

# Constraining the Residence Time of Aerosols and Air-Borne Pollutants in the Atmosphere at Xiamen, China Using $^7\text{Be}$ , $^{210}\text{Pb}$ and $^{210}\text{Po}$

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**Abstract:** Atmospheric aerosol particles have been shown to affect atmospheric circulation, the hydrological cycle, the biosphere, climate and public health. In this study, the transport and residence time of aerosols and associated air-borne pollutants were investigated using the natural radionuclides  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ . The concentrations of  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  were measured in individual rain events from March 2004 to April 2006 at Xiamen, China. Results show that the activities of  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in rainwater vary from 0.1 - 18.2, 0.01 - 2.35 Bq L<sup>-1</sup> and 0.2-30.5 mBq L<sup>-1</sup> with high and low values respectively reflecting the washout and rainout effects of aerosol/pollutant scavenging. The deposition fluxes of  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  exhibit a similar seasonal variation pattern, with a maximum in spring and minimum in autumn and winter, which is attributable to the enhanced scavenging of aerosol particles from the air in spring. The mean residence time of aerosol pollutants was estimated from the  $^{210}\text{Po}/^{210}\text{Pb}$  ratios to be shorter in spring (~ 8 days) than in other seasons (~ 14 days). The variations of  $^7\text{Be}/^{210}\text{Pb}$  ratio suggest that intensified stratosphere-troposphere air exchange (STE) occurs in spring which may also contribute the increased transport and scavenging of aerosol pollutants from the air.

**Keywords:** atmospheric pollutants; aerosols;  $^7\text{Be}$ ;  $^{210}\text{Pb}$ ;  $^{210}\text{Po}$ ; tracer

## 1 Introduction

Aerosols are mostly fine-grained particles suspended in the atmosphere, with particle diameters in the range of  $10^{-9}$ - $10^{-4}$  m<sup>[1]</sup>. We are now becoming more and more aware of the importance of aerosol particles in controlling atmospheric chemistry and physics, climatology and hydrological cycle on the earth<sup>[2-3]</sup>. Aerosols at high concentration also have health impacts particularly when they are associated with other air pollutant exposures, such as organic carbon, sulfate and nitrate etc.<sup>[4-5]</sup>. Numerous studies have reported a relationship between aerosols and respiratory disease in many cities around the world<sup>[6-7]</sup>. This relationship motivated a call for studying various characteristics of aerosol pollutants, e.g., their origin and sources, size distribution and temporal variations in urban area<sup>[8-9]</sup>. The residence time of aerosol pollutants is an important parameter describing the transport and scavenging of aerosols and aerosol-associated pollutants. In this study, this parameter will be constrained using naturally occurring particle-reactive radionuclides, e.g.,  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ , and  $^7\text{Be}$  in atmosphere.

$^{210}\text{Pb}$  ( $t_{1/2}$  = 22.3 years) and  $^{210}\text{Po}$  ( $t_{1/2}$  = 138.4 days) are daughter products of  $^{222}\text{Rn}$  emanating from soil. Once formed,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  rapidly attach to ambient aero-

sols in the atmosphere and, like most of pollutants, are mainly removed from the atmosphere by wet depositions<sup>[10]</sup>. The ratio of  $^{210}\text{Po}/^{210}\text{Pb}$  has been widely used to estimate the mean residence time of aerosol pollutants in the air<sup>[11-12]</sup>.  $^7\text{Be}$  ( $t_{1/2}$  = 53.3 days), also a particle-reactive radionuclide, is produced mostly in the upper atmosphere (stratosphere) by spallation of oxygen and nitrogen by high-energy cosmic ray particles. Contrary to  $^{210}\text{Pb}$ , it is mainly vertically transported from the stratosphere to the troposphere through stratosphere -troposphere air exchange (hereafter mentioned as STE). During spring when seasonal thinning of the tropopause takes place, it results in intensified STE and higher  $^7\text{Be}$  injection<sup>[12]</sup>. It has been suggested that  $^7\text{Be}$  can be useful to trace the vertical transport of stratospheric air mass and ozone to the troposphere during STE<sup>[13]</sup>.

