Seasonal variations of radioactive deposits were marked; we observed higher concentrations of ^{7}Be and ^{210}Pb in spring and winter, and lower ones in summer. Hirose *et al*. reported that the seasonal trends of monthly ^{7}Be deposition levels in Tsukuba and Nagasaki were higher in spring and lower in summer[18-21]. ^{210}Pb also exhibited seasonal variation, with higher deposition rates in spring and lower ones in summer[19-21]. Momoshima *et al.* reported monthly deposition rates of ^{7}Be and ^{210}Pb; these data showed a double-peak distribution pattern in spring and fall in Kumamoto[14]. The higher values of ^{7}Be and ^{210}Pb during winter (November to February) are attributed to enhanced horizontal transport from continental regions of higher latitude. The dissimilar depositional patterns in Kumamoto and this study are likely related to the high mountains located on Kyushu Island, which separate Kyushu Island meteorologically.

3.2 Correlation between atmospheric deposition of ^{7}Be or ^{210}Pb and precipitation

The variations of ^{7}Be or ^{210}Pb concentrations in atmospheric deposits and monthly total precipitation

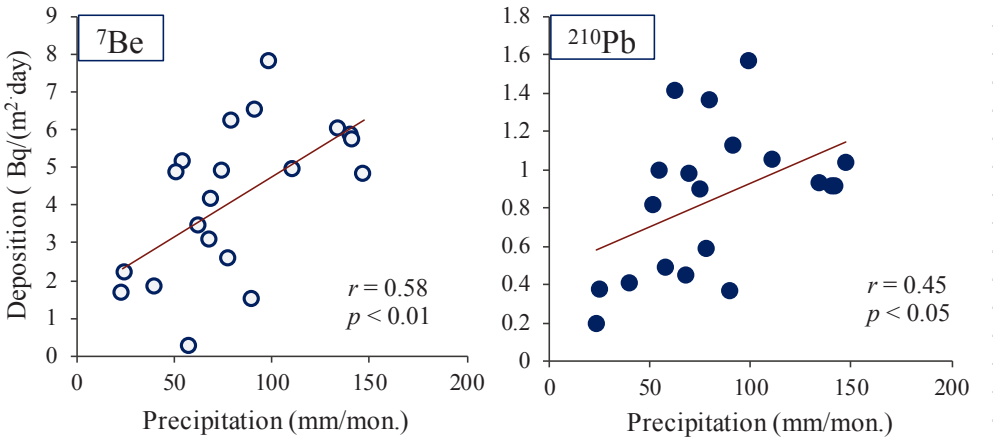

Fig. 5. Correlation between precipitation and atmospheric deposition of Be-7 (○) or Pb-210 (●). Monitoring was limited to the period between December and April..

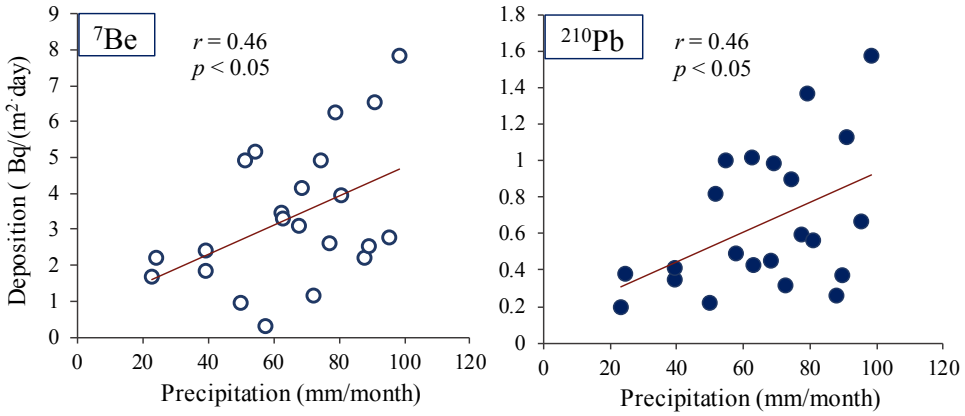

Fig. 6. Correlation between precipitation and atmospheric deposition of Be-7 (○) or Pb-210 (●). Inthis case, only results for which monthly precipitation was less than 100 mm are shown.

