Tellus (2008), 60B, 32–41
Printed in Singapore. All rights reserved

TELLUS

Analyses of ^{210}Pb concentrations in surface air and in rain water at the central Guizhou, China

By G. J. WAN[1]*, H. N. LEE[2], E. Y. WAN[1], S. L. WANG[1], W. YANG[1], F. C. WU[1], J. A. CHEN[1] and C. S. WANG[1], [1]*State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China;* [2]*Environmental Measurements Laboratory, Department of Homeland Security, New York, NY 10014-7447, USA*

(Manuscript received 11 August 2007; in final form 11 July 2007)

ABSTRACT

Samples of surface air and rain water were collected at the Mt. Guanfeng site located at Guiyang, China. The monthly mean ^{210}Pb concentrations in surface air exhibited a U-pattern with high values appearing in winter and low values in summer and early autumn. The annual mean of ^{210}Pb concentrations in surface air was estimated to be 2.7 $\pm$ 0.6 mBq m^{-3}, which was at least four times higher than the annual mean reported from a large number of monitoring stations located throughout the world. The amounts of enriched Uranium in soil at the region could significantly contribute a great deal of release of ^{222}Rn that decay to ^{210}Pb which gets attached to the atmospheric aerosols. The site was under the influence of a monsoon climate. The distribution of monthly mean of ^{210}Pb concentrations in surface air correlates well with the reciprocal of monthly average of observed air temperature and rainfall. The partition coefficients of ^{210}Pb between rain water-surface air in the winter and spring were larger than those in the summer and autumn. The type and duration of rain depending on season played key roles for the changes of partition coefficients, although the rain amount is important.

1. Introduction

In the last 30 yr, extensive studies on the changes of various environments have widely used ^{210}Pb dissolved in water for tracing aquatic sedimentation, lake catchment erosion (Robbins and Edgington, 1975; Turekian et al., 1983; Wan et al., 1987, 2003, 2005a,b; Ritchie and McHenry, 1990; Appleby, 1997; Santschi et al., 1999; Yeager and Santschi, 2003). ^{210}Pb in surface air has been used for studying the atmospheric transport processes (Feichter et al., 1991; Balkanski et al., 1993; Lee and Feichter, 1995; Rehfeld and Heimann, 1995; Graustein and Turekian, 1996; Rasch et al., 2000; Baskaran and Shaw, 2001; Lee, 2003; Lee et al., 2004, 2006, 2007).

The ^{210}Pb (half-life 22.6 a) is a decay product from ^{222}Rn, which is produced from decay of ^{238}U-series (uranium-series) in surface rocks and soils. ^{222}Rn being a noble gas, can diffuse from soils and enter into the atmosphere where it decays to ^{210}Pb. When ^{210}Pb is formed in the atmosphere, it is quickly adsorbed onto submicrometer-sized aerosols and begins its environmental biogeochemical transport. Scientists investigate the atmospheric transport and mixing processes by analysing the distributions and variations of ^{210}Pb concentrations in the atmosphere and examining the depositional fluxes of ^{210}Pb and in aerosols at the earth surface. The atmospheric airflows can mix the air masses with various species including ^{210}Pb. For example, high ^{210}Pb concentrations in the atmosphere are generally produced and transported in the continental air masses (Preiss et al., 1996). Low ^{210}Pb concentrations in the air over Finland are related to the arrival of warm and salt-enriched water vapour from the North Atlantic and the intensifying cyclone activities. The oscillations of ^{210}Pb activity concentrations in the air in Southern Finland were found to have links with the state of the northeastern part of the Atlantic Ocean (Paatero et al., 2003). Overall, the studies of ^{210}Pb concentrations have provided important information for (1) validating global transport-diffusion models of contaminants, (2) tracing and evaluating the processes of transport, sedimentation and sediment dating, (3) tracing and comparing the drainage basin erosion and finally (4) assessing the impact of natural radiation on ecological system.

The global sampling sites operated by the U.S. Department of Homeland Security's Environmental Measurements Laboratory (EML) for measuring ^{210}Pb in surface air are mostly situated in locations near the seashore or at the islands. Therefore, the measurements of atmospheric ^{210}Pb concentrations from the current site in the region of central Guizhou in inland China over Asian continent will promote scientific value for modelling transport

*Corresponding author.
e-mail: wanguojiang@vip.skleg.cn
DOI: 10.1111/j.1600-0889.2007.00316.x

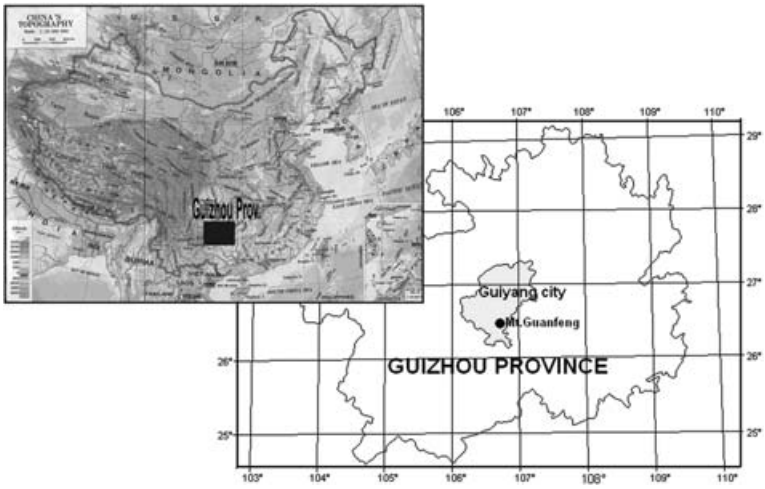

Fig. 1. Surface air sampling location at Guiyang site (circle) in China.

processes in the Asian region. The ^{222}Rn measurements in air are generally relative sparse. For the purpose of present investigation, we have used the measurements of ^{222}Rn collected previously in the Guizhou region.

Since 2001, the comprehensive measurements at Mt. Guanfeng in Guiyang of Guizhou, China have been established with the EML's support in equipment and the scientific cooperation. In this article, we use the 'Guiyang site' to refer to the Mt. Guanfeng station in Guiyang. Systematic observations of ^{210}Pb concentrations in the past four consecutive years and their analyses are presented. The motivation of the measurements in this study is to provide additional data that serves as unique in a distinctive environment for modelling studies related to the global climate and air pollution at the monsoon region in the southwestern China of the Asian continent.

