



The Elemental Composition of Atmospheric Particles at Beijing during Asian Dust Events in Spring 2004

Renjian Zhang^{1*}, Zhenxing Shen², Tiantao Cheng³, Meigen Zhang⁴, Yanju Liu⁵

¹ Key Laboratory of Regional Climate-Environment Research for Temperate East Asia, 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 Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China.

⁴ State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

⁵ Beijing Center for Physical and Chemical Analysis; Beijing 100089, China

ABSTRACT

The chemical element composition of dust particles was characterized by the ground-based samples collected at Beijing in the spring of 2004. Most of mineral and pollutant element concentrations in particles were elevated in dusty days, about 2–4 times higher than the levels in non-dusty days. Each of Si, Ca, Fe and Al accounted for over 10% of the sums of total 20 elements in mass, for example, Si was in 44.3%, 38.7% for dusty and non-dusty cases, respectively. Si, Fe, Ni or Ti can be used as an indicator of dust outflow, and Cu can be viewed as an evidence of dust particles mixing with anthropogenic contaminants as a result of coagulation processes. Mineral and pollutant elements showed a bimodal distribution in the mass particle-size distributions in both dusty and non-dusty days, but their peak concentrations fell in different size stages. Zn, Cl and Cu were mostly enriched in fine particles, Pb was enriched in intermediate sized particles, but most mineral elements, S and part of Cu were enriched in coarse particles. Mineral elements were dominated by crustal material, and pollutant elements were from non-crustal material including local and remote sources. Among the crustal material, part of Ca was originated from local construction activities. High concentration of Cu was related to the of rapidly increasing vehicles in Beijing, and the replacing of coal with diesel oil for heating fuel. Most of the mineral dust particles sampled at Beijing were originated from the Mongolian sandy soil and the Chinese loess in the spring of 2004. Using Mg/Al ratio element tracer technique method, the aerosol from outside Beijing accounted for 66.3% and 88.6% to the total mineral aerosol during dust event on 10–11 March and 28–30 March 2004, respectively.

Keywords: Dust; Chemical element composition; Asian dust event.

INTRODUCTION

Among atmospheric aerosols, mineral dust produced from windblown soils and deserts is one of the largest contributors to the global aerosol loading and has strong impacts on regional and global climates (Tegen *et al.*, 1996; Wu *et al.*, 2006), long-term climate trends (Petit *et al.*, 1990; 1999), as well as marine and terrestrial ecosystems (Martin *et al.*, 1988; Chadwick *et al.*, 1999). The climate research community has paid much attention to the long-range transport, radiative forcing and climate effects of mineral dust in the atmosphere because of their

considerable uncertainties (IPCC, 2001). Quantifying and assessing the climatic impacts of mineral dust aerosols require a knowledge of their physical, chemical, optical and radiative properties, as well as of their spatial and temporal variability (Sokolik *et al.*, 1998).

East Asia is one of the important source regions of mineral dust emission in the world (Shen *et al.*, 2006; Sun *et al.*, 2006; Zhang *et al.*, 2009). Large amounts of dust particles from the arid and semi-arid areas of Asian continent are annually emitted into the atmosphere and transported over long distance to the North Pacific and even to the North America (Iwasaka *et al.*, 1988; Duce, 1995; Wang *et al.*, 2000; Husar *et al.*, 2001; Shen *et al.*, 2005; 2007; Li *et al.*, 2006). It has been estimated that fast economic development, large areas of desert, and intensive forest and agriculture fires in this region contribute to one-fourth to one-third of the total global emissions of SO₂, organic matter, soot and dust (Chin *et al.*, 2003; Han *et al.*,

* Corresponding authors. Tel.: +86-01-62064089;
Fax: +86-01-62028604
E-mail address: zrj@mail.iap.ac.cn.

2004; Cao *et al.*, 2005; Zhang *et al.*, 2007). The regional climate effects of these aerosols are predicted to increase in the near future (Takemura *et al.*, 2001).

Beijing has about 15 million inhabitants and thus becomes one of megacities in the world who confront severe challenges of air quality management. Along with rapid economic growth and vehicle increase, the features of air pollution in Beijing are changing from typical coal-combustion pollution to a compound pollution case (Zhang *et al.*, 2004). In springtime, dust storms and local re-suspended dust due to traffic, and construction work enhance the complexity of particulate matter in Beijing (Guo *et al.*, 2004). During transport, some of the atmospheric aerosols undergo chemical modifications. For example, oxidation of gaseous material, such as SO_x and NO_x occurs on the surface of mineral dust particles (Dentener *et al.*, 1996). Also, dust particles can mix internally with sulphate and anthropogenic contaminants through coagulation process with the above types of aerosol particles (Roth and Okada, 1998; Wurzler *et al.*, 2000). As a result, dust particles change their size, shape and surface conditions, and these modifications are very important because they change both the radiative properties of dust aerosols and their ability to be a cloud condensation nucleus (Sokolik *et al.*, 2001). However, up to now, only a few measurements of aerosol mass concentration, size distribution, chemical composition, or optical properties were performed in Beijing during dust events (Bergin *et al.*, 2001; Hu *et al.*, 2002; Duan *et al.*, 2004; Wehner *et al.*, 2004; Zhang *et al.*, 2008a).

We have conducted the successive measurements and samplings of Asian dust particles in spring of Beijing since 2001. In this study, the chemical element composition of aerosol particles was characterized through the ground-based samples of size-segregated aerosols collected at Beijing during dust events in the spring of 2004.

OBSERVATIONS AND METHODS

Intensive measurements for dust particles were made at the top of a two-floor building (8 meter high above the ground) in the spring of 2004. This site was located in the Institute of Atmospheric Physics of Chinese Academy of Sciences (39.97°N, 116.37°E), near the fourth ring road, about 10 km north away from the centre of Beijing.

An eight-stage cascade impactor in a typical flow rate of 1.1 L/min (PIXE International Corp.) was employed to collect aerosol samples, with 8 size cuts in particle aerodynamic equivalent diameter (D_p) as follows: < 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, and > 16.0 μm . Millipore filters (0.4 μm porosity, Millipore Corp.) were used as the backup filters, and Mylar films (3.5 μm thickness) coated with paraffin on stage 1 and Vaseline on stage 2–7 were used for the impaction surfaces. The flow rate of air at beginning and end of each sampling intervals was recorded, and then their arithmetic average was used as the mean flow rate for each sample to calculate the volume of sampled air (Zhang *et al.*, 2005).

