Study of Elemental Mass Size Distributions of Aerosol in Lijiang, a Background Site in Southwest China

Ren-jian Zhang[1], Zhen-xing Shen[2*], Han Zou[1] , Wei Wang[3], Yuemei Han[2], Juan Zhou[2]

[1] Key Laboratory of Regional Climate-Environment Research for Temperate East Asia (REC-TEA), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

[2] Department of Environmental Science and Engineering, Xi'an Jiaotong University, Xi'An 710049, China

[3] Chinese Research Academy of Environmental Sciences, Beijing 100012, China

Abstract

Elemental mass size distributions of atmospheric aerosols over Mount Yulongxue, Lijiang, Yunnan Province, China were monitored by an 8-stage cascade sampler and analyzed using PIXE method. The results show that crustal elements such as Mg, Al, Ca, Si, K, and Fe are the main compositions of atmospheric aerosols over the Mount Yulongxue Region and account for more than 82% of a total of 20 elements determined. The concentration of soil dust in Lijiang is very low with a value of 3.22 μg/m^3. Concentration of Pb, Se, and Br in atmospheric aerosol in Lijiang is much higher than that over Mount Qomolangma Region. High enrichment factors of S, As, Se, Br, and Pb in atmospheric aerosol in Lijiang were observed. These results indicate that the impact of human activity in Lijiang can't be ignored.

Keywords：Lijiang; Atmospheric aerosols; Elemental composition.

INTRODUCTION

Lijiang, located in northwest Yunnan Province, is one of the famous historic and cultural cities in China. This region is where remnants of the ancient Dongba culture are preserved (Li *et al.*, 2001). Mount Yulongxue is set in the beautiful Jinshajiang River valley,15 km north of Lijiang. Snow-covered year round, Mount Yulongxue is 5596 m above sea level and the lowest in latitude among the Snow Mountains of China. The thirteen snow-capped peaks tower and span across about 35 km. The Yulong area has enjoyed the reputation of being one of great natural beauty. Human activity in the Lijiang area is relatively light, but the ecosystem is fragile. Recently, it has been observed that the snow-covered area has decreased rapidly, the snow line is higher

*Corresponding author.

E-*mail:* zxshen@mail.xjtu.edu.cn

and the glacier has been retreating (He and Zhang, 2004). These changes have been attributed to global climate change as well as increased human activity associated with the development of tourism in the area (He and Zhang, 2004).

Very few studies on the atmospheric environment in the Lijiang area have been published: Guo *et al.* (2004) investigated climate change and Liu and Hong (1996) analyzed the chemical composition of precipitation. Studies on the chemical composition of atmospheric aerosol in Lijiang have not been performed. Observation and analysis of changes in atmospheric chemical components could help evaluate the impact of human activities, so that the environment can be protected.

Some atmospheric observation over mountain Yulongxushan has been performed with the support of the public welfare project "The Integrated Impact of the Asian Brown Cloud over China and Corresponding Strategy" by the China Ministry of Science and Technology, The current study presents the observational results on the aerosol chemical composition. The purpose of this study was to observe and analyze the background aerosol chemical composition over the Yunan-Guizhou Plateau in southwest China, and to assess potential impacts of human activity.

METHODOLOGY

Observation site

The observation site located at the top of a two-floor building of a power transformer station (27°13′N, 100°11′E, 3100 m above sea level), 30 km north of Lijiang, near a mountain road. There are no local pollutant emission sources near the observation station. The average temperature during the sample period from December 28, 2003 to January 6, 2004) was -2℃, and the surface barometric pressure was about 700 mb.

Aerosol sampling

A cascade sampler was used to collect atmospheric aerosols in the following size ranges: < 0.25, 0.25-0.5, 0.5-1, 1-2, 2-4, 4-8, 8-16, and > 16 μm, corresponding to 8 stages ranging from 0 to 7.