Here we expand the previous analyses (Lee et al., 2004; Wan et al., 2005c) to add three more years of new data for evaluating the relationships between ^{210}Pb and ^{222}Rn measurements occurring in the region. The record of measurements on ^{210}Pb concentrations from 20 December 2001 to 7 February 2006 will be presented in this paper. The annual mean of ^{210}Pb concentrations in surface air obtained at the current Guiyang site is compared with those obtained at other stations in the world. Finally, we investigate the washout coefficients by examining the amounts of ^{210}Pb concentrations dissolved in rain water and comparing with those in surface air. The data collected at the site and the analyses presented here will help in improving the understanding of regional and global transport modelling for any species including gases and aerosols in the atmosphere and for validation of dust transport model in Asian region (Lee et al., 2006).

2. Sampling

2.1. Location

The monitoring site is situated on the piedmont of Mt. Guanfeng (26°34′19.3″ N, 106°43′22.1″ E, 1080 m above sea level), Guiyang City, Guizhou, China (Fig. 1). Guizhou is located in the middle of the Yunnan-Guizhou Plateau, and also lies on the transitional slope zone from the Qinghai-Tibet Plateau to hilly plains in the east. Stationary fronts, with an alternation of warm and cold air currents, are generally formed on the Yunnan–Guizhou Plateau due to the relatively high elevation of the site above sea level in a great geomorphic relief. The plateau region is susceptible to global climate changes and is a distinctive environmental unit with great altitude gradients that form a complex landscape with a subtropical climate influenced by monsoons.

2.2. Materials and methods

The sampling system was set up at the State Key Laboratory of Environmental Geochemistry's (SKLEG) roof, which was about 30 m above the ground surface. The Fuji-Model sampler was equipped with a rectangular filter (20.3 cm × 25.4 cm), with a sampling area of 407 cm^2. The filtering material consisted of three layers with a 100% polypropylene network film that was sandwiched between two 100% polyester protective cotton layers. The velocity of airflow in the sampler varied with a range between 0.4 and 1.6 m^3 min^{-1}. Filter samples have been collected weekly, that is, the sampling duration for each sample being about 168 h from 20 December, 2001 to 7 February, 2006. The air volume collected on each sample ranged approximately 6×10^3 to 2×10^4 Standard Cube Meter (SCM), which was calibrated to the International Standard Cube Meter by observed daily air temperature and average local pressure. Each filter sample collected on the polypropylene network filtering film was divided in half. One half was analysed by SKLEG at the Institute of Geochemistry, Chinese Academy of Sciences in China and the other half was analysed by EML in U.S. for verification of analysis.

The specific activity of ^{210}Pb for its γ-spectrum was analysed by an S-100 Series 16384 Multichannel Energy Spectrometer manufactured by U.S. Canberra Co. First, each aerosol sample

collected on the polypropylene network filtering film were sealed in a certain shape and directly put on a GC5019 co-axial HPGe detector for counting. The instrumental performance was perfectly stable without channel drift during the counting. The counting times of each individual sample ranged between 4×10^4 and 2×10^5 s. The peak position of γ-spectrum energy for ^{210}Pb is at 46.5 KeV. The uncertainties for counting errors were limited to one sigma standard deviation and the sample errors are approximately between 1.4 and 6.4%. The quality of data has been thoroughly examined by comparing the results of independent analyses of SKLEG with EML. The verifications and comparisons have been published in the previous study of Lee et al. (2004).

We have collected rain water samples from February 2002 to May 2006. In the year of 2002, a small rain collector with an area of 314 cm^2 was used. At the beginning of year 2003, we changed to use a larger rain collector with an area of 4800 cm^2 to collect more rain water. Samples collected prior to 2005 in rain were analysed by $Fe(OH)_3$ co-precipitation method. Samples collected after 2005 were vaporized at low temperature by using high-inspissation process. Because the high-inspissation process is under the weak-acid condition and the pyrogenation is used at low temperature ($\leq$60 °C). It is likely that the loss of ^{210}Pb by the wall absorption onto the container is negligible. Also, ^{210}Pb could attach tightly to the aerosol particles because abundant particles appear in the rain water sample, instead of adsorbing into the container. In addition, we have thoroughly washed the containers in the transfer of solution from one container to another. We have calibrated our instruments with radioactive standard source and the data are reproducible. Thus, we believe that the high inspissation was simple and accurate. The obtained solid samples from co-precipitation and the liquid samples from high-inspissation were counted in a co-axial HPGe detector for determining the ^{210}Pb concentrations dissolved in rain water.

Different efficiency curves were used for calibration of inhomogeneity of samples. Liquid standards (Isotope Products Laboratories, Gentech Indistries Inc., USA; Mutinuclide Standard Solution, Catalogue No. 7500, Source No.: 586-26-6) were used for the calibration of the instrument.

3. Results and discussions

3.1. Distributions of ^{210}Pb concentrations in surface air

Figure 2 shows the results of the weekly measurements of ^{210}Pb concentrations in surface air and the weekly rainfalls observed, as well as the weekly mean air temperatures measured at the Guiyang site. The first week was from December 20 to 26, 2001 and the last week 212 was from January 31 to February 7, 2006.

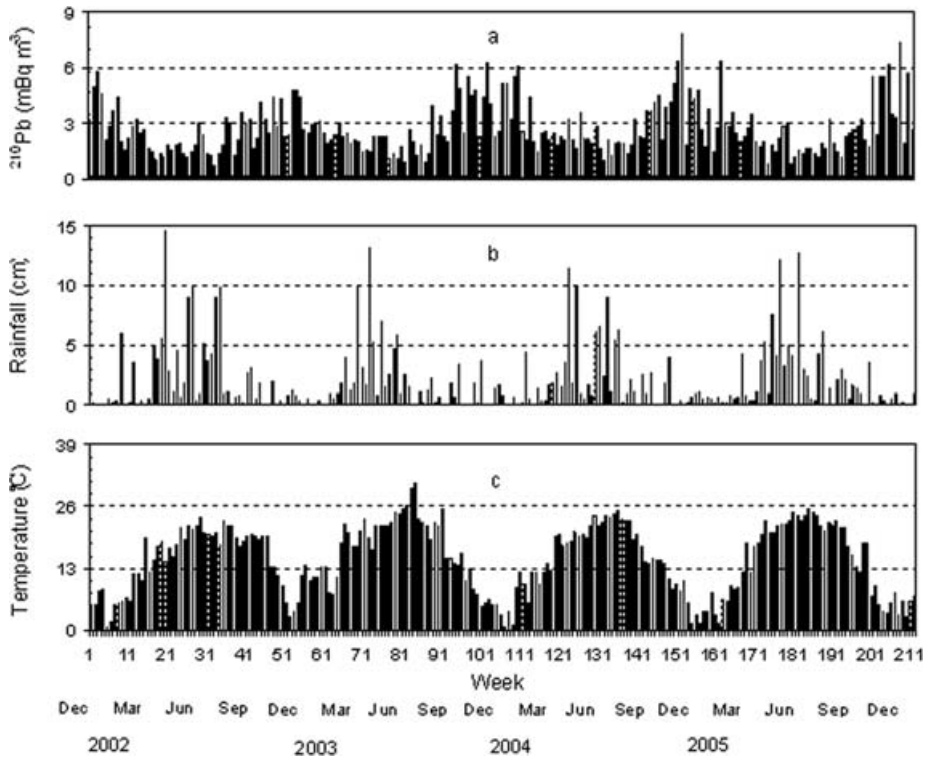

Fig. 2. Variations of the measurements from December 20, 2001 to February 7, 2006 for: (a) weekly sampling of ^{210}Pb concentrations in surface air, (b) weekly precipitation and (c) weekly mean of air temperatures.

