Temporal Variations of ^7Be and ^{210}Pb Activity Concentrations in the Atmosphere and Aerosol Deposition Velocity in Shenzhen, South China

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ABSTRACT

Naturally occurring beryllium-7 (^7Be) and lead-210 (^{210}Pb) serve as powerful tracers in atmospheric studies. In this study, ^7Be and ^{210}Pb were simultaneously measured in atmospheric aerosols and deposition samples for an entire year (from January to December 2017) in Shenzhen, South China. The activity concentrations of the airborne ^7Be and ^{210}Pb ranged from 0.33 to 9.42 mBq m⁻³ (averaging 3.23 mBq m⁻³) and from 0.59 to 4.72 mBq m⁻³ (averaging 1.58 mBq m⁻³), respectively, and were observed to be high during the winter but low during the summer. Moreover, the relatively high ^{210}Pb concentration was probably due to the elevated level of radon in this region's soil. The deposition fluxes of the ^7Be and ^{210}Pb were found to range from 0.25 to 3.04 Bq m⁻² day⁻¹ (averaging 1.57 Bq m⁻² day⁻¹) and from 0.34 to 1.31 Bq m⁻² day⁻¹ (averaging 0.73 Bq m⁻² day⁻¹), respectively. The temporal trends of these fluxes were largely influenced by rainfall and the origin and pathway of air masses, as well as by atmospheric circulation. Based on their concentrations in the aerosols and their deposition fluxes, the average deposition velocities of ^7Be and ^{210}Pb were calculated to be 0.83 and 0.62 cm s⁻¹, respectively. The deposition velocities of both radionuclides correlated well with the amount of rainfall, indicating that precipitation plays a crucial role in removing ^7Be and ^{210}Pb from the air. The activity size distributions of these nuclides combined with the characteristic meteorological conditions in this region resulted in high deposition velocities during summer and low ones during winter.

Keywords: ^7Be and ^{210}Pb ; Activity concentrations; Deposition fluxes; Deposition velocity of aerosols; Shenzhen.

INTRODUCTION

Airborne particles (aerosols) have become the major air pollutant in Chinese megacities (Liu *et al.*, 2014). As long as these particles are formed or injected into the atmosphere, they tend to get involved in physical and chemical processes in the atmosphere (Ahmed *et al.*, 2004) and ultimately sink into the terrestrial environment through dry and wet deposition. Atmospheric particles are considered the main carriers of airborne pollutants and play a crucial role both in air quality and deposition fluxes of pollutants to the earth's surface, which pose a negative effect on the ecological system and human health (Chameides *et al.*, 1999; Yadav *et al.*, 2003). To determine the transport pathway of these pollutants and to assess their environmental impact, it is essential to understand the dynamics of these airborne particles. The naturally

occurring beryllium-7 (^7Be) and lead-210 (^{210}Pb) serve as powerful tracers for atmospheric studies that include (i) air mass exchange between the stratosphere and troposphere, (ii) residence times of atmospheric aerosols, (iii) washout ratios and deposition velocity of aerosols, and (iv) source tracking of oceanic and continental air masses (Baskaran, 2011).

^7Be ($T_{1/2} = 53.3$ days) is a cosmogenic radionuclide produced by the reaction of cosmic rays with oxygen and nitrogen atoms in the stratosphere and upper troposphere (Alonso-Hernández *et al.*, 2014); the production rate of ^7Be on the earth's surface for a given latitude is independent of longitude (McNeary and Baskaran, 2003). Generally, the airborne concentration of ^7Be increases with altitude and is sensitive to meteorological conditions, such as temperature and precipitation (Gai *et al.*, 2015). A high ^7Be concentration in ambient air would indicate a large input from the upper atmosphere (Dueñas *et al.*, 2011a). At mid-latitude, the specific concentrations of ^7Be exhibit high values in spring, owing to the seasonal contraction of tropopause, which results in the enhancement of air exchange between the stratosphere and troposphere (Ali *et al.*, 2011; Gai *et al.*, 2015). ^{210}Pb

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($T_{1/2} = 22.3$ years) is a naturally occurring radionuclide, originating from the decay of ^{222}Rn ($T_{1/2} = 3.8$ days). Because ^{222}Rn is mainly emanated from the earth's surface, ^{210}Pb is considered a tracer of continental air masses (Baskaran, 2011). Once produced, ^7Be and ^{210}Pb instantaneously and irreversibly attach themselves mainly to fine particulates and are removed from air through radioactive decay, dry and wet deposition (Ali *et al.*, 2011). Because of distinctly different sources, combined measurements of ^7Be and ^{210}Pb provide a powerful tool to study the mechanisms of atmospheric processes. Furthermore, knowledge of these nuclides can help us understand the behavior of other similar contaminants in the air (McNeary and Baskaran, 2003).