Because the atmosphere in the Lijiang area is very clean, the sampling duration was about 48 hours and four groups of samples were obtained during the observation period. The timing of the 4 samples was: (1) from 1400 LST, Dec. 28 2003 to 1400 LST, Dec. 30; (2) from 1500 LST, Dec. 30 to 1300 LST, Jan. 2, 2004 (power down from 1700 LST, Dec. 30 to 1420 LST, Dec. 31); (3) from 1330 LST, Jan. 2, 2004 to 1430 LST, Jan. 4, and; (4) from 1500 LST, Jan. 4, 2004 to 1500 LST, Jan. 6, 2004. The air flow of the sampler was measured in the beginning and at the end of the sampling. The average of the flow data was set as the sample flow rate.

Element analysis

The aerosol samples collected in Lijiang were analyzed by a Particle Induced X-Ray Emission (PIXE) technique at the Institute of Low Energy Nuclear Physics, Beijing Normal University. The PIXE analyses were

carried out using 2.5 MeV proton bombardments with a beam of 30-40 nA. The concentrations of 20 elements—Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, and Pb—were determined for each sample (Zhu and Wang, 2000; Zhang *et al.*, 2001, 2003).

RESULTS AND DISCUSSION

Elemental concentration

Thirty-two samples were obtained during this study. The mean value averaged from the samples was set as the background concentration value in the Lijiang area. Table 1 presents the distribution of aerosol element mass concentration with aerosol size for the 20 listed elements. The total concentration of the 20 elements observed was 1.99 μg/m^3, which is lower than that of 2.87 μg/m^3 over Mount Qomolangma (Zhang *et al.*, 2001). The total mass concentration in Lijiang was exactly the same value as that observed in the summer of 1989 over Wudaoliang area, Qingzang plateau（35°17′N, 93°36′E, 4650 m ASL） (Yang *et al.*, 1994). The total mass concentration in Lijiang was 44% lower than the one observed over Mount Qomolangma and was about 1/15 of that observed over Beijing in December 1999 (the latter was 29.1 μg/m^3) (Zhang *et al.*, 2002). These results suggest that the aerosol concentration in Lijiang area is very low. The major elements of atmospheric aerosol in Lijiang are crustal elements (Mg, Al, Ca, Si, K, and Fe) with a concentration of about 82% of the total 20 elements determined.

Table 2 describes the ratio of aerosol elemental concentration observed in Lijiang to that over Beijing (Zhang *et al.*, 2002). The crustal elemental concentrations in Lijiang were much lower than those observed in Beijing, indicating that floating dust in Lijiang area occurred scarcely. The concentrations of V, Cr, Mn, As, Se, Br, and Pb in Lijiang were relatively high compared to the those over Beijing, suggesting that the environment in Lijiang has been polluted.

The aerosol concentrations in Lijiang area were compared with the Qomololangma background station (Zhang *et al.*, 2001). The mass concentrations of Pb, Se and Br were 17.7 ng/m^3, 5.8 ng/m^3 and 10.1 ng/m^3 in Lijiang, respectively; while the mass concentrations of Pb, Se, and Br were 2.93 ng/m^3 0.91 ng/m^3 and 1.88 ng/m^3 in Qomolangma, respectively. The concentrations of V, As, Se, Br, and Pb in the Lijiang area were evidently higher than those collected at Qomolangma. Other previous studies show that Br concentrations of 12.8 ng/m^3 and 5.2 ng/m^3 in November and December 1999, respectively, over Beijing (Zhang *et al.*, 2001), and 46.1 ng/m^3 over Taiyuan in December 2004 (Liu *et al.*, 2006), suggesting that the source of Se, Pb and Br. Pb may also come from vehicle gas emission which contains high concentrations of Pb.

Size distributions of elements

Fig. 1 illustrates the aerosol size distribution of total concentration for the 20 elements in Lijiang and Mount Qomolangma. It shows that the difference in concentration between Lijiang

Table 1. Distribution of aerosol element composition with the aerosol size in Lijiang (ng/m^3).