are shown in Fig. 4. Statistical analysis did not show any significant correlation between atmospheric radionuclide removal through deposition versus precipitation; however, this may reflect the nature of our observation period. When we limited the observation period from December to April, which corresponds to the dry season on Kyusyu Island, a higher correlation was observed between precipitation and ^{7}Be ($r = 0.58, p < 0.01$) than between precipitation and ^{210}Pb ($r = 0.46, p < 0.05$) (Fig. 5). When monthly precipitation was less than 100 mm, a moderate correlation with precipitation was obtained for both ^{7}Be ($r = 0.45, p < 0.05$) and ^{210}Pb ($r = 0.46, p < 0.05$) (Fig. 6).

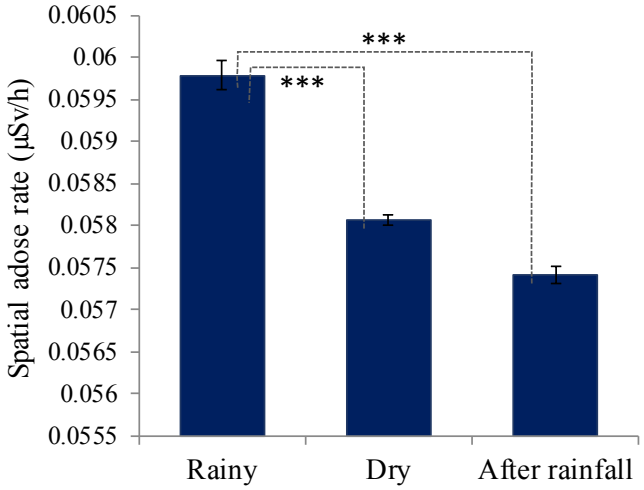

Fig. 7. Variation of spatial dose rates according to weather conditions. Daily spatial doses were distinct for rainy days, dry days, and days after rainfall. ***) p <0.001

In general, aerosols in the atmosphere settle to the ground under clear sky conditions through particle sedimentation (dry deposition) but are removed more efficiently from the troposphere by precipitation scavenging (wet deposition). Figure 7 shows the variation of spatial dose rate according to various weather conditions. Daily spatial doses from January 2015 to December 2016 were classified into three distinct categories, reflecting rainy, dry, and after rainfall conditions. The results showed that the average spatial dose rate was significantly higher on rainy days compared with other days ($p < 0.001$), and it was lowest on dry days after rainfall. These results confirm that rain is the major cause of particle deposition. However, large amounts of precipitation do not necessarily correspond with larger deposits of radionuclides. Under rainfall conditions, washouts can occur during the early stages and quickly cleanse the lower troposphere of aerosols[22]. Under fluctuating weather conditions, a high amount of deposition often corresponds to a low amount of precipitation[14]. In this study, we observed that ^{7}Be had a higher correlation with precipitation than ^{210}Pb during drier seasons. However, when monthly precipitation was less than 100 mm, correlations of ^{7}Be or ^{210}Pb with precipitation were almost equal. Some previous studies reported that a moderate correlation between radionuclide fluxes and precipitation was observed, whereby higher correlation coefficients were found for ^{7}Be than for ^{210}Pb[23-24]. In contrast, another study

