*Aerosol and Air Quality Research*, 16: 105–113, 2016

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2015.03.0182

# Atmospheric Deposition of $^{7}$Be in the Southeast of China: A Case Study in Xiamen

**Lihao Zhang[1,2], Weifeng Yang[1,2*], Min Chen[1,2], Zhou Wang[2], Peng Lin[3], Ziming Fang[2], Yusheng Qiu[1,2], Minfang Zheng[2]**

[1] *State Key Laboratory of Marine Environmental Science, Xiamen 361005, China*
[2] *College of Ocean and Earth Sciences, Xiamen University, Xiamen 361102, China*
[3] *School of Freshwater Sciences, University of Wisconsin-Milwaukee, Milwaukee, WI 53204, USA*

## ABSTRACT

Atmospheric deposition of $^{7}$Be was measured at a time-series station in the southeast of China (Xiamen) from 2011 to 2013. The deposition fluxes of $^{7}$Be ranged from 0.05 Bq m$^{-2}$ d$^{-1}$ to 7.42 Bq m$^{-2}$ d$^{-1}$, averaging 1.87 ± 0.10 Bq m$^{-2}$ d$^{-1}$. High fluxes occurred in months with northeast monsoon, and low values were observed in the southwest monsoon prevailing months. The significant correlations between $^{7}$Be deposition and precipitation, existing in both northeast and southwest monsoon seasons, suggested the dominant removal of atmospheric $^{7}$Be via precipitation. However, the correlations showed a large slope for the northeast monsoon season, indicating higher $^{7}$Be contents in the atmosphere during the northeast monsoon prevailing months, supported by the precipitation-normalized $^{7}$Be and the temporal variability of $^{7}$Be/$^{210}$Pb ratios. Such a scenario revealed more intensive exchange of air mass between the stratosphere and troposphere during the northeast monsoon prevailing months. Together with the high pollutant concentrations in ambient air observed in these seasons, the results indicated that the pollutants in Xiamen might enter into the upper troposphere via vertical air mass exchange.

***Keywords:*** $^{7}$Be; Residence time; Monsoon; Aerosol; Haze.

## INTRODUCTION

$^{7}$Be ($T_{1/2}$ = 53.4 d), a natural radionuclide, is produced through the reaction of cosmic-ray-spallation with nitrogen and oxygen in the stratosphere and upper troposphere (Lal *et al.*, 1958). Its deposition to the ground is usually influenced by latitude, sunspot numbers, and the intensity of geomagnetic field (Masarik *et al.*, 1999; Nagai *et al.*, 2000; Usoskin *et al.*, 2008; Chao *et al.*, 2012). Since the residence time of $^{7}$Be in the stratosphere is much longer than its half-life (Turekian *et al.*, 1983), $^{7}$Be usually maintains in a nearly steady state with respect to its production and removal, resulting in a nearly constant content of $^{7}$Be in the stratosphere (Dutkiewicz *et al.*, 1985). $^{7}$Be enters into the troposphere either through Brewer-Dobson circulation or through air mass exchange mainly in the mid-latitude regions during spring when the troposphere becomes thin (Kim *et al.*, 1998). Due to its particle reactivity, $^{7}$Be is readily adsorbed onto aerosols after production, and then removed from the atmosphere through both wet and dry deposition. Previous studies reported that rainfall is the predominant pathway of $^{7}$Be removal from the atmosphere (Young *et al.*, 1980; Baskaran *et al.*, 1993), and the deposition flux of $^{7}$Be in the mid-latitude regions is overall higher than that in the low-latitude regions on a global scale (Lal and Peters, 1967). However, the $^{7}$Be deposition fluxes showed significantly local and seasonal variability at a specific site, depending on both the regional climate conditions and air mass exchange between the stratosphere and troposphere. For example, the annual deposition flux was 3,783 Bq m$^{-2}$ yr$^{-1}$ at New Haven (USA), which was much higher than Westwood (USA) locating at the similar latitude (717 Bq m$^{-2}$ yr$^{-1}$) (Walton and Fried, 1962; Turekian *et al.*, 1983). In contrast, Nankang in North Taiwan and Loess Plateau in China have comparable annual deposition fluxes of 1,833 Bq m$^{-2}$ yr$^{-1}$ and 1,759 Bq m$^{-2}$ yr$^{-1}$ respectively, though the two sites have different latitudes (Su *et al.*, 2003; Zhang *et al.*, 2013b).

$^{7}$Be has been used to trace the aerosol transport processes (Viezee *et al.*, 1980; Young *et al.*, 1980; Garger, 1994). The ratio of $^{7}$Be/$^{32}$P has been used to quantify the residence time of aerosol (Lal and Zutshi, 1960; Benitez-Nelson *et al.*, 1999). The isotopic ratio of $^{10}$Be/$^{7}$Be was used to investigate the exchange of $O_3$ and $H_2O$ between the stratosphere and troposphere (Zheng *et al.*, 2011). Since pollutants cycling in the atmosphere, including residence and dispersal, are closely related to these processes, $^{7}$Be would provide valuable insights into the pollutants cycling on a timescale

* Corresponding author.
*E-mail address:* wyang@xmu.edu.cn

corresponding to its half-life. In China, a few studies on $^{7}$Be have been reported in several cities, i.e., Shanghai and Guizhou (Lee *et al.*, 2004; Du *et al.*, 2008; Kong, 2012). During the past three years, the construction and auto industry have led to heavy haze in most Chinese cities. Xiamen, one of the least polluted cities in southeastern China, also experienced the hazy weather recently (Zhang *et al.*, 2013a; Lee *et al.*, 2014). However, we have little understanding of the haze diffusion in the southeast of China.