To date, extensive studies on ^7Be and ^{210}Pb in atmospheric deposition as well as in aerosols have been conducted worldwide (Ioannidou *et al.*, 2005; Alonso-Hernández *et al.*, 2014; Du *et al.*, 2015; Gai *et al.*, 2015; Grossi *et al.*, 2016; Chham *et al.*, 2017; Dueñas *et al.*, 2017). However, combined measurements of ^7Be and ^{210}Pb in aerosols and in bulk fallout are still limited. Located at the eastern side of the Pearl River Delta (PRD) region in Guangdong Province, South China, Shenzhen is a demonstration/pilot zone for socialism with Chinese characteristics, with an area of 2,021 km² and a population approaching 20 million (Liu *et al.*, 2015). During the past three decades, Shenzhen has made rapid economic progress and has become one of the most important industrial and economic centers in China. The rapid urbanization and industrial development in Shenzhen and the PRD region have placed considerable pressure on the environment. Many studies have been conducted on air pollution in the PRD region, mostly focused on atmospheric particulate matter, heavy metals, and organic pollutants (Liu *et al.*, 2014). However, few studies have been conducted on

the atmospheric radiotracers (e.g., ^7Be and ^{210}Pb) in this region; these studies can provide insights into the mechanism of various atmospheric processes. Shenzhen features a subtropical monsoon climate, with oceanic monsoon prevailing in summer and continental monsoon prevailing in winter. The coastal or continental air masses carry the long-range-transported air pollutants to this region, particularly in winter seasons. Shenzhen thus serves as an ideal region for studying air mass transfer in the subtropical monsoon climate region.

The objectives of the current study were as follows: (1) to determine the temporal variations of ^7Be and ^{210}Pb in atmospheric aerosols in Shenzhen; (2) to study the seasonal deposition fluxes of ^7Be and ^{210}Pb in this region; (3) to investigate factors controlling ^7Be and ^{210}Pb in aerosols and deposition fluxes; and (4) to evaluate the deposition velocity of aerosols using ^7Be and ^{210}Pb .

EXPERIMENT METHODS

Sampling and Preparation

Air sampling was performed in Shenzhen University (Fig. 1), which is located in the Nantou Peninsula of Shenzhen city. The roof of the experimental building at Shenzhen University (22.5°N, 113.9°E) was selected as the sampling site. From January to December 2017, atmospheric deposition (wet + dry) samples were collected at monthly intervals by using a stainless container (0.55-m diameter and 0.6-m height). At the end of each collection, bulk fallout samples were transferred to a pre-cleaned polyethylene vessel, the container was washed with 0.2 M HCl and Milli-Q water to remove ^7Be and ^{210}Pb that are possibly adsorbed on the wall, and the combined solution was adjusted to pH 2.0 with 6 M HCl. ^7Be and ^{210}Pb were obtained through chemical co-precipitation

Fig. 1. Sampling location in Shenzhen, South China.

as described by Du *et al.* (2008). A known amount of Fe^{3+} carrier was added to the solution, and after 24 h, the solution was adjusted to pH 8.5 with NH_4OH for $\text{Fe}(\text{OH})_3$ precipitation. The precipitate was then centrifuged and transferred into a cylindrical polyethylene container ($7.5 \text{ cm} \times 7.0 \text{ cm}$) for gamma-ray measurement after drying.