In order to characterize the chemical element

composition of aerosol particles, samples were analyzed by a Particle Induced X-Ray Emission (PIXE) technique, using 2.5 MeV proton bombardments with a beam of 30–40 nA (Zhang *et al.*, 2005; 2008b). Mass concentrations of 20 elements (Al, As, Br, Ca, Cl, Cr, Cu, Fe, K, Mg, Mn, Ni, P, Pb, S, Se, Si, Ti, V, and Zn) were determined for each sample. For Mg, Al, Si, P, and Cl, the detection limits is 1 ng/m³; for other elements, the detection limits were 0.1 ng/m³. For quality control, the concentrations of 20 elements were determined by PIXE in a standard reference material from the national Bureau of chemical exploration analysis, China (GSS, 1984). The detailed quality assurance was shown in Zhu and Wang (1998).

RESULTS AND DISCUSSION

In the spring of 2004, only two moderate dust events erupted over the Asian continent, from 9 to 11 March, and 28 to 30 March, which can be demonstrated by series of PM₁₀ records. However, it is still a good opportunity to investigate the properties of Asian mineral dust outflows, and their mixture with inorganic compounds from industrial pollution, organic and black carbon, and water.

Mass Concentrations of Chemical Elements

A major objective of our studies is to characterize the chemical composition of Asian mineral dust, and for this we calculated arithmetic mean trace element concentrations (Fig. 1). In general, the mean mass concentrations of above 20 elements in all the particle samples collected during dust events were larger than those during non-dusty periods, especially for mineral elements such as Si with a maximum of 30.2 $\mu\text{g}/\text{m}^3$ in dusty days. All mineral concentrations of Si, Ca, Al, Fe, K and Mg exceeded 1.5 $\mu\text{g}/\text{m}^3$ in dusty days, approximately 3.0 times higher than those in non-dusty days (Table 1). For typical pollutant elements S, Pb, Cu, Zn, As and Cl, the concentrations were lower than 1.6 $\mu\text{g}/\text{m}^3$ in dusty days, but they were elevated about 2.0 to 4.0 times over the levels in non-dusty days. In comparison with non-dusty

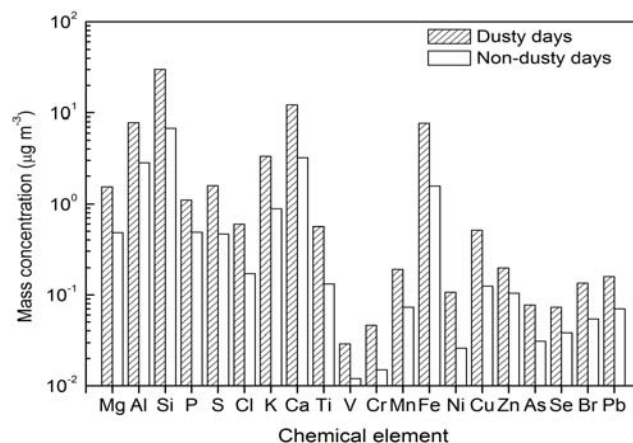


Fig. 1. Mass concentration averages of chemical elements in total size particle samples during dusty and non-dusty days.

Table 1. Elements Ratio between dusty days and non-dusty days and its mass percentage in total 20-element concentrations (pert1 and pert2 refers for dusty and non-dusty days)

Element	Ratio	Pert1 (%)	Pert2 (%)	Pert1/Pert2	Element	Ratio	Pert1 (%)	Pert2 (%)	Pert1/Pert2
Mg	3.171	2.234	2.738	0.816	P	2.279	1.619	2.762	0.586
Al	2.786	11.54	16.10	0.717	S	3.394	2.307	2.643	0.873
Si	4.447	44.26	38.67	1.144	Cl	3.474	0.879	0.983	0.894
K	3.749	4.870	5.050	0.964	Cu	4.096	0.752	0.713	1.054
Ca	3.800	17.87	18.28	0.978	Zn	1.918	0.292	0.593	0.493
Ti	4.268	0.826	0.752	1.098	As	2.535	0.113	0.174	0.652
V	2.544	0.043	0.066	0.654	Se	1.911	0.107	0.217	0.492
Cr	3.032	0.068	0.087	0.780	Br	2.507	0.198	0.307	0.645
Mn	2.622	0.282	0.418	0.675	Pb	2.283	0.233	0.397	0.587
Fe	4.965	11.35	8.886	1.277	Ni	4.054	0.157	0.151	1.043

days, the mass concentrations of other elements including Ti, Br, Mn, V, Cr, As, Se and P also displayed an increase more than 2.0 times in dusty days. This comparison revealed that during dust events, most of the chemical elements enhanced their mass concentrations in ground-based particles through different ways, for example, mineral elements came from dust outflow, and pollutant elements were from precursors, mixing and then interacting with trace gases on preexistent particle surfaces etc. Mineral elements also exhibited predominance in the sums of total 20 element concentrations in both dusty and non-dusty days (Table 1). For instance, each of Si, Ca, Fe and Al accounted for over 10.0% of the matter of total 20 elements, in particular, Si was in 44.3%, 38.7% for dusty and non-dusty cases, respectively. Compared with the non-dusty days, only Si, Ti, Fe, Ni and Cu showed an increase in the percentage of elemental concentration in total 20-element content during dusty days. Mass concentrations of other elements such as Mg, Al, V, Zn, etc. showed a decrease. This was also demonstrated by the ratios of each elemental percentage in total 20-element content during dusty days to those during non-dusty days, generally larger than 1.05 in Si, Fe, Ni, Ti and Cu (Table 1). Consequently, Si, Fe, Ni or Ti can be used as an indicator of dust outflow over Beijing, and Cu can be viewed as an evidence of dust particles mixing with anthropogenic contaminants as a result of coagulation processes.

