Continuous Measurement of Number Concentrations and Elemental Composition of Aerosol Particles for a Dust Storm Event in Beijing

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ABSTRACT

A continuous measurement of number size distributions and chemical composition of aerosol particles was conducted in Beijing in a dust storm event during 21-26 March 2001. The number concentration of coarse particles (>2 μ m) increased more significantly than fine particles (<2 μ m) during the dust storm due to dust weather, while the anthropogenic aerosols collected during the non-dust-storm period tended to be associated with fine particles. Elemental compositions were analyzed by using proton-induced X-ray emission (PIXE). The results show that 20 elements in the dust storm were much higher than in the non-dust-storm period. The calculated soil dust concentration during the dust storm was, on average, 251.8 μ g m⁻³, while it was only 52.1 µg m⁻³ on non-dust-storm days. The enrichment factors for Mg, Al, P, K, Ca, Ti, Mn, Fe, Cl, Cu, Pb, and Zn show small variations between the dust storm and the non-dust-storm period, while those for Ca, Ni and Cr in the dust storm were much lower than those in the non-dust-storm period due to significant local emission sources. A high concentration and enrichment factor for S were observed during the dust storm, which implies that the dust particles were contaminated by aerosol particles from anthropogenic emissions during the long-range transport. A statistical analysis shows that the elemental composition of particles collected during the dust storm in Beijing were better correlated with those of desert soil colleted from desert regions in Inner Mongolia. Air mass back-trajectory analysis further confirmed that this dust storm event could be identified as streaks of dust plumes originating from Inner Mongolia.

Key words: PIXE, elemental concentration, number concentration, dust storm

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1. Introduction

Aerosols are particles and/or droplets suspended in air. Aerosols may affect climate and climate change, both directly by back-scattering and absorbing incoming solar radiation, and indirectly by acting as cloud condensation nuclei and modifying cloud radiative properties (Ren et al., 1995; Sokolik and Toon, 1996; Tegen et al., 1996; Zhang et al., 2005, 2006; Huang et al., 2006). An important source of the soil-

derived component of atmospheric aerosols is wind erosion processes in the Gobi and desert regions. Arid and semiarid regions in northern and northwestern China are considered as the major sources for mineral dust by various trace elements such as iron and aluminum in the atmosphere and ocean of the North Pacific region (Duce et al., 1980; Liu and Shiu, 2001; Yuan et al., 2004, 2006). Many studies have shown that Asian dust aerosols can be transported to the North Pacific region (Hashimoto et al., 1984; Li et al., 2006) as far as to

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North America (Wang et al., 2000; Husar et al., 2001). Asian dust storms have been an important topic over the last two decades due to their possible effects on regional climate and environment (Iwasaka et al., 1983; Zhang et al., 2002; Cao et al., 2003, 2005; Zhang et al., 2004, 2007; Han et al., 2004; Shen et al., 2006, 2007). Recently, however, a significant uncertainty involving the estimation of climate effects by mineral dust (IPCC, 2001) has been highlighted. An important reason is that precise properties of aerosols on a global and regional scale are still not available. In recent years, many studies have considered the physical, chemical and optical properties of dust aerosols (Chun et al., 2001; Yabuki et al., 2002; Zhang et al., 2003).

Dust storms have occurred frequently in China from 2000 (Zhang et al., 2003). Beijing, the capital of China, is located in the transport pathway of Asian dust storms, and is easily attacked by heavy dust storms. It is important to investigate the physical and chemical properties of mineral dust and their changes at depositional areas after long-distance transport. During March 2001, a strong dust storm attacked Beijing. Here, we report a continuous observation for this dust storm event in Beijing, with the aim to characterize the physical and chemical properties of aerosol particles between the dust storm and non-duststorm periods in Beijing. Such a dataset is useful for obtaining information on the origin and contribution of high particle levels in urban areas. Furthermore, it may also be important for the evaluation of desert dust impacts on urban air quality.

2. Methods

The sampling site was located on the roof of a two-storey building, about 8 m above ground, and 50 m west of the 325-m meteorological tower (39°58′N, 116°22′E) between the North Third Ring Road and the North Fourth Ring Road of Beijing.

The aerosol samples were collected using the linear streaker sampler KE-101 (Green and Blue Corporation, Japan). The flow rate was 1 L min⁻¹. The filter was Teflon of nuclepore (Green and Blue Corporation, Japan) and its pore diameter was 1 μ m. The size of the filter is 34 mm×256 mm. Each sample in the filter was a circular orifice with a diameter of 4.7 mm. The distance between the centers of the two samples was 5 mm. The time step was set at one hour. The streaker sampler collected fine particles with a diameter of < 10 μ m. The streaker sampler can collect aerosol samples continuously, with usually 40 samples able to be obtained in a filter. A total of 105 samples from three streaker filters were obtained (from 1800 LST 21 March to 2200 LST 22 March, 1000 LST 23

March to 1900 LST 24 March, and 1900 LST 24 March to 1400 LST 26 March).