Concurrently, atmospheric aerosol samples were collected once per week using a high-volume air sampler following the procedure described in our previous work (Liu *et al.*, 2010). The sampling flow rate was $1.05 \text{ m}^3 \text{ min}^{-1}$, and the sampling time for each sample was 24 h; suspended particulates were retained on a Whatman glass microfiber filter (GFF; grade GF/A, $20.3 \text{ cm} \times 25.4 \text{ cm}$). After each sampling, filters were folded and pressed into a cylindrical polyethylene container ($7.5 \text{ cm} \times 7.0 \text{ cm}$) so that all samples had the same irradiation geometry.

Analytical Methods

The activity concentrations of ^7Be and ^{210}Pb were determined through gamma spectrometry; the methodology was described in Liu *et al.* (2015). Briefly, the gamma spectrometer was equipped with an HPGe detector (GEM-C5970; ORTEC, USA), with an energy resolution of 1.8 keV at the 1.332 MeV of ^{60}Co and the relative efficiency of 38%. The detector was enclosed in a cylindrical lead shield with 10-cm thickness and 38-cm height. The counting efficiencies of ^7Be and ^{210}Pb in filter and deposition samples were calibrated by an aerosol reference material (QRJH1204, containing ^{152}Eu and ^{137}Cs) and a soil reference material (7NTR141201, containing ^{238}U , ^{235}U , ^{226}Ra , ^{232}Th , ^{40}K , ^{241}Am , ^{137}Cs and ^{60}Co), respectively. The efficiency curves were constructed based on the full-energy peak efficiencies of these calibrated radionuclides in a cylindrical geometry ($\emptyset 75 \text{ mm} \times 70 \text{ mm}$). According to the sample-to-detector geometry, the true coincidence summing effect for the efficiency calibration with ^{152}Eu was small and could be neglected. ^7Be activity was determined through its 477.6 keV gamma-ray (yield 10.12%), and ^{210}Pb content through its 46.5 keV gamma-ray (yield 4.25%). The counting time was set as 24 h for all the samples. The GammaVision 32 software was used for the data analysis.

The activity concentrations (C) of ^7Be and ^{210}Pb were calculated as follows:

$$C_{\text{Pb/Be}} = \frac{N_s - N_b}{\varepsilon \cdot \delta \cdot V} \quad (1)$$

where N_s and N_b are the counting rates (in s^{-1}) of the sample and background, respectively; ε is the characteristic peak efficiency; δ is the branching ratio; and V is the sample volume of aerosol (in m^3). The total deposition fluxes (F) of ^7Be and ^{210}Pb were determined as follows:

$$F = \frac{A_{\text{Be/Pb}}}{S \cdot T} \quad (2)$$

where $A_{\text{Be/Pb}}$ is the total activity of ^7Be and ^{210}Pb (in Bq) deposited in the collector; S is the total surface area of the collector (in m^2); and T is the sampling duration (in days).

The final activity of ^7Be was radioactive decay corrected for the time interval between the sampling and measurement, whereas it was not performed for ^{210}Pb due to its relatively long half-life.

RESULTS AND DISCUSSION

^7Be and ^{210}Pb activity Concentration in the Air

During the sampling period from January to December 2017, the weekly activity concentration of ^7Be was in the range of $0.33\text{--}9.42 \text{ mBq m}^{-3}$, with an annual average of 3.23 mBq m^{-3} . These values were comparable with those obtained from near-surface atmospheric aerosols reported in low-latitude regions, such as Guangzhou, China (2.59 mBq m^{-3} ; Pan *et al.*, 2011), but lower than those reported from mid-latitude regions such as Beijing, China (8.39 mBq m^{-3} ; Tan *et al.*, 2013); Malaga, Spain (4.6 mBq m^{-3} ; Dueñas *et al.*, 2011b); Detroit, Michigan (4.8 mBq m^{-3} ; McNeary and Baskaran, 2003); and Brindisi, Italy (5.4 mBq m^{-3} ; Hernández-Ceballos *et al.*, 2015). The activity concentration of ^{210}Pb was in the range of $0.59\text{--}4.72 \text{ mBq m}^{-3}$, with an annual average of 1.58 mBq m^{-3} . These values were similar to those reported in the northern cities of China (Li *et al.*, 2013) but higher than in Kumamoto, Japan (0.89 mBq m^{-3} ; Momoshima *et al.*, 2006); Malaga, Spain (0.58 mBq m^{-3} ; Dueñas *et al.*, 2011b); and Detroit, Michigan (1.15 mBq m^{-3} ; McNeary and Baskaran, 2003). China is the largest coal producer and consumer in the world. During coal combustion, ^{210}Pb can be released as fly ash from a coal-fired power plant (Liu *et al.*, 2015), which in turn increases the ^{210}Pb concentration in ambient air. On the other hand, Shenzhen city is a high-radiation background region in China; the soil gas radon concentration was nearly seven times the country's average (Wang *et al.*, 2006; Liu *et al.*, 2015), which may have contributed to a high ^{210}Pb concentration in this region.

