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Variation of sources and mixing mechanism of mineral dust with pollution aerosol—revealed by the two peaks of a super dust storm in Beijing

Xiujuan Zhao [a,b,c,d], Guoshun Zhuang [a,b,c,*], Zifa Wang [b], Yele Sun [b,c], Ying Wang [a,c], Hui Yuan [c]

[a] Center for Atmospheric Chemistry Study, Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China
[b] NZC/LAPC, Institute of Atmospheric Physics, Chinese Academy of Science, Beijing 100029, China
[c] Center for Atmospheric Environmental Study, Department of Chemistry, Beijing Normal University, Beijing 100875, China
[d] Institute of Environment and Sustainable Development in Agriculture, Chinese Academy of Agriculture Sciences, Beijing 10081, China

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Abstract

The observation of the super dust storm in Beijing from 20 to 22 March 2002 with high-time resolution showed that there were two peaks of TSP of 10.9 and 5.1 mg m^{-3} with 87% and 60% of the mineral dust to TSP, respectively. The variation of sources and mixing of mineral dust with pollution aerosol was distinguished with hourly meteorological data and lidar observation and identified by horizontal visibility and chemical tracers. The dust in PI mainly originated from source I, which included west and middle regions of northern China and the nearby Gobi desert in Mongolia, and the dust in PII was mostly from source II, which mainly included the northeast of China and the southeast of Mongolia. The source I was a relatively ‘clean’ one and the source II was a ‘polluted’ one. The dust in PI mainly mixed with the pollutants from the transport pathway, and the dust in PII was rich in pollution compositions and mixed with the resuspended pollutants and the urban dust from the local area in Beijing. The mixing of the dust aerosols originated from a relatively ‘clean’ source with the pollutants on the transport pathway could carry significant amounts of pollutants downwind. The dust, which came from the ‘polluted’ source and mixed with the local resuspended pollutants, could deliver much higher content of pollutants downwind. Though the second dust peak was weaker than the first one, it would have greater impacts on the human health for the higher fraction of pollution and water-soluble components.

Keywords: Dust storm; TSP; Source; Mixing; Composition

1. Introduction

Asian dust storm, originating from the desert regions in China and Mongolia, is transported thousands of kilometers downwind by the westerly over the Asian continent and Pacific Ocean, and on occasion reaches North America (Duce et al., 1980; Uematsu et al., 2002; Cahill, 2003). The dust particles carried by dust storms

* Corresponding author. Center for Atmospheric Chemistry Study, Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China. Tel.: +86 21 55664579; fax: +86 21 65642080.
E-mail address: gzhuang@bnu.edu.cn (G. Zhuang).

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have great impacts on the global biogeochemical cycle (Zhuang et al., 1992; Johnson et al., 2003) and climate (Tegen et al., 1996). During its long-range transport the dust particles mixed with pollution aerosol by physical and chemical processes and changed the chemical compositions of the aerosol in dust storm. The ACE-Asia project made an intensive field study to characterize the dust aerosol properties in East Asia and led to more understanding of the effects of Asian dust on the Earth's climate system (Huebert et al., 2003; Seinfeld et al., 2004).

The dust particles carried by the dust storm provided surface for many physical and chemical processes and serve as carriers of anthropogenic substances. The mixing of the dust aerosol and anthropogenic pollutants has been investigated by *in situ* observation, laboratory experiments, model simulation in the last two decades (Song et al., 2005; Arimoto et al., 2004; Usher et al., 2002; Dentener et al., 1996; Okada et al., 1990; Iwasaka et al., 1988). Zhang et al. (2000) studied the mixture of Asian dust particles collected in Qingdao by X-ray (EDX) spectrometer, and found that mineral materials could enhance the formation of the particulate sulfate/nitrate and nitrate was predominant in the mixture on coarse mode particles. Ooki and Uematsu (2005) examined the ionic composition of the size-fractionated aerosols collected in the coastal region off Japan and the North Pacific Ocean in spring 2001 and 2002. They found that the concentration peaks of nss-Ca^{2+} and NO_3^- were in the same size range. The internal mixing of nss-SO_4^{2-} with mineral dust particles would increase by in-cloud collision. Usher et al. (2002) investigated the heterogeneous kinetics of SO_2 uptake on an authentic sample of China loess. The uptake of SO_2 on loess sample scaled linearly with sample mass. Adsorbed SO_2 on the surface could be oxidized with ozone to sulfate and /or bisulfate. Our group has systematically studied the dust aerosol in Beijing since 2000. Zhuang et al. (2001) investigated the composition, sources and size distribution of the aerosols in a dust storm on 6 April 2000, and pointed out that the dust storm delivered large amounts of pollutants that were either from the pollution sources on the pathway or from Beijing local pollution sources. Guo et al. (2004) found four stages of a dust storm by examining the PM10/SO2, elemental ratios and meteorology during March 2001 and March 2002 in Beijing and Shanghai, and the overlapping of stages was found to be one of the mechanisms of getting high pollution concentrations in dust storms. Wang et al. (2005a) investigated the differences of six dust episodes that occurred in Beijing in spring of 2002 using water-soluble part of the aerosols. The results showed that the mixing between mineral and pollution aerosols was ubiquitous during the dust seasons. Sun et al. (2005) put emphasis upon the impacts of source regions and transport pathways on the composition of the aerosol in Asian dust storms and the results showed that the source regions and transport pathways were two vital factors affecting chemical compositions of dust storms. The dust storm of "polluted" pathway carried more pollution elements than that of "less-polluted" one. The mixing of the dust aerosol with pollution aerosols in these studies mentioned above was investigated based on the average characteristics of bulk aerosols during the whole dust storm event, and the mixing were derived from the different dust storm events. However, the variation of the mixing of dust aerosol with pollution aerosols in one dust event has scarcely been reported. Collecting the meteorological, elemental, and ionic data with high-time resolution of a super dust storm, here we report the variation of the mixing of dust aerosol with pollution aerosol in one dust event by comparison of the source, transport, and chemical composition of two TSP peaks during a super dust storm that occurred on March 20, 2002 in Beijing.

2. Experiment

2.1. Sampling

Aerosol sampler (Beijing Geological Instrument-Dickel Co., Ltd., model (TSP/PM_{10}/$PM_{2.5}$) was employed for TSP and $PM_{2.5}$ sampling from March to April 2002 on the roof (~40 m height) of the 12th floor in the Building of Science and Technology, Beijing Normal University. Several TSP samples with each in 2–3 h continuously were collected in each dust storm day, while one TSP sample in 3-h in each non-dust day. The samples were collected on the Whatman 41 filter membrane (Whatman Company, UK) and put in polyethylene plastic bags right after sampling and reserved in a refrigerator. They were weighed after stabilizing under constant temperature (20 ± 1 °C) and humidity (40 ± 1%), using an analytical balance (model: Sartorius 2004MP) with a reading precision 10 μg. All the procedures were strictly quality-controlled to avoid any possible contamination of the samples.

2.2. Chemical analysis

2.2.1. Element analysis

The sample filters were digested at 170 °C for 4 h in high-pressure Teflon digestion vessel with 3 ml concentrated HNO_3, 1 ml concentrated HCl, and 1 ml concentrated HF. After cooling, the solutions were dried, and then diluted to 10 ml with distilled–deionized water. A total of 23 elements (Al, Fe, Mn, Mg, Ti, Sc, Na, Eu, Ce, Sr, Ca, Co, Cr, Ni, Cu, Pb, Zn, Cd, V, S, As, Se, and Sb) were analyzed by inductively coupled plasma spectroscopy and atomic emission spectroscopy (ICP-AES) (Model: ULTIMA, JOBIN-YVON Company, France). The detailed analytical procedure has been described in Zhuang et al. (2001).

