Chemical Modification of Dust Particles during Different Dust Storm Episodes

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ABSTRACT

Two dust episodes (D1: February 13 and D2: March 28–29, 2004) of long-range transport from desert and loess sources, and one dust episode (D3: March 30) mainly from local area, were investigated in spring 2004 in Beijing, China. Dust samples were collected in different dust episodes and scanning electron microscopy with X-ray energy dispersive detector (SEM/EDS) was applied to study the morphology, elemental composition, and chemical modification on the surfaces of dust particles with radius from 0.1 to 6.4 μm. The morphological features of the particles reveal that the three dust plumes carried both dust particles (90–98% by number) and anthropogenic pollutants (e.g., fly ash and soot, 2–10%). Particle compositions show that the amount of S-containing dust particles was higher in D1 and D2 than in D3. Sulfates were introduced into the dust particles through heterogeneous uptake of SO2 during their transport. In all three dust episodes, fine dust particles (radius < 1 μm) showed a higher degree of chemical modification by SO2 compared to the coarse particles (radius ≥ 1 μm). Furthermore, long-range transport dust particles were more deeply modified by SO2 than particles from local areas. The composition of the dust particles indicates that alkaline mineral components determined the level of chemical modification of individual particles. Heterogeneous uptake of SO2 on a large number of dust particles would increase mass of the internally mixed sulfates on their surfaces. The results suggest that the optical properties of dust particles during dust storm episodes are gradually altered based on how far they are transported.

Keywords: Asian dust storm (ADS); Individual dust particle; Heterogeneous reaction; Scanning electron microscopy (SEM); Alkaline mineral.

INTRODUCTION

Dust particles in the atmosphere have been recognized as an important atmospheric aerosol because they not only impose direct and indirect influence on the Earth’s radiation budget (Zhang et al., 2003b; Tegen et al., 2004; Wu et al., 2009; Wang et al., 2010b) but also link the biogeochemical cycles of land, atmosphere and ocean in East Asia and the Pacific Ocean (Buseck and Pósfai, 1999). The wind-blown dust from Gobi desert and loess plateau in northwestern China contribute significantly to the global aerosol loadings during Asian dust storms (ADS) (Sun et al., 2000; Zhang et al., 2003b; Zhang et al., 2005; Shen et al., 2007; Cao et al., 2008). The dust particles crossed continental China (Husar et al., 2001; Arimoto et al., 2004; Shi et al., 2005; Zhang et al., 2005; Gong and Zhang, 2008; Shao et al., 2008; Zhao et al., 2010), passed over Korea and Japan (Ma et al., 2001) and traveled to North Pacific in spring time (Prospero et al., 2003; Arimoto et al., 2006). Some ADS even reached North America and Greenland (Husar et al., 2001).

Numerous studies investigated the modifications of dust particles in coastal areas and over ocean during strong ADS periods and found that dust particles could absorb acidic gases and coalesce with anthropogenic particles (Zhang et al., 2003a; Lu et al., 2006; Li and Shao, 2009a; Huang et al., 2010). Heterogeneous reactions of acidic gases (i.e. NOx and SO2) can change the surface properties of dust particles (Okada et al., 2005; Li and Shao, 2009a; Dall'Osto et al., 2010). The particle surface changes can influence the climate efficiency (i.e., optical scattering and absorption) and biochemical cycle of the dust storm plumes in regional and global scales (Bauer and Koch, 2005).

In these previous studies, the unique strong dust storm episodes have been well documented because the dust particles can be transported in a global scale. In fact most
of spring dust storms in northern China are weak or middle-strong type. They are called as blowing dust and floating dust in China (Song et al., 2007). The weak dust storms usually transport over the continent and affect the downstream areas in East China (Shao et al., 2008; Zhou et al., 2008; Wang et al., 2010a). However, these weak dust storm episodes are not concerned. Whether dust particles in weak dust storm episodes are reacted with acidic gases are poorly understood. However, knowledge is essential to evaluate potential effect on regional climate and biogeochemical cycles over the China continent.