In the present study, a time-series station at Xiamen, located in the southeast of China, was selected to examine the deposition of $^{7}$Be and its potential application for constraining the diffusion and transport of air pollutants. We analyzed the intra- and inter-annual atmospheric deposition of $^{7}$Be, as well as the factors influencing its temporal variability. Additionally, the precipitation-normalized $^{7}$Be was applied to reveal the variation of $^{7}$Be contents in the atmosphere. Together with recorded hazy weather, $^{7}$Be could indicate potential dispersal of air pollutants in the southeast of China over a large spatial scope.

## MATERIAL AND METHODS

Time-series atmospheric deposition of $^{7}$Be was collected from June 2011 to December 2013 on the roof of Zengchengkui Building (24.5°N, 118°E) in Xiamen University. The durations of sampling were less than half a month for most samples, depending on the precipitation events (Table 1). To minimize the possible evaporation of rainwater, samples were gathered instantly after the rainfall events. Deposition of $^{7}$Be, including dry and wet deposition, was collected using a clean polyethylene box with an open area of 0.437 m$^2$ on its top, which allowed it to collect atmospheric deposition as much as possible. The tank below the equipment has a volume of 30 L. Before sampling, the sampler was sequentially cleaned with 2% HCl and Milli-Q water. Given sample collection, the bulk deposition was transferred into a cleaned polyethylene bottle. Part of $^{7}$Be may adsorb onto the equipment walls because it exists as BeO or $Be(OH)_2$ (Al-Azmi *et al.*, 2001). Be dissolves in acid solution. Thus, 2% HCl and Milli-Q water were used to wash $^{7}$Be possibly adsorbed on the box walls. And 6 mol L$^{-1}$ HCl was added into the combined solution until the pH value was less than 2.0, followed by the addition of $Fe^{3+}$ carrier. After 24 h, the solution was adjusted to a pH value of 8.5 with ammonia solution for the precipitation of iron hydroxide. After centrifugation and dryness of precipitate, the sample was transferred to a counting vial for gamma counting by a Canberra ultra-high purity germanium detector. The counting efficiency of $^{7}$Be was calibrated using standard material as described in Liu *et al.* (2001). Activities of $^{7}$Be were corrected to the mid-sampling time. Daily deposition flux (Bq m$^{-2}$ d$^{-1}$) was calculated based on the sampling interval and collection area. The daily precipitation (mm d$^{-1}$) was calculated according to the total rainfall and collection time. The average precipitation (mm d$^{-1}$) and deposition flux (Bq m$^{-2}$ d$^{-1}$) of $^{7}$Be in each month were calculated based on the monthly integrated equation (McNeary and Baskaran, 2007) and the specific days in each month:

$$J = \left( \sum_{i=1}^{k} n_i F_i \right) \Big/ m_d \quad (1)$$

where $J$ represents the average deposition flux of $^{7}$Be or precipitation in each month; the term $n_iF_i$ is the flux of $^{7}$Be or rainfall for a specific sampling interval, which was calculated by the specific flux or rainfall multiplied by the days during that interval; $i$ denotes the number of sampling intervals in a month; $m_d$ is the days in each month.

## RESULTS

The daily deposition fluxes of $^{7}$Be varied from 0.03 Bq m$^{-2}$ d$^{-1}$ to 45.1 Bq m$^{-2}$ d$^{-1}$ with an average of 3.49 ± 0.41 Bq m$^{-2}$ d$^{-1}$ (Table 1 and Fig. 1). On monthly timescale, the deposition fluxes of $^{7}$Be ranged from 0.05–7.42 Bq m$^{-2}$ d$^{-1}$ (Table 2), averaging 1.87 ± 0.10 Bq m$^{-2}$ d$^{-1}$, comparable to the values of 0.2–10.6 Bq m$^{-2}$ d$^{-1}$ in the low-latitude regions (e.g., Igarashi *et al.*, 1998; Hirose *et al.*, 2004). The annual fluxes of $^{7}$Be were 686.9 Bq m$^{-2}$ yr$^{-1}$ and 834.1 Bq m$^{-2}$ yr$^{-1}$ in 2012 and 2013 respectively, showing around 20% difference. Such difference revealed somewhat inter-annual variation of $^{7}$Be deposition in Xiamen, similar to the deposition of $^{210}$Po and $^{210}$Pb (Wang *et al.*, 2014).

The deposition of $^{7}$Be exhibited an intra-annual fluctuation (Fig. 2), with high fluxes (up to 7.42 Bq m$^{-2}$ d$^{-1}$) from December to May, mainly corresponding to the northeast monsoon prevailing months. In contrast, the low fluxes were observed from June to November with the lowest value in October. The higher deposition fluxes of cosmogenic $^{33}$P ($T_{1/2}$ = 25.3 d) and $^{32}$P ($T_{1/2}$ = 14.3 d) were reported from December to April in Xiamen (e.g., Zhang, 2004; Chen, 2006), showing similar intra-annual variation pattern to $^{7}$Be in the present study (Fig. 2).