2.2.2. Ion analysis

Eleven inorganic ions (SO_4^{2-}, NO_3^-, F^-, Cl^-, NO_2^-, PO_4^{3-}, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}) and 4 organic acids (acetic, formic, oxalic, and methylsulfonic acid (MSA)) were analyzed by ion chromatography (IC, Dionex 600) that consists of a separation column (Dionex Ionpac AS11 for anion and CS12A for cation), a guard column (Dionex Ionpac AG 11 for anion and AG12A for cation), a self-regenerating suppressed conductivity detector (Dionex Ionpac ED50) and a gradient pump (Dionex Ionpac GP50). The details were given elsewhere (Yuan et al., 2003).

2.3. The meteorological data and lidar observation data

The meteorological data, including wind speed, relative humidity, visibility, temperature and surface weather phenomena, etc., were obtained from the China Meteorological Administration (CMA). The hourly meteorological data of Beijing were downloaded from http://www.wunderground.com. The lidar observation in Beijing was performed at the Sino-Japan Friendship Center for Environmental Protection SJFCEP (Sugimoto et al., 2003).

3. Results and discussion

3.1. Two peaks of TSP during a super dust storm

A heavy dust storm occurred in northern China and invaded Beijing on 20 March 2002 with the highest TSP concentration of 10.9 mg m^{-3} and $PM_{2.5}$ of 1.39 mg m^{-3} (Sun et al., 2004a,b). The dust at Beijing appeared after 0900 LT in the morning of March 20. The visibility decreased to 2 km at 1100 LT and reached lowest 1.1 km at 1400 LT. After 15 LT, the dust storm started to abate, and the visibility increased to 8 km at 2000 LT. But after 2100 LT, the visibility decreased again and reached 7 km at 2300 LT. This dust storm passed out of Beijing on the early morning of March 21, and the visibility recovered to 1200 m, which was consistent with the results from our TSP measurements during the period from 1020 LT, March 20 to 1030 LT, March 21. This dust storm was one of the most severe dust events ever recorded in Beijing in history.

Two peaks of TSP concentration were observed on March 20 with the concentration of 10.9 mg m^{-3} in the first peak from 1020 to 1220 LT (named as PI) and of 5.1 mg m^{-3} in the second peak from 2025 to 2220 LT (named as PII) (Fig. 1). These two peaks could also be found in the lidar observation and the particle number measurement in Beijing during the same period (Sugimoto et al., 2003). The mineral dust in TSP was calculated as the sum of oxides of aluminum, calcium, iron, titanium, magnesium and silicon (i.e., mineral dust = 1.89Al + 2.14Si + 1.4Ca + 1.43Fe + 1.66Mg + 1.67Ti) based on our measurement of the concentration of these elementals (Taylor and Mclennan, 1985; Zhang et al., 2003; Hueglin et al., 2005). The concentration of Si was estimated according to the average ratio of Si/Al (3.9) obtained from Zhang et al. (2003). This ratio was obtained from the TSP samples from three desert sites and one loess site in China. The Si/Al was 3.9, 3.7, 3.7 and 4.2 for these four sites. The average 3.9 was used to estimate the concentration of Si. The observed TSP, the calculated concentration of the mineral dust and the ratio of the mineral dust to TSP were shown in Fig. 1. Evidently, mineral dust was overwhelming in TSP when dust storm occurred, which accounted for in average 80% of TSP. The percentages of the mineral dust in PI and PII were 87% and 60%, respectively. The difference of the mineral composition of TSP between the two peaks puzzled us. Had these dust aerosols come from different source that consists of different compositions or from variations of the mixing with the

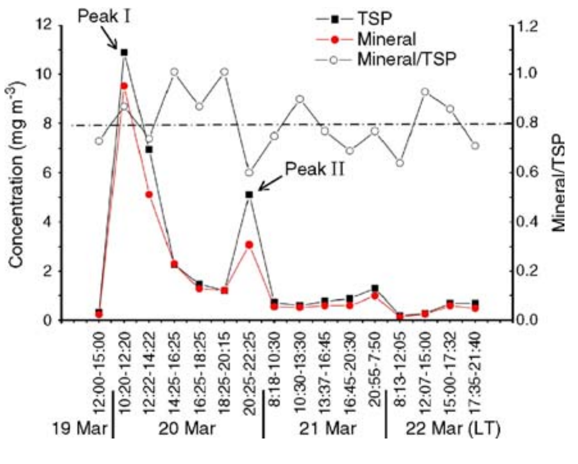

Fig. 1. Variations of concentrations of TSP and dust aerosol from 19 to 22 March 2002.

pollutants, which emitted either from the local sources in Beijing or from the transport pathway of the dust? We tried to address this question by carefully analyzing the meteorological data recorded over China with the elemental and ionic data collected during this dust storm in Beijing.

3.2. Sources of the dust in two peaks

3.2.1. Source identification by meteorological data

The source of this dust storm was well studied by back-trajectory analysis technique and model simulation (Zhang et al., 2005; Han et al., 2004; Park and In, 2003; Shao et al., 2003; Sugimoto et al., 2003). This dust storm originated from the Gobi desert near the southern China–Mongolian border on March 19, passed over Alashan Plateau and strengthened further over the southeastern China–Mongolia border on March 20, finally to Beijing along the westerly direction. However, the source of the dust in the two peaks on March 20 was not clear. Here, the visibility at 3-h interval obtained through the meteorological networks was used to distinguish the source of the two peaks. Dust events were classified into four categories of dust in suspension, blowing dust, dust storm, and severe dust storm according to the horizontal visibility of less than 10 km, 1–10 km, 500–1000 m, and less than 500 m in the surface weather records made by CMA, respectively. To show clearly, a 24-h backward isentropic air trajectory of this dust storm was calculated with HYSPLIT4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model developed by NOAA/Air Resources