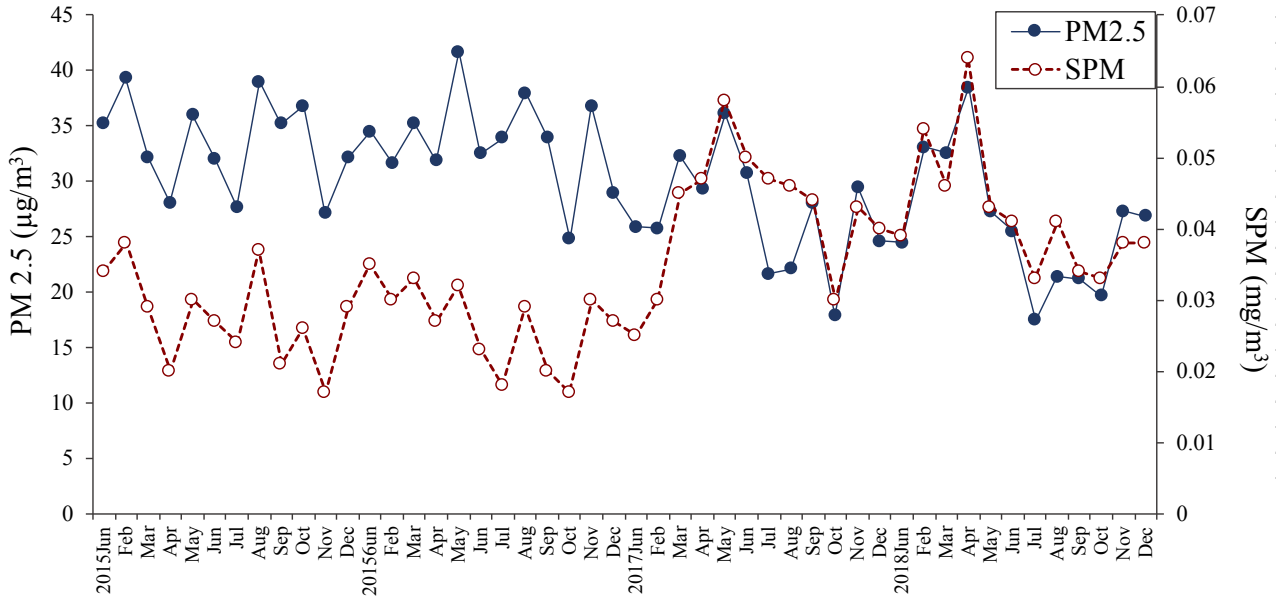

Fig. 8. Variation of PM2.5 and suspended particulate matter (SPM) concentrations in the air from 2012 to 2018. The data are shown as average concentrations per month [g].

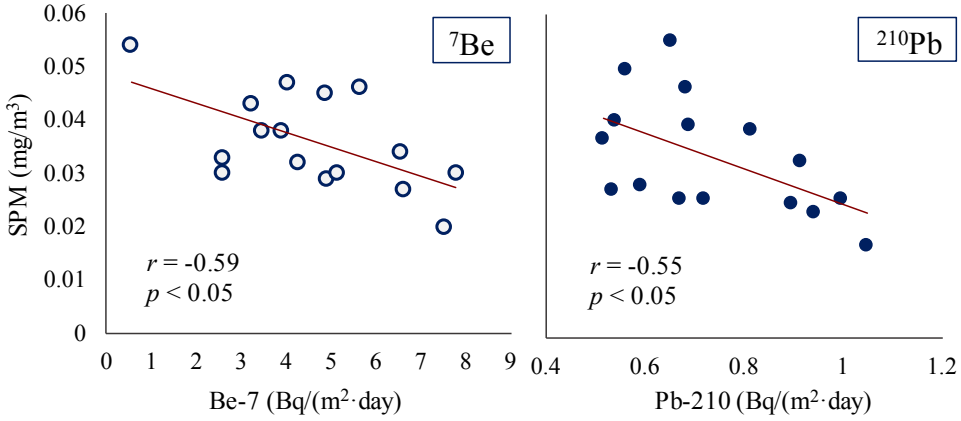

Fig. 9. Correlation between atmospheric concentrations of suspended particulate matter (SPM) and deposits of Be-7 or Pb-210. Monitoring was limited to the period between February and April.

reported that the correlation between ^{7}Be and ^{210}Pb fluxes at the same location is generally good[25]. The results obtained in this study reveal that both ^{7}Be and ^{210}Pb are removed from the air by precipitation, but that ^{7}Be is removed at a higher rate in drier seasons. Because ^{210}Pb is much heavy than ^{7}Be, dry deposition may be more effective at removing ^{210}Pb, while the wet deposition is much effective at removing ^{7}Be in dry seasons.