## DISCUSSION

### *Role of Precipitation in Removing $^{7}$Be*

During the northeast monsoon seasons (Dec.–Apr.), the significant linear correlation between the daily deposition fluxes of $^{7}$Be and precipitation ($r^2$ = 0.91, $p < 0.0001$, Fig. 3) suggested the atmospheric deposition of $^{7}$Be was tightly related to rainfall. Similar correlation was also observed from May to November ($r^2$ = 0.57, $p < 0.0001$). Hence, the precipitation was the dominant removal pathway of $^{7}$Be out of the atmosphere in Xiamen. In contrast, the daily dry deposition (averaging 0.13 ± 0.04 Bq m$^{-2}$ d$^{-1}$) only accounted for around 3.8% of the total deposition, providing additional supports for the removal of $^{7}$Be mainly via precipitation. This observation was similar to reports obtained in other regions (e.g., Young *et al.*, 1980; Baskaran *et al.*, 1993; Kim *et al.*, 2000; McNeary and Baskaran, 2003).

Although the daily deposition of $^{7}$Be was dominantly regulated by precipitation, there was evident difference in the influence of precipitation magnitude on $^{7}$Be deposition between the northeast and southwest monsoon seasons. As shown in Fig. 3, the slope of the correlation line (0.83) for the northeast monsoon months was four times the slope