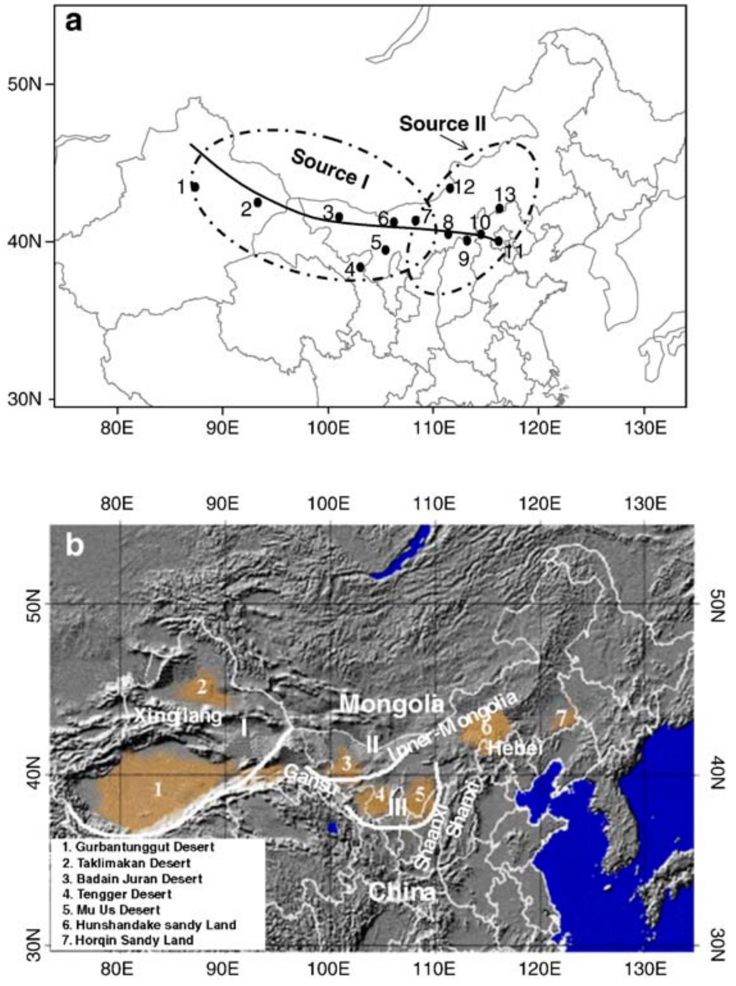

Fig. 2. (a) A 24-h back-trajectory calculation for 1500 LT on 20 March 2002 in Beijing, the source regions of this dust storm and locations of several sites: (1) Urumuchi; (2) Hami; (3) Erjinaqi; (4) Minqin; (5) Jilantai; (6) Hailisu; (7) Wulatezhongqi; (8) Huhhot; (9) Datong; (10) Zhangjiakou; (11) Beijing; (12) Erlianhaote; and (13) Duolun. (b) Map showing the main Chinese deserts and the source regions defined by Zhang et al. (1996): source regions: (I) western desert; (II) northern high-dust deserts; (III) northern low-dust deserts.

Laboratory (Draxler and Hess, 1998) using the National Centers for Environmental Prediction (NCEP) Final Analyses (FNL) meteorological database for 15 LT on 20 March 2002. The results with the seven sites along the transport pathway chosen for the source analysis were shown in Fig. 2a. The sites, Erlianhaote and Duolun (Fig. 2a), were used to analyze the possibility of these sources located to the northwest and north of Beijing, respectively. The visibilities observed at these sites from 0200 LT March 19 (the beginning of this dust storm) to 0800 LT March 21 (after ceasing of the second TSP peak) is shown in Fig. 3. To delineate the progression of this dust storm, the spatial distribution of this dust event at different time is presented in Fig. 4. This dust storm firstly appeared in southern Mongolia and the northern of Xinjiang at 0800 LT on March 19 (Fig. 4). It quickly arrived at Ejinaqi, located near Badain Juran Desert, with a visibility lower than 1 km, and then moved southeasterly to Hailisu, Jilantai, Wulatezhongqi (shade A in Fig. 3a), and Minqin, and then moved eastward to Hohhot, Datong, Zhangjiakou and finally reached Beijing (shade A in Fig. 3b, d). By carefully examining the visibility data, it was found that there was only one vale with a visibility lower than 10 km at the sites in the west and middle regions of northern China, such as Urumuchi, Ejinaqi, Minqin, Hailisu and Wulatezhongqi (Shade A in Fig. 3a) in the afternoon of March 19. This indicated that there was only one dust event that occurred in these regions from 19 to 21 march, which also could be seen from the recorded dust

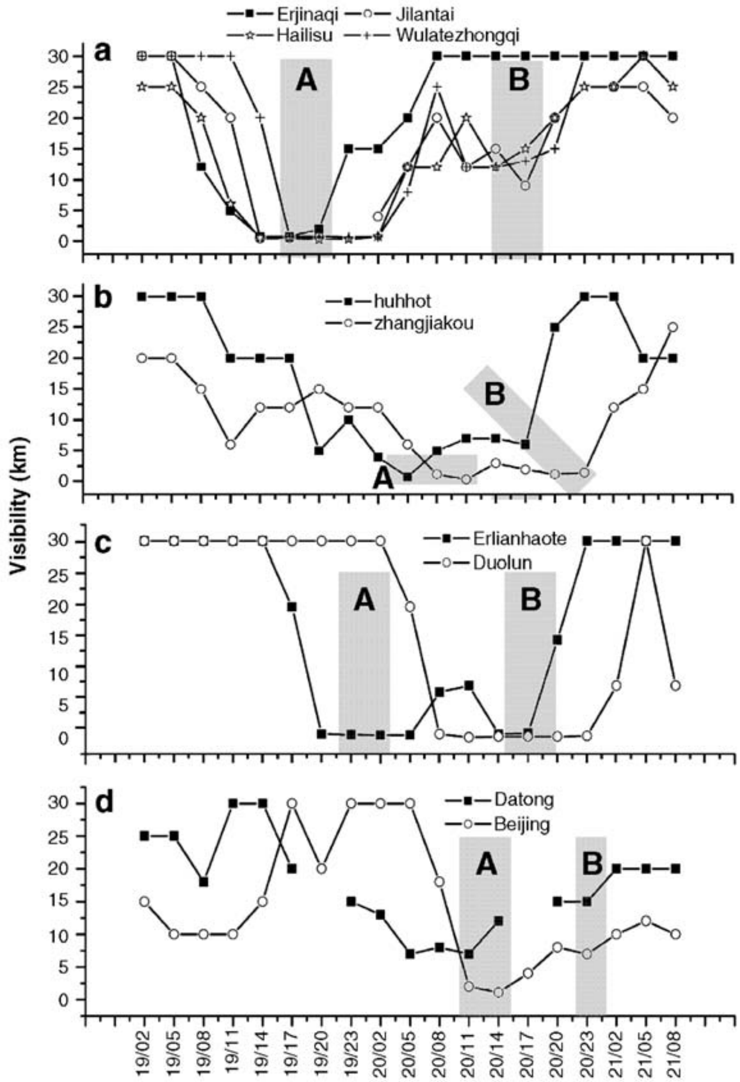

Fig. 3. Visibility observed from 0200 LT on 19 March to 0800 LT on 21 March 2002.

