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Atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po at Xiamen, China

Yong Yi, Peng Zhou, Guangshan Liu*

College of Oceanography and Environmental Science, Xiamen University,
State Key Laboratory of Marine Environmental Science, Xiamen 361005, P.R. China

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The atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po at Xiamen were measured. The samples were collected from March 2004 to April 2005 and the sampling period was one month. The ^{7}Be and ^{210}Pb activity were measured using HPGe γ-spectrometer after concentration using $Fe(OH)_3$ co-precipitation method. The ^{210}Po was counted with an α-spectrometer after the sample was digested and spontaneous plated onto a silver planchet. At Xiamen, the atmospheric deposition fluxes of ^{7}Be varied between 0.11 and 2.93 $Bq \cdot m^{-2} \cdot d^{-1}$ and the average was 1.64 $Bq \cdot m^{-2} \cdot d^{-1}$; ^{210}Pb fluxes varied between 0.04 and 0.85 $Bq \cdot m^{-2} \cdot d^{-1}$, and the average was 0.51 $Bq \cdot m^{-2} \cdot d^{-1}$; ^{210}Po fluxes varied between 0.002 and 0.133 $Bq \cdot m^{-2} \cdot d^{-1}$, and the average was 0.061 $Bq \cdot m^{-2} \cdot d^{-1}$. There were positive correlations between the deposition fluxes of ^{7}Be, ^{210}Pb or ^{210}Po and the amount of precipitation. The residence time of aerosols varied between 6.0 and 54.0 days with a mean of 27.1 days, which were calculated by ^{210}Po/^{210}Pb fluxes ratios.

Introduction

^{7}Be ($T_{1/2} = 54.3$ days) is a naturally occurring radionuclide produced by the spallation reaction of energetic cosmic rays with atmospheric oxygen and nitrogen. ^{210}Pb ($T_{1/2} = 22.3$ years) in the atmosphere is produced ultimately by the decay of precursor, ^{222}Rn ($T_{1/2} = 3.8$ days) through a series of mid-daughters, which emanates into the atmosphere from the earth's surface. ^{210}Po ($T_{1/2} = 138$ days) in the atmosphere is produced from the decay of ^{210}Pb through ^{210}Bi ($T_{1/2} = 5.0$ days).[1,2]

After produced, ^{7}Be, ^{210}Pb and ^{210}Po become associated rapidly with aerosols and remove to land and ocean through precipitation and dry deposition. Thus, these nuclides can be used to obtain information on the behavior, residence time and rate of removal of aerosols.[3–5]

^{7}Be, ^{210}Pb and ^{210}Po have been widely used in the study of atmospheric mixing and particle settling, fate of pollutants and dynamics of particle transport since their half-lives are in the same order of magnitude as the time scale of those atmospheric processes.[3–6] These three nuclides have also been widely used to marine study.[7–10] The atmospheric deposition fluxes of the three nuclides are the important parameters for verification of particle transports model. In this paper we report total monthly atmospheric deposition fluxes data of ^{7}Be, ^{210}Pb and ^{210}Po in Xiamen during March 2004 and April 2005.

Experimental

Instruments

Gamma-spectrometer: The measurement of ^{7}Be and ^{210}Pb was performed by a HPGe γ-spectrometry system (Ortec, U.S.). The detector is a Model GMX45P-S with energy resolution of 2.0 keV and relative efficiency of 45% for 1332 keV γ-rays of ^{60}Co point source located at source–detector distance of 25 cm. The lead shielding is Model NLC411. The integrated background count of system in the energy range of 10–1934 keV is 2.64 s^{-1}. The spectra were analyzed using Gamma Vision-32 Gamma-Ray Spectrum Analysis and MCA Emulator software.

Alpha-spectrometer: Octête α-spectrometer system (Ortec, U.S.) was used to measure ^{210}Po. The energy resolution of the detector is <20 keV for ^{241}Am point source when the distance of detector-to-source is >detector diameter. The detector efficiency is >25% for a distance of detector-to-source <10 mm and ^{241}Am point source. The integrated background count is <1 h^{-1}.