Level	0	1	2	3	4	5	6	7	Sum
Size (μm)	<0.25	0.25~0.5	0.5~1.0	1.0~2.0	2.0~4.0	4.0~8.0	8.0~16.0	>16.0	
Mg	32.9	8.8	8	9.5	8.5	9.6	19.4	6.6	103.2
Al	28.6	45	19.3	24.4	27.2	29	28	22.5	223.9
Si	42.3	69	67.6	76.1	90.6	110.2	84.3	78.9	619
P	24.7	15.3	13.7	14.6	12.5	11.6	14.7	12.8	120
S	29.6	19.7	6.8	2.3	1.8	5.6	1.3	4.6	71.7
Cl	9.5	2.3	1.7	0.8	1	2.3	0.8	1.2	19.5
K	20.6	9.9	5.1	5.7	7.8	11.9	6.2	6	73.2
Ca	151.9	35.7	36.3	48.5	64	109.2	74.9	59.7	580.1
Ti	8.9	2.7	0.4	0.8	1.8	1.7	1	1	18.1
V	0.6	0.7	0.3	0.2	0.3	0.4	0.3	0.3	2.8
Cr	1	0.2	0.4	0.4	0.4	0.3	0.2	0.2	3
Mn	0.8	2.2	2.2	1.2	1	0.9	0.6	0.7	9.5
Fe	2.4	5.9	5.9	9.4	14.5	23.5	12.3	9.6	83.6
Ni	0.5	0.3	0.3	0.4	0.4	0.6	0.2	0.3	2.9
Cu	0.6	1.6	1.1	0.4	0.4	0.5	0.2	0.3	5.1
Zn	7.4	1.4	1.7	0.7	0.6	0.6	0.4	0.5	13.2
As	1.2	0.6	0.7	1	0.8	0.5	0.9	0.6	6.2
Se	1.6	1.2	0.6	0.6	0.5	0.8	0.5	0.2	5.8
Br	1.8	1	1.6	1.1	1.1	1.5	1.1	0.9	10.1
Pb	2.5	2.8	2	1.7	2.2	2.8	1.8	2	17.7
Total	369.1	226.2	175.4	199.5	237.3	323.3	249.1	208.6	1988.4

Table 2. Ratios of aerosol elemental concentrations in Lijiang to Beijing.

Element	Mg	Al	Si	P	S	Cl	K	Ca	Ti	V
Ratio	13.5%	3.7%	5.8%	11.7%	4.6%	2.6%	8.2%	14.9%	7.6%	92.5%
Element	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Br	Pb
Ratio	11.0%	9.6%	3.1%	4.8%	3.0%	7.6%	11.0%	53.0%	202.0%	33.4%

and Qomolangma was not large. The mass size distribution exhibits two peaks in both areas, with maximum values occurring at aerosol sizes < 0.25 μm and 4-8 μm in Lijiang area, and at 4-8 μm and > 16 μm over Mount Qomolangma. Fig. 1 indicates that the concentration of small-size aerosol in the Lijiang area was relatively high. To a certain degree, the aerosol size distribution reflects its source; the peak values at sizes < 0.25 μm