Fig. 2 shows the monthly ^7Be and ^{210}Pb activity concentrations and precipitation in the study period. As shown in the figure, the activity concentrations of both ^7Be and ^{210}Pb showed seasonal variation; they were high in winter and low in summer, similar to those reported in Kumamoto, Japan (Momoshima *et al.*, 2006), and Qingdao, China (Yang *et al.*, 2013). Shenzhen is located in subtropical monsoon regions, which is wet and hot in summer and dry and cool in winter. During the rainy season, precipitation scavenging causes the aerosol concentration to decrease, thus reducing the attachment medium for ^7Be and ^{210}Pb in the air (Yang *et al.*, 2013). On the other hand, maritime air masses that come from the Pacific Ocean usually contain less ^7Be and ^{210}Pb , which may also be responsible for the low ^7Be and ^{210}Pb in summer. During the dry winter season, the northeast monsoon that originates from Mongolia and southeastern Siberia carries continental air masses to South China; the long-range-transported ^{210}Pb from North China may have led to the increased ^{210}Pb concentration in winter. The production rate of ^7Be is latitude dependent, and high ^7Be concentrations in aerosols have been reported in northern China (Yang *et al.*, 2013; Gai *et al.*, 2015). The air masses coming from these mid-latitude regions (enriched in ^7Be) may have contributed to the high level of ^7Be in winter

Fig. 2. Seasonal variations of ^7Be and ^{210}Pb concentrations in the air and precipitation.

when northeast monsoon prevailing. Many studies have indicated that at mid-latitude regions, an enhanced air exchange occurs between the stratosphere and troposphere in spring (Pan *et al.*, 2011); stratospheric air intrusion may have brought large amounts of ^7Be in March in this region.

Deposition Fluxes of ^7Be and ^{210}Pb

The monthly bulk deposition fluxes of ^7Be and ^{210}Pb along with the precipitation amount are shown in Fig. 3. The deposition fluxes of ^7Be and ^{210}Pb were in the range of 0.25–3.04 $\text{Bq m}^{-2} \text{ day}^{-1}$ and 0.34–1.31 $\text{Bq m}^{-2} \text{ day}^{-1}$, with an annual average of 1.57 and 0.73 $\text{Bq m}^{-2} \text{ day}^{-1}$, respectively. The deposition fluxes of ^7Be and ^{210}Pb showed seasonal variations; high deposition fluxes were observed in spring (March) for ^7Be and autumn (August and September) for both ^7Be and ^{210}Pb . The study of McNeary and Baskaran (2003) indicated that the deposition fluxes of ^7Be and ^{210}Pb on the earth's surface are largely influenced by latitude (for ^7Be), longitude (for ^{210}Pb), and local meteorological conditions. By comparison, the deposition fluxes of ^7Be in Shenzhen fall in the range of those reported in other regions of China (Yi *et al.*, 2007; Du *et al.*, 2008; Zhang *et al.*, 2013; Gai *et al.*, 2015); Cienfuegos in Cuba (Alonso-Hernández *et al.*, 2014); Geneva, Switzerland (Caillet *et al.*, 2001); Spain (Dueñas *et al.*, 2017); and Detroit, USA (McNeary and Baskaran, 2003). Table 1 summarizes the deposition fluxes of ^7Be and ^{210}Pb in 18 stations around the world in the order of latitude. As seen from Table 1, the deposition fluxes of ^7Be showed an increasing trend with latitude (from 22.2°N to 46.2°N) and were the highest in the mid-latitude regions, reflecting the well-known latitudinal dependence of atmospheric deposition fluxes of ^7Be , as have been observed in some regions (Du *et al.*, 2015; Gai *et al.*, 2015; Hernandez-Ceballos *et al.*, 2015). However, further studies of bulk deposition of ^7Be are still needed, especially in the southern hemisphere regions. For ^{210}Pb , its deposition