To better understand the chemical element composition of mineral particles and their difference under dusty and non-dusty conditions, the mass particle-size distributions (MSDs) were determined for major dust-associated elements (Fig. 2). Most of chemical elements in particles sampled in dusty days showed especially higher mass loadings in all size fractions. In general, major mineral elements exhibited similar MSDs during dusty or non-dusty days, and so did typical pollutant elements. For instance, Mg, Al, Si, Ca and Fe showed a bimodal distribution in MSDs. The major peak concentrations of these elements occurred at $D_p > 16 \mu\text{m}$ in dusty days and at 2.0–16.0 μm in non-dusty days, and their secondary peak concentrations occurred at $D_p < 1.0 \mu\text{m}$ in both dusty and non-dusty days. Also, S, Cl, Cu, Zn and Pb showed a bimodal distribution in MSDs, in which, opposite to mineral elements, the major and secondary peak

concentrations occurred at $D_p < 1.0 \mu\text{m}$, $> 16 \mu\text{m}$ respectively for both dusty and non-dusty days. As dry and wet deposition processes occurred in the dust long-range transport, these MSDs of mineral elements in downwind regions were very different to the MSDs in log-normal distributions of mineral dust previously collected in sources of arid and semi-arid regions in China (Zhang *et al.*, 2001, 2005).

Table 2 presents the percentages of each elemental content in total 20-element loading in fine ($D_p < 0.25 \mu\text{m}$), intermediate sized ($0.25 < D_p < 2.0 \mu\text{m}$) and coarse ($D_p > 2.0 \mu\text{m}$) particles. In comparison with non-dusty days, Zn, Cl, and Cu in fine particles showed an increase of 17–28 %, but the ratio of other elements were reduced or a neglectable increase in dusty days. Only Pb in intermediate sized particles ($0.25 < D_p < 2.0 \mu\text{m}$) increased over 10% in dusty days to its level in non-dusty days. For coarse particles ($D_p > 2.0 \mu\text{m}$), Al, Mg, K, Ti, Ca, and S increased their relative content over 10 %, and Si, Fe and Cu fell in an increase of 3–7 %. These results indicated that during dust events, large amounts of coarse particles were transported over Beijing along with dust outflow from remote source regions, and trace gases (for example, SO_2) from anthropogenic pollution in Beijing were absorbed and then oxidated on the surfaces of mineral dust. As a result of these chemical modifications, Zn, Cl and Cu were enriched in fine particles, Pb was in intermediate sized particles, but most mineral elements, S and part of Cu were enriched in coarse particles.

Soil Mass Concentrations

According to Malm *et al.* (1994), soil mass concentrations of aerosols can be estimated by summing the concentrations of several major elements in soil, and oxygen assuming that the involved compounds are most common oxides. The formula recommended for the calculation of soil mass concentrations by elemental concentrations is as follows:

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

where C_x represents concentration and the names of crustal elements are shown as subscripts.

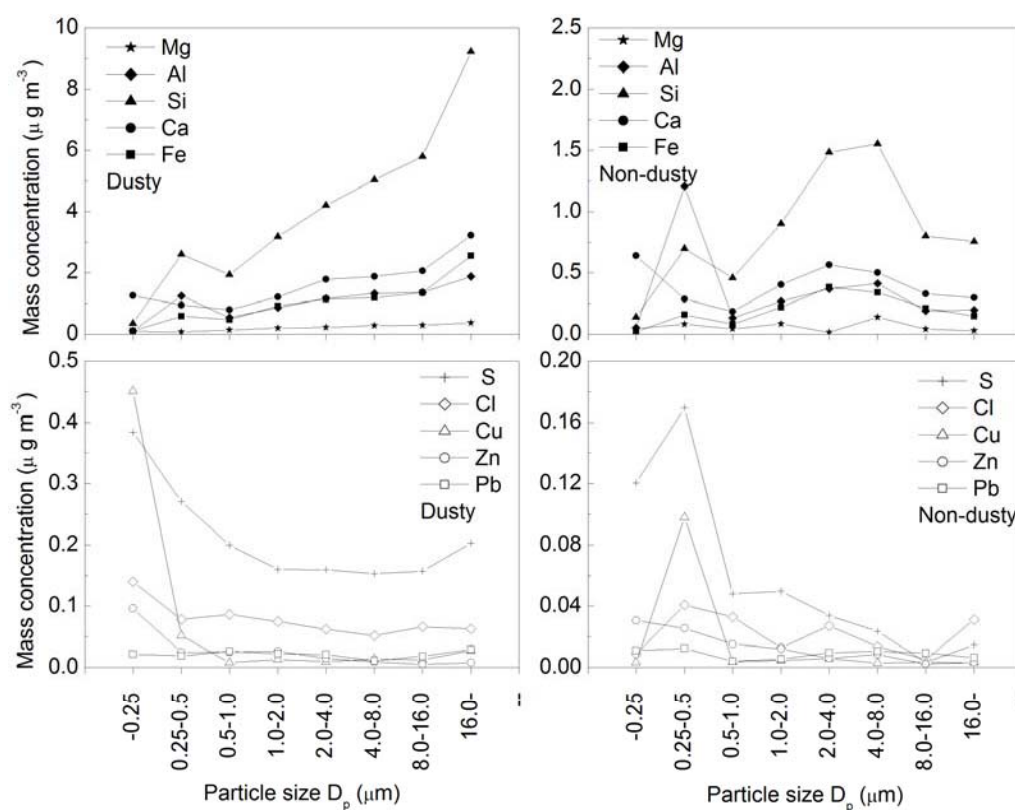


Fig. 2. Mean concentrations of dust and pollutant elements in size-segregated particle samples during dusty and non-dusty days.

Table 2. Mean percentages and enrichment factors referred to crust for major mineral and pollutant elements in fine ($D_p < 0.25 \mu\text{m}$), intermediate sized ($0.25 < D_p < 2.0 \mu\text{m}$) and coarse ($D_p > 2.0 \mu\text{m}$) particles during dusty and non-dusty days.

