Microscopy and mineralogy of airborne particles collected during severe dust storm episodes in Beijing, China

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[1] Asia dust storm (ADS) samples were collected during two severe dust storm episodes in Beijing, China. High-resolution field emission scanning electron microscopy (FESEM) and environmental scanning electron microscopy-energy dispersive X-ray detector (ESEM-EDX) coupled with an image analysis system were used to study the morphology, chemical compositions, and number-size distributions of ADS particles. FESEM analysis indicated that Beijing ADS samples were mainly composed of irregularly shaped mineral particles. More than 50% of the ADS particles were larger than 1 μm, measured as equivalent spherical diameter (ESD). Image analysis showed that when the dust storm episodes ended in Beijing, the number of anthropogenic particles, such as soot aggregates and coal fly ash, in the samples increased, and the number of mineral particles decreased. Analysis of individual ADS particles showed that about one fifth of all the particles were mineral aggregates, and at least one fourth of the particles contained sulfur. X-ray diffraction (XRD) was used to quantify the phase and the clay mineral compositions of ADS samples. Phases in the ADS sample collected during the 20 March 2002 dust storm episode included clay minerals, noncrystalline materials, quartz, calcite, plagioclase, potassium feldspar, pyrite, hornblende, and gypsum in descending order. Clay minerals were mainly illite/smectite mixed layers (78%), followed by illite (9%), kaolinite (6%), and chlorite (7%). Particulate matter (PM) less than 10 μm were enriched with clay minerals and deficient with quartz by mass compared with the total suspended particulates collected during an ADS episode. The PM less than 10 μm collected during the two severe dust storm episodes was characterized by the absence of dolomite, high quartz/clay ratio, and dominance of illite/smectite mixed layers in clay minerals.

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1. Introduction

[2] It is estimated that nearly 50% of troposphere atmospheric aerosol particles are minerals, mainly sourced from the deserts and their boundaries [*Andreae*, 1995]. Mineral aerosols affect climate directly through scattering, transmission and absorption of solar radiation, and indirectly by acting as cloud nucleation nucleus (CNN) when coated with soluble material [*Andreae*, 1995; *Levin et al.*, 1996; *Buseck and Pósfai*, 1999; *Clarke et al.*, 2001; *Sokolik et al.*, 2001]. Mineral aerosols also play an important role in removal, deposition and transport of atmospheric pollutants [*Winchester and Wang*, 1989; *Sievering et al.*, 1989; *Dentener et al.*, 1996; *Carmichael et al.*, 1996]. Mineral aerosols, especially those carried by dust storms, can be transported globally, and have significant impacts on the global environments and climate [*Buseck and Pósfai*, 1999; *Prospero*, 1999; *Clarke et al.*, 2001; *Bishop et al.*, 2002]. In addition, the identification of mineral assemblages of aerosols may be diagnostic of their sources [*Bergametti et al.*, 1989; *Merrill et al.*, 1994; *Davis and Guo*, 2000; *Ganor et al.*, 2000]. Mineral aerosol particles are highly heterogeneous, but are commonly treated as a relatively homogeneous group, and this may lead to increasing uncertainty in atmospheric chemistry and climate models [*Buseck and Pósfai*, 1999]. Therefore improving our knowledge about the mineralogy of airborne particles is an important task for atmospheric scientists.

[3] X-ray diffraction (XRD) has been used to study the mineralogy of dust storm particles [*Bergametti et al.*, 1989; *Leinen et al.*, 1994; *Merrill et al.*, 1994; *Avila et al.*, 1997; *Arnold et al.*, 1998], and urban airborne particles [*Davis and Guo*, 2000; *Shi et al.*, 2002]. However, the chemical analyses of aerosols particles are far more common than mineralogical

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analyses because they are easier to undertake on very small amounts of aerosol samples [*Leinen et al.*, 1994]. Electron microscopy coupled with energy dispersive X-ray detector (EM-EDX) were also employed to identify the mineral particles in the atmosphere [e.g., *Gaudichet et al.*, 1989; *Fan et al.*, 1996; *Mori et al.*, 1998; *Moreno et al.*, 2003]. The combination of XRD on bulk samples and EM-EDX on individual particles provides powerful means to identify the sources of aerosol particles, and to study the chemical reactions on particle surfaces [e.g., *Falkovich et al.*, 2001].

[4] Asia dust storm (ADS) can transport mineral grains thousands of kilometers to Japan, Korea, North Pacific Ocean, Hawaii and even North America [*Duce et al.*, 1983; *Uematsu et al.*, 1983; *Iwasaka et al.*, 1988; *Leinen et al.*, 1994; *Merrill et al.*, 1994; *Arnold et al.*, 1998; *Clarke et al.*, 2001; *Zhuang et al.*, 2001; *Mori et al.*, 2003; *Moore et al.*, 2003], thus having significant impacts on the global environment, climate and geochemical cycle. Intensive investigations have been conducted to study the chemical composition, sources and deposition of ADS particles [*Duce et al.*, 1983; *Uematsu et al.*, 1983; *Iwasaka et al.*, 1988; *Zhang et al.*, 1993, 2000; *Wang et al.*, 2001; *Zhuang et al.*, 2001; *Kanayama et al.*, 2002; *Mori et al.*, 2003]. Some studies have focused on individual particle characteristics [*Fan et al.*, 1996; *Zhou et al.*, 1996; *Zhang and Iwasaka*, 1999; *Ro et al.*, 2001; *Ma et al.*, 2001; *Zhang et al.*, 2003] and size distributions of ADS particles [*Chun et al.*, 2001; *Mori et al.*, 2003]. However, limited data are available on the size distributions and microscopic characteristics of ADS particles in China. In addition, the mineralogy of Africa dust has been studied by several researchers [e.g., *Bergametti et al.*, 1989; *Avila et al.*, 1997; *Ganor et al.*, 2000; *Falkovich et al.*, 2001]. The mineral compositions of aerosol particles, which possibly originated from Asia, over the North Pacific Ocean, have been analyzed by *Leinen et al.* [1994], *Merrill et al.* [1994], and *Arnold et al.* [1998]. However, data on mineral compositions of ADS particles in China has been rarely reported. This information is crucial in predicting mineral dust impacts on climate and atmospheric chemistry [*Sokolik et al.*, 2001; *Miller et al.*, 2004].