fluxes at our site were comparable to the global average value (0.53 $\text{Bq m}^{-2} \text{ day}^{-1}$) for latitude between 20°N and 30°N (Baskaran, 2011) but lower than those reported in Shanghai, China (1.31 $\text{Bq m}^{-2} \text{ day}^{-1}$; Du *et al.*, 2008), and higher than Cienfuegos, Cuba, where a value of 0.13 $\text{Bq m}^{-2} \text{ day}^{-1}$ was reported (Alonso-Hernández *et al.*, 2014), owing to the significant input of marine air in this place.

Studies have indicated that the atmospheric deposition fluxes of ^7Be and ^{210}Pb are strongly correlated with the rainfall amount (Dueñas *et al.*, 2005; Gai *et al.*, 2015; Dueñas *et al.*, 2017). To study the effect of precipitation on the removal of both nuclides in the air, the deposition fluxes of ^7Be and ^{210}Pb were plotted against the monthly rainfall amount during the sampling period. As shown in Fig. 4, a positive correlation exists between ^7Be and ^{210}Pb deposition fluxes and precipitation ($r = 0.58$ and $r = 0.47$, respectively). Furthermore, if we narrow the time scale from August to December instead of the entire year, the coefficients between rainfall and fluxes were 0.98 for ^7Be and 0.95 for ^{210}Pb . It is suggested that the removal processes of these two radionuclides from the air are similar, and rainfall is the dominant factor for the scavenging of ^7Be and ^{210}Pb in autumn and early winter. In general, the correlation coefficients between precipitation and deposition fluxes of ^7Be were higher than that of ^{210}Pb . Moreover, similar results have been observed by other authors at various stations, such as Xiamen (Yi *et al.*, 2007) and Shanghai (Du *et al.*, 2008) in China; Malaga, Spain (Dueñas *et al.*, 2005); Cienfuegos, Cuba (Alonso-Hernández *et al.*, 2014); and Detroit, USA (McNeary and Baskaran, 2003). This may be due to their distinct source; ^7Be is a cosmogenic radionuclide, whereas ^{210}Pb is mainly derived through the decay of ^{222}Rn from soil. The inventories of ^{210}Pb in the top soil layer are higher than that of ^7Be and are susceptible to the re-suspension process; thus, the dry fallout of ^{210}Pb is expected to be much higher and is less

Fig. 3. Monthly deposition fluxes of ^7Be and ^{210}Pb , and precipitation.

Table 1. Deposition fluxes of ^7Be and ^{210}Pb in different regions.

Location	Coordinates	Collection time	^7Be fluxes $\text{Bq m}^{-2} \text{ day}^{-1}$	^{210}Pb fluxes $\text{Bq m}^{-2} \text{ day}^{-1}$	Reference
Cienfuegos, Cuba	22°N, 80.3°W	2010.2–2011.1	1.89	0.13	Alonso-Hernández <i>et al.</i> , 2014
Shenzhen, China	22.5°N, 113.9°E	2017.1–2017.12	1.57	0.73	This study
Guangzhou, China	23.2°N, 113.3°E	2012.7–2013.6	1.32	–	Gai <i>et al.</i> , 2015
Xiamen, China	24.3°N, 118.5°E	2004.3–2005.4	1.64	0.51	Yi <i>et al.</i> , 2007
Shanghai, China	31.1°N, 121.2°E	2005.11–2006.10	5.68	1.31	Du <i>et al.</i> , 2008
Nagasaki, Japan	32.5°N, 129.5°E	2001.1–2001.11	4.04	0.64	Hirose <i>et al.</i> , 2004
Qingdao, China	35.8°N, 120.5°E	2012.8–2013.7	2.45	–	Gai <i>et al.</i> , 2015
Tsukuba, Japan	36°N, 140°E	2001.1–2001.11	3.63	0.48	Hirose <i>et al.</i> , 2004
Málaga, Spain	36.7°N, 4.5°W	2005–2015	3.30	0.39	Dueñas <i>et al.</i> , 2017
Norfolk, USA	36.9°N, 76.3°W	1983.1–1984.12	5.89	0.38	Olsen <i>et al.</i> , 1985
Ansan, Korea	37.2°N, 126.5°E	1992–1993	4.67	–	Kim <i>et al.</i> , 1998
Stillpond, USA	39°N, 76.1°W	1995.9–1996.8	5.94	0.36	Kim <i>et al.</i> , 2000
Beijing, China	39.5°N, 116.4°E	2012.7–2013.6	3.91	–	Gai <i>et al.</i> , 2015
New York, USA	40.9°N, 73.1°W	2008.4–2009.12	8.52	0.40	Renfro <i>et al.</i> , 2013
Boston, USA	42.3°N, 71°W	2000.9–2007.8	7.10	–	Zhu and Olsen, 2009
Detroit, USA	42.3°N, 83.1°W	1999.9–2001.3	5.20	0.64	McNeary and Baskaran, 2003
Bordeaux, France	45°N, 0.4°W	2006.1–2007.1	3.44	0.28	Sarri <i>et al.</i> , 2010
Geneva, Switzerland	46.2°N, 6.1°E	1997.11–1998.11	5.72	0.41	Caillet <i>et al.</i> , 2001