Sample collection

The sampling site was chosen on the building roof of the Department of Oceanography, Xiamen University in Xiamen (24°26′N, 118°5′E), China. Integrated monthly bulk deposition samples were collected with a plastic bucket with 56.5 cm length, 44.8 cm width and 40 cm depth from March 2004 to April 2005.

The samples were collected at the end of each month and processed immediately as follows. The sample was removed into a treating bucket from the collecting bucket and the walls of collecting bucket was rinsed with 0.2 mol/dm^{3} HCl and cleaned with a scrubbing brush and then rinsed with de-ionized water.

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* E-mail: lgshan@public.xm.fj.cn

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The washing solution was added to the sample. Upon returning the laboratory the sample was concentrated using $Fe(OH)_3$ co-precipitation method and the precipitate was transferred to a polyethylene container of 7.5 cm diameter and 5 cm height for γ-spectrum analysis of ^{7}Be and ^{210}Pb.

The precipitation data was from Xiamen Meteorological Bureau.

Measurement of ^{7}Be and ^{210}Pb using γ-spectrometry

The measurement of ^{7}Be, ^{210}Pb and ^{210}Po were performed at Isotope Marine Laboratory of Xiamen University. The ^{7}Be and ^{210}Pb were measured directly using HPGe γ-spectrometer. ^{7}Be was determined using the γ-ray peak of 477.7 keV (yield 10.3%). ^{210}Pb was analyzed by the γ-ray peak of 46.5 keV (yield 4%).

Measurement of ^{210}Po using α-spectrometry

The ^{210}Po was measured with an α-spectrometer after the sample was digested and by spontaneously plated onto a silver planchet.

The sample was transferred to a teflon beaker after the γ-spectrum analysis and a known amount of ^{209}Po spike was added to the sample for calculating the recovery. Approximately 10 ml of perchloric acid and concentrated nitric acid were dripped into the sample while heating and the sample should be dissolved perfectly. The sample solution was distilled to dryness, and then was dissolved with hydrochloric acid. After the sample was cool, the pH was raised to 2 by adding ammonia water. Adding ascorbic acid until the color disappears. Subsequently, 2 cm^3 of 20% hydroxylamine hydrochloride and 2 cm^3 of 20% sodium citrate were added in the sample and the pH was adjusted to 2.5 by ammonia water and concentrated hydrochloric acid.

The sample solution was transferred to the plating vessel. The silver planchet was mixed on the magnetic rotor with a teflon holder and put it in the vessel for spontaneous plating 180 minutes on stirring at 85–90 °C. The silver planchet plated Po was rinsed with de-ionized water and dried in the air and then stored for α-counting.

Evaluation of ^{210}Po activity in the sample

The spectrum acquisition can not be performed immediately after the sample collection because the sample need to be concentrated and had been waiting spectrometer free, so the nuclide activity at spectrum acquisition time must be corrected to sampling time. The corrections of ^{7}Be and ^{210}Pb activities are simple but it is complex for ^{210}Po which is the granddaughter of ^{210}Pb and the build-up of decay must be considered. The half-life of intermediate product ^{210}Bi is short in comparison with sampling period, so the parent ^{210}Pb and granddaughter ^{210}Po are not considered separated by intermediate product ^{210}Bi. Let A_1 and A_2 be the activities of ^{210}Pb and ^{210}Po at the time of sampling end, we can derived the ^{210}Po activity (A'_2) at any time (t_2) after sampling completed from the dynamic decay equation:

$$A'_2 = A_2 e^{-\lambda_2 t_2} + \frac{\lambda_2 A_1}{\lambda_2 - \lambda_1}(e^{-\lambda_1 t_2} - e^{-\lambda_2 t_2}) \tag{1}$$

where λ_1 and λ_2 are the decay constants of ^{210}Pb and ^{210}Po, respectively. If t_2 is the time interval from the sampling end to plating, A'_2 is the ^{210}Po activity at plating time and the A_2 is derived:

$$A_2 = A'_2 e^{\lambda_2 t_2} - \frac{\lambda_2 A_1}{\lambda_2 - \lambda_1}(e^{-(\lambda_1 - \lambda_2)t_2} - 1) \tag{2}$$