Sampling	Element	$C_{0.25}$ (%)	$EF_{0.25}$	$C_{0.25-2.0}$ (%)	$EF_{0.25-2.0}$	$C_{2.0-}$ (%)	$EF_{2.0-}$
Dusty day	Al	1.37	1.00	34.36	1.00	64.27	1.00
	Mg	11.52	4.17	29.36	0.67	63.66	0.73
	Si	1.17	1.09	26.85	0.87	71.98	1.21
	K	5.68	6.85	29.47	1.10	64.85	1.26
	Ti	12.31	14.22	23.94	0.78	63.75	1.24
	Fe	1.06	1.74	28.05	1.10	70.90	1.63
	Ca	11.32	31.89	25.27	2.48	63.81	3.16
	Zn	49.07	1223.17	32.17	38.36	18.79	8.89
	Cl	20.29	912.60	37.34	63.26	42.91	27.95
	S	22.90	1321.04	41.77	91.52	35.59	32.72
	Pb	13.70	1559.54	43.40	208.85	42.90	85.49
Non-dusty day	Cu	32.59	6217.86	41.38	39.94	26.02	16.63
	Al	1.66	1.00	52.52	1.00	45.82	1.00
	Mg	9.91	5.73	43.30	0.69	46.79	0.82
	Si	1.99	1.02	31.12	0.55	66.88	1.17
	K	9.46	7.85	35.99	1.00	54.56	1.32
	Ti	26.93	17.90	22.20	0.54	50.87	1.09
	Fe	1.20	0.68	31.07	0.72	67.73	1.49
	Ca	20.52	40.06	28.09	1.70	51.39	3.16
	Zn	31.75	976.75	48.28	76.77	19.97	22.97
	Cl	5.16	172.36	48.81	65.78	46.04	42.04
	S	26.02	1162.39	57.71	82.59	16.27	22.84
Pb	16.12	2154.49	32.00	128.80	51.88	202.90	
Cu	4.33	125.64	76.65	80.51	19.02	19.01	

The calculated soil mass concentrations in fine, intermediate sized and coarse particle samples during dusty and non-dusty days are shown in Fig. 3. The mean soil content of total size particles in dusty days ($132 \mu\text{g}/\text{m}^3$) was up to 4 times higher than that in non-dusty days ($32 \mu\text{g}/\text{m}^3$). However, it was much lower than the estimated soil mass concentrations ($288.4\text{--}350.2 \mu\text{g}/\text{m}^3$) of dusty period even the values ($113.8\text{--}257.4 \mu\text{g}/\text{m}^3$) of non-dusty period in Beijing in the dust-active year of 2001 (Zhang *et al.*, 2004). The significant change between these two literatures just showed the two dust storm events is relative weaker than those in 2001. Compared with fine particles, the soil mass concentrations of coarse particles increased from 12 times higher than non-dusty days to 13–38 times higher than dusty days. And, the ratio of intermediate sized to fine particles in soil content was about 7 in non-dusty days and increased to 12 in dusty days.

Enrichment Factor of Chemical Elements

Enrichment factor (EF) is calculated to compare the composition of trace elements in aerosol samples with that in reference materials. The EF of each element is calculated as follows:

$$EF = (C_x/C_r)_a / (C_x/C_r)_c \quad (2)$$

where C_x , C_r are the concentrations of X element and reference element respectively, the subscript 'a' refers to aerosol particles in the atmosphere, while 'c' refers to a reference material. Usually, Al, Si or Fe is chosen for the reference element. In our case, among the major mineral elements Al showed a relatively moderate variation in mass content between dusty and non-dusty days (Table 1), it is used as the reference element in EF calculations in this study. The crustal concentrations were obtained from Winchester *et al.* (1981).

The EF_{crust} values of 18 elements (As, Br, Ca, Cl, Cr, Cu, Fe, K, Mg, Mn, Ni, Pb, S, Si, Ti, V, Zn, Se) relative to the earth's upper crust for aerosols in all size stages are plotted in Fig. 4. Though in different variation ranges, the enrichment factors of most elements decreased in dusty days but increased in non-dusty days obviously. The EF_{crust} values of Mg, Si, K, Ti, Fe, Mn, V, Ca, and Cr showed negligibly small changes between the dusty samples and the non-dusty samples, all close to unity with maximum less than 6, indicating their major sources were undoubtedly dominated by crustal material even in non-dusty days. Compared with dust-derived elements, either dusty or non-dusty days, the EF_{crust} values of Zn, Cl, S, Pb, Cu, As and Br were much larger than 6, suggesting significant influences of non-crustal sources. In addition, Zn, Pb, As and Br had a great decrease in EF_{crust} during dusty days. In urban regions and suburbs around cities, it was found that pollutant elements are often enriched in ground-based particles through anthropogenic emissions (Nriagu and Pacyna, 1988; Cao *et al.*, 2009). As a result of high SO_2 concentration before and after dusty days, S enriched in dust particles was possibly attributed to the sorption of gaseous SO_2 and consequent transformation to

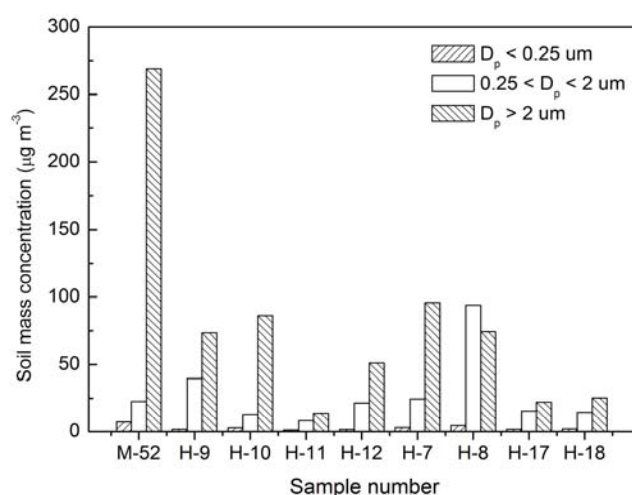


Fig. 3. Size distribution of Soil dust during dusty and non-dusty days.