Table 1. The monthly and annual mean of ^{210}Pb concentrations (mBq m^{-3}) in surface air at Mt. Guanfeng, Guiyang City from 1 January 2002 to 31 December 2005

	2002	2003	2004	2005	Mean
January	3.9 ± 0.5	4.0 ± 0.4	3.7 ± 0.5	3.6 ± 0.5	3.8 ± 0.2
February	3.1 ± 0.5	2.8 ± 0.1	4.5 ± 0.6	3.4 ± 0.7	3.5 ± 0.2
March	2.5 ± 0.2	2.4 ± 0.2	2.5 ± 0.4	3.0 ± 0.3	2.6 ± 0.1
April	1.7 ± 0.2	2.2 ± 0.1	2.3 ± 0.1	2.5 ± 0.2	2.2 ± 0.1
May	1.5 ± 0.1	1.7 ± 0.1	2.4 ± 0.2	1.6 ± 0.2	1.8 ± 0.1
June	1.5 ± 0.1	2.1 ± 0.1	2.3 ± 0.3	2.3 ± 0.3	2.1 ± 0.1
July	2.1 ± 0.2	1.2 ± 0.1	1.9 ± 0.2	1.3 ± 0.1	1.6 ± 0.1
August	1.4 ± 0.2	1.9 ± 0.2	1.8 ± 0.1	1.5 ± 0.1	1.6 ± 0.1
September	2.5 ± 0.3	2.5 ± 0.4	2.2 ± 0.3	1.9 ± 0.3	2.3 ± 0.2
October	2.7 ± 0.3	3.6 ± 0.6	3.4 ± 0.3	2.4 ± 0.2	3.0 ± 0.2
November	3.4 ± 0.3	4.4 ± 0.4	3.7 ± 0.3	3.1 ± 0.5	3.7 ± 0.2
December	3.1 ± 0.3	4.4 ± 0.5	5.2 ± 0.8	4.8 ± 0.5	4.4 ± 0.3
Mean	2.5 ± 0.5	2.8 ± 0.7	3.0 ± 0.7	2.6 ± 0.6	2.7 ± 0.6

Note: The measurement errors are expressed as 1 *SD*, but the monthly and annual mean values were based on the week-to-week weighted calculation with 95% confidence bounds.

It is seen from Fig. 2a that the ^{210}Pb concentrations show annual periodical variations with varying magnitude. The time intervals of periodical variations vary from 2 to 6 weeks. Such short-periodical variations are directly related to the meteorological conditions and the synoptic weather system passing over the site, which could develop significant rainfalls (Fig. 2b) and air temperature changes (Fig. 2c). It is generally seen that the rainfall occurring in the summertime washes out air particles to reduce the ^{210}Pb concentrations in surface air. The variations of ^{210}Pb concentrations in surface air could also be due to the variations in the sources of air masses. Intrusion of oceanic air masses could result in lowering ^{210}Pb concentrations, while intrusion of continental air masses could result in higher ^{210}Pb concentrations (Preiss et al., 1996; Paatero et al., 2003; McNeary and Baskaran, 2003). Table 1 presents the monthly and annual mean of ^{210}Pb concentrations in surface air for four consecutive years. The results are calculated based on the week-to-week weighted calculation with 95% confidence bounds. In Table 1, the mean values in the sixth column represent the average values over 4 yr. The monthly mean of ^{210}Pb concentrations in surface air exhibited a U-pattern with high values appearing in winter from November to February and low values in late spring and summer and early autumn from May to August, with a progressive increase in the autumn. The annual mean ^{210}Pb concentrations in the years of 2002, 2003, 2004 and 2005 are 2.5 ± 0.5, 2.8 ± 0.7, 3.0 ± 0.7 and 2.6 ± 0.6 mBq m^{-3}, respectively, with an annual variation of 0.5 mBq m^{-3} over these 4 yr. This annual variation could be related to changes in the exhalation of ^{222}Rn during favourable meteorological conditions. The overall average of the annual mean of ^{210}Pb concentrations in surface air over 4 yr is 2.7 ± 0.6 mBq m^{-3} as shown in the bottom row of sixth column in Table 1. The mean value is higher than the values measured at most other places (KUeR Report, 1982). For example, Fig. 3 shows the comparisons of distributions of monthly mean of ^{210}Pb concentrations with those at the Moosonee Station (51°16′N, 80°30′W, 10 m above sea level) of Ontario in Canada and at the Barrow Station (71°10′N, 156°30′W, 4 m above sea level) of Alaska in U.S. The data from Ontario and Alaska are reorganized after reference (EML website). Here, we have randomly selected Moosonee and Barrow stations for the purpose of demonstration because long records of measurements are available at these two stations. In the near future, we might choose other stations, such as the site reported in McNeary and Baskaran (2003) that may be more appropriate for comparisons in terms of latitude and elevation. It is clearly seen that the ^{210}Pb concentrations at Guiyang site are significantly larger than those at Moosonee and Barrow stations. The annual mean concentration of ^{210}Pb measured at Moosonee and Barrow stations are 0.5 and 0.4 mBq m^{-3}, respectively, that are about 1/6 of the annual mean concentration of ^{210}Pb at Guiyang site. Generally, the high concentrations of ^{210}Pb appear during the winter that usually has the lower atmospheric boundary layer for accumulating the ^{210}Pb concentrations. The low concentrations are in summer that can frequently develop the convective precipitation to reduce the ^{210}Pb concentrations in the air. In Fig. 3, the ratios of high concentration of ^{210}Pb in winter over the low concentration of ^{210}Pb in summer are about 2.2, 2.7 and 9.9 for Guiyang, Moosonee and Barrow stations, respectively. During winter months in Barrow, the presence of arctic haze tends to retain ^{210}Pb in aerosols and hence the activity in aerosols decreases from winter to summer (Baskaran and Shaw, 2001). It is clearly shown that the ratio at Guiyang site is much smaller than the one at Barrow station. It means that the ^{210}Pb concentrations at Barrow station are significantly reduced. This might suggest that the less reduction