[5] Airborne particulate matter (PM) samples were collected during two severe dust storm episodes on 6 April 2000 and 20 March 2002, and a less severe dust storm episode on 16 March 2002 in Beijing. A Tapered Element Oscillating Microbalance (TEOM) 1400a with a PM10 (particulate matter with an aerodynamic diameter less than 10 microns) inlet recorded an instantaneous peak PM10 mass of 5100 $\mu g\ m^{-3}$ during the 20 March 2002 severe dust storm. The morphologies, chemical compositions, size distributions and mineralogical compositions of the ADS particles were investigated using high-resolution field emission scanning electron microscopy (FESEM), environmental scanning electron microscopy (ESEM) coupled with energy dispersive X-ray (EDX) detector, an image analysis system, and the XRD.

2. Materials and Measurements

2.1. Sample Collection

[6] The collection site (N39°59′37.1″, E116°20′45.6″) was located in northwestern Beijing around 1 km from the northern fourth ring road of the Beijing City. The samplers

Table 1. Sample Information

Sample	Collection Date	Collection Time, LT	Type	Sampler/Filter	Mass Concentration, $\mu g\ m^{-3}$	Average Temperature, °C	Average Humidity, %	Startup Wind Speed, mph	Final Wind Speed, mph	Startup Wind Direction[a]	Final Wind Direction[a]
ADS-T1[b]	6 April 2000	1000–2200	severe dust storm	HT-1000/glass fiber							
ADS98	16 March 2000	1000–2200	dust storm	Negretti/polycarbonate	864	15	18	>10	>10	W	W
ADS99	16–17 March 2000	2200–0800	post dust storm	Negretti/polycarbonate	81	12	19	10	breeze	W	
ADS100	17 March 2000	1500–2030	post dust storm	Negretti/polycarbonate	94	16	19	>10	>10	NW	
ADS1[c]	20 March 2000	1000–1420	severe dust storm	Negretti/polycarbonate	2600	10	32	8	8	W	W
ADS1a[c]	20 March 2000	1000–1420	severe dust storm	HT-1000/glass fiber		10	32	8	8	W	W
ADS2[d]	20 March 2000	1420–1550	post dust storm	Negretti/polycarbonate	797	12	24	8	>10	W	W
ADS4	20–21 March 2000	2200–0810	post dust storm	Negretti/polycarbonate	171	8	20	8	6	W	W

[a]Note that wind the direction "W" means from west or "eastward".
[b]ADS-T1 was TSP and all other samples are PM10s.
[c]The ground is wet because of the rain the night before. During the dust storm, the sky is yellow and the visibility is less than 200 m. The air was filled with dust taste.
[d]The visibility increased gradually.

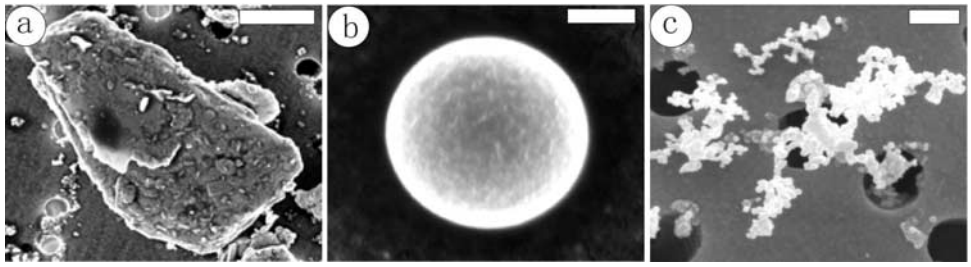

Figure 1. FESEM of typical particle types. (a) Mineral dust, represented by its irregular shape; (b) coal fly ash, characterized by spherical shape; (c) soot aggregates, developing from small groups or chains into larger chains.

were mounted on the fifth floor of the main building on campus of China University of Mining and Technology, 100 m west from the Xueyuan Road; a major road in Beijing. A Negretti selective inlet head (UK) was used to collect PM10 samples on polycarbonate filters (Millipore, UK) with 0.67 μm pore sizes. A XH-1000 high-volume sampler (Hebai Sailhero Inc. Ltd.) was also used to collect the Total Suspended Particulates (TSP) and PM10 samples using glass fiber filters. PM samples were collected during two severe dust storm episodes on 6 April 2000 (TSP) and 20 March 2002 (PM10) and a less severe dust storm episode on 16 March 2002 (PM10). Sample information was listed in Table 1.

2.2. Electron Microscopy

[7] A 4 mm (internal diameter) copper washer was glued to a standard 12.5 mm aluminum SEM stub using epoxy resin (Araldite). Approximately one eighth of each polycarbonate filter was cut and then mounted onto the copper washer using epoxy resin. Two specimens were prepared for each sample. For each sample, one specimen was gold coated to a thickness of 20 nm and another was uncoated. The high-resolution morphologies of the coated specimens were obtained using a Philips XL30 FESEM at Cardiff University (UK). Uncoated specimens were directly put into a low vacuum ESEM chamber for the elemental analysis of individual particles. This excluded interference from the coating elements during the analyses.

