Separated status of the natural dust plume and polluted air masses in an Asian dust storm event at coastal areas of China

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[1] Asian dust particles usually refer to mineral particles which originate from arid and semiarid areas in the Asian continent and disperse eastward in a wide range in the atmosphere. Their appearances in the downstream marine areas are always accompanied by high concentrations of sulfate and nitrate, suggesting their significance as a medium for pollutant conversion and transportation. However, analysis of particles collected at a coastal site in east China before and after the cold front arrival of a heavy dust–loading low-pressure system in this study revealed that air masses polluted by anthropogenic emissions from local or regional areas and the natural Asian dust plume from northwest China were in different air parcels. Anthropogenic pollutants were in the prefrontal air while the so-called Asian dust particles were in the postfrontal air. There was a large amount of anthropogenic mineral particles in the polluted air masses, and the particles were abundant in sulfur, sodium, and calcium, while the so-called Asian dust particles were very "clean" (meaning lack of anthropogenic pollution). Continuous records of PM_{10}, SO_2, and O_3 concentrations indicated that the cold front was the boundary layer between the polluted air masses and the clean dust plume. Hence, at the coastal areas the Asian dust plume and polluted air masses did not mix with each other and were separated in two consecutive air parcels by the cold front although they were in the same low-pressure system. In addition, the analysis also revealed that calcium was not a good indicator for Asian dust from natural sources because the anthropogenic mineral particles contained significant calcium.

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1. Introduction

[2] Asian dust frequently transports to coastal areas of China, Japanese archipelago, the northern Pacific Ocean, and sometimes to North America [*Duce et al.*, 1980; *Uematsu et al.*, 1983; *Iwasaka et al.*, 1988; *Husar et al.*, 2001]. In addition to gravitational settlement, dust particles usually experience various variations such as surface uptake of ambient gaseous species and coagulations with other particles during their long-range transport from source areas to remote marine atmosphere. It has been realized that Asian dust may have significant influences on global geochemical mass cycles, atmospheric radiation transfer and marine ecosystem [*Intergovernmental Panel on Climate Change*, 2001; *Martin et al.*, 1989; *Bishop et al.*, 2002]. To evaluate these potential influences, physical and chemical variations of dust particles during their long-range dispersion, in particular their interaction with anthropogenic pollutants, need to be identified.

[3] Asian dust plumes are initially stimulated by cold fronts of low-pressure systems (cyclone systems) in northwestern and northern China and parts of Mongolia, and move eastward following the fronts [*Fang et al.*, 1997]. As they arrive at eastern and northeastern China, where significant anthropogenic pollutants are constantly emitted into the atmosphere [*Akimoto and Narita*, 1994; *Guttikunda et al.*, 2003; *Streets et al.*, 2003], the dust plumes meet polluted air masses. Then the plumes and polluted air masses move out of the continent. Observations at Korea, Japan and the northern Pacific revealed that Asian dust could enhance particulate sulfate and nitrate formation [*Prospero et al.*, 1985; *Kim et al.*, 1998; *Kanamori et al.*, 1991]. Similar enhancement was not found at and near the dust source regions [*Okada and Kai*, 1995; *Trochkine et al.*, 2003], which is consistent with the fact that there are not anthropogenic emissions in synoptic scales there [*Streets et al.*, 2003]. However, observations at Beijing and coastal areas of China, where dust plumes were expected to have

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Table 1. Summary of Sampling Time and Weather Conditions

Sampling Time					Wind			
Date	Period, BST[a]	Weather	Temperature, °C	Relative Humidity, %	Direction	Speed, ms^{-1}	Visibility, km	Remarks
20 March 2002	1400–1500	fog	13.8	85	SSW	6.0	~5	prefrontal
21 March 2002	1000–1100	dust	8.5	53	W	3.5	~4	postfrontal
19 Feb. 2002	1000–1100	clear	3.5	59	NW	2.5	~15	nondust

[a]BST: Beijing Standard Time (UTC plus 8 hours).