The filter samples were analyzed by proton induced X-ray emission (PIXE) at the Institute of Low Energy Nuclear Physics, Beijing Normal University. The PIXE analyses were carried out by 2.5 MeV proton bombardments with a beam of 30–40 nA. Elemental concentrations of particles for Mg, Al, Si, P, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, and Pb were determined. The quality control and accuracy tests showed that both the precision (10%) and accuracy (15%) were satisfactory (Zhang et al., 2002).

The BCJ-1 optical particle counter (OPC) (Suzhou, China) was used to measure the variation in size distributions of aerosol particles. This counter measures six channels of aerodynamic diameters as follows: 0.3–0.5, 0.5–0.7, 0.7–1.0, 1.0–2.0, 2.0–5.0, and $> 5.0~\mu m$. The airflow rate was 2.83 L min⁻¹. The OPC instrument was calibrated before observation, detailed descriptions of which can be found in Zhang et al. (2001).

3. Results and discussion

3.1 Variation of number concentrations

According to the standard of the China Meteorological Administration (CMA) (Zhou, 2001), dust events are classified into three categories: fugitive dust, floating dust, and dust storm. These categories are defined based on horizontal visibility. The horizontal visibilities for fugitive dust, floating dust, and dust storm are <10 km, 1–10 km, and < 1 km, respectively. On 21–23 March 2001, a dust storm event attacked Beijing with strong winds.

Figure 1 illustrates the number size distribution of particles in the dust storm on 21–25 March 2001. The number concentrations for coarse particles (diameter $> 2 \mu m$) and fine particles (diameter $< 2 \mu m$) during the dust storm were 4.56 cm^{-3} and 22.9 cm^{-3} respectively, i.e., the number of fine particles was about five times the number of coarse particles. This result shows that fine fraction is the main constituent of aerosol particles after long-distance transport. Compared with the dust storm observation, during the non-dust-storm period the number concentrations for coarse and fine fractions were 0.72 cm⁻³ and 18 cm⁻³ respectively. The fine fraction was nearly 25 times the coarse fraction during the non-dust-storm period. Comparing the number size distribution between the two conditions, it is shown that number concentrations of coarse particles increased more significantly than those of fine particles in the dust storm due to the dust weather. The coarse fraction had a low level, while the fine particles were highly abundant during the non-dust-storm

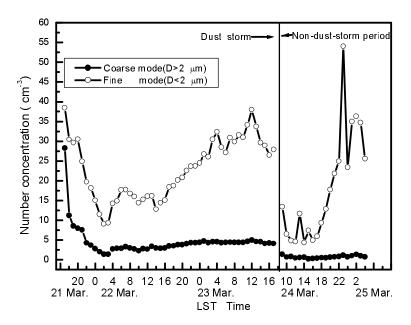


Fig. 1. Number size distributions for aerosol particles on 21–25 March 2001 in Beijing.

period, which indicates pollution aerosols tend to be associated with finer particles. A previous study of the chemical composition for TSP and $PM_{2.5}$ in Beijing in spring (Wang et al., 2005) drew similar conclusions to our study. As we know, these fine particles are more harmful to human health and have a greater influence on visibility during dust storm events.

3.2 Mass concentration of elements and soil dust

The statistical descriptions of 20 elements are summarized in Table 1. Chemical results show that high elemental concentrations were mainly for the six typical dust elements during the dust storm event, which are Al, Si, Mg, Ca, Fe, and K. The mean concentrations for these dust elements were 16.1, 63.2, 5.3, 14.7, 13.4, and 8.87 $\mu g \text{ m}^{-3}$, respectively. High S and Cl were also observed during the dust storm event, which revealed that anthropogenic aerosols mixed with soil dust on the transport pathways and arrived in Beijing. Previous studies have also reported this point (Arimoto et al., 2004; Zhang et al., 2005; Wang et al., 2005; Shen et al., 2006), that mineral dust mixes with pollution aerosols at source and in downwind regions. Compared to the chemical composition during the dust storm, elemental concentrations during nondust-storm days are obviously different. Lower dust element concentrations were monitored in contrast to those of the dust storm period. Dust elemental concentrations of Al, Si, Fe, K, and Mg during calm days were only one fifth or lower than those during the dust storm event. A remarkable difference was observed for

elemental composition between dust storm and calm days. S concentration was higher than the dust elements of Al, Mg, K, and Fe during calm days. Since the element S is associated with anthropogenic sources,

Table 1. Concentrations of elements and soil dust for a dust storm (DS) and non-dust-storm days (NDS) observed in Beijing (units: $\mu g m^{-3}$).