controlled by precipitation compared with ^7Be . Although the deposition fluxes of ^7Be are mainly controlled by precipitation, its fallout has a certain limit, regardless of the rainfall amount. As shown in Fig. 3, the deposition fluxes of ^7Be remained relatively constant from April to July, with which rainfall fluctuated greatly; the maximum monthly precipitation occurred in June (363.6 mm), but the highest deposition fluxes of ^7Be were observed in March. Gai *et al.* (2015) observed a negative correlation between the annual deposition fluxes of ^7Be and the average annual precipitation at different latitudes; thus, it is indicated that the fallout of ^7Be

in certain latitude does not increase with continuing rainfall due to its latitude dependence.

Wet precipitation, dry deposition, atmospheric circulation, continental or oceanic air masses are considered main factors that influence the deposition fluxes of ^7Be and ^{210}Pb . Dry deposition is estimated to contribute to 14% of the global ^{210}Pb fallout (Balkanski *et al.*, 1993), but it could be much higher in semi-arid areas and even more important than wet fallout (Rastogi and Sarin, 2008). As shown in Figs. 2 and 3, the monthly rainfall was low in January and February, whereas the deposition fluxes of ^7Be and ^{210}Pb were still

Fig. 4. Correlation between deposition fluxes of ^7Be and ^{210}Pb , and precipitation.

relatively high, along with high concentrations of ^7Be and ^{210}Pb in the air. It is suggested that dry deposition may play an important role in winter months in this region. In addition to dry and wet deposition, the deposition fluxes of ^7Be and ^{210}Pb are also affected by the origin and pathway of air masses as well as atmospheric circulation. During fall when northeast monsoon prevailing, the continental air masses (rich in ^7Be and ^{210}Pb) could have led to high deposition fluxes of ^7Be and ^{210}Pb in August and September, with plenty of precipitation during these periods. In Shanghai, China, high deposition fluxes of ^{210}Pb were ascribed to storm dust from northern and northwestern China (Du *et al.*, 2008), reflecting the influence of air masses on local ^{210}Pb deposition. During spring, as the mid-latitude troposphere narrows, the enhanced air exchange between the stratosphere and troposphere may have caused the high deposition fluxes of ^7Be in March.

Total Deposition Velocity of Aerosols Using ^7Be and ^{210}Pb

Atmospheric deposition is one of the key processes that remove pollutants from air and is considered the major source of nutrients and toxic substances to ecosystems. The deposition velocity of aerosol particles can be used to estimate the deposition fluxes of air pollutants and to assess environmental impact. The total deposition velocity (V_d) of any radionuclide in air can be determined as follows (McNeary *et al.*, 2003; Dueñas *et al.*, 2005):

$$V_d = \frac{F}{C} \quad (3)$$

where F and C are the total deposition flux (wet + dry) and the activity concentration of this radionuclide in aerosols, respectively. In this study, the total deposition velocity of ^7Be and ^{210}Pb were calculated as the ratio of the deposition fluxes of ^7Be and ^{210}Pb to the monthly average concentrations

of ^7Be and ^{210}Pb in the air.