The ^{210}Po activity in the silver planchet is not affected by ^{210}Pb and varies on exponential law with time as:

$$A''_2 = A'_2 e^{-\lambda_2 t_3} \tag{3}$$

where t_3 is the time interval from plating to spectrum acquisition and A''_2 is the ^{210}Po activity at the spectrum acquisition. From Eq. (3) the A'_2 is derived:

$$A'_2 = A''_2 e^{\lambda_2 t_3} \tag{4}$$

Let N_2 and N_3 are peak areas of ^{210}Po and ^{209}Po in sample spectrum and A_3 is the activity of ^{209}P spike added then:

$$\frac{N_2}{N_3} = \frac{A''_2}{A_3} \tag{5}$$

and, hence

$$A''_2 = \frac{N_2}{N_3} A_3 \tag{6}$$

Substituting Eq. (6) into Eq. (4) and then into Eq. (2) gives:

$$\begin{aligned} A_2 = A_3 \frac{N_2}{N_3} e^{\lambda_2 (t_2 + t_3)} - \\ - \frac{\lambda_2 A_1}{\lambda_2 - \lambda_1}(e^{-(\lambda_1 - \lambda_2)t_2} - 1) \end{aligned} \tag{7}$$

Atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po

Assume that the atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po are constant during the sampling period, the mass balance equations for ^{7}Be and ^{210}Pb activity in sample can be written as;

$$\frac{dA_1}{dt} = SI_1 - \lambda_1 A_1 \tag{8}$$

where A_1 is the activity of ^{7}Be or ^{210}Pb in the sample which are the function of time t, I_1 is the deposition fluxes of ^{7}Be or ^{210}Pb, S is the area of collecting vessel. The solution of Eq. (8) is:

$$A_1 = \frac{SI_1}{\lambda_1}(1 - e^{-\lambda_1 t}) \tag{9}$$

Atmospheric deposition fluxes of ^{7}Be and ^{210}Pb was evaluated by:

$$I_1 = \frac{\lambda A_1}{S} \cdot \frac{e^{\lambda t_2}}{1 - e^{-\lambda t_1}} \tag{10}$$

The half-life of ^{210}Pb is long enough in comparison with the sampling period, therefore, to simple Eq. (10) gives:

$$I_1 = \frac{A_1}{St_1} \tag{11}$$

where A_1 is ^{7}Be or ^{210}Pb activity at spectrum acquisition, I_1 is the atmospheric deposition flux, t_1 is the sampling period, t_2 is the time interval from the end of sampling to the spectrum acquisition and S is the mouth area of the sampling vessel.

The mass balance equations for ^{210}Po activity in sample can be written as:

$$\frac{dA_2}{dt} = SI_2 - \lambda_2 A_2 + \lambda_2 A_1 \tag{12}$$

where I_1 and I_2 are the deposition fluxes of ^{210}Pb and ^{210}Po, respectively. Substituting Eq. (9) into Eq. (12) and integrating gives:

$$A_2 = \left(\frac{SI_2}{\lambda_2} + \frac{SI_1}{\lambda_1}\right)(1 - e^{-\lambda_2 t}) + \\ + \frac{\lambda_2 SI_1}{(\lambda_2 - \lambda_1)\lambda_1}(e^{-\lambda_2 t} - e^{-\lambda_1 t}) \tag{13}$$

where t is the sampling period. The atmospheric deposition flux of ^{210}Po is:

$$I_2 = \frac{\lambda_2}{S(1 - e^{-\lambda_2 t})} \\ \left[A_2 - \frac{SI_1}{\lambda_1}(1 - e^{-\lambda_2 t}) - \frac{\lambda_2 SI_1}{\lambda_1(\lambda_2 - \lambda_1)}(e^{-\lambda_2 t} - e^{-\lambda_1 t})\right] \tag{14}$$

Equation (14) needs the ^{210}Pb flux and the ^{210}Po activity at the end of sampling for calculating the ^{210}Po flux.