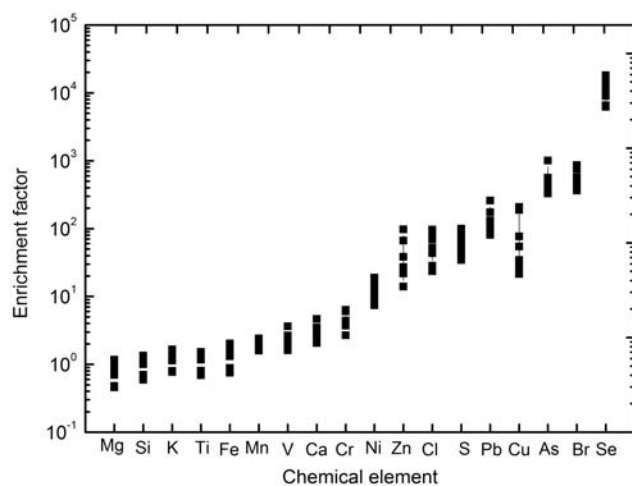


Fig. 4. Enrichment factors of elements referred to crust dust in total size particle samples.

sulfate on preexistent particle surfaces, or the mixture of sulfate aerosols with suspended dust particles (Arimoto *et al.*, 2004). The EF_{crust} of Ca was enhanced from non-dusty to dusty days, suggesting that part of Ca originated from local sources, most likely local construction activities. The EF_{crust} and concentrations of Se in dusty days were much higher than those in non-dusty days. Actually it has the highest EFs among all elements. On the basis of back trajectory analyses, Zhang *et al.* (2005) pointed that Se, as a major impurity in coal, was emitted from coal combustion including local and remote sources. The high EF_{crust} and mass concentrations of Cu were related to the emission of rapidly increasing vehicles in Beijing, and the replacing of coal with diesel oil for heating fuel since 1999 (Zhang *et al.*, 2004).

As showed in Table 2, for dusty or non-dusty days, the EF_{crust} values of Al, Si, and Fe in fine particles were similar to those in intermediate sized and coarse particles, commonly less than 2. However, Ca and Ti in fine particles had relatively high EF values larger than those in

intermediate sized and coarse particles, indicated that part of them associated to fine particles came from non-crustal sources. On the other hand, for dusty or non-dusty days, typical pollutant elements of Zn, Cl, S, Pb, and Cu in all size particles were larger than 10, and their EF values in fine fraction were remarkable higher than those of in intermediate and coarse size particles. Previous study reported that Zn, Cl, S, and Pb were mainly from industry, and Cu was from oil combustion (Zhang *et al.*, 2002; Zhang *et al.*, 2003). During dusty days, the EF_{crust} values of above five pollutant elements in fine particles increased at least 14 times higher than those in intermediate sized particles, and at least 32 times higher than those in coarse particles. Especially, in comparison with non-dusty days, the EF_{crust} of S in fine particles severely increased and elevated to a maximum 50 times higher than that of dusty days, indicated that Cu was much enriched on fine particle in dusty days. Previous study reported that the pollution materials accumulated before dust outbreaks, and then mixed with dust particles causing high concentration of pollution species, especially in weak dust storm events (Wang *et al.*, 2005).

It is known that mineral dust particles are emitted from the arid and semi-arid lands linked to surface soil erosion, vegetation sparse, damaged even destroyed under certain circumstances (Tegen *et al.*, 2004). To identify the primary sources of mineral dust particles qualitatively, we calculated the EFs of 20 chemical elements referred to crust, Mongolian sandy soil, Chinese soil and Chinese loess (Fig. 5, Sun *et al.*, 2004). In both dusty and non-dusty days, the EFs of major mineral elements were commonly lower than 10 in intermediate sized and coarse particles, with the minimal values to Mongolian sand and loess. However, part of these elements such as Ca, Ti, Mn and Mg were slightly enriched in fine particles, suggesting that their emissions may include non-crustal sources. Typical pollutant elements of Cu, Zn and Pb were commonly enriched in particles during both dusty and non-dusty days, especially in fine particles, with the highest EFs related to Mongolian sand and loess. These results revealed that most of the mineral dust particles sampled at Beijing were originated from the Mongolian sand soils and the Chinese loess in the spring of 2004. However, investigation on exact sources and corresponding emissions of ground-based aerosol particles is beyond the scope of this study.

Contribution of Local and Non-Local Sources to Beijing Mineral Dust

The ratios of Mg/Al element tracer technique were estimated to present the contribution of local and non-local sources into Beijing mineral dust (Sun *et al.*, 2004). The method is shown in Eq. (3)

$$(Mg/Al)_{\text{aerosol}} = m \times (Mg/Al)_{\text{local}} + n \times (Mg/Al)_{\text{non-local}} \quad (3)$$

$$m + n = 1$$

where

- $(Mg/Al)_{\text{aerosol}}$: Average ratio of Mg/Al in aerosol,
 $(Mg/Al)_{\text{local}}$: Average ratio of Mg/Al in local soil sample (in this case, the value of 0.46 was used (Zhou *et al.*, 2007)
 $(Mg/Al)_{\text{non-local}}$: Average ratio of Mg/Al in non-local soil sample.

24-hour mass back trajectories arriving at 1000 m above ground level (at 06 UTC) were calculated for Beijing using the NOAA HYSPLIT 4 trajectory model to investigate the soil dust transport pathways. The results showed the air masses passed from north to Northwest through Gobi and desert regions Inner Mongolia, and then moving to Beijing (the figure was omitted). In this case, the value of 0.12 in Duolun desert in south Inner Mongolia can represent the average ratio of Mg/Al in non-local soil sample (Sun *et al.*, 2004). Using above Mg/Al ratio element tracer technique method, the aerosol from outside Beijing accounted for 66.3% and 88.6% to the total mineral aerosol during dust event on 10–11 March and 28–30 March 2004, respectively. These results clearly indicated that the sources from outside Beijing contributed significantly to the mineral aerosol in Beijing, and much more in spring than that in summer/winter (Sun *et al.*, 2004). As during dust period in spring in Beijing, more frequent and stronger north or northwest wind bring much more dust particles from outside Beijing and result in the much greater contributions of non-local sources to aerosols in Beijing.

CONCLUSIONS

The ground-based samples collected at Beijing in the spring of 2004 offered an opportunity to examine the chemical element composition of mineral dust after long-range transport over the heavily populated city of eastern China. During dust events most of chemical elements in particles were enhanced in mass concentration. Mineral elements exhibited a predominance in the sums of total element loadings either dusty or non-dusty days. Si, Fe, Ni or Ti can be used as an indicator of dust outflow, and Cu can be viewed as an evidence of dust particles mixing with anthropogenic contaminants. Zn, Cl and Cu were mostly enriched in fine particles, Pb was enriched in intermediate sized particles, but most mineral elements, S and part of Cu were enriched in coarse particles.