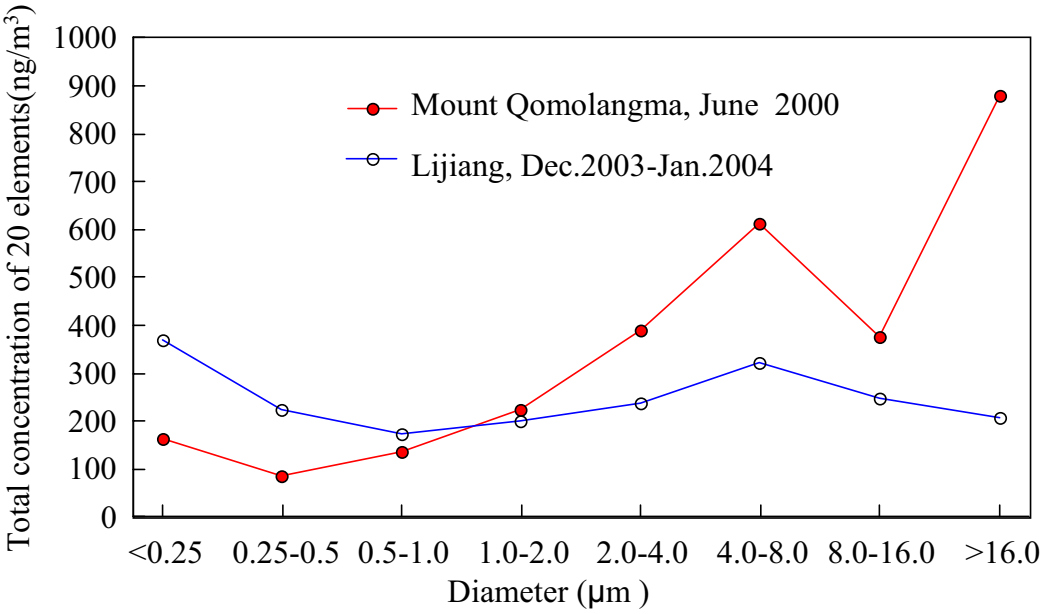

Fig. 1. Aerosol size distribution of total concentration for the 20 aerosol elements in Lijiang and Mount Qomolangma.

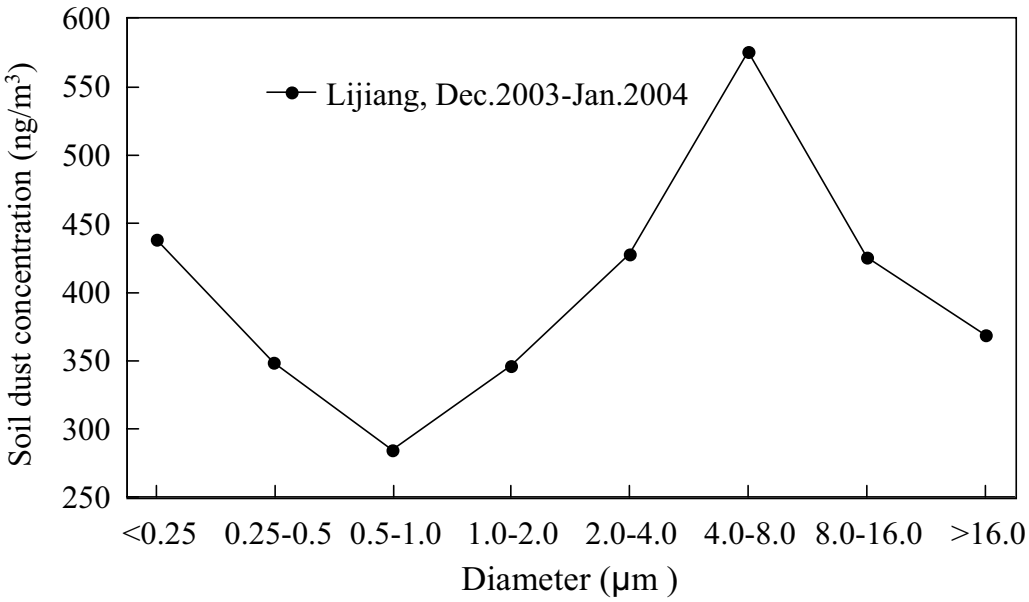

Fig. 2. Size distribution of soil mass concentration of aerosols in Lijiang.

and 4~8 μm suggest coal burning and local soil dust sources.