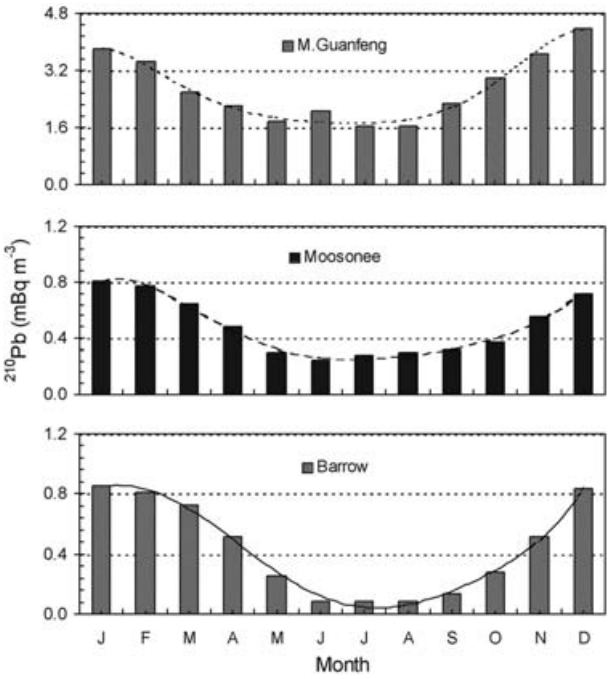

Fig. 3. Comparisons of monthly mean of ^{210}Pb concentrations in surface air at Guiyang site (January 2002–December 2005) with those at Moosonee Station in Ontario, Canada (August 1974–August 1976 and October 1981–August 1999) and at Barrow Station at Alaska, U.S.A. (September 1975–Auguast 1976 and October 1981–September 1999).(The data from Ontario and Alaska are reorganized by monthly averages after EML website at http://www.eml.doe.gov/databases/sasp/sasp_data_search.htm)

of the ^{210}Pb concentrations at Guiyang site could be controlled by the significant emissions of ^{222}Rn in the region that had distinctive features of geological and atmospheric environment. Even though those stations are much different in terms of latitude, longitude and elevation, but their annual distributions of monthly mean of ^{210}Pb concentrations showed a parallel trend exhibiting a U-pattern with high values appearing in winter and low values in summer and early autumn in Fig. 3. The release of ^{222}Rn that decays to ^{210}Pb increases the ^{210}Pb concentrations in the atmosphere.

Comparing the ^{210}Pb concentrations in surface air at Guiyang site with those at Freiburg in Switzerland, which has similar topography and landforms as Guiyang, we have found that the ^{210}Pb concentrations at Freiburg site are small with the average of annual mean concentrations being 0.44 $\pm$ 0.02 mBq m^{-3}, based on the measurements taken from the years of 1973 to 1981 (KUeR Report, 1982). It is approximately equal to 1/6 of the average of annual mean of ^{210}Pb concentrations at Guiyang site. Also, the average value of annual mean of ^{210}Pb concentrations obtained from a large number of monitoring stations throughout the world ranged from 0.02 mBq m^{-3} to 0.71 mBq m^{-3}, based on the EML archived dataset accumulated for many years in the past. At Guiyang site, the average annual mean of ^{210}Pb concentrations of 2.7 $\pm$ 0.6 mBq m^{-3} is clearly at least four times higher than the value reported from other stations in the world. Such high concentrations of ^{210}Pb at Guiyang site agree with the results of model simulations of high concentrations and depositions of ^{210}Pb in the southwesten region of China (e.g. Lee et al., 2004). High concentrations of ^{210}Pb in surface air can lead to high depositions of ^{210}Pb during the precipitation, particularly for the amount and duration of precipitation that will be further discussed in a later section.

3.2. *Geological environment of the region and its effects on the distributions of ^{210}Pb concentration in surface air*

There are many factors that can contribute to high concentrations of ^{210}Pb in the air in the central part of Guizhou province. One of the main factors is the sources of ^{222}Rn that decays to ^{210}Pb. The release rate of ^{222}Rn depends on the concentration of ^{226}Ra in the soil, which is derived ultimate from the decay of ^{238}U. In the central Guizhou, the soils generally contain yellow soil and limestone rocks with 78.1 $\pm$ 3.5 and 79.7 $\pm$ 5.2 Bq kg^{-1}, respectively, of high specific activity of ^{226}Ra (Chen and Zheng, 1985). The average specific activity of ^{226}Ra in the sediments of Lake Hongfeng in the central part of Guizhou province is estimated to be 66.1 $\pm$ 8.8 Bq kg^{-1} (Wan et al., 1990). These significantly elevated amounts of ^{226}Ra can contribute a great deal of ^{222}Rn exhalation to the atmosphere in the region of study. Another factor that could contribute to the high concentrations of ^{210}Pb in surface air is the dissolution and erosion of carbonate rocks. In the region of central Guizhou, there is a widespread of carbonate rocks. As U content in these rocks are not high (e.g. 1.14 mg kg^{-1} for limestone and 0.92 mg kg^{-1} for dolomite), carbonate rocks can easily dissolve and erode. The rate of chemical weathering for carbonate rocks in the central Guizhou may reach 0.06 mm a^{-1} (Wan et al., 1995) that is approximately two times faster than the rate in siliceous rocks. Hence, the elements in the uranium-series could release ^{222}Rn into the groundwater through the processes of dissolution and erosion. The average specific activity of ^{226}Ra in the groundwater is about 40.6 $\pm$ 8.6 Bq m^{-3} (Chen and Zheng, 1985). The increase of ^{222}Rn in the water column would increase the ^{222}Rn concentrations in the atmosphere, but the ^{222}Rn emission from water is much smaller than the one from soil. The third factor that could contribute high concentrations of ^{210}Pb in surface air is due to the exploitation of coal and phosphorous resources for use in homes and manufacturers. Coal and phosphorus are the two dominant mineral resources in Guizhou Province. Exploiting the coal, particularly for the coal burning, may lead to the release of ^{226}Ra or ^{222}Rn, which has a high concentration of ^{238}U of about 0.2 $\pm$ 0.1 g kg^{-1} of uranium contents. Phosphorous ores in Guizhou province contain uranium from 0.1 g kg^{-1} up to 0.9 g kg^{-1}. The exploitations and processes of coal and phosphorus resources would necessarily