**Table 1.** Daily deposition of $^{7}$Be and precipitation (P) in Xiamen from May 2011 to December 2013.

| Sampling date | $^{7}$Be flux (Bq m$^{-2}$ d$^{-1}$) | P (mm d$^{-1}$) | Sampling date | $^{7}$Be flux (Bq m$^{-2}$ d$^{-1}$) | P (mm d$^{-1}$) |
|---|---|---|---|---|---|
| 2011/05/17–2011/05/23 | 3.31 ± 0.29 | 9.28 | 2012/08/02–2012/08/05 | 1.72 ± 0.15 | 2.40 |
| 2011/05/23–2011/05/24 | 2.23 ± 0.44 | 10.20 | 2012/08/05–2012/08/12 | 0.97 ± 0.08 | 5.04 |
| 2011/05/24–2011/06/02 | 0.75 ± 0.08 | 1.89 | 2012/08/12–2012/08/18 | 4.95 ± 0.40 | 12.97 |
| 2011/06/02–2011/06/14 | 0.63 ± 0.06 | 0.88 | 2012/08/18–2012/08/29 | 0.58 ± 0.05 | 0.79 |
| 2011/06/14–2011/06/21 | 0.10 ± 0.03 | 0.03 | 2012/08/29–2012/09/04 | 2.13 ± 0.18 | 1.60 |
| 2011/06/21–2011/07/07 | 2.51 ± 0.20 | 5.05 | 2012/09/04–2012/09/11 | 3.34 ± 0.28 | 6.31 |
| 2011/07/07–2011/07/16 | 1.62 ± 0.13 | 4.52 | 2012/09/11–2012/09/25 | 0.50 ± 0.04 | 0.75 |
| 2011/07/16–2011/07/17 | 2.15 ± 0.30 | 21.27 | 2012/09/25–2012/10/24 | 0.09 ± 0.01 | 0 |
| 2011/07/17–2011/07/18 | 1.85 ± 0.21 | 24.67 | 2012/10/24–2012/10/31 | 1.24 ± 0.11 | 1.59 |
| 2011/07/18–2011/08/10 | 0.94 ± 0.08 | 3.77 | 2012/10/31–2012/11/12 | 0.23 ± 0.02 | 0 |
| 2011/08/10–2011/09/05 | 3.16 ± 0.26 | 5.63 | 2012/11/12–2012/11/26 | 1.13 ± 0.09 | 1.20 |
| 2011/09/05–2011/09/21 | 0.11 ± 0.01 | 0 | 2012/11/26–2012/11/30 | 4.52 ± 0.37 | 23.68 |
| 2011/09/21–2011/09/28 | 0.16 ± 0.02 | 0 | 2012/11/30–2012/12/02 | 4.49 ± 0.38 | 18.00 |
| 2011/09/28–2011/10/12 | 0.19 ± 0.02 | 0.11 | 2012/12/02–2012/12/07 | 3.75 ± 0.31 | 1.73 |
| 2011/10/26–2011/11/07 | 0.08 ± 0.03 | 0 | 2012/12/07–2012/12/21 | 0.75 ± 0.06 | 0.12 |
| 2011/11/07–2011/11/11 | 2.70 ± 0.37 | 22.03 | 2012/12/21–2012/12/29 | 2.37 ± 0.19 | 0.86 |
| 2011/11/11–2011/11/20 | 4.63 ± 0.42 | 13.61 | 2012/12/29–2013/01/02 | 4.64 ± 0.38 | 5.72 |
| 2011/11/20–2011/12/20 | 0.15 ± 0.02 | 0.03 | 2013/01/02–2013/01/17 | 0.08 ± 0.01 | 0 |
| 2011/12/20–2012/01/19 | 2.14 ± 0.17 | 1.45 | 2013/01/17–2013/01/26 | 0.18 ± 0.03 | 0 |
| 2012/01/19–2012/02/19 | 0.91 ± 0.07 | 1.33 | 2013/01/26–2013/02/06 | 2.46 ± 0.20 | 1.41 |
| 2012/02/19–2012/03/15 | 4.32 ± 0.35 | 1.80 | 2013/02/06–2013/02/21 | 0.23 ± 0.02 | 0 |
| 2012/03/15–2012/03/31 | 0.61 ± 0.06 | 0.16 | 2013/02/21–2013/02/27 | 1.16 ± 0.10 | 0.42 |
| 2012/03/31–2012/04/11 | 2.90 ± 0.24 | 1.35 | 2013/02/27–2013/03/03 | 1.18 ± 0.12 | 0 |
| 2012/04/11–2012/04/16 | 1.51 ± 0.13 | 1.37 | 2013/03/03–2013/03/20 | 0.59 ± 0.5 | 0 |
| 2012/04/16–2012/04/18 | 5.78 ± 0.48 | 4.10 | 2013/03/20–2013/03/27 | 7.10 ± 0.57 | 3.99 |
| 2012/04/18–2012/04/21 | 6.39 ± 0.52 | 7.03 | 2013/03/27–2013/03/29 | 19.37 ± 1.56 | 12.87 |
| 2012/04/21–2012/04/26 | 4.58 ± 0.38 | 3.06 | 2013/03/29–2013/04/03 | 6.26 ± 0.51 | 3.89 |
| 2012/04/26–2012/04/27 | 45.01 ± 3.67 | 52.20 | 2013/04/03–2013/04/06 | 30.94 ± 2.50 | 34.32 |
| 2012/04/27–2012/04/28 | 15.30 ± 11.27 | 10.10 | 2013/04/06–2013/04/12 | 5.96 ± 0.48 | 3.18 |
| 2012/04/28–2012/05/02 | 2.33 ± 0.20 | 2.33 | 2013/04/12–2013/04/21 | 2.84 ± 0.23 | 0.95 |
| 2012/05/02–2012/05/04 | 5.82 ± 0.49 | 5.35 | 2013/04/21–2013/05/01 | 5.51 ± 0.44 | 4.23 |
| 2012/05/04–2012/05/05 | 8.47 ± 0.74 | 6.86 | 2013/05/01–2013/05/17 | 8.57 ± 0.72 | 14.59 |
| 2012/05/05–2012/05/10 | 1.38 ± 0.12 | 2.40 | 2013/05/17–2013/06/13 | 3.91 ± 0.34 | 10.17 |
| 2012/05/10–2012/05/14 | 1.53 ± 0.14 | 1.77 | 2013/06/13–2013/07/02 | 1.65 ± 0.16 | 3.95 |
| 2012/05/14–2012/05/16 | 1.60 ± 0.15 | 10.85 | 2013/07/02–2013/07/17 | 0.03 ± 0.06 | 15.16 |
| 2012/05/16–2012/05/18 | 3.75 ± 0.33 | 16.30 | 2013/07/17–2013/07/20 | 15.31 ± 1.29 | 67.22 |
| 2012/05/18–2012/05/20 | 3.63 ± 0.31 | 7.35 | 2013/07/20–2013/08/03 | 0.21 ± 0.02 | 0.28 |
| 2012/05/20–2012/05/26 | 0.04 ± 0.03 | 0 | 2013/08/03–2013/08/27 | 1.03 ± 0.09 | 1.87 |
| 2012/05/26–2012/05/28 | 6.85 ± 0.62 | 45.00 | 2013/08/27–2013/09/01 | 3.47 ± 0.30 | 15.82 |
| 2012/05/28–2012/06/09 | 0.09 ± 0.02 | 0.04 | 2013/09/01–2013/09/15 | 1.09 ± 0.09 | 0.03 |
| 2012/06/09–2012/06/13 | 1.14 ± 0.11 | 4.43 | 2013/09/15–2013/09/23 | 1.34 ± 0.11 | 3.46 |
| 2012/06/13–2012/06/17 | 3.04 ± 0.27 | 20.68 | 2013/09/23–2013/10/08 | 0.05 ± 0.01 | 0 |
| 2012/06/17–2012/06/20 | 1.86 ± 0.19 | 14.30 | 2013/10/08–2013/10/22 | 0.03 ± 0.01 | 0 |
| 2012/06/20–2012/06/23 | 2.93 ± 0.25 | 6.87 | 2013/10/22–2013/11/05 | 0.40 ± 0.04 | 0.65 |
| 2012/06/23–2012/06/25 | 10.32 ± 0.87 | 19.45 | 2013/11/05–2013/11/18 | 0.09 ± 0.02 | 0 |
| 2012/06/25–2012/07/06 | 0.20 ± 0.02 | 0.14 | 2013/11/18–2013/11/25 | 0.90 ± 0.08 | 0.96 |
| 2012/07/06–2012/07/18 | 0.56 ± 0.05 | 1.69 | 2013/11/25–2013/12/11 | 0.16 ± 0.02 | 0 |
| 2012/07/18–2012/07/23 | 1.37 ± 0.12 | 6.52 | 2013/12/11–2013/12/18 | 5.16 ± 0.42 | 6.83 |
| 2012/07/23–2012/08/02 | 0.31 ± 0.03 | 0.44 | | | |