3.3 Variation of atmospheric deposition and SPM or PM2.5

Radionuclides in atmosphere attach rapidly to aerosols. It has been reported that around 90% or more of total ^{7}Be deposits in temperate zones reflect wet deposition[26-29]. To monitor aerosols in the atmosphere, we recorded atmospheric concentrations of suspended particulate matter (SPM) and particulate matter less than 2.5 microns in diameter (PM2.5). Figure 8 shows the monthly variation of the two radionuclides from 2015 to 2018. At first, it appears that the two aerosol measurements have similar increases/decreases; however, statistical methods indicate that there is no correlation (Table 1). When

Table 1. Results of each correlation coefficent

comparisoned items	monitoring period	*rs*	*p*
PM2.5 - SPM	2014-2018	0.0959	n.s.
	Feb-Apr	0.465	< 0.05
	May-Jan	0.0147	n.s.
Be-7 - SPM	2014-2018	-0.1495	n.s.
	Feb-Apr	-0.6823	< 0.05
	May-Jan	-0.1386	n.s.
Pb-210 - SPM	2014-2018	-0.1185	n.s.
	Feb-Apr	-0.5188	< 0.05
	May-Jan	-0.1207	n.s.
Be-7 - PM2.5	2014-2018	0.1777	n.s.
	Feb-Apr	-0.2507	n.s.
	May-Jan	0.2333	n.s.
Pb-210 - PM2.5	2014-2018	0.2226	n.s.
	Feb-Apr	0.1143	n.s.
	May-Jan	0.1664	n.s.
Be-7 - precipitation	2014-2018	0.191	n.s.
	Feb-Apr	0.3833	n.s.
	May-Jan	0.259	n.s.
Pb-210 - precipitation	2014-2018	-0.0191	n.s.
	Feb-Apr	-0.1824	n.s.
	May-Jan	0.1393	n.s.

Note: Correlation coefficients calculated in this study. Note: *P*-values show significant differences between samples; "*rs*" = correlation coefficient; "ns" = not significant.

we limited the observation period from February to April, the concentrations of SPM and PM2.5 showed moderate correlations ($r = 0.465$, $p < 0.05$). SPM had a negative correlation with both ^{7}Be and ^{210}Pb during the period from February to April ($r = -0.6823$ and $r = -0.5188$, respectively) (Fig. 9). This suggests deposition rates of radionuclides increase as atmospheric concentrations of SPM decrease. There was no correlation between PM2.5 and these radionuclide deposition rates.

Airborne yellow sand, derived from arid areas of continental China, is a characteristic phenomenon in East Asia in spring. From February to April, desert sands cause extensive material damage and health issues in Japan[30-31]. Clearly, SPM contains yellow sand, and the density of SPM becomes higher during spring dust storms[32]. Interestingly, our data indicate an increasing tendency of radionuclide deposition with decreasing concentrations of SPM in atmosphere. Meanwhile, radionuclides in dry deposition were observed during February and April, when there were low levels of precipitation, but abundant dust. This is because radionuclides in the atmosphere attach to aerosols and are removed from the atmosphere by gravitational settling or precipitation processes[17]. This process is enhanced when yellow sand is airborne over Japan. Therefore, the negative correlation between radionuclides and SPM is difficult to explain, perhaps an effective concentration of aerosols is required for dry deposition of radionuclides. We still need to investigate the relationship between SPM and radionuclide deposition rates at this site.

4. Conclusions

This study detected seasonal variations in atmospheric deposition rates of ^{7}Be and ^{210}Pb and correlated them with aerosols at Fukuoka in Japan. For the period from 2014 to 2018, the average annual total deposits of ^{7}Be and ^{210}Pb in Fukuoka were 1,509 Bq/m^2 and 256 Bq/m^2, respectively. The deposition velocities of ^{7}Be and ^{210}Pb were strongly correlated. The seasonal variations of radioactive deposition were higher in spring and winter, and lower in summer. SPM had a negative correlation with ^{7}Be and ^{210}Pb between February to

April. Analysis of the deposition velocities revealed that ^{7}Be and ^{210}Pb were almost equally removed from the air by precipitation, but that ^{7}Be was removed at a higher rate in drier seasons. In drier seasons, wet deposition was more effective at removing ^{7}Be while dry deposition process was more effective at removing ^{210}Pb from the atmosphere.

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