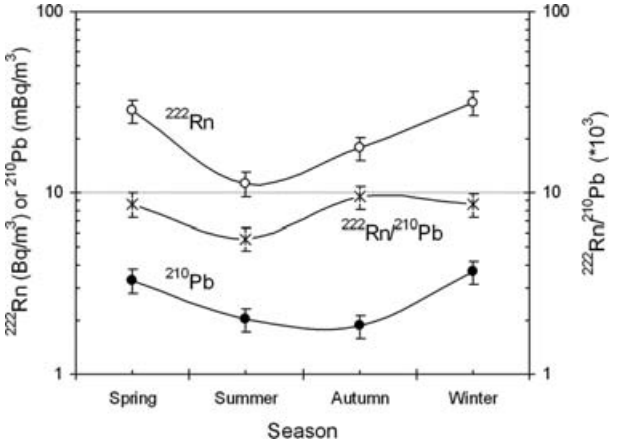

Fig. 4. Comparisons of seasonal variations of ^{210}Pb concentrations in surface air from January of 2002 to December of 2005 with the history data of ^{222}Rn concentrations taken from Chen and Zheng (1985) at Guiyang site.

give rise to the release of uranium and other members in the ^{238}U series. Especially during the process and production of phosphorite containing uranium to produce the phosphorous fertilizers in Guizhou, the specific activity of ^{226}Ra may reach $(2 \sim 5) \times 10^3$ Bq kg^{-1}.

Although there exists a geochemical background of high uranium-series elements as mentioned above in the region of study, the major factor controlling the high ^{210}Pb concentrations in surface air is the exhalation of ^{222}Rn from the limestone rocks and/or soils containing high ^{226}Ra. Generally, the variations of ^{210}Pb concentrations in surface air should reflect the ^{222}Rn concentrations in the air. Chen and Zheng (1985) have reported the seasonal variations of atmospheric ^{222}Rn concentrations measured at the Guizhou area. In this study, we have compared their ^{222}Rn concentrations with our measurements of ^{210}Pb concentrations. The results of the comparisons of seasonal variations of ^{210}Pb and ^{222}Rn concentrations are shown in Fig. 4. We have seen that the seasonal variations of ^{210}Pb concentrations are consistent with those in ^{222}Rn concentrations. The ^{222}Rn concentrations are high in winter and spring, so are the ^{210}Pb concentrations. The ratios of ^{222}Rn to ^{210}Pb concentrations are 8.6×10^3, 5.6×10^3, 9.5×10^3 and 8.6×10^3 in the spring, summer, autumn and winter, respectively. The summer minimum in both ratio and value is seen due to the frequently convective precipitation in the summer, leading to preventing the release of ^{222}Rn and scavenging of radionuclide aerosols in the atmosphere.

3.3. Effects of meteorological conditions on the distributions of ^{210}Pb concentrations in surface air

The exhalation rate of ^{222}Rn is greatly influenced by the meteorological conditions (Li, 2000). Generally, in the hottest months of each year from June to August, ^{222}Rn concentrations in the air

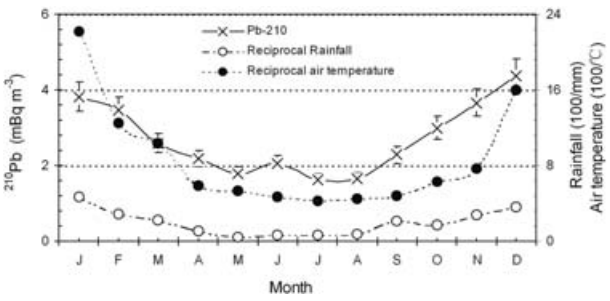

Fig. 5. Comparisons of monthly mean of ^{210}Pb concentrations in surface air with the reciprocals of monthly average of air temperatures and rainfalls over the years from 2002 to 2005 at the Guiyang site.

are the lowest because of the atmospheric high-pressure system suppressing the release of ^{222}Rn from surface soil. If the vertical mixing is active in the summer months, the ^{222}Rn-enriched (as well as ^{210}Pb-enriched) air masses could move vertically upward to increase ^{222}Rn concentration and, reversely, the ^{222}Rn-depleted air masses from stratosphere could move down to lower the concentration. In contrast, the coldest months of each year from December to January are generally with the highest ^{222}Rn concentrations (Cheng et al., 2001). Figure 5 shows the distributions of the monthly mean of ^{210}Pb concentrations with the monthly average of air temperatures and the rainfalls observed over the years from 2002 to 2005. It is seen that the variations of ^{210}Pb concentrations in surface air are well fitted with the reciprocal of air temperatures and rainfalls.

In order to examine the correlations of ^{210}Pb concentrations with the air temperatures and the rainfalls, we have performed the correlation analysis. Figure 6 shows the correlations of monthly mean concentration of ^{210}Pb corresponding with the rainfalls from December 2001 to March 2006. In the central Guizhou, the monthly mean of ^{210}Pb concentrations in surface air decreases along with the increase of the rainfall. The trend of decrease is in a negative power function of rainfall as shown in the legend of Fig. 6. Once the ^{210}Pb concentrations in surface air are below

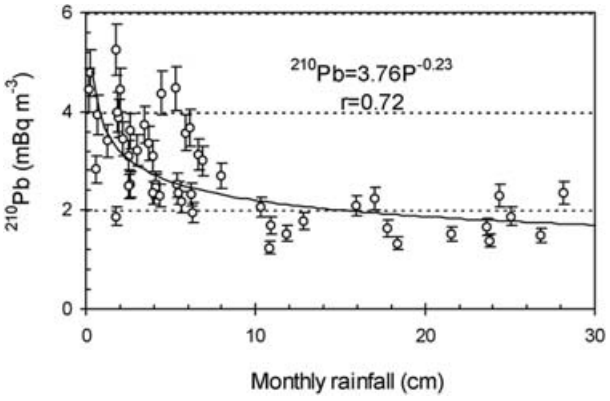

Fig. 6. Correlation of monthly mean ^{210}Pb concentrations with the rainfalls.