In this study, the concentrations of  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in rainwater were measured from individual rain events during March 2004 to April 2006 at Xiamen, China. The measurements would yield information about air mass flow from the land surface or from the upper atmosphere and the scavenging or removal process of aerosols and aerosol-associated pollutants. The data thus obtained are expected to address the following questions:

- 1) The temporal variations in fluxes of  $^7\text{Be}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  and the possible mechanisms controlling such

variations.

- 2) The source and origin, either local or remote, and scavenging behavior of aerosols pollutants.
- 3) The residence time of aerosol pollutants in the atmosphere.

## 2 Materials and method

Ten rain collectors (78.4 dm<sup>3</sup> polyethylene tank with surface area of 0.24 m<sup>2</sup>) were deployed on the roof floor of ZengChengKui building (~24 m high above sea level) at Xiamen University, Xiamen, China (24°26.132' N, 118°05.416' E, Fig.1). Sampler was opened from the end of the last precipitation event and closed in the end of the next event except the periods of the two dry deposition samples collection. The location is 50 m away from the coastline of the Taiwan Strait. The annual average precipitation is around 1100 mm and the dominant wind direction is southeast during the summer with maritime and humid air, and northeast during the winter with dry air. The climate is semitropical monsoon, with contrasting wet (mostly in February- September) and dry (mostly in October-January) seasons. Samples were collected by draining the tanks into poly-ethylene buckets after each individual precipitation events, and the tanks were cleaned repeatedly with conc. HCl in order to prevent adsorption of radionuclides on the wall of the tank. The rinses were combined with the rain sample. Then, the samples were acidified with concentrated HCl, and the chemical yield tracer (stable Pb and Be) and spike of <sup>209</sup>Po were added. Samples were stirred and then equilibrated for over 6 h.

The subsequently processes are depicted about the radionuclides of <sup>7</sup>Be, <sup>210</sup>Pb and <sup>210</sup>Po concentrated by co-precipitation with Fe hydroxides. Shortly, 60~90mg Fe (in the form of FeCl<sub>3</sub>) was added to <sup>7</sup>Be, <sup>210</sup>Pb and <sup>210</sup>Po samples. These radionuclides were concentrated by co-precipitation by raising the pH to 8~10 with NH<sub>4</sub>OH. The Fe(OH)<sub>3</sub> precipitate was allowed to settle for at least 24 h, and then the precipitate was removed separated by centrifuge. Finally, the samples were frozen-dried before radio-analysis. In the dry season, only dry deposition samples were collected per month from October 2004 to November 2004. The samples were processed and measured for <sup>7</sup>Be and <sup>210</sup>Pb according to the method described by Jia et al<sup>[14]</sup>. In brief, the sampler was cleaned with 0.2 M HCl and pure water to obtain the dry fallout. After collection, the chemical yield tracer (stable Pb and Be) and FeCl<sub>3</sub> were added. Then the dry fallout and Fe(OH)<sub>3</sub> precipitates were removed by centrifugal method.

The activities of <sup>210</sup>Pb (46.5 keV) and <sup>7</sup>Be (477.6 keV) nuclei in the co-precipitation sample were measured by a low-level gamma spectrometry with HPGe detector (Canberra GX30- 20), having resolution of 1.91keV at

1.33 MeV of <sup>60</sup>Co. The efficiency calibration and the calculation of <sup>7</sup>Be and <sup>210</sup>Pb concentrations have been previously described<sup>[15]</sup>. An aliquot of the sample was also measured for stable Be and Pb using the gravimetric and the atomic absorption spectroscopy (AAS) method, respectively, to determine the chemical yield.

The analytical procedures for <sup>210</sup>Po were described in Yang et al<sup>[16]</sup>. In brief, the Fe(OH)<sub>3</sub> precipitates were dissolved with 1:1 HCl, and then transferred into a 100 cm<sup>3</sup> Teflon beaker. Ascorbic acid was then added until the yellow color (Fe<sup>3+</sup>) disappeared. 1ml of 20% hydroxylamine hydrochloride and 1 cm<sup>3</sup> of 25% sodium citrate solutions were added and the pH of solution was adjusted to 1.5. Polonium was deposited on a silver disc at 85-90 °C by using a magnetic stirrer for 4 h, then the silver disc was rinsed with pure water and ethanol, and dried naturally. The activities of <sup>210</sup>Po and <sup>209</sup>Po were determined by alpha spectrometer (Octète<sup>TM</sup> PC, EG&G ORTEC). All activities were corrected for radioactivity decay to the mid-point of collection. The propagated uncertainties are smaller than 15% for <sup>7</sup>Be and 20% for <sup>210</sup>Pb and <sup>210</sup>Po.