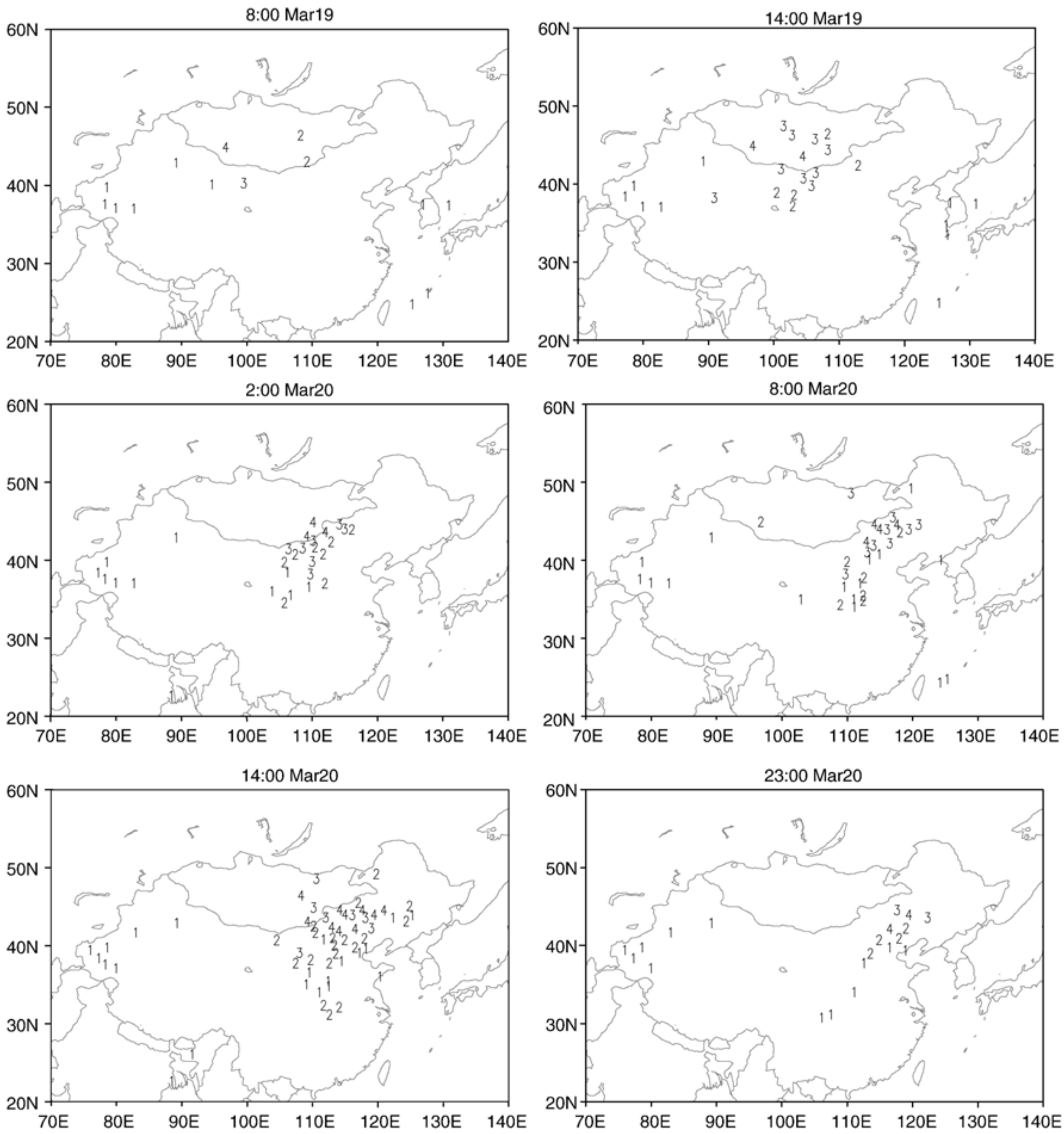

Fig. 4. Spatial distributions of observed surface dust event from 19 to 20 March 2002 (1, 2, 3, 4 represents dust in suspension, blowing dust, dust storm, and severe dust storm, respectively).

events in surface observation as shown in Fig. 4. This region and the nearby Gobi desert in Mongolia were defined as source I of this dust storm, which covered the northern part of the western desert source, the whole northern high-dust source and west part of northern low-dust source (Zhang et al., 1996). This result was also consistent with that derived from model simulation mentioned above. However, from Hohhot to Beijing along the transport pathway and to the northwest of Beijing (at Erlianhaote), the visibility had two vales that appeared both on March 20 (Shade A and B in Fig. 3b–d). The first was much lower than the second one in Hohhot and Beijing but almost same in Zhangjiakou and Erlianhaote, indicating that the dust storm was stronger in the first episode than that in the second one. This agreed well with the TSP observation in Beijing as described in Section 3.1. The variation of the visibility indicated that the first dust episode was injected new dust in the morning on March 20, which was uplifted from the region defined as source II of this dust storm

(Fig. 2a), and another weak dust episode started in this region in the afternoon of March 20. Source II was one of the main sources of the Asian dust storm, which included the east part of the northern low-dust source (Zhang et al., 1996), the northeast of China and the southeast of Mongolia. The dust storm arrived at Duolun a little earlier than Beijing, but it stood to 2300 LT in the same intensity (Fig. 3c). The dust that originated from this region could barely contribute to PI in Beijing, but could provide a certain amount of dust to PII by the northwest wind. It could be merged into source II. From the timing and intensity of the dust storm that occurred in sources I and II, it could be found that the dust storm originated from source I on March 19 and was injected with new dust when it passed over source II. It reached Beijing in the morning on March 20 that produced the first TSP peak (Fig. 4). The dust of PII was mainly from the source II, where the second deflation of dust occurred in the afternoon and evening on March 20.

3.2.2. Source identification by chemical tracers

Elements that are useful as a reference for crustal material include Si, Al, Fe, and Ti, in which Al is most frequently used as the reference element for the mineral component. The mass percentage of Al in TSP was 6.8% (Table 1) in PI, close to that of 7% in source I (Zhang et al., 2003). The mass percentage of Al in PII was 4.5%, which was evidently much lower than 7% and close to that of 4.0–5.7% in the soil of source II in China (Zheng et al., 1994) and 3.9% in TSP in non-dust days in Beijing in March and April. These results supported the above conclusion to some extent that the dust in PI mostly came from source I and the dust in PII mainly originated from the source II, the northwest region to Beijing.

The ratios of main crustal elements, such as Fe, Mg, Sc and Ca, to Al are used as tracers to identify the source of dust storm (Chester et al., 1984). The ratios of Fe, Mg, Ca and Na to Al were 0.5, 0.24, 1.04, 0.23 in PI and 0.72, 0.18, 1.15, 0.25 in PII. Zhang et al. (1996, 1997) collected 120 aerosol samples at 12 sites in Chinese deserts using eight-stage cascade impactors, which were analyzed by a particle-induced X-ray emission (PIXE) technique. Using four interelement ratios, they found three discriminable dust sources in deserts of North China, namely, the western desert source, the northern high-dust source and the northern low-dust source (Fig. 2b). Their results showed that the ratio of Fe/Al was 0.65 in the northern high dust source and 0.44 in the north low dust source. In the surface soil of Duolun Fe/Al was 0.9. The Fe/Al of 0.5 in PI was between that of the northern high dust source and that of the northern low dust source, while in PII of 0.72 was between that of the northern high dust source and Duolun. This indicated that the dust in PI mainly came from the west source I and mixed with a certain amount of dust from source II, while the dust in PII mainly originated from source II, which has higher Fe/Al. The Mg/Al is also an indicator to be used to identify the origin of the dust from different areas (Zhang et al., 1996; Han et al., 2005). The ratio of Mg/Al in TSP was 0.24 in the first peak and 0.18 in the second one. The Mg/Al for dust aerosol and surface soil in typical sites in China

Table 1
Enrichment factors of the elements and percentages of the elements and ions in TSP in the two peaks