^7Be and ^{210}Pb serve as good tracers for studying the total deposition velocity of aerosols (McNeary *et al.*, 2003). First, the activity concentrations and deposition fluxes of ^7Be and ^{210}Pb in ambient air can be easily determined. Second, the production rates of ^7Be and ^{210}Pb at any given site remain constant over a long period. Third, the size distributions of ^7Be and ^{210}Pb in aerosols are similar to other atmospheric particulate pollutants; thus, the deposition velocity of ^7Be and ^{210}Pb can be used to determine the deposition fluxes of these pollutants to the earth's surface.

Table 2 presents the monthly average deposition velocity of ^7Be and ^{210}Pb , together with the precipitation amount, the number of rainy days, and atmospheric fine particulate matter ($\text{PM}_{2.5}$) during the sampling period. The total deposition velocity of ^7Be at our site ranged from 0.04 to 2.06 cm s^{-1} , with an average value of 0.83 cm s^{-1} . The corresponding values for ^{210}Pb varied from 0.09 to 1.24 cm s^{-1} , with an average value of 0.62 cm s^{-1} . Moreover, both ^7Be and ^{210}Pb deposition velocity displayed seasonal variations, and they were high in summer and low in winter. Limited data are available on the simultaneously measured deposition velocity for ^7Be and ^{210}Pb . Table 3 lists the deposition velocity of ^7Be and ^{210}Pb calculated in previous studies. The deposition velocity of ^7Be and ^{210}Pb at our site were similar to those reported in Detroit, Michigan (McNeary *et al.*, 2003), and Norfolk, Virginia (Todd *et al.*, 1989). The deposition velocity of ^7Be was higher than that of ^{210}Pb , except in a study from Málaga, Spain (Dueñas *et al.*, 2005). Considering ^{210}Pb is mainly derived from the decay of ^{222}Rn , the activity concentration of ^{210}Pb in aerosol near the ground is expected to be high; thus, the total deposition velocity of ^{210}Pb is expected to be low (McNeary *et al.*, 2003). The strong positive correlation ($r=0.80$; Fig. 5) between ^7Be and ^{210}Pb deposition velocity indicates that, irrespective of the different source, both radiotracers are attached to similar size aerosols and they are removed from the atmosphere by similar mechanisms.

Table 2. Deposition velocity of ^7Be and ^{210}Pb together with precipitation amount, rainy days, and $\text{PM}_{2.5}$ concentrations in the air.

Month	^7Be velocity (cm s^{-1})	^{210}Pb velocity (cm s^{-1})	Precipitation (mm)	Rainfall days (days)	$\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$)
Jan	0.42	0.24	6.1	10	36.9
Feb	0.45	0.52	29.9	8	34.8
Mar	0.52	0.56	250.8	9	29.8
Apr	0.85	1.24	130.8	11	26.2
May	1.23	0.50	163.0	10	25.9
Jun	1.60	0.95	363.6	21	11.4
Jul	1.56	0.91	303.6	16	12.9
Aug	2.06	1.19	300.8	17	16.6
Sep	0.77	0.62	173.1	13	24.2
Oct	0.34	0.39	74.6	7	29.5
Nov	0.04	0.19	9.9	3	35.9
Dec	0.09	0.09	49.9	4	36.2

Table 3. Deposition velocity of aerosols in different locations using ^7Be and ^{210}Pb .

Location	Coordinates	Collection time	^7Be (cm s^{-1})	^{210}Pb (cm s^{-1})	Reference
Bermuda	33°N, 64°W	1979.5–1979.6	4.0	1.0	Turekian <i>et al.</i> , 1983
New Haven, Connecticut	41°N, 73°W	1977.2–1978	2.8	0.95	Turekian <i>et al.</i> , 1983
Quillayute, Washington	47°N, 124°W	1976–1977	1.0	–	Crecelius, 1981
Norfolk, Virginia	35°N, 84°W	1983–1984	1.3	0.7	Todd <i>et al.</i> , 1989
Neuherberg, Germany	48°N, 11°E	1981–1999	–	0.7–1.7 (1.0)	Winkler and Rosner, 2000
Detroit, Michigan	42°N, 83°W	1999.9–2001.02	0.17–6.05 (1.6)	0.04–3.64 (1.1)	McNeary and Baskaran, 2003
Malaga, Spain	36°N, 4°W	1992.1–1999.12	0.07–1.2 (0.4)	0.2–4.3 (1.5)	Dueñas <i>et al.</i> , 2005
Shenzhen, China	23°N, 113°E	2017.1–2017.12	0.05–2.4 (0.96)	0.22–1.44 (0.78)	This study

Fig. 5. Deposition velocity of ^7Be versus ^{210}Pb .