(0.19) obtained during the southwest monsoon months, indicating more atmospheric $^{7}$Be carried by the same amount of rainwater during the northeast monsoon season. Thus, besides precipitation, the contents of $^{7}$Be in the air could be the other important factor affecting the $^{7}$Be deposition flux. Such seasonal difference reflected the higher $^{7}$Be contents in the atmosphere during the northeast monsoon months. Similar phenomenon was also reported by Su *et al.* (2003) based on a time-series observation of $^{7}$Be deposition in Taiwan. Since the troposphere usually becomes thinner in

the mid-latitude regions in late winter and early spring, the air mass exchange between the troposphere and stratosphere is much active (Kim *et al.*, 1998), leading more $^{7}$Be to enter into the troposphere (Yong *et al.*, 1980; Stohl *et al.*, 1999;

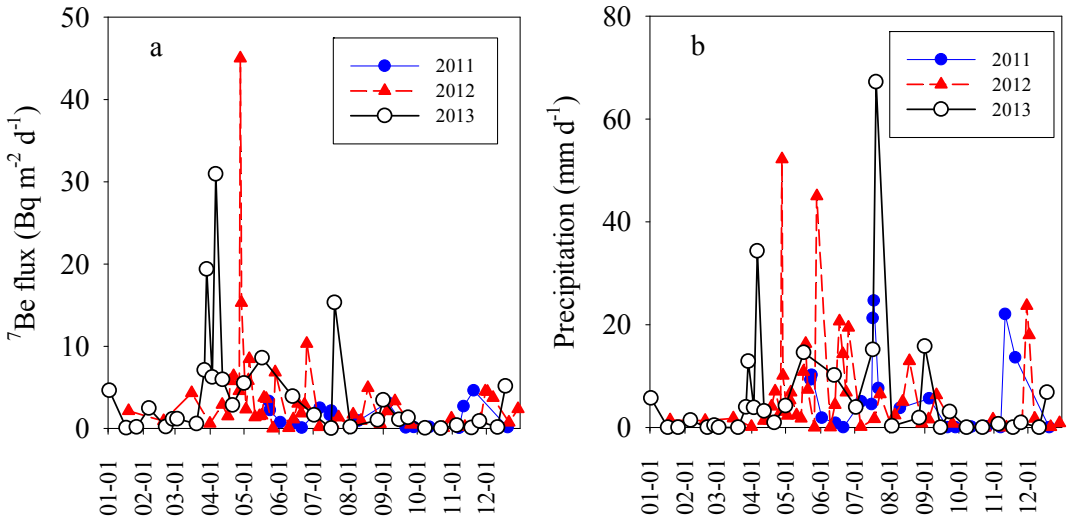


**Fig. 1.** Daily deposition fluxes of $^{7}$Be (a) and precipitation (b) in Xiamen from May 2011 to December 2013.

**Table 2.** Deposition of $^{7}$Be in each month from June 2011 to December 2013.

| Month | $^{7}$Be flux (Bq m$^{-2}$ d$^{-1}$) | | | Precipitation (mm d$^{-1}$) | | |
|---|---|---|---|---|---|---|
| | 2011 | 2012 | 2013 | 2011 | 2012 | 2013 |
| Jan. | - | 1.63 ± 0.10 | 0.79 ± 0.04 | - | 1.40 | 0.60 |
| Feb. | - | 2.21 ± 0.22 | 0.91 ± 0.05 | - | 1.51 | 0.38 |
| Mar. | - | 2.29 ± 0.16 | 3.69 ± 0.17 | - | 0.90 | 1.98 |
| Apr. | - | 5.35 ± 0.18 | 7.42 ± 0.23 | - | 4.52 | 6.01 |
| May | - | 2.18 ± 0.07 | 6.24 ± 0.40 | - | 6.39 | 11.93 |
| Jun. | 1.08 ± 0.07 | 1.79 ± 0.08 | 2.63 ± 0.17 | 2.00 | 6.80 | 6.65 |
| Jul. | 1.23 ± 0.06 | 0.55 ± 0.03 | 1.68 ± 0.13 | 5.16 | 1.84 | 14.19 |
| Aug. | 2.44 ± 0.18 | 1.77 ± 0.08 | 1.26 ± 0.08 | 5.03 | 4.33 | 3.51 |
| Sep. | 0.63 ± 0.04 | 1.27 ± 0.07 | 0.99 ± 0.05 | 0.95 | 1.98 | 1.36 |
| Oct. | 0.05 ± 0.02 | 0.32 ± 0.03 | 0.14 ± 0.01 | 0.13 | 0.36 | 0.19 |
| Nov. | 1.82 ± 0.14 | 1.22 ± 0.07 | 0.34 ± 0.02 | 7.03 | 3.72 | 0.33 |
| Dec. | 0.85 ± 0.06 | 2.04 ± 0.09 | - | 0.53 | 2.05 | - |