2.3. Image Analysis

[8] Electronic images saved as high-resolution TIF files from the FESEM were analyzed with an Image Analysis System (Leica), with a dedicated software programmed for airborne particles. The particles were identified according to their morphologies. By targeting each of the particle groups the custom-written program automatically calculated the equivalent spherical diameter (ESD) of each particle based on its projected area on filter. For each sample, ten images were obtained for images analysis. More than 1000 particles for each sample were targeted for number-size distribution and number percentage analyses. The data were then downloaded into Microsoft Excel for further processing. The high-resolution images allowed us to recognize the submicron particles.

2.4. XRD Analysis

[9] A D/MAX-250 (Rigaku, Japan) X-ray diffractometer scanning with Cu Kα radiation at 40 kV, 125 mA was used for mineralogical analysis. The scanning speed was $-3°$ (2θ) min^{-1} and the sampling distance was 0.01° (2θ). Because the mass of PM collected during the two severe dust storm episodes was more than 3 g, it was possible to scrape the PM samples from filters directly. The specimen was put in a small cuboid trough made by aluminum substrate for XRD analysis. The XRD scanned on the specimen only. This made phase composition quantification possible. For the 6 April 2000 severe dust storm, PM less than 10 μm was also separated from the TSP sample by sedimentation in water based on Stokes' settling law. Thus only water insoluble minerals were examined for the PM less than 10 μm during the 6 April 2000 severe dust storm episode. Mineral types were identified by means of JCPDS cards based on the d value and diffraction intensity. Weight percentages of different phases in PM samples were quantified based on the Petroleum Industry Standard Methods of China (FY-T5263-1995). For samples collected when no dust storm was occurring (non-ADS) on the polycarbonate and glass fiber filters, about one fourth of the filters were cut and adhered to aluminum substrate for XRD analysis directly. To analyze clay mineral compositions, the PM less than 2 μm was also separated by sedimentation in water. To identify and quantify clay mineral species, firstly, the original <2 μm samples were scanned with XRD; then the samples were saturated with glycol and analyzed with XRD again; finally, the samples were heated at 450°C for 2 hrs and analyzed with XRD again. Quantification of clay mineral species from XRD patterns was also based on Petroleum Industry Standard Methods of China (FY-T5263-1995).

3. Results

3.1. Microscopic Morphology and Number-Size Distribution of ADS Particles

3.1.1. Microscopic Morphology

[10] Three major categories of particles have been differentiated under the FESEM: mineral particles (Figure 1a), coal fly ash (Figure 1b), and soot aggregates (Figure 1c). Soot aggregates were characterized by chain-like and "fluffy" appearance, coal fly ash by a smooth spherical shape and mineral particles by irregular shape.

[11] The PM10 (ADS98, Table 1) collected during the 16 March 2002 dust storm episode was dominated by irregularly shaped mineral grains with trace amounts of soot aggregates adhering onto the surfaces of these minerals (Figure 2a). The PM10 (ADS99, Table 1) collected after the