The mass concentration of soil dust can be estimated by summing up the concentration of several elements predominantly associated with soil plus oxygen, assuming the compounds involved are the most common oxides (Malm *et al.*, 1994). The formula to calculate the soil dust concentration is given as:

$$C_{soil} = 2.2C_{Al} + 2.49C_{Si} + 1.63C_{Ca} + 2.42C_{Fe} + 1.94C_{Ti} \quad (1)$$

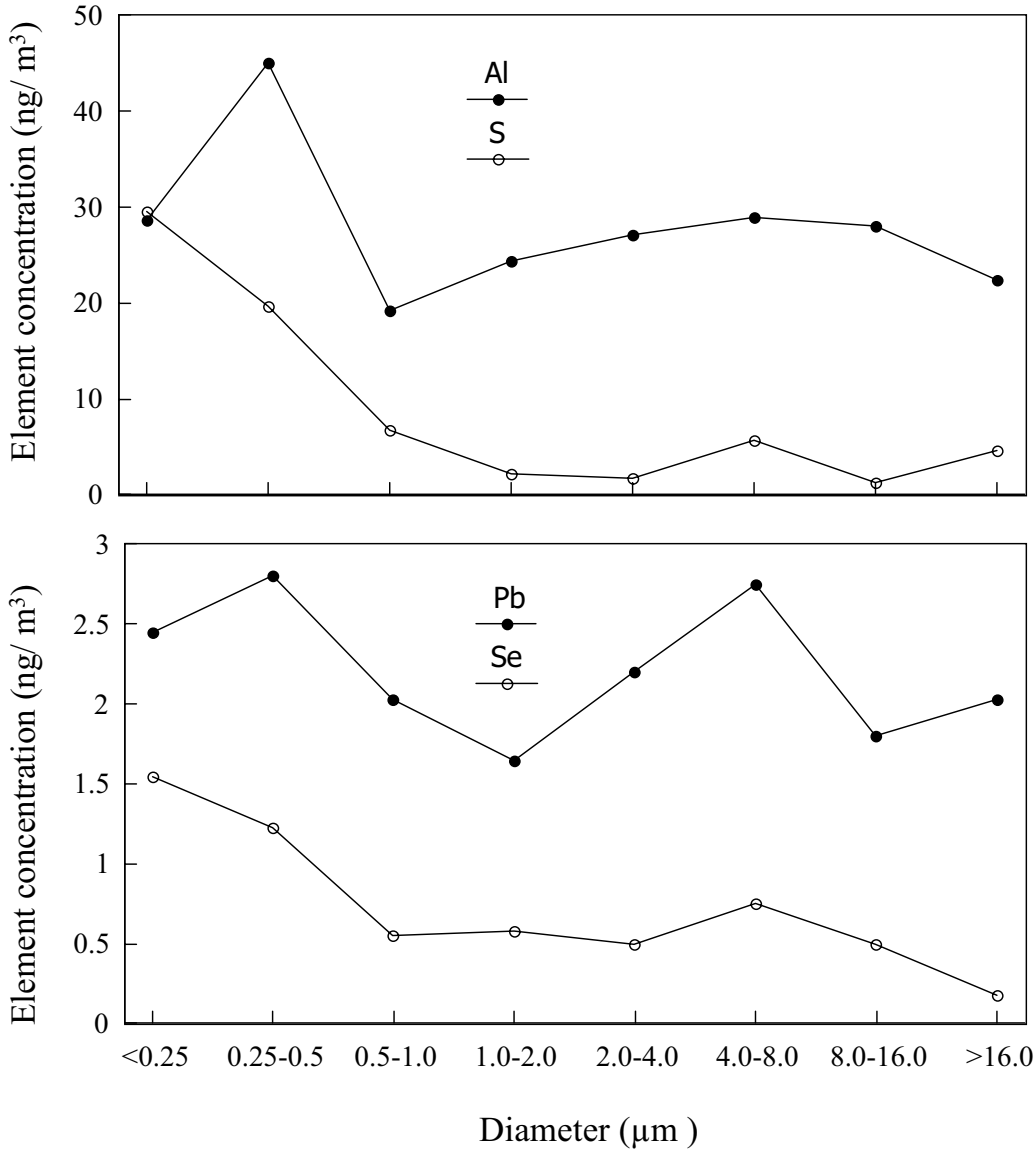

Fig. 3. Size distributions of elemental concentrations of Al, S, Se, and Pb.