Mineral elements in particles were mainly originated from crustal material, and typical pollutant elements were from anthropogenic pollution emissions. S enrichment was possibly attributed to the sorption of gaseous SO_2 and consequent transformation to sulfate on preexistent particle surfaces, or the mixture of sulfate aerosols with suspended dust particles. Part of Ca was originated from local sources, most likely local construction activities. Se was emitted from coal combustion including local and remote sources. The high EF_{crust} and mass concentrations of Cu were related to the emission of rapidly increasing vehicles, and the replacing of coal for heating fuel with diesel oil. Mineral dust particles were most possibly originated from the Mongolian sand soils and the Chinese loess in the spring of 2004.

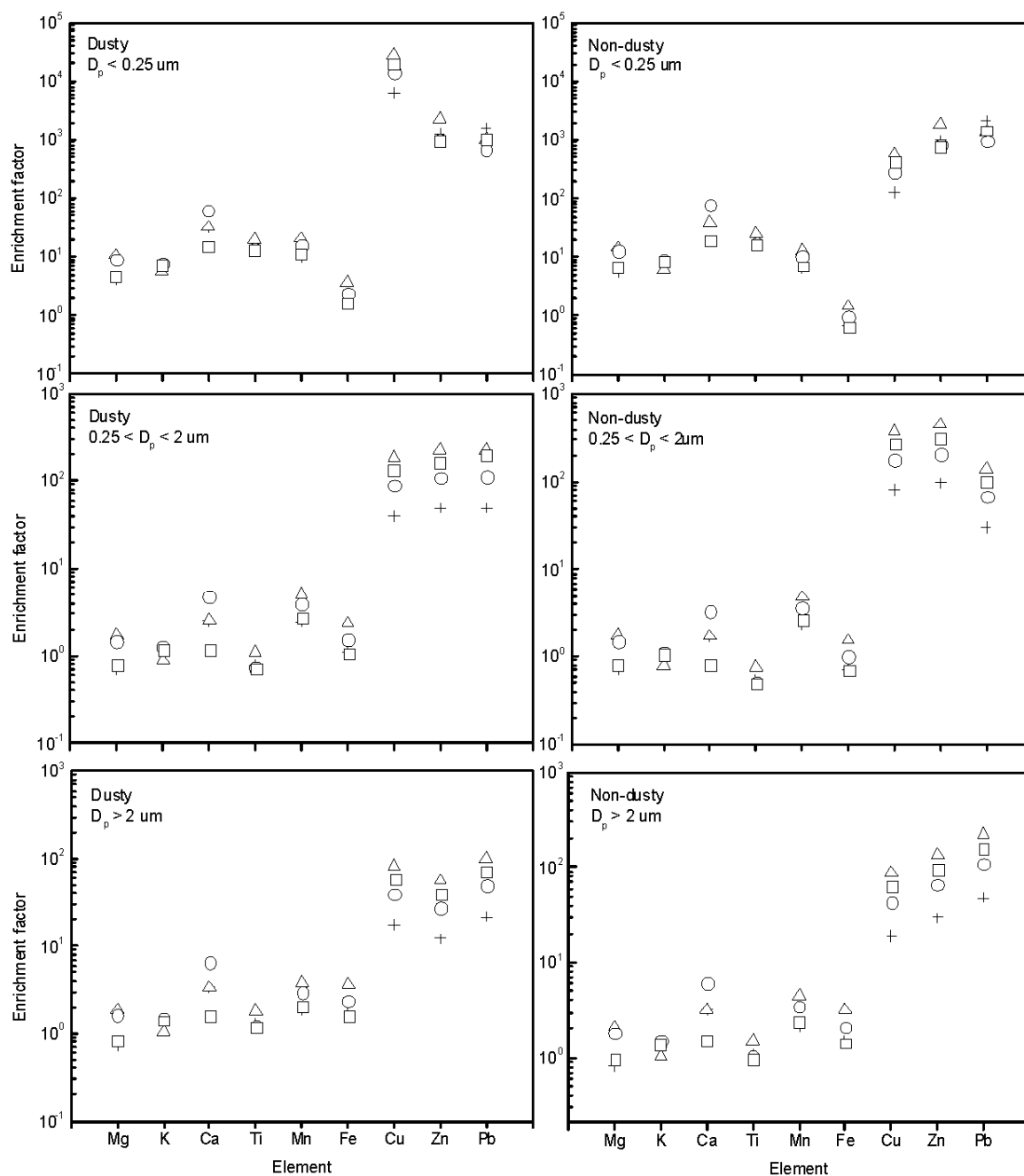


Fig. 5. Enrichment factors of dust and pollutant elements referred to crust (cross), Mongolian sandy soil (triangle), Chinese soil (circle) and Chinese loess (square) in different size particles during dusty and non-dusty days.

Using Mg/Al ratio element tracer technique method, the aerosol from outside Beijing accounted for 66.3% and 88.6% to the total mineral aerosol during dust event on 10-11 March and 28-30 March 2004, respectively.

ACKNOWLEDGMENTS

This research is supported by National Basic Research Program of China (No.2007CB407300), the Pilot Project of Knowledge Innovation Program of the Chinese Academy of Sciences (KZCX2-YW-Q11-03), Financial Project of the Beijing Municipal Financial Bureau

(No.PXM2008_178305_06995), and the Hundred Talents Program (Aerosol Characteristics and its Climatic Impact) of the Chinese Academy of Sciences.

REFERENCES

Arimoto, R., Zhang, X.Y., Huebert, B.J., Kang, C.H., Savoic, D.L., Prospero, J.M., Sage, S.K., Schloesslin, C.A., Khaing, H.M. and Oh, S.N. (2004). Chemical composition of atmospheric aerosols from Zhenbeitai, China, and Gosan, South Korea, during ACE-Asia. *J. Geophys. Res.* 109: D19S04.