Figure 1. Location for sampling collection

## 3 Results and discussion

### 3.1 The activities and fluxes of radionuclide

Samples from 68 rain events were collected from March 2004 to April 2006. The activities of <sup>7</sup>Be, <sup>210</sup>Pb have a wide range, 0.1 - 18.2 Bq L<sup>-1</sup> with a mean value of 3.2±0.4 Bq L<sup>-1</sup> and 0.01 - 2.35 Bq L<sup>-1</sup> with a mean value of 0.35±0.04 Bq L<sup>-1</sup>, respectively. These activities are comparable to those found in other areas, i.e. 0.35-24.0 Bq L<sup>-1</sup> for <sup>7</sup>Be, and 0.03-2.67 Bq L<sup>-1</sup> for <sup>210</sup>Pb<sup>[17-18]</sup>. The activities of <sup>210</sup>Po are 0.2-30.5 mBq L<sup>-1</sup>, with a mean value of 7.2±1.3 mBq L<sup>-1</sup>, which are consistent with those observed in Australia, i.e., 0.8-67 mBq L<sup>-1</sup><sup>[19]</sup>.

The radionuclide concentrations in precipitation de-

pend upon various factors, such as the aerosol particle size, the captured mechanism by precipitation (washout or rainout), the amount of precipitation and the vertical mixing of air masses<sup>[20]</sup>. To investigate the rainfall effect on the concentrations of <sup>7</sup>Be and <sup>210</sup>Pb, we plot the nuclide concentrations versus rainfall in the individual rain events (Fig. 2), showing higher concentrations of <sup>7</sup>Be and <sup>210</sup>Pb at low rainfalls (group A) and a slight decrease with increasing rainfall (group B). The high concentrations observed in low rainfalls (group A) indicate the preponderance of washout and/or possible contribution of dry deposition (to be discussed later). Under heavier rainfall, the aerosol pollutants in lower atmosphere is quickly washed out and the relative importance of washout becomes negligible (group B) compared to the rainout. Despite different origins of <sup>7</sup>Be and <sup>210</sup>Pb, the relationships between the nuclide concentrations and rainfall are very similar (Fig. 2), suggesting the similar scavenging mechanisms for these nuclides and pollutants (i.e., the similar rainout or washout effect).

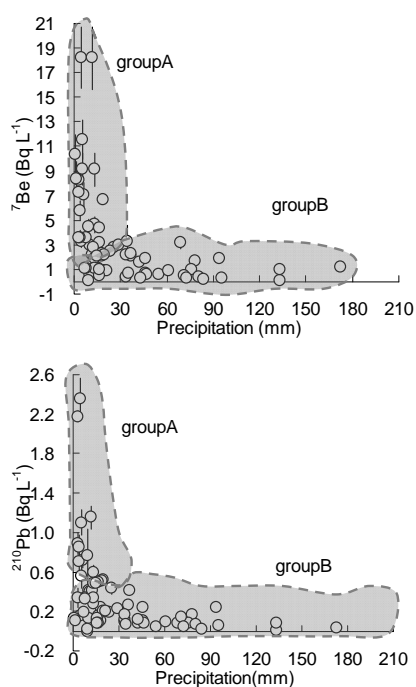


Figure 2. Correlation between the activities of <sup>7</sup>Be and <sup>210</sup>Pb and rainfall

Seasonal variation patterns in the fluxes of <sup>7</sup>Be, <sup>210</sup>Pb and <sup>210</sup>Po are shown in Fig. 3. The annual fluxes of <sup>7</sup>Be vary from 0.01 to 1.12 Bq cm<sup>-2</sup> y<sup>-1</sup> with an annual mean value of 0.29±0.04 Bq cm<sup>-2</sup> y<sup>-1</sup>, which are slightly lower than that observed at other coastal locations in Texas (~0.39 Bq cm<sup>-2</sup> y<sup>-1</sup>) and New Haven (~0.38 Bq cm<sup>-2</sup> y<sup>-1</sup>)<sup>[21-22]</sup>, but significantly higher than the global mean value of 0.10 Bq cm<sup>-2</sup> y<sup>-1</sup><sup>[23]</sup>. This discrepancy may be

attributed to the regional difference in <sup>7</sup>Be production and/or STE transport.