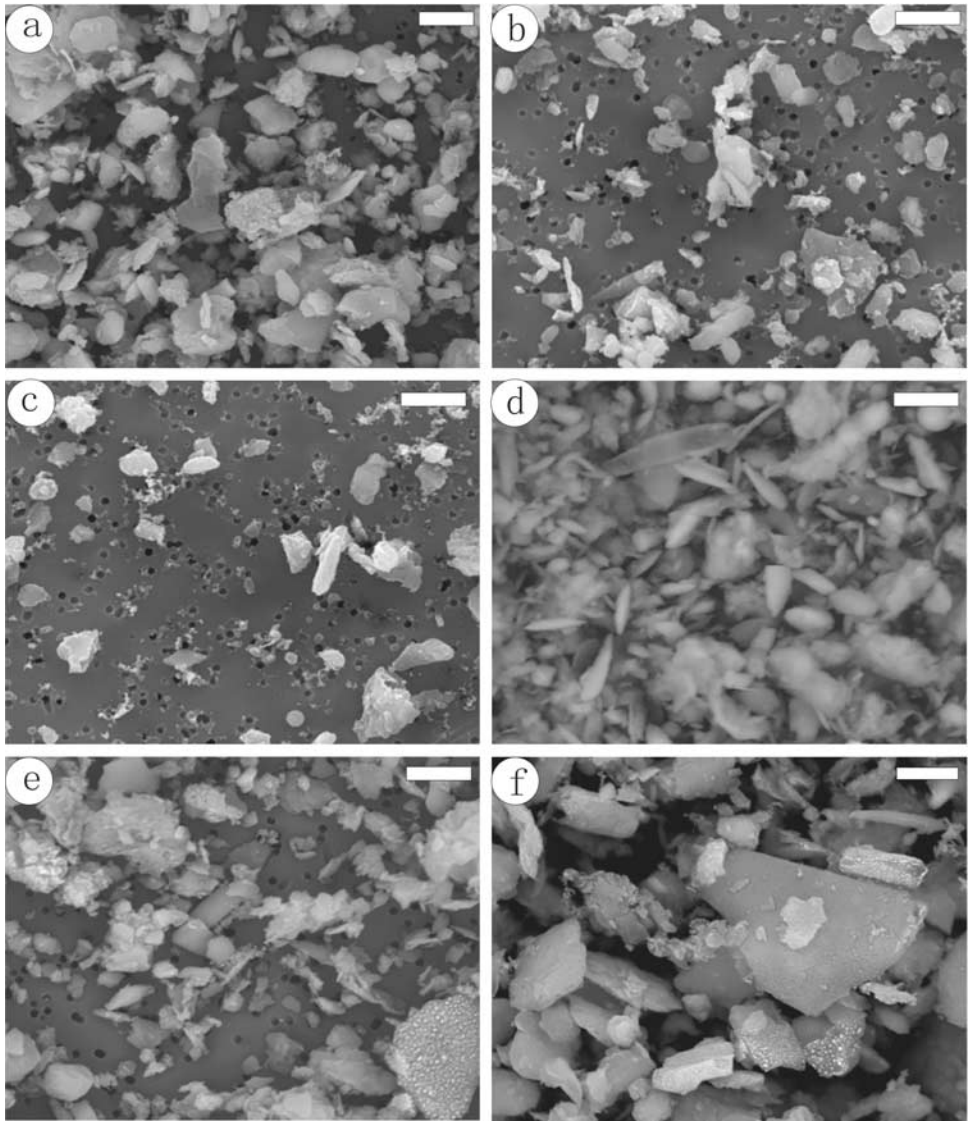

Figure 2. FESEM of PM10 samples collected during and after the dust storm episodes (scale bar 5 μm). (a) ADS98 during the 16 March 2002 dust storm episode, dominantly mineral particles with trace amounts of soot aggregates; (b) ADS99 collected after the 16 March 2002 dust storm episode, mainly mineral particles with small amounts of soot aggregates and trace amounts of coal fly ash; (c) ADS100 collected after the 16 March 2002 dust storm episode, mainly soot aggregates and coarse mineral particles with trace amounts of coal fly ash; (d) ADS1 collected during the 20 March 2002 severe dust storm episode, predominantly mineral particles in which about 30% are bar shaped; (e) ADS2 collected after the 20 March 2002 severe dust storm episode, mainly mineral particles; (f) ADS4 collected after the 20 March 2002 severe dust storm episode, mainly mineral particles with a few soot particles.

dust storm was also mainly composed of minerals with small amounts of fine soot aggregates and trace amounts of coal fly ash (Figure 2b). However, in the PM10 (ADS100, Table 1) collected the following day, many more soot aggregates and ultrafine particles (<100 nm) were observed (Figure 2c). This indicated an increasing influence from anthropogenic emissions.

[12] The sample designated ADS1 (see Table 1) collected during the 20 March 2002 severe dust storm episode was almost completely composed of minerals (Figure 2d). A distinct characteristic of this sample was that many bar-shaped particles were observed, which were not commonly seen in the 16 March 2002 dust storm sample (Figure 2a). ADS2 was also dominated by minerals (Figure 2e). Sample ADS4 collected on 20–21 March 2002 was still dominated by mineral particles (Figure 2f) due to the strong wind during our sampling period (Table 1), which inhibited anthropogenic particle accumulation.

3.1.2. Number Percentages and Number-Size Distributions of Soot Aggregates, Coal Fly Ash, and Minerals

[13] Number percentages and number-size distributions of different types of particles in the PM samples were obtained based on image analysis results. Mineral particles

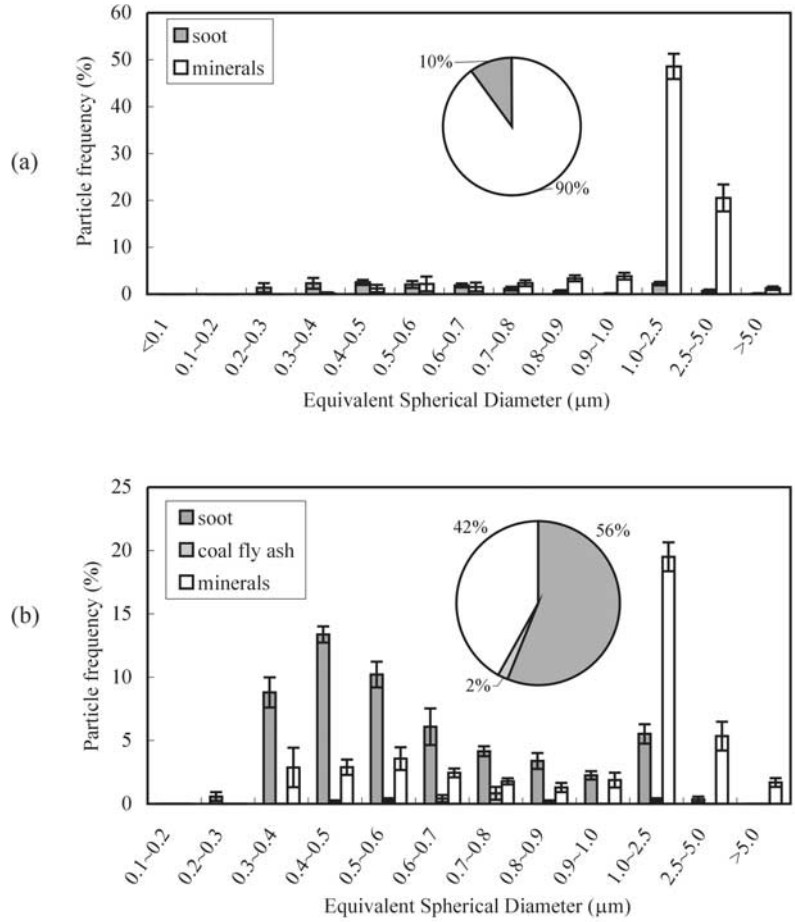

Figure 3. Number-size distribution (bar graph) and percentage (pie chart) of PM10 collected during and after the 16 March 2002 dust storm episode. (a) PM10 collected during the dust storm episode (sample ADS98); (b) PM10 collected after the dust storm episode (sample ADS100). The legends for the bar graph and pie chart are the same.

occupied 90% by number (the pie chart in Figure 3a) of the total PM10 collected during the 16 March 2002 dust storm episode. Soot aggregates accounted for only 10% by number (the pie chart in Figure 3a). No coal fly ash was observed. In the PM10 (ADS100) collected one day after the 16 March 2002 dust storm episode, the percentages of soot aggregates and coal fly ash increased to 56% and 2% by number (the pie chart in Figure 3b), respectively, while the percentage of the mineral particles decreased to 42% by number (the pie chart in Figure 3b). Thus contributions from anthropogenic emissions after the peak dust storm episode increased significantly.