Aerosol residence time

If all ^{210}Po is produced from the decay of ^{210}Pb in the atmosphere, the change of ^{210}Po concentration may be expressed by the mass balance equation:[4,5]

$$\frac{dC_2}{dt} = \lambda_2 C_1 - (\lambda_2 + \lambda_R)C_2 \tag{15}$$

where C_1 and C_2 are the concentrations of ^{210}Pb and ^{210}Po in the air, λ_2 is the decay constant of ^{210}Po, and λ_τ is the first order rate constant for removal of aerosol. If the system is in the steady state, $dC_2/dt = 0$, we can derive:

$$\lambda_2 C_1 = \lambda_2 C_2 + \lambda_R C_2 \tag{16}$$

The apparent mean resident time is given by:

$$\tau_R = \frac{1}{\lambda_R} = \frac{1}{\lambda_2}\frac{C_2 / C_1}{1 - C_2 / C_1} \tag{17}$$

Assuming that the activity ratios of ^{210}Po and ^{210}Pb in the air are equal to that in atmospheric deposition, to substitute the concentration ratio by the flux ratio I_2/I_1 of ^{210}Po and ^{210}Pb in Eq. (17), we get:

$$\tau_R = \frac{1}{\lambda_R} = \frac{1}{\lambda_2}\frac{I_2 / I_1}{1 - I_2 / I_1} \tag{18}$$

Results and discussion

Atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po in Xiamen

The atmospheric deposition fluxes of ^{7}Be, ^{210}Pb, ^{210}Po at Xiamen during March 2004 to April 2005 are given in Table 1 and Fig. 1. The flux of ^{7}Be varied between 0.11 and 2.93 $Bq \cdot m^{-2} \cdot d^{-1}$, with a mean of 1.64 $Bq \cdot m^{-2} \cdot d^{-1}$. The flux of ^{210}Pb varied between 0.04 and 0.85 $Bq \cdot m^{-2} \cdot d^{-1}$, with a mean of 0.51 $Bq \cdot m^{-2} \cdot d^{-1}$. The flux of ^{210}Po varied between 0.002 and 0.133 $Bq \cdot m^{-2} \cdot d^{-1}$, with a mean of 0.061 $Bq \cdot m^{-2} \cdot d^{-1}$. The flux ratios of ^{7}Be/^{210}Pb varied between 0.45 and 5.53, with a mean value of 3.20. The flux ratio of ^{210}Pb/^{210}Po varied between 0.03 and 0.21, with a mean value of 0.11. The precipitation at Xiamen varied between 0.7–293.8 mm/month, with a mean value of 94.6 mm/month.

During the sampling period, high precipitation occurred from February to September. The atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po were constant, high from February to September while low during October to January (winter) when the precipitation was low. High ^{210}Po was in April, May, September 2004, and February and March 2005 but it was low in November and December.