The annual depositional pattern of <sup>7</sup>Be is quite similar in the past 2 years (see solid circles, Fig. 3). After monthly averaging, higher values of <sup>7</sup>Be flux occur in spring (March-May), medium values in summer (June-August), and lower in autumn (September- November) and winter (December-February). The higher deposition fluxes of <sup>7</sup>Be in spring may be caused by the intensified STE, which often happens in spring when the tropopause folds at mid-latitudes and enhances transfer of stratospheric <sup>7</sup>Be into the troposphere<sup>[23-24]</sup>. The similar STE enhancement is also seen from other two cosmogenic radionuclides tracers, <sup>33</sup>P and <sup>32</sup>P, which show higher <sup>33</sup>P/<sup>32</sup>P ratio in the spring samples<sup>[25]</sup>. In other seasons, the depositional fluxes of <sup>7</sup>Be are more likely to be controlled by the rainfall intensity.

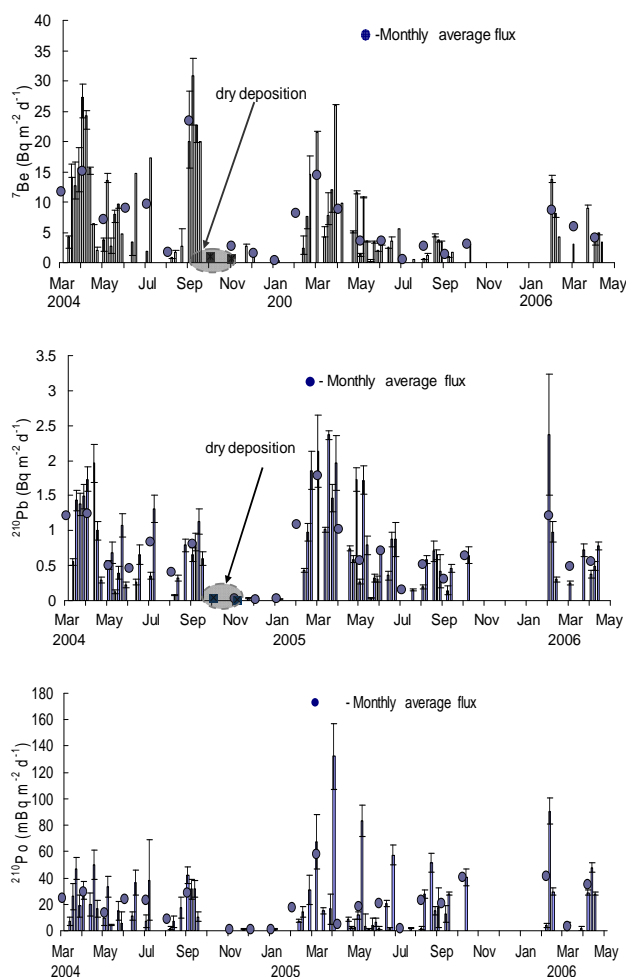


Figure 3. Daily (shaded bars) and monthly average (circles) fluxes of <sup>7</sup>Be, <sup>210</sup>Pb and <sup>210</sup>Po at Xiamen. The fluxes are derived by multiplying the cumulative rainfall by the specific activity found in the sample and dividing by the number of days in the collection period.