where C_{soil} is the calculated soil dust concentration, and C_{Al}, C_{Si}, C_{Ca}, C_{Fe}, and C_{Ti} represent the elemental concentrations of Al, Si, Ca, Fe, and Ti in aerosols. Fig. 2 presents size distribution of soil mass concentration of aerosols in Lijiang, showing a double peak in fine mode (< 0.25 μm) and coarse mode (4-8 μm). The concentrations of soil dust in fine mode (d < 2 μm), coarse mode (d > 2 μm) and total aerosols are 1.42, 1.8, and 3.22 μg/m^3, respectively. The concentration of soil dust is very low in Lijiang.

Fig. 3 shows the size distributions of elemental concentrations of Al, S, Se, and Pb: the distribution of Al was mainly in fine mode; while most S and Se mass were in fine mode as well, reflecting a possible coal-burning source. The concentration of Pb

Table 3. Enrichment Factors of aerosols in Lijiang.

Element	Mount Yulongxue		Beijing area (Zhang *et al.*, 2002)	
	Fine aerosol	Coarse aerosol	Fine aerosol	Coarse aerosol
Mg	0.55	1.59	0.44	0.7
Al	0.28	0.99	2.72	1.77
S	43.98	38.78	649.11	76.91
Cl	21.54	30.36	790.28	50.52
K	0.31	0.93	1.95	0.72
Ca	1.46	6.37	2.06	2.83
Ti	0.57	0.93	0.88	1.47
V	2.43	6.15	1.44	0.44
Cr	3.73	8.12	13.59	5.92
Mn	1.31	2.52	7.16	1.99
Fe	0.09	0.91	1.48	1.36
Ni	3.79	14.1	31.07	18.54
Cu	13.1	18.88	395.48	30.66
Zn	31.17	21.85	319.06	24.93
As	375.89	1133.01	2697.35	509.48
Se	15297.12	29080.55	15536.72	4141.57
Br	431.46	1389.82	233.05	23.67
Pb	134.64	509.85	522.87	40.96

was low and its size distribution showed a double peak mode.

Element enrichment analysis

An element enrichment analysis was used to identify the origins of elements from natural or anthropogenic sources. The enrichment factor is defined as (Winchester *et al.*, 1981; Liu *et al.*, 2006):

$$EF = (C_{element} / C_{refercence})_{air} / (C_{element} / C_{refercence})_{crust} \quad (2)$$

where $C_{element}$ is the concentration of any element, $C_{refercence}$ is the concentration of reference element. Generally, Al, Fe, or Si are chosen as reference elements, and in the case of this study, Si was chosen. The crustal concentrations were obtained from Winchester *et al.* (1981).

Table 3 presents the element enrichment factors in Lijiang. Enrichment factors of crustal elements, such as Al, Ca, Fe, and Ti, were not very large; however, the element enrichment factors of S, As, Se, Br, and Pb, which are closely related with human activities, were very high. The aerosol element S in the Lijiang area mainly results from the chemical

reaction in heterogeneous phase of SO_2, produced by burning coal. The enrichment factors of elements Se, Br, and Pb were higher than those in Beijing, suggesting that the impact of human activities (especially coal burning) on the environment in Lijiang cannot be ignored. The total soil mass concentration in Lijiang was very low.