met anthropogenic polluted air masses, indicated that dust particles were not apparently influenced by anthropogenic pollutants [*Zhang and Iwasaka*, 1999; *Zhang et al.*, 2003]. These results leave a question: what are the mutual states of dust plumes and polluted air masses in eastern China before the plumes leave the continent? In respect to that cold and dry air masses and warm and humid air masses in low-pressure systems are usually separated by the cold fronts of the systems and prefrontal warm air masses are pushed forward or lifted upward by postfrontal cold air masses as the systems move from west to east, it seems that dust plumes behind cold fronts and polluted air masses before cold fronts cannot easily mix with each other in the continental atmosphere. A recent study of continuous records of PM_{10} and anthropogenic air pollutants at Beijing by *J. Guo et al.* [2004] showed that atmospheric dust loads anticorrelated to anthropogenic pollutants during dust events. *Z. Guo et al.* [2004], after analyzing the integrated $PM_{2.5}$ aerosol samples of Asian dust episodes at Qingdao, reported that high concentrations of local pollutants appeared before the arrival of long-distance transported materials, showing the possibility that dust plumes and anthropogenic pollutants were not in the same air parcels. Despite these facts, a number of numerical model studies have been conducted with the assumption that dust particles and pollutants mixed homogeneously. Therefore it is necessary to make clear whether and how Asian dust particles mix with anthropogenic pollutants in the continental atmosphere and take them to downstream areas.

[4] In this paper, status of the dust plume and polluted air masses in a dust-loading low-pressure system at coastal areas of China is described and discussed on the basis of the analysis of mineral particles which were collected at a coastal site in eastern China in two periods when the system passed there. Continuous monitoring records of air quality near the site are also included to show the evolution of air pollution during the event. Analysis results of mineral particles from a nondust case study are included for comparisons of long-range transported particles and local particles.

2. Particle Collection and Analysis

[5] A severe dust storm occurred in east Asia from 18–23 March 2002 (http://info.nies.go.jp:8094/AsiaNet/). Samples were obtained at Qingdao, a coastal city in the eastern China, on 20–21 March when the event passed there. Nondust samples were collected on 19 February 2002, when no dust was reported. The observation site was on the roof of a 35-m building of the First Institute of Oceanography of the State Oceanic Administration (36°06′N, 120°27′E), which is located at the coastal line near the Yellow Sea. Particles were collected onto electron microscope Ti grids (3 mm diameter) using a single-stage cascade impactor. The pump rate was 5 l min^{-1}. The 50% cutoff diameter of the impactor is 0.25 μm at this rate if the density of particles is 2.3 g cm^{-3}. The collection time for each grid was about 2 min. After collection, each grid was reserved in a plastic capsule, which, in turn, was sealed in plastic bags together with paper-packaged silica gel. During each observation campaign, three Ti grids were applied to collect particles and the time discrepancy between two samples was about 15 ~ 20 min. Table 1 lists the sampling time and weather conditions at the site when particles were collected.

[6] Particles on the grids were investigated and photographed using the scanning electron microscope (SEM; Hitachi S-3000N) of the Solar Terrestrial Environment Laboratory of Nagoya University for the observation of particle morphology and size. Photographs covered the central regions where most particles were captured on the grids. After a photograph was taken, the elemental composition of every particle in the photograph was determined using the energy dispersive X-ray (EDX) spectrometer (Horiba EMAX-500) attached to the SEM. With this SEMEDX system, we are able to quantitatively detect the relative weight and atom ratios of elements with atom number $Z > 5$ except nitrogen in a single particle down to 0.1 μm. However, we did not estimate the relative weight ratios of carbon (C) and oxygen (O) in this study since their contents were always largely overestimated by the system. The SEM was operated at 20 keV accelerating voltage and 80 μA filament current. The X-ray spectrum of a particle was generated from a square covering the particle and was integrated for 50 live seconds with probe current around 0.3 nA. The atom number fractions and weight fractions of different elements in individual particles were calculated using ZAF (Z: element atomic number; A: X-ray absorption; F: X-ray fluorescence) matrix correction. To avoid underestimating the content of sodium (Na), low-energy correction was not applied in the ratio calculation, which led to the ratios of Na were somewhat overestimated. In terms of elemental composition, mineral particles were those mainly containing silicon (Si) together with/without other mineral elements, such as aluminum (Al) and iron (Fe).

3. Weather and Trajectories

[7] Figure 1 shows the surface weather charts around the sampling time and 3-hour temperature variations recorded at Qingdao Meteorological Observatory from 800 BST on 20 March to 2000 BST on 21 March 2002. The observatory is located about 10 km west of the observational site and in the urban areas of Qingdao. The temperature variations indicate that the cold front causing the dust storm passed the observatory between 1400 and 1700 BST on 20 March. The weather charts indicate the particle collection on 20 March was carried out just before the cold front arrived