The annual fluxes of  $^{210}\text{Pb}$  are estimated to range from 0.7 - 86.3  $\text{mBq cm}^{-2} \text{y}^{-1}$ , or about one to two orders of magnitude higher than those of  $^{210}\text{Po}$  (i.e., 0.03-3.31  $\text{mBq cm}^{-2} \text{y}^{-1}$ ). An average flux of  $28.3 \pm 4.2 \text{ mBq cm}^{-2} \text{y}^{-1}$  is estimated for  $^{210}\text{Pb}$ , which is comparable to the global average  $^{210}\text{Pb}$  deposition of 21.8 - 31.8  $\text{mBq cm}^{-2} \text{y}^{-1}$ [26]. The temporal patterns of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  monthly fluxes show a larger flux in spring than in other seasons, consistent with those of  $^7\text{Be}$ . The maximum fluxes in spring are attributed to the following factors:

- 1) The average flux of  $^{222}\text{Rn}$  from continental soil surface to atmosphere is approximately 15-48  $\text{mBq m}^{-2} \text{s}^{-1}$ [27], in a close balance with the average removal flux of  $^{210}\text{Pb}$  of about 21.9-70  $\text{mBq m}^{-2} \text{y}^{-1}$ [28]. Pan et al.[29] have reported that the activities of  $^{222}\text{Rn}$  in Fujian Province (including Xiamen area) were highest in China (48.7  $\text{Bq m}^{-3}$ ). These higher emanation of  $^{222}\text{Rn}$  from soil might produce higher fluxes of  $^{210}\text{Pb}$ , 86.3  $\text{mBq m}^{-2} \text{y}^{-1}$  (the largest value in our rain samples), for example.
- 2) Additionally, high  $^{210}\text{Pb}$  deposition flux might link with high aerosol concentrations in spring. Chen et al.[30] found that the annually averaged aerosol optical depth (AOD) was higher in spring than in other seasons in the Taiwan Strait from 2002 to 2004, indicating increased aerosol abundance in the atmosphere which would make more particle-active nuclides and pollutants to be scavenged.
- 3) In spring, higher  $^7\text{Be}$  flux is found in the rainwater, which shows that intensified STE in spring increases the scavenging efficiency of aerosols, making a clean sweep of all kinds of aerosol pollutants from the atmosphere. The higher  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  scavenging fluxes may occur in this event.

In summer, the humid air masses above Xiamen are derived chiefly from the adjacent oceans[31], which are typically depleted in  $^{222}\text{Rn}$  (e.g.,  $\sim 0.72 \text{ mBq m}^{-2} \text{s}^{-1}$ ) [27,32] compared to the continental areas (e.g., 15-48  $\text{mBq m}^{-2} \text{s}^{-1}$ ). Thus, the air masses are also depleted in its daughter product, including  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , resulting in the lower fluxes of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in summer rainfall. During the fall-winter, the depositional fluxes of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are lowest due to the lowest rainfall.

The importance of dry deposition in scavenging radionuclides and pollutants is evaluated by collecting two single dry deposition samples in October and November 2004, respectively. The dry depositional fluxes for  $^7\text{Be}$  are measured to vary between 0.02 and 0.03  $\text{Bq cm}^{-2} \text{y}^{-1}$  (the square in Fig.3), which account for only  $\sim 8\%$  of the total annual depositional fluxes of  $\sim 0.29 \text{ Bq cm}^{-2} \text{y}^{-1}$ . The  $^{210}\text{Pb}$  dry deposition collected in October and November 2004 varies from 1.67 to 3.83  $\text{mBq cm}^{-2} \text{y}^{-1}$  (the square in Fig. 3), or about 10% of the total depositions during the study periods. To conclude, the dry depositions of  $^{210}\text{Pb}$

and  $^7\text{Be}$  could not be a major contributor to the annual fluxes and the wet deposition have to be a dominant process in scavenging and removal of aerosol pollutants from the atmosphere.

### 3.2 Seasonal variation of $^7\text{Be}/^{210}\text{Pb}$ ratios

The measured activity ratios of  $^7\text{Be}/^{210}\text{Pb}$  in rain water vary between 2.06 and 40.0, with a mean value of  $10.9 \pm 2.3$  (Fig. 4) comparable to the global average value of  $\sim 12.7$ [26]. Figure 4 shows that the ratios are particularly higher during spring or during the heavy rainfall events (e.g., typhoon) in other seasons. The higher  $^7\text{Be}/^{210}\text{Pb}$  ratios further confirms that STE is intensified in spring or when typhoon occurs, which may contribute the increased vertical transport and scavenging of aerosol pollutants from the air.