Table 1. Atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po in Xiamen

Sampling date	^{7}Be, $Bq{\cdot}m^{-2}{\cdot}d^{-1}$	^{210}Pb, $Bq{\cdot}m^{-2}{\cdot}d^{-1}$	^{210}Po, $Bq{\cdot}m^{-2}{\cdot}d^{-1}$	Precipitation, mm/month	^{7}Be/^{210}Pb, activity ratio	^{210}Po/^{210}Pb, activity ratio	τ_R, days
March, 2004	2.78 ± 0.04	0.67 ± 0.04	0.020 ± 0.006	46.2	4.16	0.03	6.0
April, 2004	0.69 ± 0.04	0.81 ± 0.04	0.106 ± 0.023	88.8	0.85	0.13	30.3
May, 2004	2.75 ± 0.05	0.50 ± 0.04	0.105 ± 0.018	154.1	5.53	0.21	53.0
June, 2004	0.93 ± 0.13	0.24 ± 0.04	0.051 ± 0.012	67.4	3.89	0.21	54.0
July, 2004	2.63 ± 0.08	0.61 ± 0.04	0.026 ± 0.011	75.5	4.34	0.04	8.8
August, 2004	2.67 ± 0.05	0.64 ± 0.04	0.046 ± 0.012	197.4	4.19	0.07	15.3
September, 2004	2.90 ± 0.05	0.85 ± 0.04	0.133 ± 0.010	293.8	3.41	0.16	36.7
October, 2004	0.71 ± 0.02	0.42 ± 0.03	0.037 ± 0.005	0.7	1.69	0.09	19.1
November, 2004	0.17 ± 0.04	0.04 ± 0.02	0.002 ± 0.001	2.4	4.64	0.05	9.9
December, 2004	0.17 ± 0.02	0.07 ± 0.03	0.005 ± 0.001	18.2	2.58	0.08	18.1
January, 2005	0.11 ± 0.07	0.23 ± 0.07	0.023 ± 0.004	2.0	0.45	0.10	22.0
February, 2005	2.93 ± 0.05	0.83 ± 0.03	0.091 ± 0.038	93.2	3.54	0.11	24.5
March, 2005	2.24 ± 0.07	0.66 ± 0.04	0.131 ± 0.011	219.6	3.37	0.20	48.9
April, 2005	1.24 ± 0.06	0.57 ± 0.08	0.080 ± 0.020	65.6	2.18	0.14	32.6
Range:	0.11–2.93	0.04–0.85	0.002–0.133	0.7–293.8	0.45–5.53	0.03–0.21	6.0–54.0
Mean:	1.64	0.51	0.061	94.6	3.20	0.12	27.1