CONCLUSIONS

Chemical composition of atmospheric aerosols and their size distributions over Mount Yulongxue, Lijiang, Yunnan Province, China were observed by cascade sampler and analyzed by using PIXE method. These background data were compared with those observed over Mount Qomolangma and the Beijing area. The main results are as follows:

(1) The total concentration of 20 elements in Lijiang area was lower than that of those collected in Qomolangma and 1/15 of those collected in Beijing. The main elements in Lijiang aerosol were crustal elements, Mg, Al, Ca, Si, K, and Fe, which account for more than 82% of the total 20 elements.

(2) The concentration of soil dust in Lijiang is very low, with a value of 3.22 $\mu g/m^3$.

(3) The enrichment factors of aerosol elements, S, As, Se, Br, and Pb, were high in the Lijiang area, which suggests that the impact of human activities cannot be ignored.

ACKNOWLEDGEMENTS

This research is supported by National Basic Research Program of China (No. 2006CB400501), National Science Foundation of China (No. 40675074 and 40599422), and the Knowledge Innovation Project (No. KZCX3-SW-231).

REFRENCES

Guo, S., Chang, Y., Chen, H., Zhang, X., Chen, Z. and Yan, H. (2004). The Multifractal Characteristics of Climate Change in Yunnan, China, *J. of Yunnan University,* 26: 325-330. (In Chinese)

He, Y. and Zhang, D. (2004). Climatic Warming is the Major Reason for Glacier Retreat on Mt. Yulong, China, *J. Glaciology and Geocryology,* 26: 230-231. (In Chinese).

Liu, J. and Hong, Y. (1996). Analysis of Chemical Composition of Precipitation in a Typical Background Site, *J. Environ. Chem.* 15: 391-398. (In Chinese).

Li, J., Hu, Y. and Zhu, K. (2001). Study of the Climatic Resources for Tour in the Lijiang Area, *J. Chengdu University of Information Technol.* 16: 179-182. (In Chinese).

Liu, X., Wang, X., Zhang, R., Zhang, H., Hu, L., Zhu, L. and Shi, Lei. (2006). Analysis on the sources and characteristics of particles in winter in Taiyuan, *Journal of Graduate School of the Chinese Academy of Sciences*. 23: 494-499. (In Chinese).

Malm, W. C., Sisler, J. F., Huffman, D., Eldred, R.A. and Cahill, T.A. (1994). Spatial and Seasonal Trends in Particle Concentration and Optical Extinction in the United States, *J. Geophys. Res.* 99 (D1):

1347-1370

Winchester, J.W., Lu, W., Ren, L., Wang, M. and Maenhau, W. (1981). Fine and Coarse Aerosol Composition from a Rural Area in Northern China, *Atmos. Environ.* 15: 933-937.

Yang, L., Wang, M., Lu, G. and Gong, Y. (1994). The Observation and Research for the Continental Aerosol Background Characteristics in the Northern Part of the Qinghai-Xizhang Plateau, *Plateau Meteorology,* 13: 135-143. (In Chinese).

Zhang, R., Wang, M., Hu, F. and Xu, Y. (2002). Elemental Concentration and Distribution of Atmospheric Particles Before and During Heating Season in Beijing, *J. of Graduate School of the Chinese Academy of Sciences.* 19: 75-81. (In Chinese).

Zhang, R., Zou, H., Wang, M., Zhou, L. and Zhu, G. (2001). Observation and Analysis on Elemental Composition of Atmospheric Aerosols Over the Mount Qomolongma Region, *Plateau Meteorology,* 20: 234-238. (In Chinese).

Zhang, R., Wang, M., Zhang, X. and Zhu, G. (2003). Analysis on the Chemical and Physical Properties of Particles in a Dust Storm in Spring in Beijing, *Powder Technol.* 137: 77-82.

Zhu, G. and Wang, G. (2000). Cross-Check of PIXE Analysis between Three International Laboratories, *Environ. Climate Res.* 5: 80-84. (In Chinese)

Received for review, June 29, 2008
Accepted, July 22, 2008