For this purpose, we examined properties of dust particles in weak dust storm episodes observed in one downstream urban site. Morphologies, size distribution and composition of individual dust particles were obtained using scanning electron microscopy with an energy dispersive X-ray spectrometry (SEM/EDS). The contents of Ca and S in dust particles with different radii suggest the possible heterogeneous uptake of SO$_2$ on particle surfaces.

EXPERIMENTS

Sample Collection

Beijing city with a population of more than 22 million inhabitants is located within the downstream areas of transport pathways of most ADS. The sampling site (E116°20'45.6", N39°59'37.1") is located in northwestern urban Beijing, 100 m west from the Xueyuan Road, a major road in Beijing. The samplers were mounted on the top of a building at the campus of China University of Mining and Technology (Beijing) (CUMTB), about 18 m above ground. There were a few construction sites over western direction although we could not look at them at the sampling site. The sampling site represents one urban Beijing resident area. MiniVol samplers (Airmetrics, USA) with a flow rate of 5 L/min were used to collect PM$_{10}$ on polycarbonate filters 47 mm in diameter (Millipore, UK, pore size 0.6 μm). Meteorological data including wind speed, wind direction, relative humidity, barometric value, and ambient temperature, were automatically recorded every 30 minutes by Kestrel 4000 pocket weather tracker (Nielsen Kellerman, USA) during sampling periods.

The Descriptions of Three Dust Storm Episodes

A total of five samples (B1–B5) were collected during three dust storm episodes (D1–D3): B1 and B2 for the D1, B3 and B4 for the D2, and B5 for the D3 in spring 2004 (Table 1). To avoid the overlapping of particles on the filter, each sampling duration was estimated according to the visibility and the possible loading and color of filter after one or two hours sampling.

The D1 occurred on February 13, 2004. Before the dust storm, a haze with the relatively high humidity (60–94%) and low wind (~1 m/s) covered northern China on February 12. The dust storm reached Beijing at 10 a.m. on February 13, 2004. Although the wind speed was low, the color of the sky changed from white to light yellow. Visibility was down to 1 km. The B1 sample was collected during the sky color changing period. The B2 sample was collected from over 4½ hours (1550 until 2000 BST), while the wind speed increased from ~6 m/s and the humidity decreased to ~14%. During this period, the ADS completely occurred in Beijing city.

The D2 occurring in Beijing from March 28 to 29, 2004 was a light and long dust episode (Table 1). The strong wind (3–7 m/s) and low humidity (~10%) were monitored and a yellowish sky was observed during the B3 and B4 sampling periods.

The D3 was a sudden dust episode with low humidity (9%) and strong wind speed (9–12 m/s) on March 30, 2004 (Table 1). The particles were mainly the re-suspended dust particles from the local soil according to our observations in Beijing and reports from Beijing Meteorological Bureau.

Transport Pathways of the Three Dust Storm Episodes

Isentropic backward trajectories of the three dust storm episodes were calculated using the HYSPLIT (Hybrid Sing-Particle Lagrangian Integrated Trajectories) Model (Draxler and Rolph, 2003). Three 24-hour backward trajectories started at the altitude of 50 m above the sampling site (Fig. 1).

The D1 mainly passed over desert and semidesert areas in the east of Inner Mongolia and Mongolia and loess areas in Hebei province. The D2 mainly passed over desert and loess areas in Xinjiang and the west of Inner Mongolia. Air masses of the D1 and D2 trajectories indicate that dust particles had the potential to be transported a long distance to sampling site. Air mass on the D3 trajectory moved over only a short distance during the same period, suggesting that dust particles were from local areas and around Beijing. These results suggested in Fig. 1 are consistent with our meteorological observations at the sampling site. The meteorological scenarios can cause different dust episodes (i.e., regional, local, and global) (Coz et al., 2010).