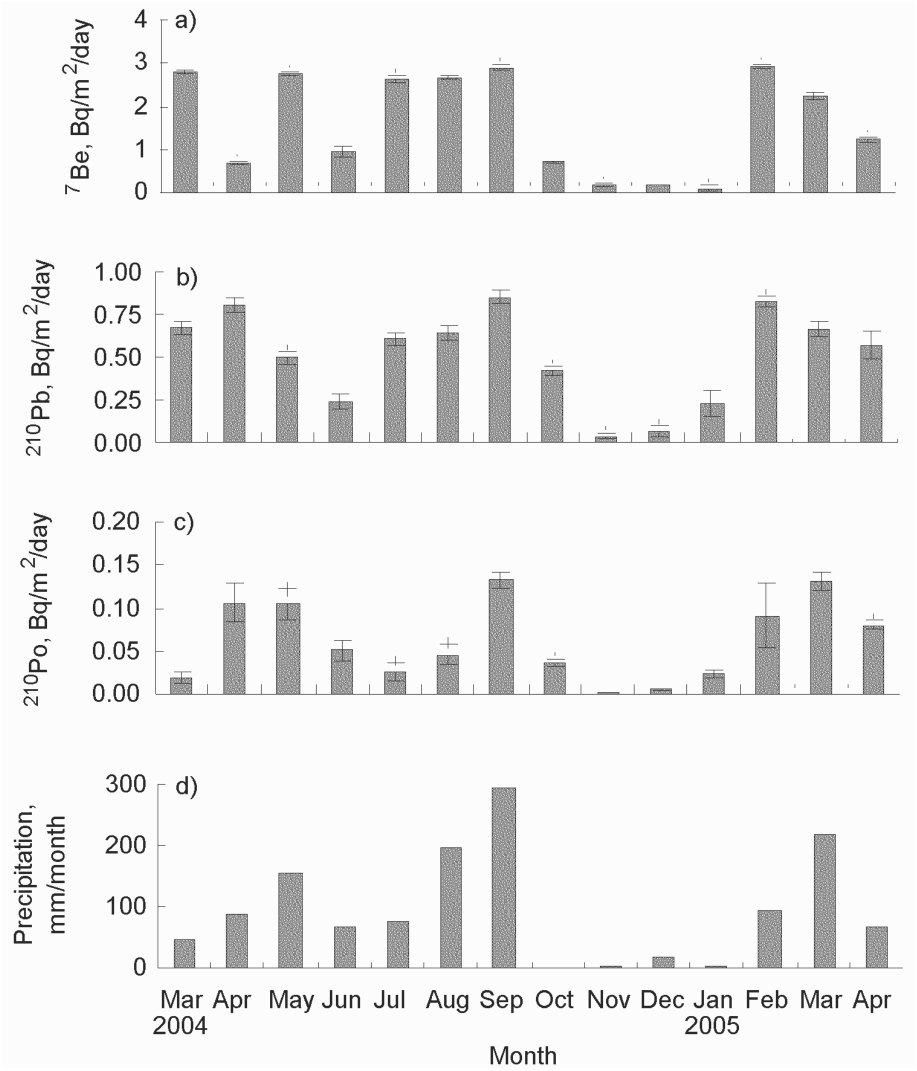

Fig. 1. Atmospheric deposition fluxes of (a) ^{7}Be, (b) ^{210}Pb, (c) ^{210}Po and (d) precipitation during March, 2004 to April, 2005 at Xiamen

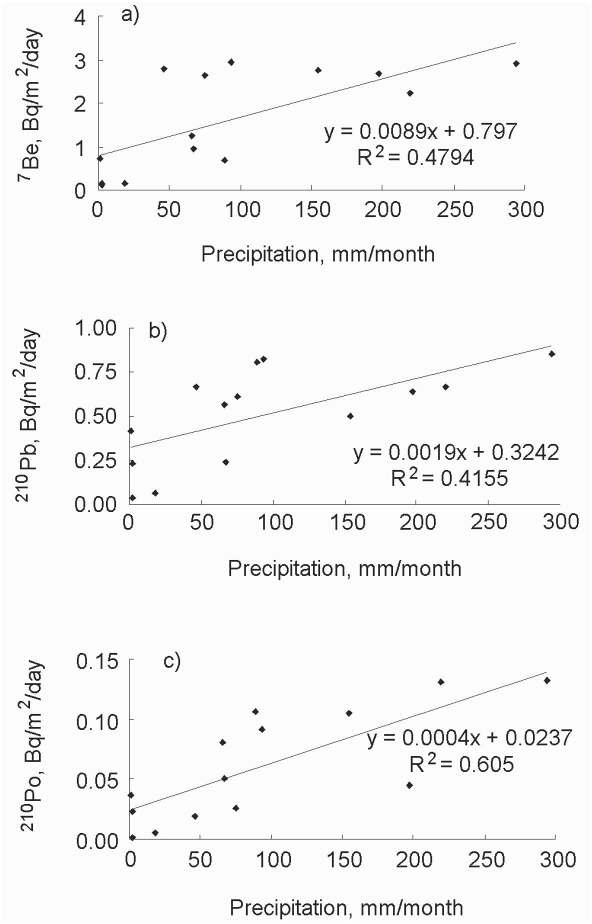

Fig. 2. Correlation plots of atmospheric deposition fluxes of (a) ^{7}Be, (b) ^{210}Pb and (c) ^{210}Po against the precipitation. The straight lines in the figures were fitted by the least squares method