In summer, the high ratios associated with the heavy rainfall may be attributed to the intrusion of oceanic air masses (which is depleted in  $^{210}\text{Pb}$ ). In fall and winter, when the rainfall is generally low, the washout effect becomes more important, causing more efficient scavenging of the nuclides (e.g.,  $^{210}\text{Pb}$ ) and aerosol pollutants from the lower atmosphere and the low  $^7\text{Be}/^{210}\text{Pb}$  ratios observed in rainwater [33].

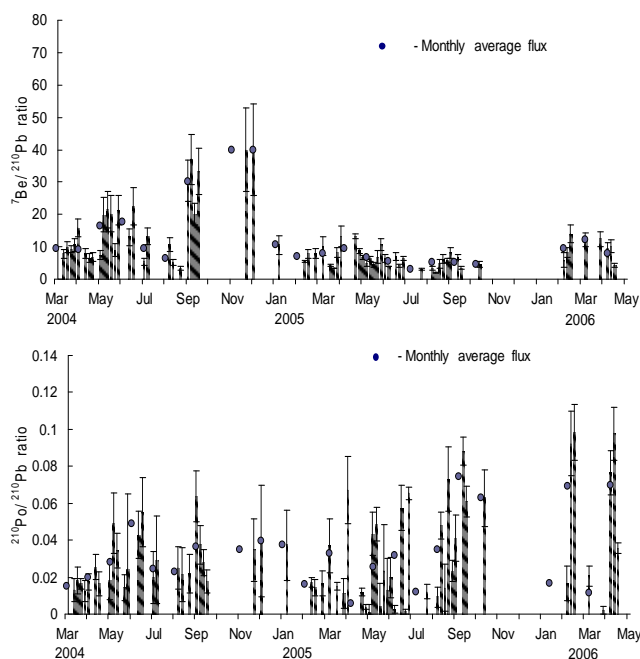


Figure 4. Ratios of  $^7\text{Be}/^{210}\text{Pb}$  and  $^{210}\text{Po}/^{210}\text{Pb}$  in rain events (shaded bars) and monthly average ratios (circles) at Xiamen.

### 3.3 The residence time of aerosols

The activity ratios of  $^{210}\text{Po}/^{210}\text{Pb}$  vary 0.01 to 0.10 (mean value =  $0.03 \pm 0.01$ , Fig.4). These ratios are in the



lower range of those reported previously for other areas (i.e., ~ 0-0.2)<sup>[12]</sup>. Yang et al.<sup>[34]</sup> have observed a slightly higher ratios of 0.06 to 0.11 (averaging ~0.09) at the same study area. This discrepancy may be explained by the variations in rainfall and/or in the origins of air masses. High ratios (e.g., <sup>210</sup>Po/<sup>210</sup>Pb ≥ 1) are reported in the literature for some areas and are generally attributed to the additional sources of <sup>210</sup>Po, such as soil particles from dust storms, coal-burning from the power plants, <sup>210</sup>Po emission from volcano eruptions etc.<sup>[12,35]</sup>. Apparently, our data suggest no significant contributions of <sup>210</sup>Po from these additional sources. As the observed <sup>210</sup>Po/<sup>210</sup>Pb ratios in our samples are much smaller than the secular equilibrium value (=1) in soil particles, the contribution of <sup>210</sup>Po by local re-suspended soil particles during the dry periods (autumn and winter) will be negligible.

Using the measured ratio of <sup>210</sup>Po/<sup>210</sup>Pb in rainwater, it is possible to estimate the mean residence time of aerosol pollutants<sup>[12]</sup>. As shown earlier, <sup>210</sup>Pb and its daughter product <sup>210</sup>Po are particle-active and are mainly scavenged from the atmosphere via wet precipitation following their adsorptions onto the aerosol particles. Assuming that (1) all the <sup>210</sup>Pb and its daughter products in the atmosphere are carried by aerosols and scavenged by rainfall precipitation and (2) the only source of <sup>210</sup>Po is the radioactive decay of <sup>210</sup>Pb in the atmosphere, the following equation can be used to describe the change rate of the daughter nuclide (<sup>210</sup>Po) concentration in the air:

$$dN_d / dt = \lambda_p N_p - N_d (\lambda_d + \lambda_r) \quad (1)$$

where  $N_p$  and  $N_d$  refer to the concentrations of parent and daughter products,  $\lambda_p$  and  $\lambda_d$  are their respective reciprocal lifetimes,  $\lambda_r$  is the removal rate constant. Under a steady-state condition, the <sup>210</sup>Po growth is balanced by their scavenging and radioactive decay. So the residence time ( $\tau = 1/\lambda_r$ ) can be determined from Eq. (2)<sup>[36]</sup>:

$$\tau = \left[ -b + (b^2 - 4ac)^{1/2} \right] / 2a \quad (2)$$

where  $a = A_{pb} - A_{po}$ ,  $b = -A_{po}(\tau_{Bi} + \tau_{po})$ ,  $c = -A_{po} \tau_{Bi} \tau_{po}$ , and  $\tau_{po}$  and  $\tau_{Bi}$  are their respective mean lifetimes,  $A_{pb}$  and  $A_{po}$  are the activities of <sup>210</sup>Pb and <sup>210</sup>Po respectively.

Using Eq. (2), the residence time of aerosol pollutants is estimated to range from 1.4 to 28.1 days, with the smaller values mostly occurring in spring (~8 days) or in the heavy-rainfall events of other seasons, and larger values in other seasons (~14 days). In literature, higher estimates have been made based on <sup>210</sup>Po/<sup>210</sup>Pb ratios for the atmospheric aerosols in other mid-latitude areas: e.g., ~ 13 - 33 days at Colorado and ~ 33 - 66 days for many cities in North America<sup>[36-37]</sup>. Apparently, the shorter residence times of aerosol pollutants in the Xiamen area

are likely to be associated with the higher and more frequent rainfall precipitations.

It has been reported that for the same samples, the residence time of aerosols estimated from <sup>210</sup>Po/<sup>210</sup>Pb is generally longer than that based on <sup>210</sup>Bi/<sup>210</sup>Pb<sup>[37]</sup>. This difference can be attributed to the fact that relative to <sup>210</sup>Po, <sup>210</sup>Bi is more like to be enriched in the lower atmosphere where coarser-grained aerosol particles with shorter residence time are dominant<sup>[37,38]</sup>. This difference becomes more obvious in the lower atmosphere or in the case of sand storms where there may be a broader spectrum of aerosol grain size. The difference may be diminished in the upper atmosphere where the aerosols may be dominated by fine-grained particles. Taking this factor into account, we suggest that the <sup>210</sup>Po/<sup>210</sup>Pb-derived residence time in this study is more likely to reflect the residence time of the fine-grained aerosols and aerosol-associated pollutants in the atmosphere.

#### 4. Conclusions

This study investigated the temporal variations of the concentrations and fluxes of <sup>7</sup>Be, <sup>210</sup>Pb, and <sup>210</sup>Po in rainwater and reached the following conclusions:

- (1) The wet precipitation plays an important role in scavenging aerosols and aerosol-bounded radionuclides and pollutants from the atmosphere, with the washout being a dominant scavenging mechanism during the small-rainfall events.
- (2) The depositional fluxes of <sup>7</sup>Be, <sup>210</sup>Pb and <sup>210</sup>Po radionuclides show a similar seasonal variation pattern, with a maximum in spring and minimum in autumn and winter, in spite of their different origins and sources. This similarity indicates that the radionuclides are scavenged by the same mechanisms, i.e., through rapid adsorption of radionuclides onto aerosol particles followed by rainfall scavenging of aerosol-bounded radionuclides and pollutants.
- (3) The contributions from dust storms, coal-burning from the power plants, volcano emission, etc. may not be important sources for radionuclides <sup>7</sup>Be, <sup>210</sup>Pb and <sup>210</sup>Po of the rain waters in the study area.
- (4) The ratios of <sup>210</sup>Pb/<sup>7</sup>Be show that intensified STE occurs in spring, which may cause more efficient scavenging of aerosol pollutants from the air.
- (5) The residence time of aerosol pollutants was estimated to range from 1.4 to 28.1 days, showing smaller values in spring than in other seasons, which are also consistent with the increase in radionuclide fluxes and STE intensity in spring.

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