Elements	EF			Percentage (%)			Ions	Percentage (%)		
	PI	PII	PII/PI	PI	PII	PII/PI		PI	PII	PII/PI
As	15.2	101.8	6.7	0.002	0.010	4.4	Na^+	0.15	0.77	5.1
Pb	3.7	11.7	3.2	0.004	0.009	2.1	NH_4^+	0.02	0.13	6.4
Cd	11.0	23.4	2.1	0.0001	0.0002	1.4	K^+	0.04	0.18	4.4
Cr	0.98	1.2	1.2	0.008	0.006	0.8	Mg^{2+}	0.04	0.18	4.5
Zn	1.7	2.3	1.3	0.01	0.009	0.9	Ca^{2+}	0.55	1.92	3.5
Sr	0.79	0.87	1.1	0.024	0.018	0.7	F^-	0.007	0.01	1.4
Ni	0.53	0.84	1.6	0.004	0.004	1.0	Cl^-	0.21	0.07	0.4
Co	1.03	1.5	1.5	0.002	0.002	1.0	NO_2^-	0.006	0.015	2.3
Fe	0.73	1.1	1.4	3.4	3.2	1.0	NO_3^-	0.028	0.03	1.2
Mn	0.70	0.9	1.3	0.054	0.047	0.9	SO_4^{2-}	0.74	0.38	0.5
Mg	1.5	1.1	0.7	1.6	0.8	0.5	CH_3COO^-	0.0074	0.0003	0.0
V	1.0	1.4	1.3	0.010	0.009	0.9	$HCOO^-$	0.0021	0.0004	0.2
Ca	2.1	2.3	1.1	7.1	5.2	0.7	Total	0.18	0.4	2.1
Cu	0.6	0.94	1.6	0.003	0.003	1.0				
Na	0.8	0.89	1.1	1.6	1.1	0.7				
S	15.9	16.3	1.0	0.459	0.313	0.7				
Al	1	1	1.0	6.8	4.5	0.7				

Table 2
The ratios of Mg/Al in soil and aerosol at different sites

Sampling	Location	Type	Mg/Al	Literature
PI	Beijing	Aerosol	0.24	This study
PII	Beijing	Aerosol	0.18	This study
Dunhuang in Gansu Province	SI	Aerosol	0.16	Zhang et al. (1996)
Jiayuguanin Gansu Province	SI	Aerosol	0.3	Zhang et al. (1996)
The top of the mountain in Gansu Province	SI	Soil	0.29	Han et al. (2005)
Minqin in Gansu Province	SI	Aerosol	0.22	Zhang et al. (1996)
Jilantai in Inner Mongolia	SI	Aerosol	0.32	Zhang et al. (1996)
Heiquan in Inner Mongolia	SI	Aerosol	0.31	Zhang et al. (1996)
Baotouin Inner Mongolia	SI	Soil	0.16	Nishikawa et al. (1991)
Taiyuan in Shanxi Province	SII	Soil	0.22	Nishikawa et al. (1991)
Yulin in Shaanxi Province	SII	Aerosol	0.17	Zhang et al. (1996)
Dingling in suburb of Beijing	SII	Soil	0.23	Han et al. (2005)
TSP at Duolun in Inner Mongolia	SII	Aerosol	0.13	Han et al. (2005)
PM2.5 at Duolun	SII	Aerosol	0.16	Han et al. (2005)
Fengning in Hebei Province	SII	Soil	0.15	Han et al. (2005)
Duolun	SII	Soil	0.12	Han et al. (2005)

was listed in Table 2. It can be found that the ratios of Mg/Al in source I ranged from 0.16 to 0.32, while in source II that varied from 0.12 to 0.23. Clearly, the ratios of Mg/Al were higher in source I than that in source II in average. The ratio of Mg/Al in PI was 0.24, close to the average ratio of 0.26 (the standard deviation was 0.07) in source I. In PII, the ratio of Mg/Al was 0.18, which was very close to the average ratio of 0.17 in source II (the standard deviation was 0.04). These results further supported the source identification mentioned above, i.e., most of the dust in PI originated from the Gobi desert and part of loess plateau in source I, and the dust in PII mainly came from the nearby areas to the northwest of Beijing.

The different sources of dust in PI and PII should firstly be responsible for the different mineral content in PI and PII. However, during transport from the source regions, the dust aerosols mixed with the pollutants on the pathway and mixed the local pollutants when arrived at Beijing. The different mixing of the dust aerosols in these two peaks with pollutants could also cause the different composition of PI with PII. The mixing between dust and pollution aerosols during this dust storm event is discussed further in the following section.

3.3. The mixing between dust and pollution aerosols

Source I mainly consisted of Gobi and deserts, where there are few cities, the pollution emission was less in that region. Thus, it could be seen as relatively 'clean' source. However, in source II, there are many big coal mines plus some heavy polluted cities, such as Hohhot, Erlianhaote, Xilin Haote, and Datong, etc., where coal have extensively been used in heating and industry, which are regarded as the sources of the pollution elements, such as As, Pb, S, etc. (Borbély-Kiss et al., 1998). Also, the road dust from urban area is often enriched with traffic-related elements (Al, Ti, Pb, etc) (Hien et al., 2001; Borbély-Kiss et al., 1998), coal and oil combustion components (S, As, Se, etc.) (Morawska and Zhang, 2002) and construction species (Ca^{2+}). Thus, this source could be seen as 'polluted' source. The dust that originated from source I carried few pollutants, while the dust originated from source II would be rich with pollution elements (As, Pb and S, etc.), and the pollution particles previously deposited in that region would be uplifted by the strong wind with the dust particles and transported to Beijing. However, the dust aerosols in PI traveled through part of the regions of source II, and then mixed with the pollutants emitted by the pollution sources and the dust uplifted in source II along the transport pathway, and finally carried them to Beijing. The dust particles firstly arrived at Beijing at the high layer with a maximum at 1800 m at 0500 LT on March 20, which was observed by lidar (Fig. 5). Then

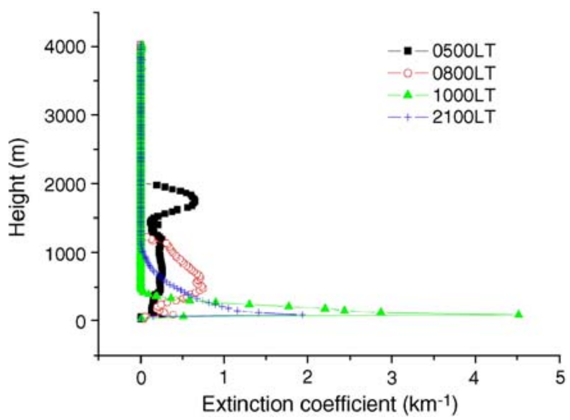

Fig. 5. Extinction coefficient of dust aerosol derived from lidar observation on March 20.