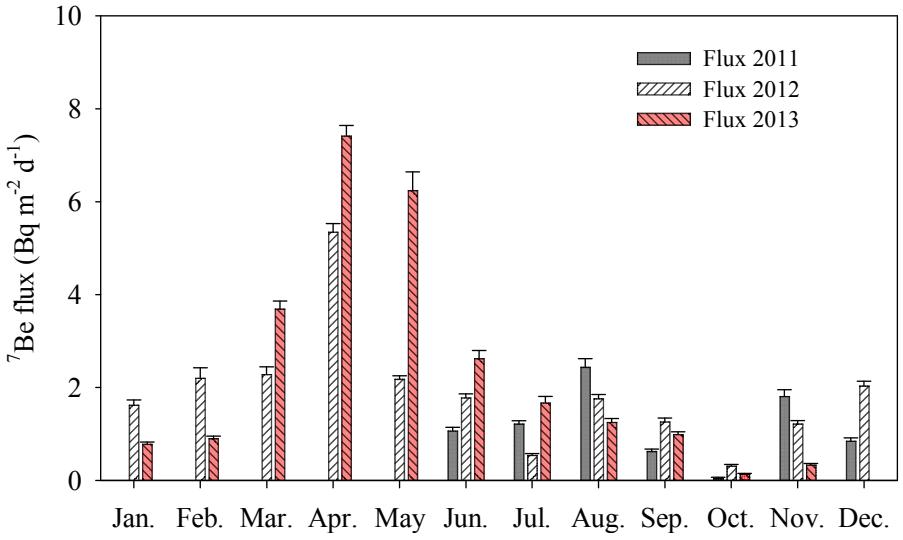


**Fig. 2.** Average deposition fluxes of $^{7}$Be in each month in Xiamen from 2011 to 2013.

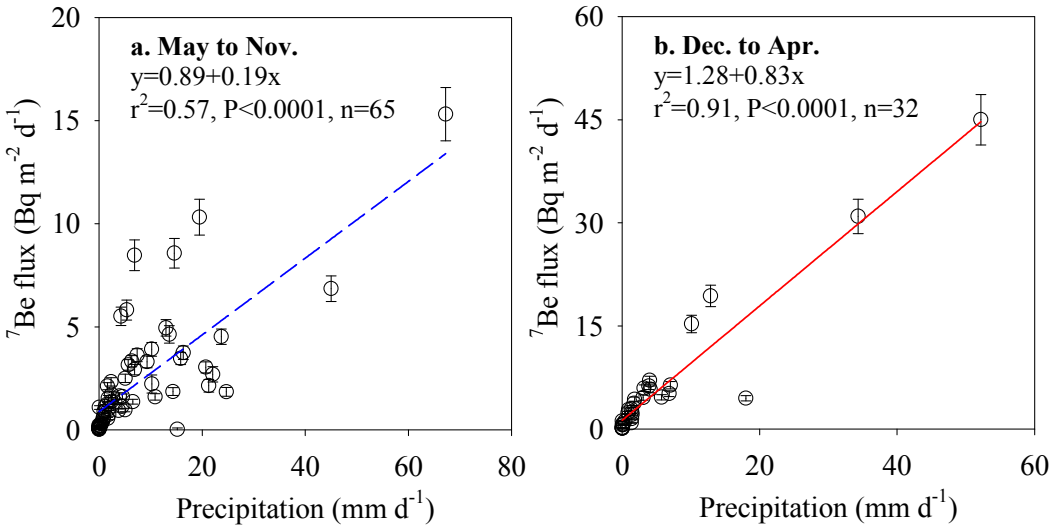


**Fig. 3.** Relationships between the daily deposition of $^{7}$Be and precipitation from (a) May to November, and (b) December to April.

Tan *et al.*, 2013). In contrast, aerosols in Xiamen are mainly derived from ocean during the southwest monsoon season (Cai *et al.*, 2000), which contain less $^{7}$Be. Consequently, the contents of $^{7}$Be in the atmosphere are higher during the northeast monsoon season but lower in the southwest monsoon months.

The different influence of precipitation on $^{7}$Be deposition can be quantified by the precipitation-normalized enrichment factor $\alpha$ (Baskaran, 1995), defined as:

$$\alpha = (F_s/F_t)/(P_s/P_t) \quad (2)$$

where $F_s$ and $P_s$ represent the deposition fluxes of $^{7}$Be and the amount of rainfall during a specific month, respectively. $F_t$ and $P_t$ are the corresponding flux and precipitation. Given a certain amount of rainwater, more $^{7}$Be is removed from the atmosphere than the annual average when the $\alpha$ value is higher than unity. From December to April, the average $\alpha$ values were 2.07 ± 0.11 and 2.39 ± 0.14 in 2012 and 2013 respectivily, with an average of 2.23 ± 0.09, indicating that the same amount of rainfall removed more $^{7}$Be from the atmosohere within the northeast monsoon season. In comparison, the values from May to November in 2012 (0.57 ± 0.04) and 2013 (0.62 ± 0.05) were significantly lower than that obtianed between December and April, demonstrating the contrasting difference in removing $^{7}$Be between the northeast and southwest monsoon seasons.

### *Exchange of Air Mass between the Stratosphere and Troposphere*

Since $^{7}$Be in surface air is from the upper troposphere and stratosphere, the higher contents of $^{7}$Be between December and April probably indicated intensively vertical exchange of air mass in the northeast monsoon season. Usually, the timescale of exchange between the stratosphere and surface air is 15–20 days (Baldwin *et al.*, 2001). The transport of cosmogenic phosphorus (i.e., $^{32}$P and $^{33}$P) from stratosphere to surface air is less than 28 days in Xiamen (Chen, 2006). The residence times of surface aerosol are around 20 days in Xiamen based on the $^{210}$Po/$^{210}$Pb ratios (Wang *et al.*, 2014). Thus, the resident timescale allows aerosols in surface air in Xiamen to be transported into the upper troposphere and stratosphere via vertical mixing.