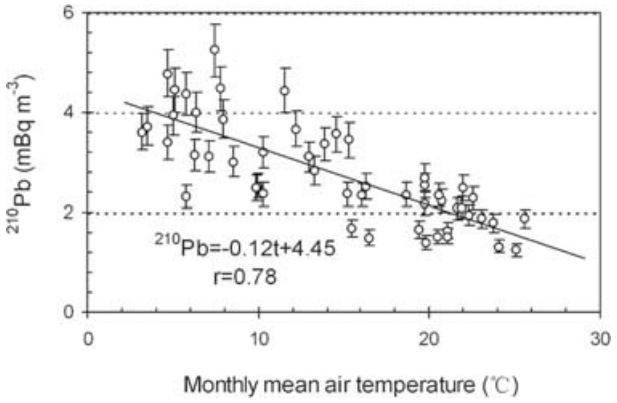

Fig. 7. Correlation of monthly mean ^{210}Pb concentrations with the monthly mean air temperature.

2 mBq m^{-3}, the rainfalls would not make much further reduction of the concentrations. This is an observation result only. Perhaps, the reason has been from the equilibrium relationship of ^{210}Pb in the surface air and in the rainfall. Figure 7 shows the monthly mean ^{210}Pb concentrations corresponding with the monthly mean air temperatures over the same period. It is seen in Fig. 7 that the ^{210}Pb concentrations fit well in a negative linear curve with the air temperature data. The correlations of ^{210}Pb concentrations with the local air temperatures are consistent with the warm summer season for a relatively low release of ^{222}Rn from soils as mentioned before. Therefore, the characteristics of the distributions of monthly mean of ^{210}Pb concentrations are clearly constrained by the atmospheric precipitation and air temperature in the region. The feature of high ^{210}Pb distributions in surface air at the Guiyang site reflects the characteristics of joint influences of the monsoon climate with the inland–plateau features of a low diffusion surrounded by the lower atmospheric boundary layer and high emissions of ^{222}Rn at the site.

3.4. *Comparisons of ^{210}Pb concentrations dissolved in rain water with ^{210}Pb concentrations in surface air*

Atmospheric precipitation can wash out the atmospheric aerosols and the amount of precipitation plays a very important role on the removal of radionuclides from the air. On the other hand the precipitation can prevent the release of ^{222}Rn from soils. Consequently, the rainfall removing ^{210}Pb aerosols in the rain water may exert a great influence on the measurements of ^{210}Pb concentrations in the air. Figure 8 shows the relationships of the ^{210}Pb concentrations dissolved in rain water with the ^{210}Pb concentrations in surface air from February 2002 to May 2006. Note that the logarithm scale in y-coordinate is used in Fig. 8. We have classified the data into two groups according to the dissolved ^{210}Pb concentrations in rain water. In Fig. 8, group 1 (opened circles) is for the low concentrations of ^{210}Pb in rain water, that is, C_w being below 0.2 Bq L^{-1}, while group 2 (solid circles) is for the high concentrations of ^{210}Pb in rain water which is C_w above 0.2 Bq L^{-1}. It is clearly seen that the ^{210}Pb concentrations dissolved in rain water increase with the increase of the ^{210}Pb concentrations in surface air during the cold months from October to May as shown in a dashed line. Note that the boundary value of 0.2 Bq L^{-1} is an observation result only. Perhaps, the reason has been from the equilibrium relationship of ^{210}Pb in the surface air and in the rainfall. However, the dissolved ^{210}Pb concentrations in rain water remain low for the low concentrations of ^{210}Pb in surface air during the warm months from April to November as shown in a thin solid line. Note that C_w could fluctuate during the seasonal changes in April/May from cold to warm months and in October/November from warm to cold months. For instance, if C_w in April is above 0.2 Bq L^{-1}, it belongs to group 2. Otherwise, it is in group 1. Hence, the fluctuation in April is included in both groups as shown in the legend of figure. It is also seen in Fig. 8 that the concentrations of ^{210}Pb resolved in rain water are generally smaller than the concentrations of ^{210}Pb in surface

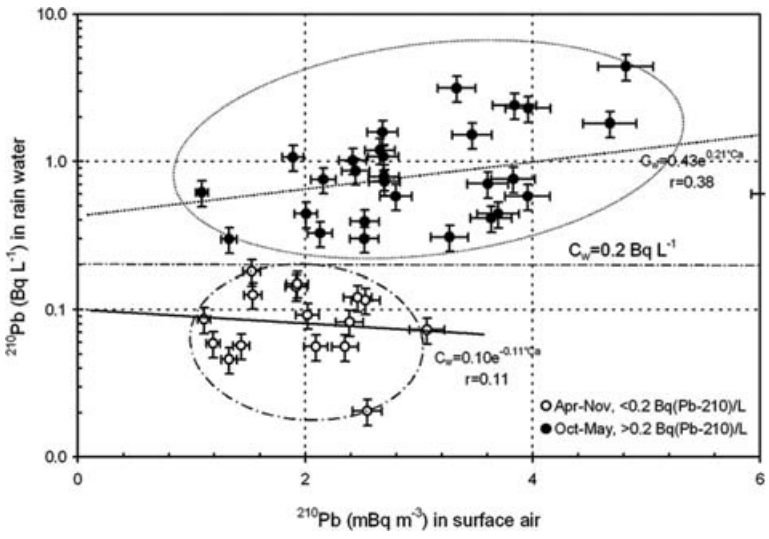

Fig. 8. Relationships of ^{210}Pb concentrations dissolved in rain water with ^{210}Pb concentrations in surface air. The data is divided into group 1 (opened circles) for the period from April to November and group 2 (solid circles) for the period from October to May. Least square fit is used for group 1 (thin solid line) and group 2 (dashed line).

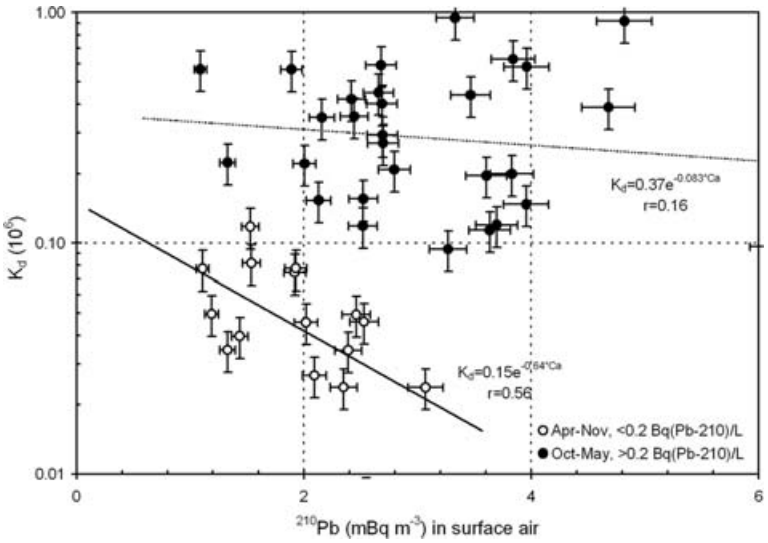

Fig. 9. The partition coefficient (K_d) versus ^{210}Pb concentrations in surface air. The data is divided into group 1 (opened circles) for the period from April to November and group 2 (solid circles) for the period from October to May. Least-square fit is used for group 1 (thin solid line) and group 2 (dashed line).

air. In the near future, we will examine and assess if there is any correlation of aerosol mass concentration in the atmosphere with the ^{210}Pb concentration in the rain water during the process of wet scavenging.