<table>
<thead>
<tr>
<th>Dust Episode</th>
<th>Sample ID</th>
<th>Sampling Time</th>
<th>Meteorological Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Date</td>
<td>Period, BST$^a$</td>
</tr>
<tr>
<td>D1</td>
<td>B1</td>
<td>February 13, 2004</td>
<td>1000–1530</td>
</tr>
<tr>
<td></td>
<td>B3</td>
<td>March 28, 2004</td>
<td>0815–1115</td>
</tr>
<tr>
<td>D2</td>
<td>B4</td>
<td>March 29, 2004</td>
<td>1115–2015</td>
</tr>
<tr>
<td></td>
<td>B5</td>
<td>March 30, 2004</td>
<td>1400–2030</td>
</tr>
</tbody>
</table>

$^a$BST: Beijing Standard Time (UTC plus 8 hours).

T-temperature, H-relative humidity, WS-wind speed, WD-wind direction.
Fig. 1. Isentropic backward trajectories of three dust episodes. Circles on the trajectories mark the D1 (blue), D2 (black), D3 (red), respectively, and they indicate the 6-hour interval. The latitude-longitude data of isentropic backward trajectories were calculated using the HYSPLIT model (http://www.arl.noaa.gov/ready). The back trajectories at the altitude from 50 m to 2000 m above the sampling site (E116°20', N39°59') show same pathway. Loess, semidesert, and desert regions in northwest China were marked by yellow, deep yellow, and yellowish.

Electron Microscopy

Approximately one eighth of each polycarbonate filter was cut and then mounted onto a copper washer using epoxy resin. Two specimens were prepared for each sample. One specimen was gold-coated to a thickness of 20 nm and another remained uncoated. High-resolution images of the coated specimens were obtained using a Philips XL30 FESEM. Morphologies of about 6,600 particles were totally observed in the five samples. Uncoated specimens were directly put into JEOL JSM-7600F SEM coupled with an Energy Dispersive X-ray Spectrometry (EDS) chamber for the elemental analysis of individual particles. The non-coated sample was used to eliminate any interference in the identification of sulfur from the gold coating. Manual operation was conducted to obtain the chemical compositions of 488 individual dust particles with different sizes. The relative percentages of the elements heavier than C (Z ≥ 6) were estimated based on the X-ray spectra acquired through the INCA software (Oxford Instruments). Elemental weight percents were calculated using standard ZAF (atomic number, absorption and fluorescence) corrections and backscattered imaging obtained particle morphology. Operation conditions were 20 keV accelerating voltage and 600 pA beam current with spectral acquisition times of 80 s. The diameter of spot of electron beam was 1 μm.

Image Analysis

To obtain an estimate of the radius of the individual dust particles analyzed, the electron images from the SEM were further analyzed with the Qwin image processing and analysis system (Leica, UK) (Shi et al., 2003). By drew one circle along with the edge of each analyzed particle, the Qwin software can automatically calculated its equivalent spherical diameter (ESD). In this paper, the size range is divided into two bins of fine and coarse mode particles: 1) fine particle is defined as the submicron particles with the radius from 0.1 to 1 μm; 2) coarse particle is larger than 1 μm in radius.

RESULTS

Morphologies of Aerosol Particles

Mineral particles, fly ash and soot particles with different morphologies were clearly identified in FESEM images (Shi et al., 2005; Shao et al., 2007). Most mineral particles show irregular shapes or not well-defined shapes, while a small amount of gypsum particles that formed in the atmosphere display regular shapes (Fig. 2). Fly ash particles from coal combustion usually show a spherical shape and mainly contain Si and Al (data in the next section), and soot particles from coal combustion and vehicle display a chain-like shape.

For the D1, the B1 and B2 samples contain more than 90% (by number) of dust particles with irregular shapes. Some soot aggregates and regular mineral particles (rectangle shapes) were detected in the B1 sample, whereas a few fine chain-like soot aggregates occurred in the B2 sample. Fly ash (~5%) was observed in the B1 and B2 samples (Fig. 2). For the D2, the B3 and B4 samples contain approximate 98% and 80% dust particles, respectively (Fig. 2(B3) and (B4)). Abundant soot and fly ash particles were observed in the B4 sample. For the D3, the B5 sample exhibited approximate 97% dust particles (Fig. 2(B2)). These fine fly ash and soot particles from anthropogenic sources were possibly brought from the transport pathway of the dust plume (Li and Shao, 2009b).