	DS $(n = 61)$		NDS	NDS(n = 44)	
	Mean	SD	Mean	SD	
Mg	5.25	2.20	0.69	0.59	
Al	16.12	6.44	2.93	1.42	
Si	63.17	25.57	11.53	5.58	
Р	0.85	0.50	0.17	0.16	
S	5.92	2.78	3.55	0.69	
Cl	2.24	1.28	0.46	0.27	
K	8.87	4.18	1.52	0.69	
Ca	14.74	7.68	6.13	2.71	
${ m Ti}$	1.41	0.63	0.25	0.13	
V	0.03	0.04	0.00	0.00	
Cr	0.01	0.02	0.00	0.01	
Mn	0.28	0.12	0.05	0.03	
Fe	13.35	5.96	2.69	1.26	
Ni	0.12	0.06	0.04	0.02	
Cu	0.25	0.14	0.05	0.03	
Zn	0.67	0.56	0.10	0.06	
As	0.08	0.11	0.02	0.02	
Se	0.29	0.17	0.02	0.02	
Br	0.23	0.16	0.08	0.04	
Pb	0.36	0.36	0.06	0.08	
Soil dust	251.84	103.83	52.12	24.50	

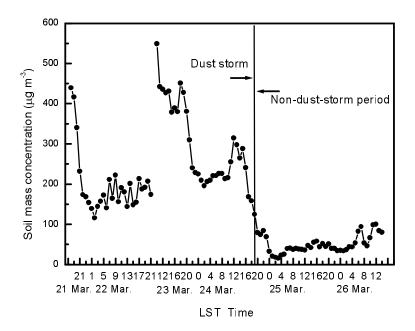


Fig. 2. Temporal variations of soil dust concentrations in Beijing on 21–25 March 2001.

it shows that coal burning is one of the main sources of fine particles in Beijing.

According to Malm et al. (1994), the soil mass concentration of aerosols can be estimated by summing up the elements predominantly associated with soil, plus oxygen for the normal oxides, and the formula for the calculation of soil mass concentrations by elemental concentrations is:

$$[Soil] = 2.2[Al] + 2.49[Si] + 1.63[Ca] +$$

$$2.42[Fe] + 1.94[Ti].$$
(1)

Calculated soil mass concentrations are also listed in Table 1. Figure 2 also shows the temporal variations of soil dust during dust storm and calm days. The soil mass concentrations ranged from 115.7 to 548.9 μg m⁻³ in the dust storm period from 1800 LST 21 March to 1900 LST 24 March 2001, while the soil mass concentrations ranged from 16–100.5 μg m⁻³ in the non-dust-storm period from 2000 LST 24 March to 1400 LST 26 March 2001. The mean soil mass concentrations in the dust and the non-dust-storm periods were 251.8 and 52.1 μg m⁻³ respectively, i.e., in the dust storm it was almost five times than in the non-dust-storm period. Therefore, dust storm events can cause serious air pollution to urban regions.

3.3 Enrichment factors

The enrichment factor $(f_{\rm E})$ of an element in aerosols is an important index to study the origin of aerosols (Zhang et al., 2003). The $f_{\rm E}$ of elements in

aerosols is defined as:

$$f_{\rm E} = (C_x/C_{\rm r})_{\rm a}/(C_x/C_{\rm r})_{\rm c}$$
 (2)

where C_x is the concentration of element x, C_r is that of the reference element, the subscript "a" indicates aerosol, and "c" reference material. Here, Si was selected as the reference element to calculate the enrichment factors of the elements in particles collected in Beijing (Winchester et al., 1981). The results are shown in Fig. 3. $f_{\rm E}$ of Mg, Al, P, K, Ca, Ti, Mn, and Fe were in the range of one to four and they showed little variation between the dust and non-dust-storm periods. Thus, the major source for these elements mainly came from crustal materials. Based on the trajectory analysis, one would expect that deserts in western and northwestern China, and possibly deserts in Mongolia and Inner Mongolia, can be considered as the sources of these crustal elements. More discussion about the source of this dust storm will be made later. $f_{\rm E}$ of Ca in the dust storm were lower than in the non-dust-storm period, possibly suggesting that Ca had local sources, most likely from local construction activities (Wang et al., 2005). $f_{\rm E}$ of S in the non-dust-storm period were much higher than that in the dust storm, implying that S was mainly from local sources, most likely associated with local high SO₂ emissions. It is the same for Ni, Br, and Cr, which were also due to significant local emission sources. $f_{\rm E}$ of Cl, Cu, Zn, and Pb were much higher than those of crustal elements, but showed small differences in the dust and non-dust-storm periods.