The concentrations of ^{210}Pb dissolved in precipitation rain can provide useful information about the partition coefficient that is used in modelling transport for estimating the amount of substances in the air being washed out by precipitation (McNeary and Baskaran, 2003). Some part of ^{210}Pb dissolved in rain water is derived from the original raindrop (cloud and/or mist), but other part of ^{210}Pb dissolved in rain water is from atmospheric aerosols during the washout precipitation. In order to differentiate the 'washout coefficient', we have use the 'partition coefficient'. If C_w and C_a indicates the concentrations of ^{210}Pb dissolved in the rain water and in the air contemporaneously, the partition coefficient, K_d of ^{210}Pb in water–air is:

$$K_d = C_w/C_a. \quad (1)$$

Figure 9 shows the results of calculations for K_d, based on the analysis of ^{210}Pb concentrations dissolved in rain water for the period from February 2002 to May 2006. It is seen in Fig. 9 that the water-air partition coefficient, K_d is generally high in winter and spring from October to May (solid circles). During the wintertime, the long duration of rain can continually dissolve atmospheric aerosols, which are attached with high concentrations of ^{210}Pb. The values of C_w could reach more than 1.0 Bq L^{-1} and K_d could be higher than 0.4×10^6 during these months. In summer and autumn from April to November, C_w is lower than 0.2 Bq L^{-1} (see Fig. 8) and K_d is lower than 0.1×10^6 (see Fig. 9). Such low values of C_w and K_d may be attributed to the convective precipitations in response to the influence of monsoons during the summertime. The convective precipitation can effectively wash out atmospheric aerosols to reduce the values of C_a. Once C_a in the atmosphere is low, the amount of ^{210}Pb dissolved in the rain water will be lower during the continued precipitation. Thus, the values of C_w do not increase dramatically for long duration of rain during the summer months. Therefore, the type of rain and the duration of precipitation play important roles for the changes of K_d values. These illustrate the seasonal differences with respect to the concentrations of ^{210}Pb dissolved in rain water and in surface air caused by the atmospheric precipitation. Overall, the concentrations of ^{210}Pb in rain water and in surface air are not only dependent on the duration of rain in various seasons, but also on the intensity and the type of rain. A large body of literature already exists on this subject matter reporting similar observations (Baskaran, 1995; McNeary and Baskaran, 2003).

4. Summary

Activity concentrations of ^{210}Pb have been widely used as a tracer for studying the transport processes related to catchment erosion, lake sedimentation and atmospheric circulation. Better understanding of the variations of ^{210}Pb concentrations is the key to improve the studies of tracing lake sedimentation and catchment erosion in the soil-sediment system. The average of annual mean of ^{210}Pb concentration over 4 yr is estimated to be 2.7 $\pm$ 0.6 mBq m^{-3} which is at least four times larger than the values reported from a number of monitoring stations throughout the world. The principal factor of producing such high ^{210}Pb concentrations in surface air over this region was the significant ^{222}Rn gas exhalation from soil that is enriched with ^{226}Ra elements from the uranium-series. We found that the ^{210}Pb concentrations in surface air were high in winter and low in summer and early autumn. We also found that the distributions of ^{210}Pb concentrations in surface air in the central Guizhou were well correlated with the reciprocal of regional air temperature and precipitation. In this study, by comparing the ^{210}Pb concentrations dissolved in rain water with the ^{210}Pb concentrations in surface air, we found

that the type and duration of rain depending on the season were important factors for the changes in calculating the water–air partition coefficients. The information obtained from the calculations of partition coefficients would be useful for improving the modelling in wet scavenging schemes used in global models.

Finally, the variations of ^{210}Pb concentrations in surface air appear periodically in yearly basis and do not change significantly in magnitude. This could be due to the fact that the region contains relatively constant ^{226}Ra sources and it has the relatively stable release of ^{222}Rn from the decay of ^{226}Ra to the atmosphere, in addition to the steady influences of continent and oceanic air masses in the region.

5. Acknowledgments

This research project was financially supported jointly by National Natural Science Foundation of China (Grant No. 40773071, 40373038, 40525011, 40632011) and the Chinese Academy of Sciences (Grant No. kzcx2-yw-102), and the EML of the United States of America. The authors would also like to thank Dr. Baskaran and other anonymous reviewers for their reviews and comments.

References

Appleby, P. G. 1997. Sediment records of fallout radionuclides and their application to studies of sediment-water interactions. *Water, Air Soil Pollut*. **99**, 573–586.

Balkanski, Y. J., Jacob, D. J., Gardner, G. M., Graustein, W. C. and Turekian, K. K. 1993. Transport and residence times of tropospheric aerosols inferred from a global three-dimensional simulation of ^{210}Pb. *J. Geophys. Res*. **98**, 20 573–20 586.

Baskaran, M. 1995. A search for the seasonal variability on the depositional fluxes of ^{7}Be and ^{210}Pb, *J. Geophys. Res*. **100**, 2833–2840.

Baskaran, M. and Shaw, G. E. 2001. Residence time of arctic haze aerosols using the concentrations and activity ratios of ^{210}Po, ^{210}Pb, and ^{7}Be. *J. Aerosol Sci*. **32**(4), 17–26.

Chen, Z. Y. and Zheng, Z. H. 1985. The environmental radioactivity level and the forecast of its contaminated trend at Guizhou area (in Chinese). In: *The Environmental Forecast for 2000 AD at Guizhou Area*. Special Issue, No. 21, 14.

Cheng, Y. X, Wang, N. P, Hou, S. L. and Liu, Q. C. 2001. Theoretical research on Radon from earth into air (in Chinese). *Radioprotect. Commun*. **21**(2), 15–18.

EML website at http://www.eml.doe.gov/databases/sasp/sasp_data_search.htm.

Feichter, J., Brost, R. A. and Heimann, M. 1991. Three-dimensional modeling of the concentration and deposition of ^{210}Pb aerosols. *J. Geophys. Res*. **96**, 22 447–22 460.