Types of Individual Dust Particles

A criterion, P(X), is used to represent the weight ratio of element “X”, such as Na, Mg, Al, Si, S, Cl, K, Ca, Ti, and Fe. This criterion P(X) is defined as follows (Okada et al., 2005):

\[
P(X) = \frac{X}{\text{Na} + \text{Mg} + \text{Al} + \text{Si} + \text{S} + \text{Cl} + \text{K} + \text{Ca} + \text{Ti} + \text{Fe}} \times 100\%
\]  

According to the P(X) values, a total of 488 particles with the radius from 0.1 to 6.4 μm were classified into six
“Si-rich” particles account for 76.1% (by number) of the analyzed particles in the five samples (Table 2). They are considered to be quartz, feldspar, and clay minerals (Shao et al., 2007). “Ca-rich” particles account for 12.3% of the analyzed particles. According to their compositions, these particles are possible calcite, dolomite, and their mixtures with silicates.

“S-rich” particles account for 8.3% of the analyzed particles. S in individual mineral particles has strong relationship with Ca. The “S + Ca” sub-type, which was mainly gypsum, was the most abundant sub-type, accounting for up to 7.6%. Other sub-types including the “S + Na” and “S + Si” accounted for 0.4% in all the dust samples.

“Fe-rich”, “K-rich”, and “Na-rich” particles were also detected in the five samples. They should consist of some trace minerals such as hematite and halite (Okada et al., 2005).

**Distribution of Calcium in Individual Dust Particle**

The calcium present in dust particles is an important alkaline element of the atmospheric aerosols. The dust particles containing calcium exhibit extensive reactivity with SO₂ and NO₂ or nitric acid and sulfur acid (Li and Shao, 2009a; Coz et al., 2010). The mixing degree of Ca in the dust particle directly affects chemical reactions with acidic gases or acids in the air (Usher et al., 2003). Previous studies indicated that Ca-containing dust particles show a high number percentage and a high weight proportions not only in the loess soils in China (Liu, 1985) but also in the aerosol samples collected in the loess plateau in China (Okada and Kai, 2004; Okada et al., 2005). The mixing degree of Ca in dust particles of long-range transport over the continent has not been yet clarified.

The dust particles in the D1 and D2 were mainly collected in the dust storm episodes of long-range transport. Fig. 3 shows the relative weight ratio of calcium (P(Ca)) in the individual particles as a function of the radius. P(Ca) in fly ash particles was in the range of 2–7% and their radii are about 0.3 μm. P(Ca) in regular mineral particles was in the range of 27–34%, and their radii varied from 0.5 to 1.2 μm. The results indicate that the fine fly ash in the samples contain the low weight of calcium, whereas the regular particles contain a high level of calcium. Moreover, P(Ca) in irregular dust particles in the B1–B4 samples was in the range of 0–78% and their radii varied from 0.1 to 6.4 μm. For compared to the previous results from Okada and Kai (2004) and Okada et al. (2005), we also calculated P(Ca) and P(S) in individual dust particles. In this study, dust particles with P(Ca) greater than 10% and 2% in the whole particle size in the B1–B4 samples were present in 39% and 91%, respectively. The previous results show that dust particles with P(Ca) greater than 10% and 2% were present in only 12% and 19% in the Zhangye and Qira regions near the southern border of the Badain-Jaran Desert and in 25% and 49% at Hohhot, respectively (Okada et al., 2005). Therefore, we can assume that P(Ca) from the area downstream of ADS is higher than that in dust samples collected around desert sources. A possible reason might be that the dust storms of
Table 2. Number of different types of dust particles in five samples.