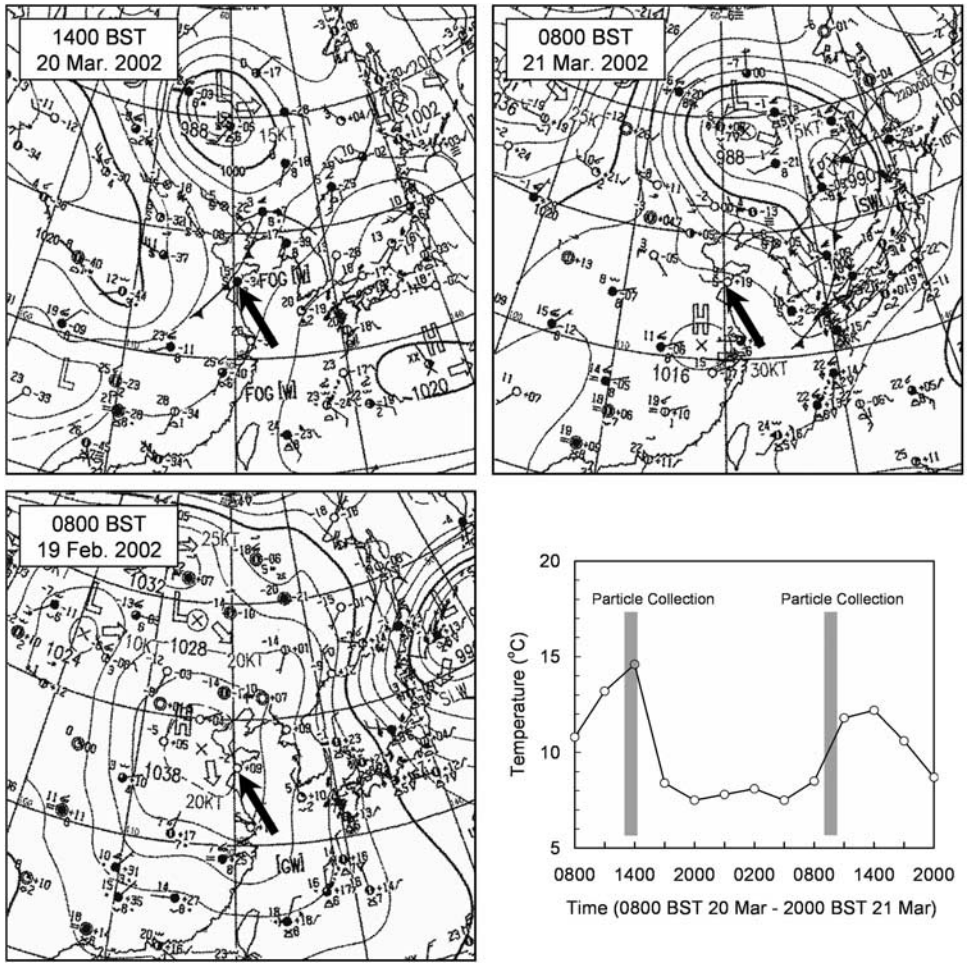

Figure 1. Surface weather charts when particles were collected and the 3-hour temperature variations from 800 BST on 20 March to 2000 BST on 21 March 2000 (BST is UTC plus 8 hours) recorded at the Qingdao Meteorological Observatory, which is located about 10 km west from the particle collection site. Qingdao is marked by arrows in the surface charts. Durations corresponding to the air masses in which particles were collected are indicated by shaded bars. The durations were estimated with the assumption that the cold front moved at a speed of 5 m s^{-1} (see Table 1) from the monitoring station to the particle collection site.

at the observational site and on 21 March the whole Qingdao area was covered by the postfrontal air. Thus particles collected on 20 March were from the prefrontal air and particles collected on 21 March were from the postfrontal air. As shown in Table 1, the prefrontal air was warm and humid while the postfrontal air was cold and dry, further confirming that the particles were collected in different type air parcels.

[8] Figure 2 shows the isentropic backward and forward trajectories for the three observation cases, which were calculated online using the NOAA HYSPLIT model at http://www.arl.noaa.gov/ready/hysplit4.html. In the case of the dust event, the postfrontal air came from the northwestern China in the west, where are the main source regions of Asian dust. However, the prefrontal air came from the eastern areas of China in the south, where no Asian dust sources are anticipated [*Duce*, 1995]. Both prefrontal and postfrontal air moved fast when approaching Qingdao, and then they moved eastward out of the continent consecutively as in one dust pall. On 19 February, Qingdao was in a weak high-pressure system (Figure 1) and the weather was clear (Table 1). The trajectory indicates that the air masses on that day moved slowly. Thus particles collected in that case should be from local areas.