[14] The number-size distributions of PM10s (ADS98 and ADS100) collected during and after the 16 March 2002 dust storm episode are shown in the bar graphs in Figures 3a and 3b. The particle morphologies of these two samples are shown in Figures 2a and 2c. In ADS98, particles larger than 1 μm account for 73% by number (Figure 3a). This is very different from the number-size distribution patterns of PM10s collected during nondust storm episodes, with the latter being dominated by soot aggregates smaller than 1 μm [*Shi et al.*, 2003]. In ADS100, however, particles larger than 1 μm only accounted for 33% by number (Figure 3b). Soot aggregates of ADS100 had an obvious peak in the 0.4∼0.5 μm size range (Figure 3b), indicating an increasing influence from anthropogenic emissions.

3.2. Mineralogy of ADS Particles

3.2.1. Mineral and Chemical Compositions of Individual ADS Particles

[15] Using ESEM-EDX, we have identified individual plagioclase, clay mineral, quartz and calcite particles in the 20 March 2002 dust storm sample (ADS1). A typical ESEM-EDX chemical spectrum of the bar-shaped mineral particles is shown in Figure 4a; these particles were clay minerals and their morphology is shown in Figure 2d. However, some individual particles were mineral aggregates composed of two or more minerals, for example, aggregates of aluminosilicate and gypsum, aluminosilicate and calcite, aluminosilicate and Fe oxides, aluminosilicate and Ti oxides and quartz and calcite, rather than pure minerals. This is in agreement with observations showing that wind-blown dust particles sourced from highly arid regions tend to be aggregated [*Buseck and Pósfai*, 1999; *Prospero*, 1999; *Falkovich et al.*, 2001; *Gao and Anderson*, 2001].

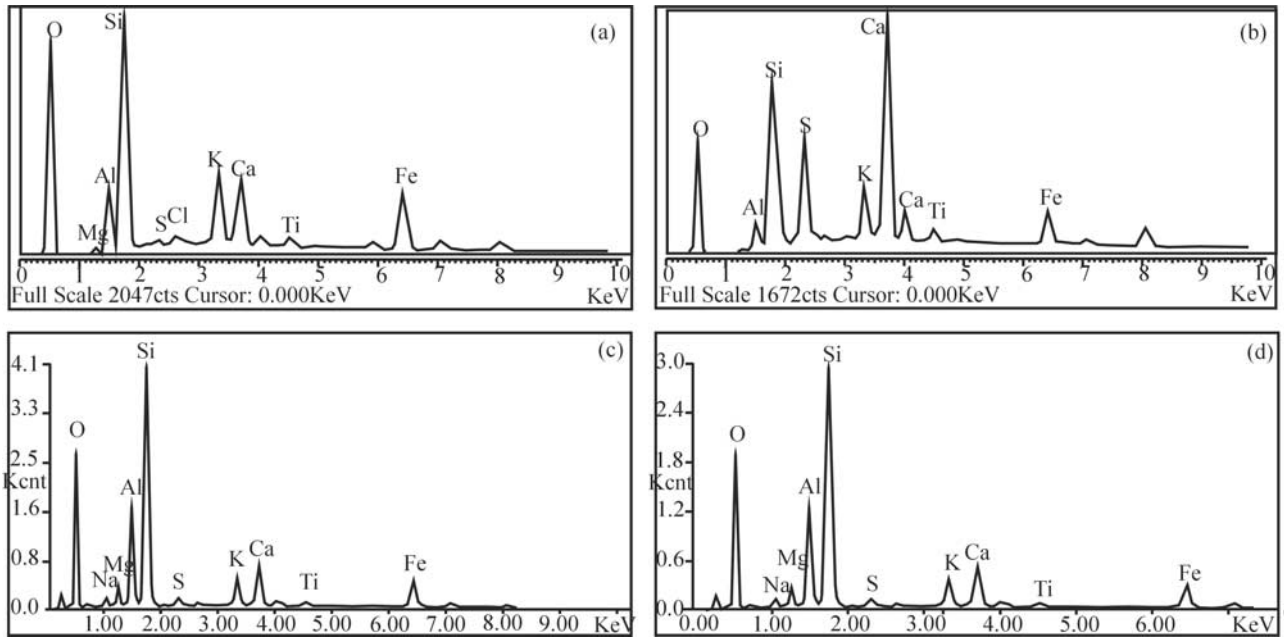

Figure 4. ESEM-EDX spectra of mineral particles collected during the 20 March 2002 severe dust storm. (a) Typical spectrum of a bar-shaped mineral particle; (b) spectrum of a mineral aggregate composed of aluminosilicate and gypsum; (c) spectrum of a microarea (bulk), showing a sulfur peak; (d) spectrum of a sulfur-containing individual particle.