- Bergin, M.H., Cass, G.R., Xu, J., Fang, C., Zeng, L.M., Yu, T., Salmon, L.G., Kiang, C.S., Tang, X.Y., Zhang, Y.H. and Chameides, W.L. (2001). Aerosol Radiative, Physical, and Chemical Properties in Beijing during June 1999. *J. Geophys. Res.* 106: 17969-17980.
- Cao, J.J., Lee, S.C., Zhang, X.Y., Chow, J.C. An, Z.S., Ho, K.F., Watson, J.G., Fung, K., Wang, Y.Q. and Shen, Z.X. (2005). Characterization of Airborne Carbonate over a Site on Asian Dust Source Regions during 2002 Spring and Its Climatic and Environmental Significance. *J. Geophys. Res.* 110: D03203.
- Cao, J.J., Shen, Z.X., Chow, J.C. Qi, G.W. and Watson, J.G. (2009). Seasonal Variations and Sources of Mass and Chemical Composition for PM₁₀ Aerosol in Hangzhou, China. *Particuology*. 7: 161-168.
- Chadwick, L.A., Derry, O.A., Vitousek, P.M., Huebert, B.J. and Hedin, L.O. (1999). Changing Sources of Nutrients during Four Million Years of eEcosystem Development. *Nature*. 397: 491-497.
- Chin, M., Ginoux, P., Lucchesi, R., Huebert, B., Weber, R., Anderson, T., Masonis, S., Blomquist, B., Bandy, A. and Thornton, D. (2003). A Global Aerosol Model Forecast for the ACE-Asia Field Experiment. *J. Geophys. Res.* 108: 8654.
- Dentener, F.J., Carmichael, G.R., Zhang, Y., Lelieveld, J. and Crutzen, P.J. (1996). Role of Mineral Aerosol as a Reactive Surface in the Global Troposphere. *J. Geophys. Res.* 101: 22869-22889.
- Duan, F., Liu, X., Yu, T. and Cachier, H. (2004). Identification and Estimate of Biomass Burning Contribution to the Urban Aerosol Organic Carbon Concentrations in Beijing. *Atmos. Environ.* 38: 1275-1282.
- Duce, R.A. (1995). In—*Sources, Distributions, and Fluxes of Mineral Aerosols and Their Relationship to Climate, in Aerosol Forcing of Climate*, Charlson, R.J. and Heitzenberg, J. John (Eds.), Wiley, New York, pp.43-72.
- Guo, J., Rahn, K.A. and Zhuang, G. (2004). A Mechanism for the Increase of Pollution Elements in Dust Storms in Beijing. *Atmos. Environ.* 38: 855-862.
- Han, Z., Ueda, H., Matsuda, K., Zhang, R., Arao, K., Kanai, Y. and Hasome, H. (2004). Model Study on Particle Size Segregation and Deposition during Asian Dust Events in March 2002. *J. Geophys. Res.* 109: D19205.
- Hu, M., Zhou, F., Shao, M., Zhang, Y., Tang, X. and Slanina, J. (2002). Diurnal Variations of Aerosol Chemical Compositions and Related Gaseous Pollutants in Beijing and Guanzhou. *J. Environ. Sci. Health. Part A*, 37: 479-488.
- Husar, R.B., Tratt, D.M., Schichtel, B.A., Falke, S.R., Li, F., Jaffe, D., Gasso, S., Gill, T., Laulainen, N.S., Lu, F., Reheis, M.C., Chun, Y., Westphal, D., Holben, B.N., Gueymard, C., McKendry, I., Kuring, N., Feldman, G.C., McClain, C., Frouin, R.J., Merrill, J., DuBois, D., Vignola, F., Murayama, T., Nickovic, S., Wilson, W.E., Sassen, K., Sugimoto, N. and Malm, W.C. (2001). Asian dust events of April 1998. *J. Geophys. Res.* 106: 18317-18330.
- IPCC (2001). *Climate Change 2001: The Scientific Basis—Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge University Press, New York.
- Iwasaka, Y., Yamato, M., Imasu, R. and Ono, A. (1988). Transport of Asian Dust (Kosa) Particles; Importance of Weak Kosa Events on the Geochemical Cycle of Soil Particles. *Tellus*. 40B: 495-503.
- Li, X.X., Shen, Z.X., Cao, J.J., Liu, S.X., Zhu, C.S. and Zhang, T. (2006). Distribution of Carbonaceous Aerosol during Spring 2005 over the Horqin Sandland in Northeastern China. *China Particuology*. 4: 316-322.
- 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: 1347-1370.
- Martin, J.H. and Fitzwater, S.E. (1988). Iron Deficiency Limits Phytoplankton Growth in the North-East Pacific Subarctic. *Nature*. 331: 341-343.
- Nriagu, J.O. and Pacyna, J.M. (1988). Quantitative Assessment of Worldwide Contamination of Air, Water and Soils by Trace Metals. *Nature*. 333: 134-139.
- Petit, J.R., Mounier, L., Jouzel, J., Korotkevich, Y.S., Kotlyakov, V.I. and Lorius, C. (1990). Palaeoclimatological and Chronological Implications of the Vostok Core Dust Record. *Nature*. 343: 56-58.
- Petit, J.R., Jouzel, J., Raynaud, D., Barkov, N.I., Barnola, J.M., Basile, I., Benders, M., Chappellaz, J., Davis, M., Delayque, G., Delmotte, M., Kotlyakov, V.M., Legrand, M., Lipenkov, V.Y., Lorius, C., Pépin, L., Ritz, C., Saltzman, E. and Stievenard, M. (1999). Climate and Atmospheric History of the Past 420,000 Years from the Vostok Ice Core, Antarctica. *Nature*. 399: 429-436.
- Roth, B. and Okada, K. (1998). On the Modification of Sea-Salt Particles in the Coastal Atmosphere. *Atmos. Environ.* 32: 1555-1569.
- Shen, Z.X., Cao, J.J., Zhang, X.Y., Arimoto, R., Ji, J.F., Balsam, W.L., Wang, Y.Q., Zhang, R.J. and Li, X.X. (2006). Spectroscopy Analysis of Iron-Oxide Minerals in Aerosol Particles and Its Environmental Significance in North China. *Sci. Total Environ.* 36: 899-907.
- Shen, Z.X., Cao, J.J., Arimoto, R., Zhang, R.J., Jie, D.M., Liu, S.X. and Zhu, C.S. (2007). Chemical Composition and Source Characterization of Spring Aerosol over Horqin Sand Land in Northeastern China. *J. Geophys. Res.* 112: D14315.
- Shen, Z.X., Li, X.X., Cao J., Caquineau, S., Wang, Y. and Zhang, X. (2005). Characteristics of Clay Minerals in Asian Dust and Their Environmental Significance. *China Particuology*. 3: 260-264.
- Shen Z.X., Cao, J.J., Tong, Z., Liu, S.X., Reddy, L.S.S., Han, Y.M., Zhang, T. and Zhou, J. (2009). Chemical Characteristics of Submicron Particles in Winter in Xi'an. *Aerosol Air Qual. Res.* 9: 80-93.
- Sokolik, I.N., Toon, O.B. and Bergstrom, R.W. (1998). Modeling the Radiative Characteristics of Airborne Mineral Aerosols at Infrared Wavelengths. *J. Geophys. Res.* 103: D8, 8813-8826.
- Sokolik, I.N., Winker, D.M., Bergametti, G., Gillette, D.A.,