the dust at a lower layer, which was restricted below 1000 m with maximum at 500 m at 0800LT, together with the surface dust, was transported towards Beijing after 1000 LT. Before the surface dust reached Beijing, there was light rain in the early morning on March 20 (about 0600 LT). When the dust storm reached Beijing from the higher layer about 0800 LT, the relative humidity was about 80% and kept above 50% before the main body of the dust storm reached Beijing at 1000 LT (Fig. 6). The low wind speed and the moist earth surface provided little resuspended pollutants and local dust to the TSP in PI. The high relative humidity could lead to the surface of moist dust particles. The moist and alkaline surface could more effectively adsorb the pollution gases and particles in atmosphere, such as SO_2/SO_4^{2-}, NO_2/NO_3^-, and organic compounds before the strong wind arrived. The high EF of S (15.9 in PI, Table 1) and much higher mass percentage of SO_4^{2-} (0.74% in PI, Table 1) than that in Gobi soil (0.01%, Nishikawa et al., 1991) evidently indicated the pollution sources of S, which mixed with the crustal source. The results were similar to Nishikawa et al. (1991), in which it was reported that the SO_4^{2-} in the surface soils of arid area of China could not account for the increase of SO_4^{2-} in dust aerosol. The SO_4^{2-} in coarse size fraction of the dust aerosol was introduced as a surface deposit during transport. The analysis of aerosol data collected near Asia during TRACE-P also provided the evidence of uptake of NO_3^- and SO_4^{2-} on dust surfaces (Jordan et al., 2003). In general, the average concentration of SO_2 in atmosphere was much higher in winter and early spring than that in other seasons for using coal in winter heating in the northern of China (Hu et al., 2002). During the transport of the dust that originated from the source, it could mix with more pollutants containing S because of the high concentration of SO_2 in the atmosphere and the high alkaline surface of dust particles that were beneficial to the absorption of SO_2, which, in turn, was oxidized to be sulfate on the dust surface. On the other hand, the dust particles fell from the higher layer before the surface dust arrived could also absorb acid gases and particles in Beijing under the high relative humidity condition. These results indicated that the pollution composition in PI was mostly from the mixing with the pollutants during the transport to Beijing rather than from the local resuspension. After the dust storm arrived, the blowing dust weather phenomena was recorded at 1400 LT, as the wind speed increased rapidly and the Earth's surface became dry with the sharp decrease of relative humidity. As the sandy ground surface in Beijing area accounted for 14.2% of the total plain area (Beijing Sand Drift Research Group of the Institute of Desert Research, 1987), the sandy land, local road dust and the deposited dust from PI on the ground surface made the Beijing local area become a potential source of dust before the second dust episode arrived. After 2000 LT, the dust from 'polluted' source II carried more pollutants from the source region and arrived to

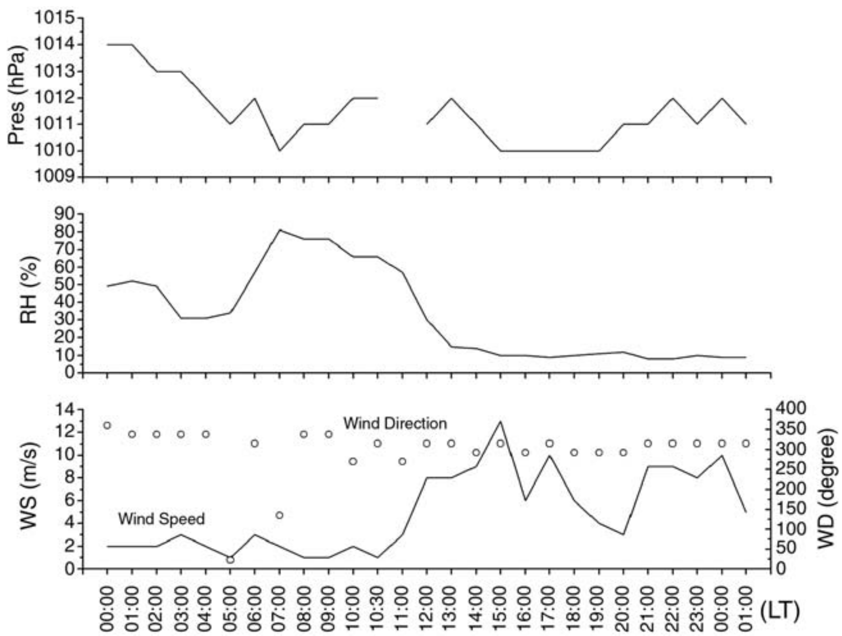

Fig. 6. Variations of pressure, relative humidity, wind speed and wind direction on March 20 at Beijing.

Beijing along surface. With the arrival of dust, the increased wind speed (>8 m s^{-1}) in PII exceeded the threshold wind speed (5 m/s) for Beijing local dust source (Beijing sand drift research group of the institute of desert research, 1987), which could lift the dust from the local area in Beijing to atmosphere. These local dust aerosols mixed with that came from source II contributed to the second TSP peak. The ratio of Ca^{2+}/Al could be used to explain this process. Al is a typical crustal component and it has been used widely as a tracer for suspended soil and the mineral aerosols from the long-range transport. Ca^{2+} could be derived from both soil dust and the construction materials in urban aerosols, and it could be used as an indicator for construction dust in Beijing (Zhang and Iwasaka, 1999). Ca^{2+}/Al ratio could be seen as a good tracer for the mixing of soil dust with the suspended construction materials in urban aerosols (Wang et al., 2005b). The ratio of Ca^{2+}/Al in TSP was 0.08 in PI, much lower than the ratio of 0.43 in PII, which was close to 0.6, the ratio of Ca^{2+}/Al in the TSP collected in the non-dust days in Beijing. The high ratio of Ca^{2+}/Al in PII could indicate the contribution of construction dusts in the cities in source II and in the local area at Beijing. Compared with the Ca^{2+}/Al in non-dust days, the lower ratio in PII may indicate the mixing of construction dust with the soil dusts from source II. These results suggested that the dust from 'polluted' source II carried more pollutants from the source region and arrived to Beijing. These dust aerosols mixed with the resuspended local dust in Beijing by the increased wind, while the dust storm passed Beijing.

The source region of the dust storm and the mixing between dust and pollutants are two vital factors effecting the composition of dust storms. The transport and mixing process of dust will be sure to change the composition of the aerosols in dust storms. The following discussion will focus on the differences of PI and PII and their impact on the composition of TSP.

3.4. Elements in the two peaks

3.4.1. Crustal elements

The concentrations of the major crustal elements, including Al, Ca, Fe, Mg, and Na in the two peaks were presented in Fig. 7. Two peaks of crustal elements, as that of TSP, were observed on March 20. The concentrations of the crustal elements increased substantially in PI, and then decreased rapidly in the afternoon but moderately increased again in PII. The concentrations of Al, Ca, Fe, Mg, and Na were 740, 771, 369, 177, and 170 μg m^{-3} in PI and 231, 266, 166, 41, and 59 μg m^{-3} in PII, respectively. The enrichment factors and percentages of the major crustal elements in the two peaks were presented in Table 1. The EFs of Ca, Fe, Mg, Na, and Mn, using Al as a reference element ($EF(X)=[X/Al]_{aerosol}/[X/Al]_{crust}$), were 2.1, 0.7, 1.5, 0.8, 0.7 in PI and 2.3, 1.1, 1.1, 0.9, 0.9 in PII, respectively. The EFs of the main crustal elements basically unchanged in PI and PII. These results indicated that the crust was the dominant source of these elements in both peaks. The mass percentages of Fe were 3.4% in PI and 3.2% in PII, both were close to the value of 3.5% in crust (Taylor and Mclennan, 1985). However, the mass percentages of Ca, Mg, and Al decreased from 7.1%, 1.6% and 6.8% in PI to 5.2%, 0.8% and 4.5% in PII, respectively. Zhang et al. (2003) reported mass percentages of Ca, Mg, and Al were 7%, 2%and 7% in the northern high-dust source (source I of this dust storm), which were very close to that in PI. The soil environmental background value of source II in China was used to compare with the mass percentage of the crustal elements in PII (Zheng et al., 1994). The abundance of Ca, Mg, and Al in the soil of the part of source II in China were 0.9–5.03%, 0.46–1.12% and 4.0–5.7%, which were lower than that in the northern high-dust source but close to that in PII. These results suggested further that the different sources could lead to the change of the contents of crustal elements in TSP. The mixing of dust aerosol from source region with the pollutants and dust from the pathway and Beijing local area scarcely affected these crustal elements.