Based on the relations between the deposition fluxes and rainfall, the influence of rainfall magnitude on $^{7}$Be fluxes could be mostly eliminated by normalizing the flux of $^{7}$Be to the precipitation. As shown in Fig. 4, the normalized $^{7}$Be indicated that the $^{7}$Be contents were much higher from December to April. Generally, the vertical exchange of air mass depends on the eddy-induced mean zonal force between the stratosphere and troposphere (Haynes *et al.*, 1991). Since the strongest eddy-induced forces appear in the northern hemisphere winter, the most extensive air mass exchanges between the troposphere and stratosphere have been also observed in this season (Holton *et al.*, 1995; Yang *et al.*, 2003). This mechanism might be responsible for the higher $^{7}$Be contents in the atmosphere between December and April in Xiamen. Actually, previous studies in other regions also attributed the high atmospheric $^{7}$Be contents to the enhanced air exchange between the stratosphere and troposphere during the season transition from winter to spring (Yong *et al.*, 1980; Kim *et al.*, 1998; Stohl *et al.*, 1999; Tan *et al.*, 2013). Comparatively, the lower atmospheric 7Be contents from May to November resulted from the relatively weaker exchange of air mass during these seasons. Therefore, the precipitation-normalized deposition of $^{7}$Be in Xiamen seemed to indicate the air mass exchange between the stratosphere and troposphere.

The flux ratios of $^{7}$Be to $^{210}$Pb obtained concurrently also provided evidence for $^{7}$Be as a proxy of air mass exchange between the troposphere and stratosphere. $^{210}$Pb in the atmosphere is produced through the decay of $^{222}$Rn in the troposphere (Peirson *et al.*, 1966; Turekian *et al.*, 1977; Papastefanou, 2009a; Ali *et al.*, 2011), while $^{7}$Be is mainly from the stratosphere. Hence, the $^{7}$Be/$^{210}$Pb ratios in the air could indicate the air mixing intensity between the

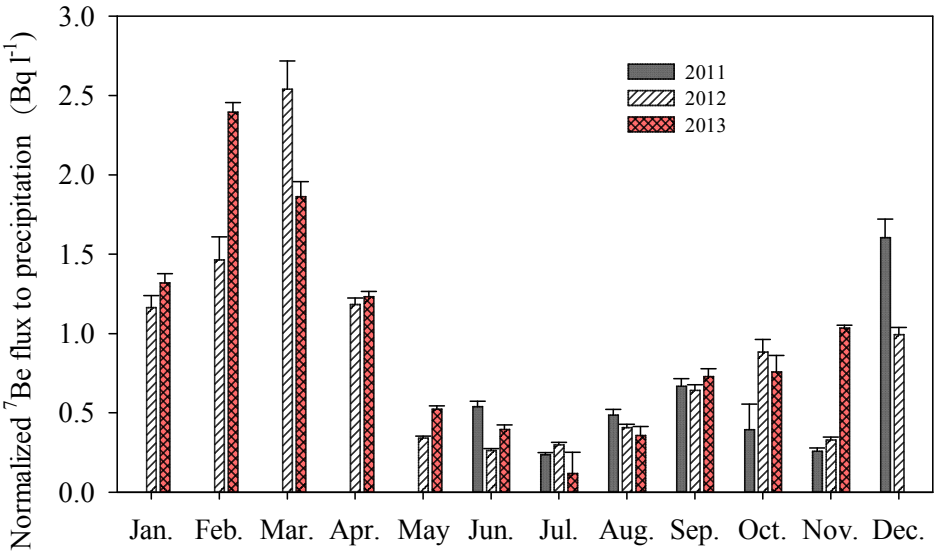


**Fig. 4.** Intra-annual variation pattern of atmospheric $^{7}$Be abundance based on precipitation-normalized $^{7}$Be flux.