4. Results and Discussion

[9] Figure 3 illustrates examples of electron microphotographs of particles collected on Ti grids on 20 and 21 March together with X-ray spectra of particles marked in the microphotographs. A large number of particles which had irregular shapes were captured in both the prefrontal (20 March) and postfrontal air (21 March) and their X-ray spectra indicate most of them were mineral particles. Since the postfrontal air came from Asian dust source areas, mineral particles enclosed in the air were believed to be the so-called Asian dust particles. However, mineral particles in the prefrontal air should be originated from local or regional anthropogenic emissions because no significant natural sources of mineral particles were expected in east China.

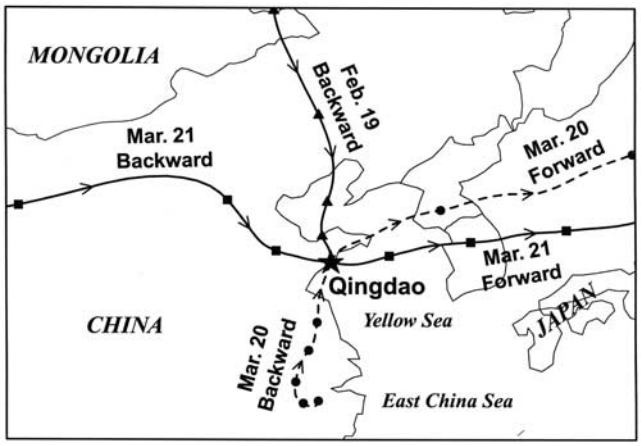

Figure 2. Isentropic backward (19 February, 20 March, and 21 March) and forward (20 March and 21 March) trajectories. The calculation was performed online by the HYSPLIT model of NOAA Air Resources Laboratory's Web server at http://www.arl.noaa.gov/ready/. The trajectories start at 500 m above the sampling site and from the sampling time. Arrows on the trajectories indicate moving directions of the air parcels, and squares, dots, and triangles indicate 12-hour increments for each case.

[10] In order to compare mineral particles in the prefrontal and postfrontal air, number fractions of collected mineral particles containing different elements in the prefrontal and postfrontal air and in the nondust case were estimated. The results are shown in Figure 4. The number fractions of mineral particles containing different elements in the postfrontal air (21 March) were approximately at the same levels of dust particles previously observed at dust source areas, Beijing and Qingdao [*Iwasaka et al.*, 2003; *Zhang and Iwasaka*, 1999; *Zhang et al.*, 2003], suggesting the postfrontal particles did not change apparently yet during their transport to the coastal areas. In other words, the Asian dust particles had not been modified or interacted by anthropogenic pollutants apparently yet. On the contrary, the fractions of particles containing C, Na, sulfur (S) and calcium (Ca) in prefrontal mineral particles (20 March) were much larger than those in postfrontal particles, and even quite larger than those of nondust case except C-containing ones. Particles containing Mg, phosphor (P), chlorine (Cl), potassium (K) and Fe, had similar features but no significant differences. Therefore the prefrontal particles originally contained or were heavily polluted by anthropogenic pollutants such as sulfur- and carbon-containing compounds, which is consistent with that the prefrontal and postfrontal particles had different sources.

[11] A possible source for the Na, S and Ca in the prefrontal particles was sea salt because the prefrontal air masses passed ocean areas just before arriving at Qingdao (Figure 2). In order to investigate sea salt contributions, the weight ratios of Na:S:Cl in the particles were compared with that of fresh sea salt (Figure 5a). Na:S:Cl in most particles was quite different from that of fresh sea salt. Many particles contained extra S even assuming that Na in the particles was totally from sea salt (particles of (S:Na) >