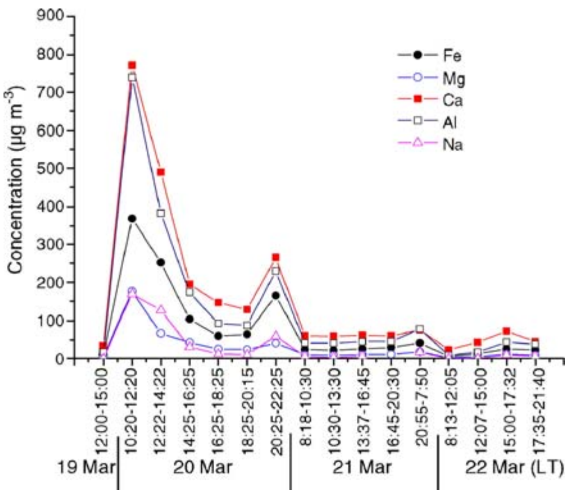

Fig. 7. Concentrations of crustal elements in TSP from 19 to 22 March 2002.

3.4.2. Pollution elements

Dust storms not only carry large amounts of mineral aerosols but also delivered significant amounts of pollution aerosols. The concentrations of major pollution

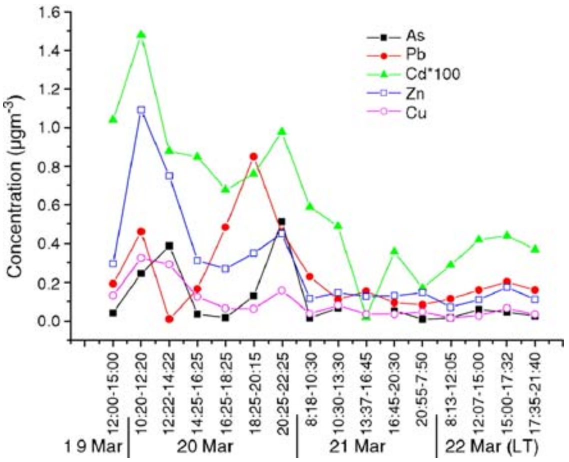

Fig. 8. Concentrations of pollution elements in TSP from 19 to 22 March 2002.

elements, including As, Pb, Cd, Zn, and Cu were presented in Fig. 8. The Zn and Cu generally represent the sources from motor vehicles emission and metallurgical processes (Pacyna, 1986; Lee et al., 1999). However, their variations were similar to that of the crustal elements. They also had the same two peaks as TSP and the mineral elements, with concentrations of 1.1, 0.33 μg m^{-3} and 0.5, 0.16 μg m^{-3} in PI and PII, respectively. The EFs of Zn and Cu were 1.7, 0.6 in PI and 2.3, 0.9 in PII, which were close to 1. These results indicated that most of Zn and Cu were from the crustal source. The concentrations of As, Pb, and Cd, which mostly originated from coal combustion and fossil fuel burning (Borbély-Kiss et al., 1998), also increased during the dust storm. The variations of As and Pb were different from that of crustal elements. The maximum of As of 0.51 μg m^{-3} appeared in PII instead of in PI. Pb reached the maximum 0.85 μg m^{-3} a little earlier than As. The EFs of As, Pb, Cd were 15.2, 3.7, 11 in PI and 101.8, 11.7, 23.4 in PII, respectively. The EFs in PII were much higher than that in PI by a factor of 6.6, 3.2 and 2.1, and the mass percentages of them were also increased in PII by a factor of 4.4, 2.2 and 1.5 compared with that in PI. Furthermore, the pollution contributions of As, Pb and Cd was calculated using a simple formula ($X_{pollution} = 1 - (Al/X)(X/Al)_{crust}$), where $(X/Al)_{crust}$ is the average concentration ratio of X to Al in crust (Taylor and Mclennan, 1985). The pollution contributions accounted for 93%, 72% and 91% of As, Pb and Cd in PI and 99%, 92% and 97% in PII, respectively. These results strongly indicated that As, Pb and Cd in both PI and PII mainly came from pollution sources and there were more pollutants in PII than that in PI. The pollutants in PI were mainly delivered by mixing with dust from source I on the transport pathway. The pollution materials exited in the atmosphere of Beijing also contributed to part of pollutants in PI. However, the dust originated from 'polluted' source that mixed with pollutants resuspended from Beijing local area led the higher content of pollution elements in PII. This mainly attributed to the coal combustion and industrial activities in source II.

It is interesting to note that the characteristics of S, which was much different from other pollution elements in both dust peaks. The enrichment factor of S was very similar in these two peaks while the mass percentage of S was lower in the second one compared with that in the first one, which was the same as those crustal elements. These results associated with the strong correlation (0.97) between S and Al (from 20 to 22 March) indicated that part of S had a crustal source. However, when compared with the crustal abundance 0.035% of S (Taylor and Mclennan, 1985), the high enrichment factor of S and the high mass percentage 0.46% and 0.31% in the two peaks clearly showed that S was affected by pollution sources. The crustal and non-crustal fraction of *S* were calculated by the formulas: $S_{crust} = Al_{total} \times (S/Al)_{crust}$ and $S_{pollution} = S_{total} - S_{crustal}$. In PI and PII, the crustal S was about 3.1 and 1.0 μg m^{-3}, respectively. The non-crustal S was 46.9 and 15.0 μg m^{-3} in PI and PII, which accounted for 93.7% and 93.9% of total S, respectively. If all of S existed in aerosols as the soluble species, i.e., sulfate, the ratio of $[SO_4^{2-}]/S$ should be 3.0. The ratio of 1.8 and 2.3 for sulfate/S in P I and P II indicated that about half of S existed as insoluble species, which was most likely from dust soils in source regions or from the mixing with aerosols containing insoluble S, possibly the combustion residues or the construction dusts from the transport pathway or/and from Beijing local area. For lacking SO_2 data, more work needs to be done to reveal the source and mixing process of the non-crustal S in future.

3.5. Ions of the two peaks

The variations of concentrations of major ions in TSP are shown in Fig. 9. The concentrations of Ca^{2+}, K^+, Mg^{2+}, Cl^-, SO_4^{2-}, and F^- increased significantly in the dust storm. There were also two peaks of these ions, the same as that shown in TSP. The major water-soluble ions were classified into three groups according to their correlation with the aerosol particles and the sources of these ions (Wang et al., 2005a). Ca^{2+}, Na^+ and Mg^{2+} were in the 'crust' group. The concentrations of these ions were 60, 16.3, 4.2 μg m^{-3} in PI and 98.2, 39.3,

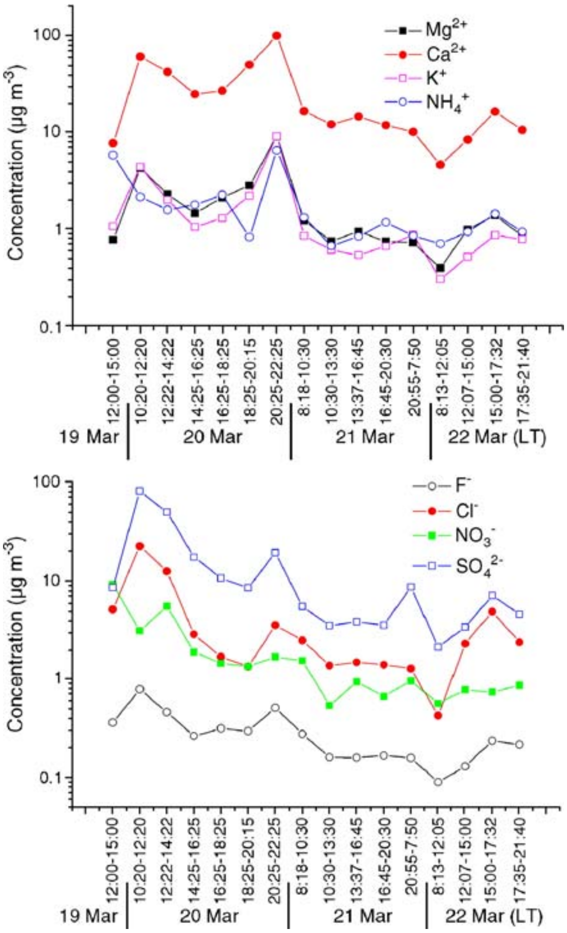

Fig. 9. Concentrations of ions in TSP from 19 to 22 March 2002.