<table>
<thead>
<tr>
<th>Types</th>
<th>Major elements</th>
<th>B1 Sample</th>
<th>B2 Sample</th>
<th>B3 Sample</th>
<th>B4 Sample</th>
<th>B5 Sample</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si-rich</td>
<td>Si, Al, Mg, Ca, S, Fe</td>
<td>46[5]a</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ca-rich</td>
<td>Ca, Si, Mg, S</td>
<td></td>
<td>9</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>S-rich</td>
<td>S, Ca, Na</td>
<td></td>
<td>37[2]b</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe-rich</td>
<td>Fe, Si, S</td>
<td></td>
<td>4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>K-rich</td>
<td>K, Si, Al</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na-rich</td>
<td>Na, S, Mg</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a The number of spherical particles is indicated in square bracket.

b The number of secondary mineral particles is indicated in square bracket.

Fig. 3. Relative weight ratio of calcium (P(Ca)) in individual particles as a function of radius. N represents the total number of particles (488) detected in dust samples. The spherical particles (fly ashes) and regular particles (regular mineral particles) are marked by closed circles and squares, respectively. The B1–B4 samples collected during the D1 and D2 periods represent dust particles of long range transport, B5 collected during the D3 period represent dust particles from Beijing area or some areas nearby Beijing area.

long-range transport lifted ground loess particles containing abundant calcium in northern China, these loess particles then mingled with the dust particles from the deserts, prior to arriving at Beijing (Fig. 1). The dust particles in the B5 sample with P(Ca) greater than 10% and 2% were present in 51% and 100%, respectively.

**Distribution of Sulfur in Individual Dust Particles**

In order to evaluate the chemical interactions between dust particles and the anthropogenic pollutants, we focus on the sulfur contents in individual particle during three dust storm episodes. Fig. 4 shows the relative weight ratios of sulfur (P(S)) in individual particles as a function of their radius. The P(S) of fly ash particles with radii of around ~0.3 μm was in the range of 3–11%. The P(S) of regular mineral particles was in the range of 36–42%, and their radii were from 0.5 to 1.2 μm. The results display that fly ash particles from the coal combustion contain little sulfur, and the regular minerals are gypsum (Shi et al., 2003). The dust particles with P(S) greater than 10% and 2% during the D1–D3 are present in 36% and 62% (D1), 9% and 58% (D2), and 7% and 49% (D3), respectively.

The distribution of Al, S, and Ca can suggest the mixing degree of sulfur in individual dust particles (Okada and Kai, 2004). For a better understanding of the mixing degree of sulfur in individual dust particles, the distribution of the relative weight ratios of Al, S, and Ca is shown in Fig. 5. The percentages of S-containing dust particles collected during the three dust storm episodes are between 56% and 64%. In the D1, 61% of fine particles are distributed in the area defined by the Al-S line and the Al-CaSO₄ line, thus indicating high relative weight ratios of sulfur (Fig. 5(a)). 23% of coarse particles are distributed in the area defined by the Al-CaSO₄ line and the Al-Ca line (Fig. 5(b)). In the D2 dust samples, S/Ca ratios of 8% fine particles are higher than that of CaSO₄ (0.8), and the entirety of coarse particles are distributed in the area defined by the Al-CaSO₄ line and the Al-Ca line (Figs. 5(c) and (d)). In the D3 dust sample, 3% of fine particles and 4% of coarse particles have a higher S/Ca ratio than that of CaSO₄ (0.8) (Figs. 5(e) and (f)).
**DISCUSSION**