- Carmichael, G., Kaufman, Y.J., Gomes, L., Schuetz, L. and Penner, J.E. (2001). Introduction to Special Section: Outstanding Problems in Quantifying the Radiative Impacts of Mineral Dust. *J. Geophys. Res.* 106: 18015-18027.
- Sun, J.H., Zhao, L.N., Zhao, S.X. and Zhang, R.J. (2006). An Integrated Dust Storm Prediction System Suitable for East Asia and Its Simulation Results. *Global Planet. Change.* 52: 71-87.
- Sun, Y., Zhuang, G., Wang, Y., Han, L., Guo, J., Dan, M., Zhang, W., Wang, Z. and Hao, Z. (2004). The Air-Borne Particulate Pollution in Beijing-Concentration, Composition, Distribution and Sources. *Atmos. Environ.* 38: 5991-6004.
- Takemura, T., Nakajima, T., Nozawa, T. and Aoki, K. (2001). Simulation of Future Aerosol Distribution, Radiative Forcing, and Long-Range Transportation in East Asia. *J. Meteor. Soc. Japan.* 79: 1139-1155.
- Tegen, I., Lacis, A. and Fung, I. (1996). The Influence of Mineral Aerosol from Distribution Soils on the Global Radiation Budget. *Nature.* 380: 419-422.
- Tegen, I., Werner, M., Harrison, S.P. and Kohfeld, K.E. (2004). Relative Importance of Climate and Land Use in Determining Present and Future Global Soil Dust Emission. *Geophys. Res. Lett.* 31: L05105.
- Wang, Y., Zhuang, G.S., Sun, Y.L. and An, Z.S. (2005). Water-Soluble Part of the Aerosol in the Dust Storm Deason—Evidence of the Mixing between Mineral and Pollution Aerosols. *Atmos. Environ.* 39: 7020-7029.
- Wang, Z.F., Hiromasa, U. and Huang, M.Y. (2000). A Deflation Module for Use in Modeling Long-Range Transport of Yellow Sand over East Asia. *J. Geophys. Res.* 105: 26947-26959.
- Wehner, B., Wiedensohler, A., Tuch, T.M., Wu, Z.J., Hu, M., Slanina, J. and Kiang, C.S. (2004). Variability of the Aerosol Number Size Distribution in Beijing, China: New Particle Formation, Dust Storms, and High Continental Background. *Geophys. Res. Lett.* 31: L22108.
- Winchester, J., Wang, M.X., Lu, W.X. and Ren, L.X. (1981). Fine and Coarse Aerosol Composition from a Rural Area in Northern China. *Atmos. Environ.* 15: 933-937.
- Wu, J., Xu, Y.Y., Fu, C.B., Zhang, R.J., Dai, M. and Zhu, Y. (2006). Comparison of Simulating Mineral Dust Aerosols in East Asia by Two Emission Schemes. *China Particology.* 4: 293-299.
- Wurzler, S., Reisin, T.G. and Levin, Z. (2000). Modification of Mineral Dust Particles by Cloud Processing and Subsequent Effects on Drop Size Distributions. *J. Geophys. Res.* 105: 4501-4512.
- Zhang, M.G., Han, Z.W. and Zhu, L.Y. (2007). Simulation of Atmospheric Aerosols in East Asia Using Modeling System RAMS-CMAQ: Model Evaluation. *China Particology.* 5: 321-327.
- Zhang, R.J., Wang, M.X. and Xia, X.G. (2002). Chemical Composition of Aerosols in Winter/Spring in Beijing. *J. Environ. Sci.* 14: 7-11.
- Zhang, R.J., Xu, X.F. and Han, Z.W. (2003). Inorganic Chemical Composition and Source Signature of PM_{2.5} in Beijing during ACE-Asia Period. *Chin. Sci. Bull.* 48: 1002-1005.
- Zhang, R.J., Xu, Y.F., and Han, Z.W. (2004). A Comparison Analysis of Chemical Composition of Aerosols in the Dust and Non-Dust Periods in Beijing. *Adv. Atmos. Sci.* 21: 300-305.
- Zhang, R.J., Arimoto, R., An, J., Yabuki, S. and Sun, J. (2005). Ground Observations of a Strong Dust storm in Beijing in March 2002. *J. Geophys. Res.* 110: D18S06.
- Zhang, R.J., Han, Z.W., Shen, Z.X. and Cao, J.J. (2008a). Continuous Measurement of Number Concentrations and Elemental Composition of Aerosol Particles for a Dust Storm Event in Beijing. *Adv. Atmos. Sci.* 25: 89-95.
- Zhang, R.J., Shen, Z.X., Zou, H., Wang, W., Han, Y.M. and Zhou, J. (2008b). Study of Elemental Mass Size Distributions of Aerosol in Lijiang, a Background Site in Southwest China. *Aerosol Air Qual. Res.* 8: 339-347.
- Zhang, R.J., Han, Z.W., Cheng, T.T. and Tao, J. (2009). Chemical Properties and Origin of Dust Aerosols in Beijing during Springtime. *Particology.* 7: 61-67.
- Zhang, X.Y., Arimoto, R., Cao, J.J., An, Z.S. and Wang, D. (2001). Atmospheric Dust Aerosol over the Tibetan Plateau. *J. Geophys. Res.* 106: 18471-18476.
- Zhou, J.M., Zhang, R.J., Shi, L. and Liu, Y. (2007). Chemical Characterization of Dust in a Floating Dust Event over Beijing. *Journal of Graduate School of the Chinese Academy of Sciences.* 24: 720-723. (In Chinese with English Abstract)
- Zhu, G.H. and Wang, G.F. (1998). Investigation of the Particulate Derived from Indigenous Zinc Smelting Using PIXE Analytical Technique. *Nucl. Instrum. Methods Phys. Res., Sect. B.* 136: 966-969.

Received for review, June 2, 2009

Accepted, September 25, 2009