9.0 μg m^{-3} in PII. The mass percentages of Ca^{2+}, Na^{+} and Mg^{2+} increased in PII by a factor of 3.5, 5.1, and 4.5 compared with that in PI, respectively. The local road dust and the dust from cities in source II might contribute to the increase of the 'crust' ions in PII.

SO_4^{2-}, Cl^- and F^- were in the second group. The variations of these ions were similar to that of the TSP and the crustal elements. As shown in Fig. 10, the correlation coefficients of SO_4^{2-}, Cl^- and F^- with Al were 0.99, 0.94, and 0.93, respectively, in this dust storm from March 20 to 22. The strong correlations between these ions with Al indicated that SO_4^{2-}, Cl^- and F^- had, at least partially, the same source as Al, namely, the crustal source. However, the mass percentages of SO_4^{2-}, Cl^- were 0.74%, 0.2% in PI and 0.4%, 0.1% in PII, which were both much higher than that in soils of the Gobi desert (0.01% for SO_4^{2-} and less than 0.01% for Cl^-) and Loess Plateau (0.03% for SO_4^{2-} and 0.02% for Cl^-) (Nishikawa et al., 1991). This indicated that besides the crustal source, there must be other sources of SO_4^{2-} and Cl^- in this dust storm. As mentioned above, the SO_4^{2-} might partially form the adsorption of SO_2/SO_4^{2-} on the dust surface and the mixing of dust aerosol with the pollution aerosols that contained SO_4^{2-} from the pathway in transport and from the local areas of Beijing. The mass percentage of F^- was slighter higher in PII than that in PI. The Cl^- and F^- were mainly associated with the waste incineration and part of the coal burning in winter (Sun et al., 2004a,b; Yao et al., 2002). The different sources and the mixing progresses showed little effect on these ions in these two peaks.

The third group contained NH_4^+, NO_3^-, and K^+. The concentrations of NH_4^+ and NO_3^- decreased significantly when the dust storm arrived at Beijing and the correlation coefficients of NH_4^+/NO_3^- with Al were only 0.17 and 0.26 in this dust storm from March 20 to 22. This result suggested that NH_4^+ and NO_3^- mainly originated from the local pollution sources and diluted by the invaded dust storm. However, the concentrations of NH_4^+ and NO_3^- increased in PII, similarly to that of TSP, and NH_4^+ reached its maximum of 6.5 μg m^{-3} in PII during this dust storm. The mass percentage of NH_4^+ in PII was higher than that in PI by a factor of 6.5. It was clear that over the surrounding regions of Beijing (in source II), such as Hebei Province and Shanxi Province, there are many fields for agricultural cultivation, and the chemical nitrogenous fertilizers, such as carbamide, NH_4HCO_3, NH_4NO_3, and NH_4Cl, are the prevailing fertilizers (Tang et al., 2004). In spring, the dry weather with strong wind could lead these lands to be the seasonal aerosol sources, from which those aerosols originated would contain more ammonium and easily transport to Beijing. Furthermore, the heterogeneous reaction between NO_x and dust is stronger in the morning and night than that at noontime (Tang et al.,

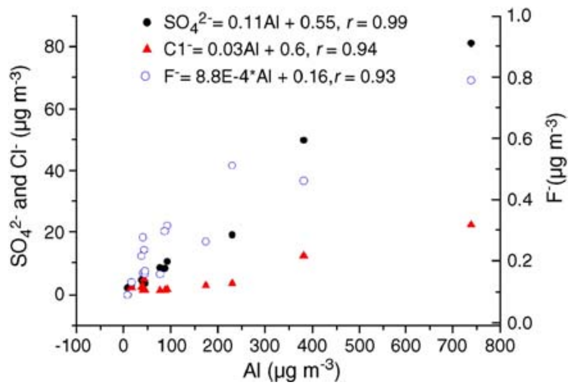

Fig. 10. X (SO_4^{2-}, Cl^- and F^-) vs. Al in TSP samples collected from 20 to 22 March 2002. Correlations (r-values) are significant, $P<0.01$.

2004), which is probably another reason why there was a high concentration of NH_4^+ and NO_3^- in PII in Beijing local area, as it occurred at the night time. The dust originated from the surrounding area mixed with the local dust reacted with NO_x led the increase of NH_4^+ and NO_3^- in PII. The concentration of K^+ was 4.9 and 9.0 μg m^{-3} in PI and PII with mass percentage 0.04% and 1.17%, respectively. The correlation between K^+ and Al was relatively poor with correlation coefficient 0.5. This might indicate that K^+ partly came from crustal source and partly from pollution source. The dust form source II mixed with the resuspended pollutants and the dust from the local area had higher content of K^+ than that from the source I mixed with pollutants on the pathway.

The total ions in these two peaks were also compared at last. The mass concentrations of total ions were 204 and 196 μg m^{-3} in PI and PII and the mass percentages in TSP were 1.9% and 3.8% in PI and PII, respectively. The mass percentages of the water-soluble part of TSP in PII was twice higher than that in PI. These results suggested that the fraction of water-soluble part in the dust, which originated form source II and mixed with the resuspended pollutants and the road dust in the local area, was evidently higher than that in the dust, which came from the source I and mixed with pollutants on the pathway.

4. Conclusions

A super dust storm invaded Beijing on March 20, 2002. The observation of this dust storm from 20 to 22 March 2002 with high-time resolution showed that there were two peaks of TSP of 10.9 and 5.1 mg m^{-3} with 87% and 60% of the mineral dust to TSP, respectively. The dust in these two peaks originated from different sources, which were identified by horizontal visibility and chemical tracers. The dust in PI mainly originated from source I, which included west and middle regions of northern China and the nearby Gobi desert in Mongolia, and the dust in PII was mostly from source II, which mainly included the northeast of China and the southeast of Mongolia. Source I was a relatively 'clean' one and source II was a 'polluted' one. The dust in PI mainly mixed with the pollutants from the transport pathway, and the dust in PII was rich in pollution aerosols, and mixed with the resuspended pollutants and the urban dust from the local area of Beijing. The mixing of the dust aerosol that originated from a relatively 'clean' source with the pollutants on the transport pathway could carry significant amounts of pollutants downwind. The dust, which came from the 'polluted' source and mixed with the local resuspended pollutants, could deliver much higher content of pollutants downwind. Though the second dust peak was weaker than the first one, it would have greater impacts on the human health for the higher fraction of pollution and water-soluble components.

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