Graustein, W. and Turekian, K. K. 1996. ^{7}Be and ^{210}Pb indicate an upper troposphere source for elevated ozone in the summertime subtropical free troposphere of the eastern North Altantic. *Geophys. Res. Lett*. **23**(5), 539–542.

KUeR (**K**ommission zur **U**berwachung der **R**adioaktivitat) Bericht. 1982. 25 Jahre Radioaktivitat Uberwachung in der Schweiz (in German). Phys. Inst., Univ. of Freiburg, Freiburg, Switzerland, 1982, 1–46.

Lee, H. N. 2003. Issues and challenges of using natural radionuclides as tracers for atmospheric studies, *Global Atmosphere Watch Report No. 155*, World Meteorological Organization TD No. 1201, 30–34.

Lee, H. N. and Feichter, J. 1995. An intercomparison of wet precipitation scavenging schemes and the emission rates of ^{222}Rn for simulation of global transport and deposition of ^{210}Pb. *J. Geophys. Res*. **100**, 253–270.

Lee, H. N., Wan, G. J., Zheng, X. D., Sanderson, C. G., Josse, B. and co-authors. 2004. Measurements of ^{210}Pb and ^{7}Be in China and their analysis accompanied with global model calculations of ^{210}Pb. *J. Geophys. Res*. **109**, D22203, doi:10.1029/2004JD005061.

Lee, H. N., Y. Igarashi, M. Chiba, Aoyama, M. Hirose, K. and co-authors. 2006. Global model simulations of the transport of Asian and Sahara dust: total deposition of dust mass in Japan, *Water, Air, Soil Pollut*. **169**, 137–166.

Lee, H. N., Tositti, L., Zheng, X. and Bonasoni, P. 2007. Analyses and comparisons of variations of ^{7}Be, ^{210}Pb and ^{7}Be/^{210}Pb with ozone observations at two GAW stations from high mountains. *J. Geophys. Res*. **112**, D05303, doi:10.1029/2006JD007421.

Li, R. J. 2000. Determination of radon eduction rate and exploration on its influence factors (in Chinese). *Uranium Mining and Metallurgy* **19**(1), 56–61.

McNeary, D. and Baskaran, M. 2003. Depositional characteristics of ^{7}Be and ^{210}Pb in southeastern Michigan. *J. Geophys. Res*. **108**(D7), 4210, doi:10.1029/2002JD003021.

Paatero, J., Hatakka, J., Holmén, K., Eneroth, K. and Viisanen, Y. 2003. Lead-210 concentration in the air at Mt. Zeppelin, Ny-Ålesund, Svalbard. *Phys. Chem. Earth Parts A/B/C*, **28**(28–32), 1175–1180.

Preiss, N., Mélières, M. A., Pourchet, M. 1996. A compilation of data on lead-210 concentration in surface air and fluxes at the air-surface and water-sediment interfaces. *J. Geophys. Res*. **101**, 28 847–28 862.

Rasch, P. J., Feichter, J., Law, K., Mahowald, N., Penner, J. and co-authors. 2000. A comparison of scavenging and deposition processes in global models: results from the WCRP Cambridge Workshop of 1995. *Tellus* **52B**, 1025–1056.

Rehfeld, S. and Heimann, M. 1995. Three dimensional atmospheric transport simulation of the radioactive tracers ^{210}Pb, ^{7}Be, ^{10}Be and ^{90}Sr. *J. Geophys. Res*. **100**, 26141–26161.

Ritchie, J. C. and McHenry, J. R. 1990. Application of radioactive fallout Cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: a review. *J. Environ. Qual*. **19**, 215–233.

Robbins, J. A. and Edgington, D. N. 1975. Determination of recent sedimentation-rates in Lake Michigan using Pb-210 and Cs-137. *Geochimica et Cosmochimica Acta* **39** (3), 285–304.

Santschi, P. H., Guo, L., Walsh, I. D., Quigley, M. S. and Baskaran, M. 1999. Boundary exchange and scavenging of radionuclides in continental margin waters of the Middle Atlantic Bight: implications for organic carbon fluxes. *Continental Shelf Research* **19**, 609–636.

Turekian, K. K., Benninger, L. K., Dion, E. P. 1983. ^{7}Be and ^{210}Pb total deposition fluxes at New Haven, Connecticut and at Bermuda. *J. Geophys. Res*. **88**(C9), 5411–5415

Wan, G. J., Santschi, P., Sturm, M., Farrenkothen, K., Lueck, A. and co-authors. 1987. Natural (^{210}Pb, ^{7}Be) and fallout (^{137}Cs, $^{239,\ 240}$Pu, ^{90}Sr) radionuclides as geochemical tracers of sedimentation in Greifensee, Switzerland. *Chem. Geol*. **63**, 181–196.

Wan, G. J., Huang, R. G., Wang, C. S. and Rong, J. 1990. A variance of vertical profile for $^{210}Po_{ex}$ at the top of sediment in Hongfeng Lake. *Chin. Sci. Bull*. **35**(22), 1910–1914.

Wan, G. J., Bai, Z. G., Zhu, L. J., Yuan, D. X., Rong, Q. T. and co-authors. 1995. *The Carbonate Rocks and the Environments (in Chinese)*. Seismological Press, Beijing, 16–40.

Wan, G. J., Bai, Z. G., Qing, H., Mather, J. D., Huang, R. G. and co-authors. 2003. Geochemical records in recent sediments of Lake Erhai: implications for environmental changes in a low latitude-high altitude lake in southwest China. *J. Asian Earth Sci*. **21**(5), 489–502.

Wan, G. J., Chen, J. A., Wu, F. C., Xu, S. Q. and Santschi, P. H. 2005a. Sudden enhancement of sedimentation flux of $^{210}Pb_{ex}$ as an indicator of lake productivity as exemplified by Lake Chenghai, *Science in China (Series D)* **48**(4), 484–495.

Wan, G. J., Chen, J. A., Wu, F. C., Xu, S. Q., Bai, Z. G. and co-authors. 2005b. Coupling between $^{210}Pb_{ex}$ and organic matter in sediments of a nutrient-enriched lake: An example from Lake Chenghai, China. *Chem. Geol*. **224**(4), 223–236.

Wan, G. J., Yang, W., Wang, S. L., Wan, E. Y., Wu, F. C. and co-authors. 2005c. Characteristics of U-pattern distribution of high-concentration ^{210}Pb in surface air at central Guizhou, China. *Chin. Sci. Bull*. **50**(16), 1750–1755.

Yeager, K. M. and Santschi, P. H. 2003. Invariance of isotope ratios of lithogenic radionuclides: more evidence for their use as sediment source tracers. *J. Environ. Radioact*. **69**(3), 159–176.