Figure 4b shows the EDX spectrum of a mineral particle which was an aggregate of aluminosilicate and gypsum. We have analyzed 132 individual ADS particles, of which about one fifth are mineral aggregates.

[16] A randomly selected portion of the 20 March 2002 dust storm sample was analyzed with ESEM-EDX, and the chemical spectrum is shown in Figure 4c. A sulfur peak was evident in Figure 4c. By ZAF correction, the weight percentage of each element present in the spectrum could be quantified. If normalized to 100% for Si, O, Al, Ca, K, Fe, Mg, Na, S and Ti, sulfur weight percentage was 0.7%. Figure 4d shows the EDX spectrum of a sulfur-containing particle. About one fourth of the 132 individual particles analyzed contained sulfur. Although there were particles in which sulfur was the major element such as that in Figure 4a, sulfur was a minor element for most of the sulfur-containing particles.

3.2.2. Semiquantitative Results of Mineral Species in Beijing ADS Particles

[17] The XRD analyses demonstrated that quartz, K-feldspar, plagioclase, calcite, pyrite, hornblende, illite/smectite mixed layers, illite, kaolinite and chlorite were present in the 6 April 2000 dust storm sample (ADS-T1) (Figure 5a). The same mineral assemblage, together with hematite, was also identified in the PM10 (ADS1) collected during the 20 March 2002 severe dust storm episode (Figure 5b). For comparison, Figure 5c showed a typical XRD pattern of a non-ADS sample. Illite, gypsum, quartz, dolomite, calcite, and hematite were identified in the non-ADS sample.

[18] The phase compositions of the PM samples collected during the two severe dust storm episodes were further quantified based on XRD patterns. By weight percentages, the TSP sample collected during 6 April 2000 dust storm episode was dominated by clay minerals (35.5%) and quartz (30.3%), followed by calcite (14.0%), noncrystalline materials (10.1%), plagioclase (7.0%), K-feldspar (1.7%), pyrite (1.0%) and hornblende (0.4%). In particles smaller than 10 μm separated from TSP, the major phases by weight were clay minerals (39.0%), quartz (20.7%) and noncrystalline materials (20.0%), with certain amount of calcite (9.1%), plagioclase (7.8%), pyrite (1.0%), K-feldspar (2.1%) and hornblende (0.3%) (Figure 6a).

[19] Quantification of the PM10 collected during the 20 March 2002 severe dust storm episode is shown in Figure 6b. By weight, this sample was dominated by clay minerals, accounting for 40.1%, and the other phases included noncrystalline materials (20.5%), quartz (19.5%), plagioclase (8.4%), calcite (7.5%), K-feldspar (1.5%), hematite (0.9%), pyrite (0.9%), hornblende (0.4%) and gypsum (0.3%).

[20] The XRD measurements of both the TSP and PM10 collected during the 6 April 2000 severe dust storm episode showed that quartz was mostly enriched in TSP and deficient in PM10, while clay minerals were mainly concentrated in PM10 and deficient in TSP. Percentages by weight of other minerals were similar in TSP and PM10. For the 20 March 2002 dust storm samples, quartz was relatively concentrated in PM10 while clay minerals were enriched in PM less than 2 μm. Because the fine ADS particles could be transported a longer distance, the clay minerals may have greater impacts on the global environment and climate. The results of *Arnold et al.* [1998] supported this inference.

3.2.3. Semiquantitative Results of Clay Mineral Species in ADS Particles

[21] Because clay minerals were the most abundant species collected during the two severe dust storm episodes, they were separated from the whole samples, and then