The deposition velocity of atmospheric aerosols depends on the particle size and meteorological conditions. ^7Be and ^{210}Pb in air are mainly attached to fine particles, and the most likely route for their removal is rainout. For a better understanding of the rainout effect, the deposition velocity of ^7Be and ^{210}Pb versus precipitation and the number of rainy days are plotted in Fig. 6. As seen in the figure, both ^7Be

and ^{210}Pb deposition velocity showed a significant positive correlation with the rainfall amount and the number of rainy days. It is suggested that precipitation is the key factor in affecting the deposition velocity of ^7Be and ^{210}Pb and plays a crucial role in removing these radionuclides from air. The deposition velocity of radionuclide is a function of its deposition fluxes and the activity concentration in air.

Fig. 6. Deposition velocity of ^7Be and ^{210}Pb in correlation with precipitation and the number of rainy days.

Atmospheric fine particulate matter, the transfer medium of particle-active radionuclides, is an important factor in controlling the activity concentration of radionuclides in air, which in turn affect their deposition velocity. As seen in Fig. 7, strong negative correlations were observed between ^7Be ($r = -0.91$) and ^{210}Pb ($r = -0.78$) deposition velocity, and $\text{PM}_{2.5}$ content in the air. Similar results were reported in Dueñas *et al.* (2005) and McNeary and Baskaran (2003), with negative correlations observed between the two deposition velocity and total suspended particle (TSP), and weak correlations between ^7Be and ^{210}Pb deposition velocity and the particulate matter collected on the filter membranes. Shenzhen features a subtropical monsoon climate; the summer monsoon occurs annually, and typhoons are common in this region. During the rainy season, the lower layer of the atmosphere is rapidly washed and the deposition velocity of ^7Be and ^{210}Pb are expected to be high; as seen from Table 2, the deposition velocity of ^7Be or ^{210}Pb displayed the highest values in summer, with an average of 1.74 and 1.02 $\text{cm}\cdot\text{s}^{-1}$, respectively. During the dry winter season, $\text{PM}_{2.5}$ concentrations in air were high ($> 30\ \mu\text{g}\cdot\text{m}^{-3}$). Considering that ^7Be and ^{210}Pb are mainly attached to fine particles ($< 1\ \mu\text{m}$) and are feebly affected by gravitational settling, the dry fallout of ^7Be or ^{210}Pb is expected to be low, which results in the low deposition velocity of these

radionuclides in winter.

CONCLUSION

The atmospheric activity concentrations and bulk deposition fluxes of ^7Be and ^{210}Pb were simultaneously measured for a whole year in Shenzhen, South China. Owing to the elevated ^{222}Rn level in this region's soil, relatively high ^{210}Pb concentrations in the atmospheric aerosols were observed. The activity of both airborne ^7Be and ^{210}Pb and the bulk deposition fluxes showed seasonal variations, with higher winter values and lower summer ones. Precipitation and the origin and pathway of air masses, in addition to atmospheric circulation, are considered the primary controlling factors for ^7Be and ^{210}Pb 's activity concentrations in the air and their deposition fluxes in the studied region. Using these two radiotracers to calculate the deposition velocity of aerosols, we discovered that the deposition velocities of ^7Be and ^{210}Pb were strongly positively correlated with the rainfall amount but negatively correlated with the airborne $\text{PM}_{2.5}$ concentration. Precipitation plays a critical role in removing these radionuclides from the air, and the deposition velocities were high during the summer and low during the winter due to the activity distributions of ^7Be and ^{210}Pb and the meteorological conditions in this region.

Fig. 7. Deposition velocity of ^7Be and ^{210}Pb in correlation with $\text{PM}_{2.5}$ in the air.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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