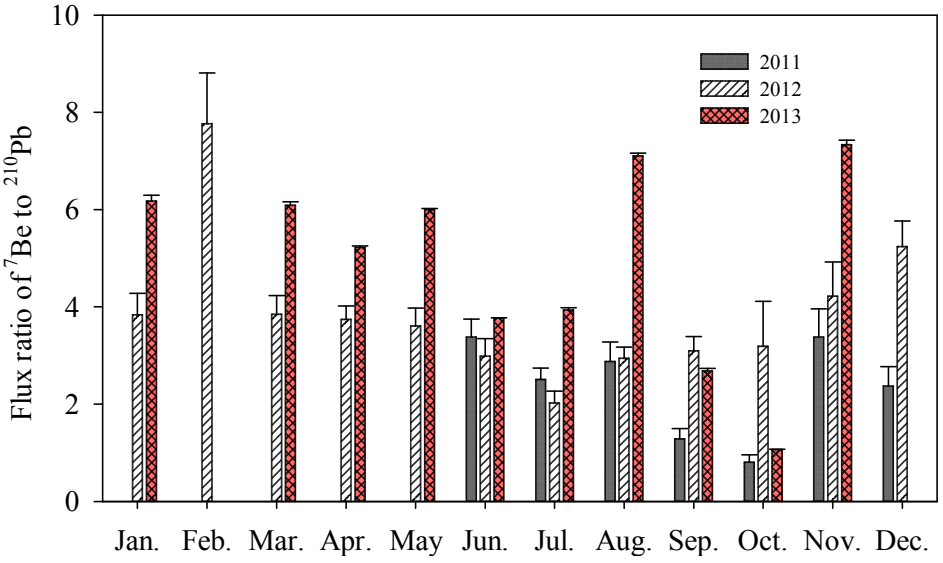


**Fig. 5.** Intra-annual variation patterns of $^{7}$Be to $^{210}$Pb flux ratios.

stratosphere and troposphere (Baskaran, 1995). The higher $^{7}$Be/$^{210}$Pb ratios suggest more extensive mixing of air mass between the stratosphere and troposphere. Based on the similar removal pathway of both $^{7}$Be and $^{210}$Pb (Fig. 3; Baskaran *et al.*, 1993; Kim, 2000; Huh *et al.*, 2006; Wang *et al.*, 2014), it is reasonable to use the flux ratios of $^{7}$Be to $^{210}$Pb to substitute their activity ratios as an alternative strategy (Baskaran, 2011; Yang *et al.*, 2012; Wang, 2013). The annual average $^{7}$Be/$^{210}$Pb ratio was 4.42 ± 0.07. From December to April, the average $^{7}$Be/$^{210}$Pb ratio (5.72 ± 0.30) was higher than the mean value (Fig. 5), while the ratios were lower than the average in other months (3.73 ± 0.09). Overall, this temporal pattern of $^{7}$Be/$^{210}$Pb ratio was similar to the trend derived from precipitation-normalized $^{7}$Be, supporting either the stronger air exchange in the northeast monsoon seasons or the application of $^{7}$Be to constrain the air exchange in Xiamen.

## CONCLUDING REMARKS

Tens of hazy days each year have been reported during the last three years in Xiamen (Fig. 6; Zhang *et al.*, 2013a), mainly resulting from the increasing abundance of < 2.5 μm aerosol ($PM_{2.5}$). Previous study indicated that $^{7}$Be in the aerosol was dominantly concentrated in submicron aerosol (0.4–2.0 μm, Papastefanou, 2009b). The present research revealed the concurrently high contents of $^{7}$Be and haze in Xiamen (Figs. 2, 4, and 6). Together, these results lent supports to $^{7}$Be as a tracer of constraining the fate of haze in Xiamen. $PM_{2.5}$ collected in Xiamen shows high concentrations of polycyclic aromatic hydrocarbon (PAH) (Wu *et al.*, 2009; Lee *et al.*, 2014) and Pb (Zhang *et al.*, 2013a). Thus, $^{7}$Be cycling could also provide insights into the dispersal of atmospheric pollutants adsorbed on the submicron aerosol over a large spatial scale.

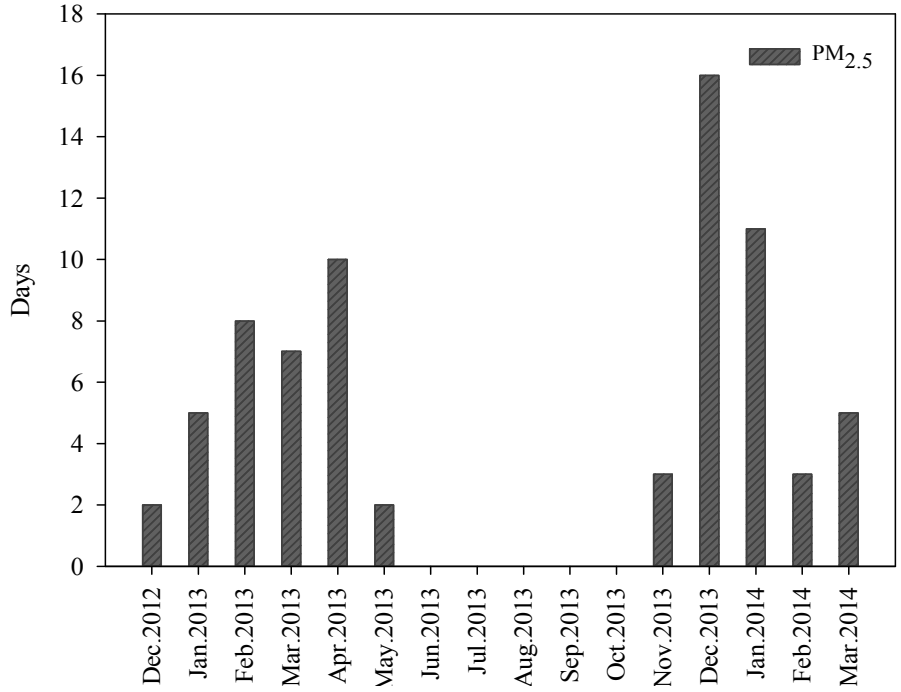


**Fig. 6.** Recorded days for hazy weather (indexed by $PM_{2.5}$) from Dec. 2012 to Mar. 2014 in Xiamen (data from Xiamen Environmental Monitoring Station at 24°28′N, 118°05′E).

## ACKNOWLEDGEMENTS

We appreciate Dr. L.-C. Wang, S. Gilardoni, Katsura Hidemitsu, and an anonymous reviewer for their constructive suggestions during the manuscript preparation. We also thank Raechel Huff, University of Wisconsin, for polishing the manuscript. This study was supported in part by NSFC (41076043 and 41125020), State Oceanic Administration of China (GASI-03-01-02-02), and the Fundamental Research Funds for the Central Universities (Xiamen University: 20720140508).

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*Received for review, March 23, 2015*
*Revised, June 21, 2015*
*Accepted, September 28, 2015*