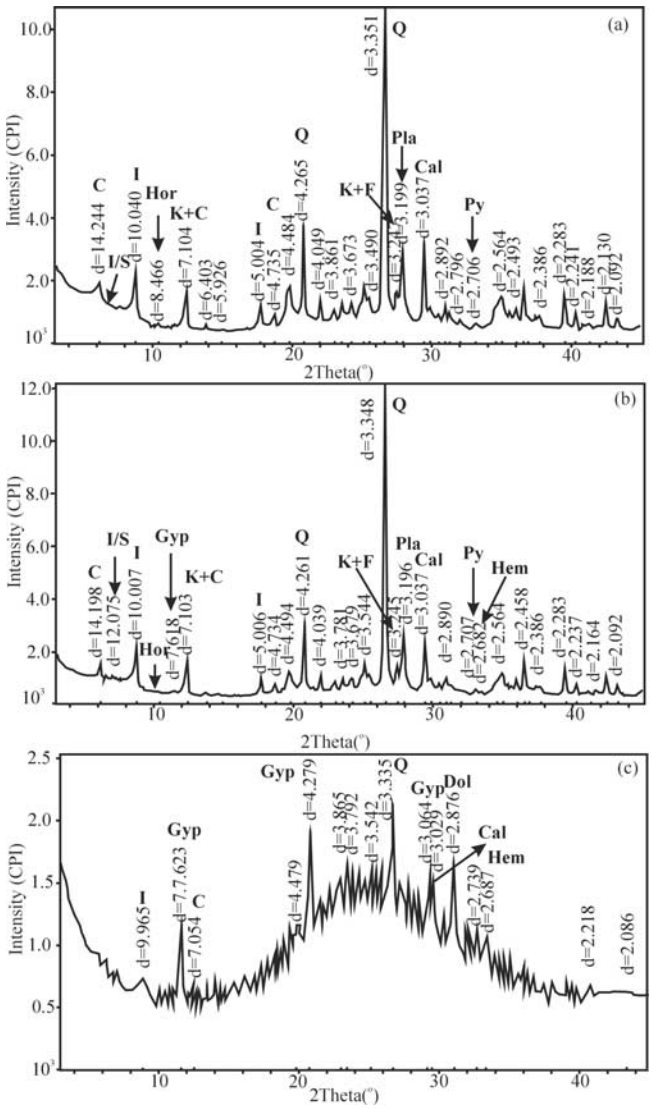

Figure 5. XRD patterns of PM10 samples collected during dust storm and nondust storm episodes. (a) PM less than 10 μm (by sedimentation in water from TSP, ADS-T1) collected during the 6 April 2000 severe dust storm episode; (b) PM10 (ADS1a) collected during the 20 March 2002 severe dust storm episode. (c) An example of nondust storm PM10 collected on glass fiber filter (28 March 2002 at 2100 LT until 29 March 2002 at 0900 LT). The dolomite peak is obvious; this pattern was used for comparison with those of ADS samples only. I, illite; Hor, hornblende; Q, quartz; C, chlorite; K, kaolinite; K+C, kaolinite+chlorite; I/S: illite/smectite; Cal, calcite; Gyp, gypsum; Py, pyrite; K+F, potassium feldspar; Pla, plagioclase; Hem, hematite.

analyzed and quantified with the XRD. Figure 7 is the XRD pattern of clay minerals in PM10 collected during the 20 March 2002 severe dust storm episode. The quantitative analysis (Table 2) showed that clay mineral assemblages of PM less than 2 μm during both severe dust storm episodes were similar. Illite/smectite mixed layers were the major clay mineral species, accounting for about 78%, followed by illite (9%), chlorite (6%) and kaolinite (7%). In addition, smectite occupied 40% in the illite/smectite mixed layers of the particles less than 2 μm for the two severe dust storms (Table 2).

4. Discussion

4.1. Sulfur Element in the Individual Dust Storm Particles

[22] The individual particle analysis has revealed that about one fourth of the 20 March 2002 ADS particles collected in Beijing contained sulfur element. *Falkovich et al.* [2001] reported that about 65% of the African dust storm particles collected in Israel contained sulfur, but sulfur was from soil-derived processes in the source regions and not from atmospheric processes. The ESEM-EDX analyses showed the presence of irregularly shaped gypsum in the 20 March 2002 dust storm sample. This provides evidence that at least some of the sulfur in the 20 March 2002 dust storm sample was from processes in the source region. Using S/Al ratio as an indicator, *Guo et al.* [2004] suggested that some of sulfur in the 20 March 2002 dust storm sample in Beijing was of crustal origin. *Zhang and Iwasaka* [1999] also found that 14.6% of the particles showed a sulfur peak in EDX spectra although few of the dust particles collected in Beijing contained water-soluble sulfate. The results in our study also showed that no morphological modification, as that for ADS particles collected in Japan [*Iwasaka et al.*, 1988; *Zhou et al.*, 1996; *Fan et al.*, 1996], could be observed in single ADS particles collected in Beijing (Figures 2a and 2c). On the other hand, Ion Chromatography (IC, Dionex 100, US) analysis in this study showed that the water-soluble sulfates account for 0.55% of the mass of PM10 collected during the 20 March 2002 dust storm episode. The sources of the water-soluble sulfates are still unknown. Therefore further work on individual particle analysis, as well as bulk chemical analysis of more ADS samples during different dust storm episodes, needs to be conducted to elucidate whether and how much the atmospheric processes contribute to the sulfur content in ADS samples collected in the Asian continent.

4.2. Comparison of Mineral Compositions of Dusts From Different Regions

[23] For comparison, 10 non-ADS samples were also analyzed with XRD. Dolomite was present in all of the non-ADS samples, as seen in Figure 5c. *Davis and Guo* [2000] and *Shi et al.* [2002] also reported the presence of dolomite in Beijing non-ADS samples. However, dolomite was not identified by the XRD in the two ADS samples (Figures 5a and 5b). Dolomite was also an important mineral species in dust storm samples originated from other regions; for example, Western Sahara, Moroccan Atlas and Central Algeria [*Avila et al.*, 1997], African, southern Algeria and northern Chad [*Falkovich et al.*, 2001]. Thus the absence of dolomite was an important characteristic of the two severe dust storm samples.

[24] It has been previously suggested that the mineralogy of dust storm particles may be more diagnostic of their sources [*Bergametti et al.*, 1989; *Merrill et al.*, 1994; *Davis and Guo*, 2000; *Ganor et al.*, 2000]. *Gomes and Gillette* [1993] also showed that calcite/clay and quartz/clay ratios could be used as tracers to identify the origins of the dusts. The ranges of calcite/clay and quartz/clay ratios of silt size