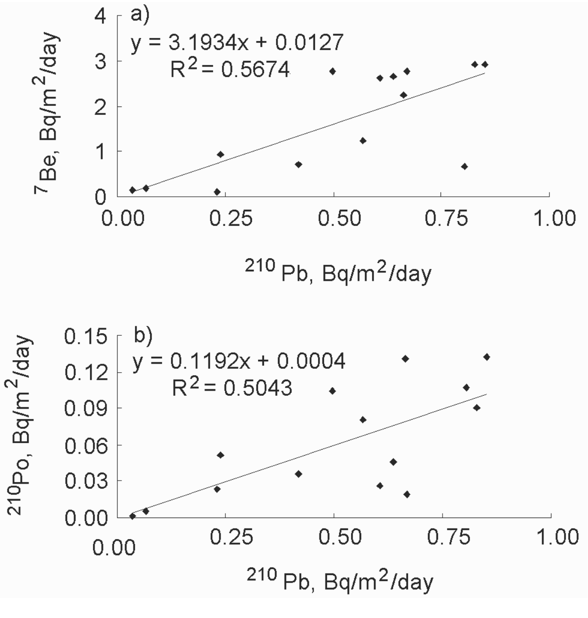

Fig. 3. Correlation of the deposition fluxes between ^{7}Be and ^{210}Pb (a) and between ^{210}Po and ^{210}Pb (b)

Correlation of deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po with the amount of precipitation

The atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po at Xiamen during the sampling period have been plotted against the amount of precipitation (Fig. 2). There are good positive correlations between the deposition fluxes of ^{7}Be or ^{210}Pb or ^{210}Po and the amount of precipitation suggested that the precipitation dominate the ^{7}Be, ^{210}Pb and ^{210}Po deposition fluxes. The sampling period was a month, therefore, the deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po are averages over a month period, so the variations of the deposition fluxes of ^{7}Be and ^{210}Pb rise mainly from seasonal effect of precipitation.

Correlations between deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po

At Xiamen, the atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po were higher in spring, summer and early autumn but lower during late autumn and winter that presented a good correlation between the fluxes of the three nuclides (Fig. 3). The variation of the three nuclide fluxes shows a similar trend (Fig. 1) demonstrating that the ^{7}Be, ^{210}Pb and ^{210}Po fluxes were controlled by the same mechanism and process.

The residence time of aerosols

The residence time of aerosols based the ^{210}Po/^{210}Pb fluxes ratio is given in Table 1. The residence time of aerosols of Xiamen varied between 6.0 and 54.0 days with a mean of 27.1 days. To calculate this residence time we assumed that the ^{210}Po in the atmospheric deposition collected for this study was derived from the decay of ^{210}Pb only, and there was no other source of ^{210}Po. The residence time of aerosols in this study is quite comparable to that obtained by ^{210}Po/^{210}Pb activity ratio in the aerosols but higher than that evaluated by measuring ^{210}Bi/^{210}Pb activity ratio in aerosol.[4,7]

Conclusions

The atmospheric deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po were measured at Xiamen during March 2004 to April 2005. The fluxes of ^{7}Be varied between 0.11 and 2.93 Bq·m^{-2}·d^{-1}, with a mean of 1.64 Bq·m^{-2}·d^{-1}. The fluxes of ^{210}Pb varied between 0.04 and 0.85 Bq·m^{-2}·d^{-1}, with a mean of 0.61 Bq·m^{-2}·d^{-1}. The flux of ^{210}Po varied between 0.006 and 0.133 Bq·m^{-2}·d^{-1}, with a mean of 0.061 Bq·m^{-2}·d^{-1}.

There are good positive correlations between the deposition fluxes of ^{7}Be, ^{210}Pb or ^{210}Po and the amount of precipitation suggesting that the precipitation dominate the ^{7}Be, ^{210}Pb and ^{210}Po deposition fluxes. There are good correlations between the deposition fluxes of ^{7}Be, ^{210}Pb and ^{210}Po.

The residence time of aerosols at Xiamen evaluated by ^{210}Po/^{210}Pb fluxes ratio varied between 6.0 and 54.0 days with a mean of 27.1 days.

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