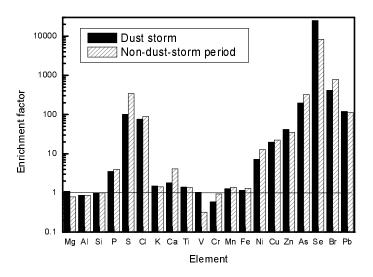


Fig. 3. Enrichment factors for elements of particles in Beijing on 21–25 March 2001.

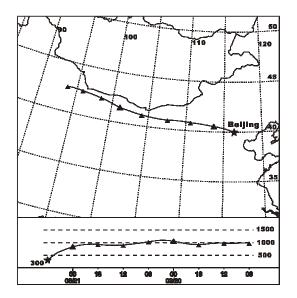


Fig. 4. A 48-hour back-trajectory calculation for 1400 LST 21 March 2001, started from the observation site in Beijing, China. Note that the time is in UTC, not LST.

It is very interesting that $f_{\rm E}$ of Se in the dust storm were a factor of three relative to those in the non-dust-storm period, which implies the dust particles were obviously contaminated by industrial pollution from coal combustion when dust particles were transported to Beijing.

A 48-hour back-trajectory was calculated by using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler, 1996) with the FNL meteorological database starting at 1400 LST 21 March 2001. The trajectory of air masses was analyzed at a height of 300 m over Beijing to indicate

the air mass transported to Beijing. Figure 4 shows that the dust particles in Beijing on 21–23 March 2001 were transported from southern Mongolia, central Inner Mongolia, and passed through the northern part of Shaanxi Province, then turned to Beijing. Shaanxi Province is abundant in coal, and many large coal mines are located in the northern part of the province. Therefore, coal is the widely consumed fuel for cooking, heating and power plants in this region, emitting large quantities of anthropogenic aerosols which result in seriously bad air quality for most cities in the province. So, one can speculate that dust parcels through Shaanxi Province might mix with such anthropogenic aerosols and cause higher concentrations and $f_{\rm E}$ for polluting elements such as S and Se during the dust storm observed in Beijing.

3.4 Correlation between chemical composition of dust particles in Beijing and desert soil in Inner Mongolia

As mentioned above, this dust storm event originated from the desert region of Inner Mongolia and then moved on to Beijing, so it is useful to identify the source regions by comparing the elemental composition of dust particles collected in Beijing with surface soil from the deserts of Inner Mongolia. Concentrations of 13 elements (Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, and Zn) in desert soil sampled in the Mu-US desert (38°23′N, 109°2′E, west of Beijing) in Inner Mongolia were analyzed by PIXE. The correlation coefficients between the chemical composition of particles in Beijing and desert soil in Inner Mongolia were calculated and are presented in Fig. 5. The average correlation coefficients are 0.96 for the dust storm and 0.87 for the non-dust-storm period, which

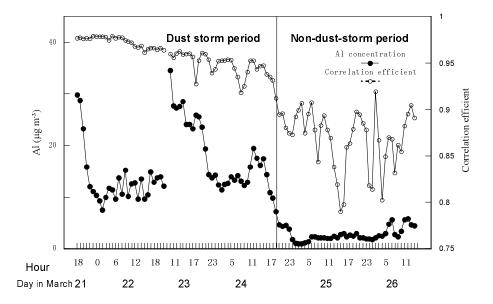


Fig. 5. Correlation between chemical composition of dust aerosols in Beijing and desert soil in Inner Mongolia.

indicates that the chemical composition of particles in the dust storm correlated better with desert soil than in the non-dust-storm period. This result, therefore, possibly points towards the fact that the desert region in Inner Mongolia was the source of this dust storm observed in Beijing.

4. Summary

By continuous measurement of a dust storm event during 21–26 March 2001 in Beijing, the following conclusions can be drawn:

- (1) The number concentration of coarse particles in the dust storm was 6.3 times that of the non-dust-storm period, but only 1.3 times for fine particles, which is due to dust weather and anthropogenic pollution aerosols tending to reside in fine fraction.
- (2) Concentrations for elements and soil dust in the dust storm were much higher than those in the non-dust-storm period. $f_{\rm E}$ of Mg, Al, P, K, Ca, Ti, Mn, and Fe were in the range of one to four and no notable variation was found between the dust storm and non-dust-storm periods. $f_{\rm E}$ of Ca, Ni, and Cr in the dust storm were lower than in the non-dust-storm period due to their local emission sources. The high concentrations and $f_{\rm E}$ of Se in the dust storm reveals that the dust particles were mixed with anthropogenic aerosols during long-range transport.
- (3) Correlation analysis shows that the elemental concentration of particles in the dust storm in Beijing are better correlated to desert soil in Inner Mongolia than in the non-dust-period, which implies desert regions in Inner Mongolia were probably the major

source of this dust storm event.

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