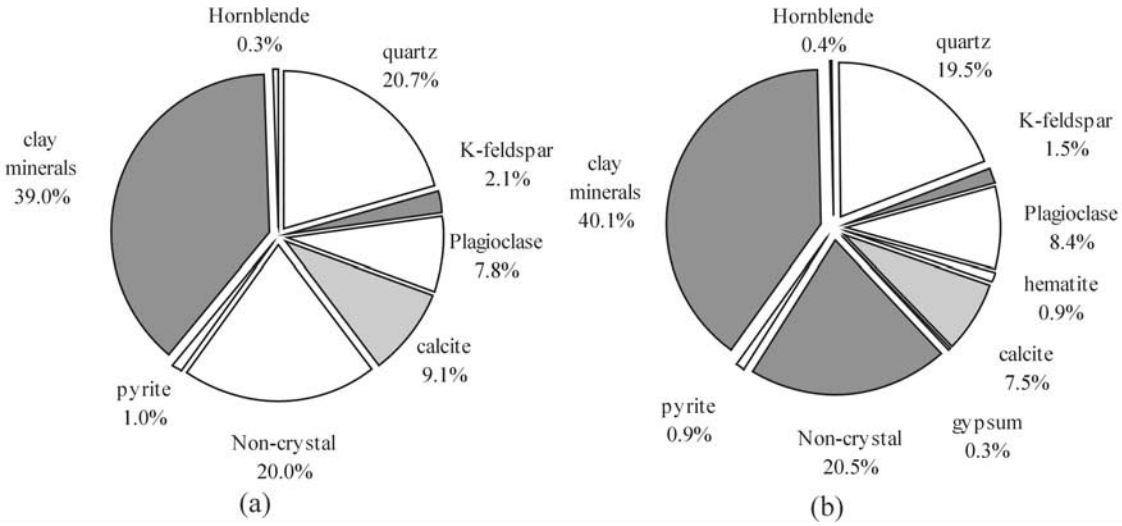

Figure 6. Weight percentages of different phases in PM10 samples collected during the severe dust storm episodes in Beijing: (a) 6 April 2000 (by sedimentation in water from TSP) and (b) 20 March 2002.

mineral aerosol sampled in northern Sahara, Soviet Central Asia and southwestern United States were also provided by *Gomes and Gillette* [1993, Figure 3]. Calcite/clay and quartz/clay ratios of the PM10 collected during the 20 March 2002 ADS episode were 0.19 and 0.49, respectively, while those of the PM less than 2 μm separated from the PM10 collected during the 20 March 2002 ADS episode were 0.19 and 0.24, respectively. Calcite/clay and quartz/clay ratios of the TSP collected during the 6 April 2000 ADS episode were 0.2 and 0.85, respectively, while those the PM less than 10 μm separated from TSP collected during the 6 April 2000 ADS episode were 0.14 and 0.40, respectively. Therefore proportions of different mineral species in PM samples vary significantly with the sizes of the particles. Unfortunately, mineral dusts were generally sampled as silt and clay sizes [e.g., *Gomes and Gillette*, 1993; *Leinen et al.*, 1994; *Arnold et al.*, 1998]. This made it difficult to compare the calcite/clay and quartz/clay ratios of ADS samples in this study with those of dust samples collected in other regions. However, even with clay minerals being enriched in the finer dust sizes, the quartz/clay ratios in PM less than 10 μm collected during the two severe dust storm episodes in this study were still higher than those of silt size mineral aerosols from northern Sahara, Soviet Central Asia and southwestern United States. The quartz/clay ratios of PM in these regions are equal to or less than 0.36 [*Gomes and Gillette*, 1993].

5. Conclusions

[25] (1) ADS particles collected in Beijing were mainly composed of irregularly shaped minerals.

[26] (2) More than 50% of ADS particles were larger than 1 μm in ESD.

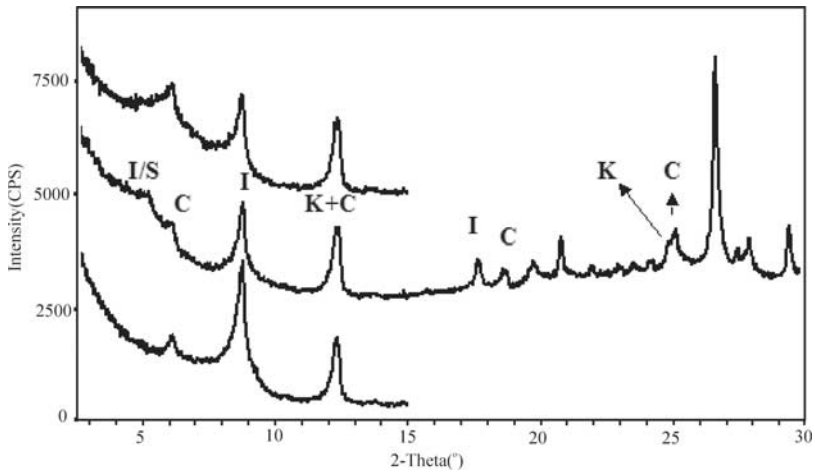

Figure 7. XRD pattern of clay minerals collected during the 20 March 2002 severe dust storm episode (<2 μm by sedimentation in water from PM10). I/S, illite/smectite mixed layers; C, chlorite; I, illite; K+C, kaolinite +chlorite; K; kaolinite.

Table 2. Quantitative Results of Clay Mineral Species in PM Samples Collected During the Severe Dust Storm Episodes

	Illite/Smectite	Illite	Koalinite	Chloride	Illite/Smectite Ratio
6 April 2000	79	10	5	6	1.5
20 March 2002	78	9	6	7	1.5

[27] (3) About 20% of individual particles collected during the 20 March 2002 dust storm were mineral aggregates and at least 25% of the individual particles contained sulfur. Sulfur in some of the ADS particles was originated from processes in the source region.

[28] (4) The XRD analyses of phase compositions of PM10 samples from two severe dust storm episodes showed that clay minerals were major mineral species, accounting for more than 40%, followed by noncrystalline materials and quartz (both around 19%), with small amounts of calcite, plagioclase, K-feldspar, pyrite and other trace minerals (all less than 10%).

[29] (5) Clay minerals in the PM samples collected during the two severe dust storm episodes were mainly illite/smectite mixed layers, with small amounts of illite, kaolinite and chlorite.

[30] (6) XRD, FESEM and ESEM-EDX analyses allowed us to distinguish the two ADS samples from dust generated in other regions. PM10 samples collected during the severe dust storm episodes were characterized by the absence of dolomite, high quartz/clay ratio as well as dominance of illite/smectite mixed layers in clay mineral species.

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