Morphologies of the analyzed particles indicate that dust samples collected in the three dust storm episodes include small amounts of soot and fly ash particles and abundant mineral particles (Fig. 2). The result indicates that these weak and regional dust storms carried anthropogenic pollutants during their transports. Because only small amounts of soot and fly ash particles were found in the dust episodes, they could not affect the surfaces of large number of dust particles. The backward trajectories and their meteorological observations show that the weak D1 and D2 transported a...
Fig. 5. Relative weight ratios of Al, S and Ca for the mineral particles collected in D1, D2 and D3. N represents the number of particles detected by EDS in dust samples. The sum of the relative weight of Al, S, and Ca is equal to 1 (Al represents the dust particles. The distribution of S and Ca can affect chemical modification of individual dust particle.). (a) and (b) represent D1 dust particles of long range transport. (c) and (d) represent D2 dust particles of long range transport. (e) and (f) represent D3 dust particles of non-long range transport. (a) 139 particles with the radius of 0.1–1 μm, (b) 62 particles with the radius > 1 μm, (c) 171 particles with the radius of 0.1–1 μm, (d) 30 particles with the radius > 1 μm, (e) 61 particles with the radius of 0.1–1 μm, (d) 25 particles with the radius > 1 μm. Fly ash and secondary particles are marked by closed circles and squares.
long distance and that the D3 transported only a short distance (Fig. 1). Based on compositions of dust particles in different dust episodes, S-rich particles only occurred in the D1 (Table 2). However, sulfate particles or their aggregations on the surface of individual dust particles were not clearly observed through FESEM images. The only explanation might be that dust particles reacted with SO$_2$ or sulfuric acid on their surfaces and the secondary sulfates didn’t enough change the features of dust particles. The similar conclusions were found by numerous studies, which indicate that heterogeneous reactions on mineral dust can convert gas SO$_2$ into sulfates on dust particles in the atmosphere (Zhang et al., 2000; Okada and Kai, 2004; Zhang et al., 2005).

Number percentages of the fine S-containing dust particles are larger than these of the coarse S-containing dust particles in the D1–D3 (Fig. 5). The results can be explained as follows: (1) Larger size particles modified by acidic components were more readily deposited on the ground (Bauer and Koch, 2005). In contrast, the fine dust particles move long distance over Asian continent during dust storm periods. (2) The study found that the percentage of dust particles with P(Ca) greater than 10% are larger than those of the coarse dust particles during the Asian dust episodes (Fig. 3). The fine dust particles can offer a large surface of heterogeneous chemical reactions for the acidic materials in the atmosphere during long range transport over the continent. Therefore, number percentages of dust particles with S decreasing from the D1 to the D3 suggested that dust storms of long-range transport enhanced heterogeneous reactions of dust particles with SO$_2$ in the atmosphere.

Minor S peak is clearly shown in the EDS spectra of most dust particles (Fig. 6). These mineral particles with sulfur often include abundant calcium. Laboratory experiments suggest that atmospheric chemistry of individual dust particles strongly depend on alkaline mineral components (e.g., calcite, dolomite, and halite) (Usher et al., 2003; Zhao et al., 2011). As a result showing in Figs. 3 and 4, we can conclude that alkaline mineral components within dust particles mainly determine their heterogeneous reactions.

Light chemical modification shown in Fig. 6 indicates that these dust particles still have great potential to further absorb the acidic materials in the downstream air. Dust particles heterogeneous uptake of SO$_2$ to their surfaces can change SO$_2$ distribution in the atmosphere (Zhang et al., 2000; Usher et al., 2003; Li and Shao, 2009a; Coz et al., 2010). The anthropogenic sulfate forcing is estimated to decrease because of the reduced load of externally mixed sulfate aerosols (Bauer and Koch, 2005).

CONCLUSIONS

Five dust samples in three weak dust storm episodes were collected in Beijing in February and March, 2004. Air mass backward trajectories indicate that the D1 and D2 were transported for a long distance, and that the D3 trajectory moved only for a short distance. FESEM analyses show that all these three weak dust storms carried anthropogenic pollutants (e.g., soot and fly ash) to downstream areas during their transports.

Based on compositions of individual dust particles, they were classified into six classes. In five samples, S-rich particles only occurred in the D1. Difference of number percentage in dust particles containing S between the D1-2 and the D3 indicate that calcium sulfate in dust particles were possibly formed through heterogeneous reactions with SO$_2$ during their transports. Fine dust particles in the ADS tend to react with SO$_2$ in the atmosphere because they can transport long distance and supply large reacted surfaces. The long-transport dust particles can enhance heterogeneous reactions with SO$_2$ in the atmosphere. In addition, alkaline mineral components within dust particles mainly determine their heterogeneous reactions. Heterogeneous uptake of SO$_2$ on surfaces of the dust particles transported in regional scale could change their optical properties in the dust episodes.

![Fig. 6. Backscatter electron image and the EDS spectra of individual dust particles in the B4 sample. The number represents the area of EDS analysis.](image-url)
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REFERENCES


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