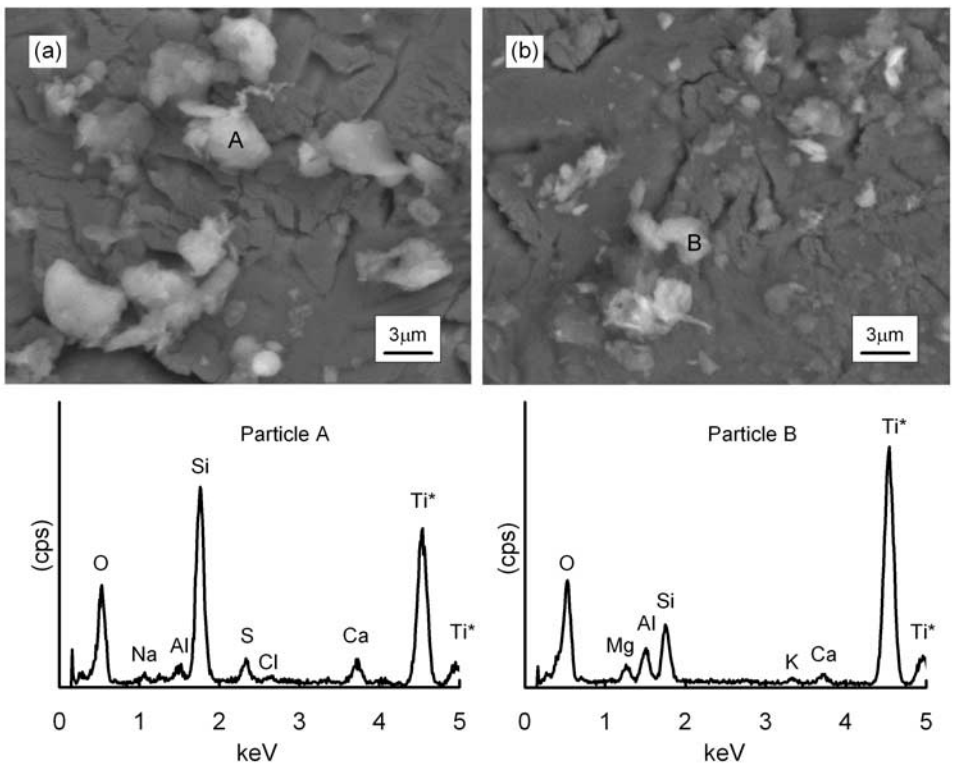

Figure 3. Electron microphotographs of particles collected on Ti grids (a) on 20 March and (b) on 21 March and the EDX spectra of marked particles in the photographs. Peaks of Ti in the spectra were caused by the grids.

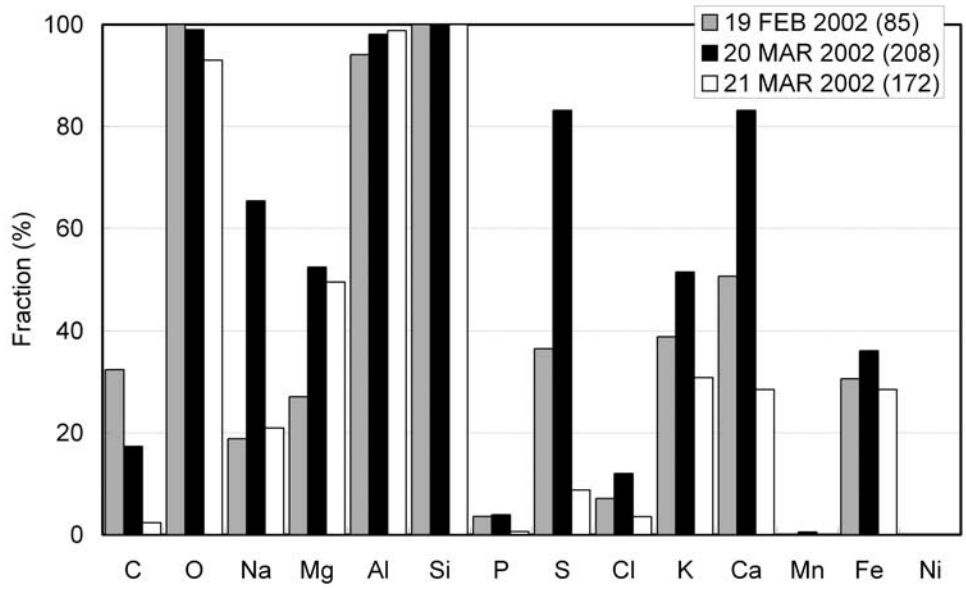

Figure 4. Number fractions of particles containing different elements in detected mineral particles. Numbers in parentheses are the numbers of detected mineral particles in each case.

$(S:Na)_{Na2SO4}$: they are in the left side of line Na_2SO_4–sea salt in the triangle of Figure 5a). A similar investigation of Na:S:Ca in the particles (Figure 5b) revealed that a large number of prefrontal particles contained significant non-sea salt Ca. Therefore S and Ca in the prefrontal particles must have other substantial sources besides sea salt. A number of particles appear on the line of Cl = 0 in Figure 5a, and this is also the reason why the number fraction of Cl-containing mineral particles in the prefrontal air was much smaller than that of Na (Figure 4). If Na in these particles was from sea

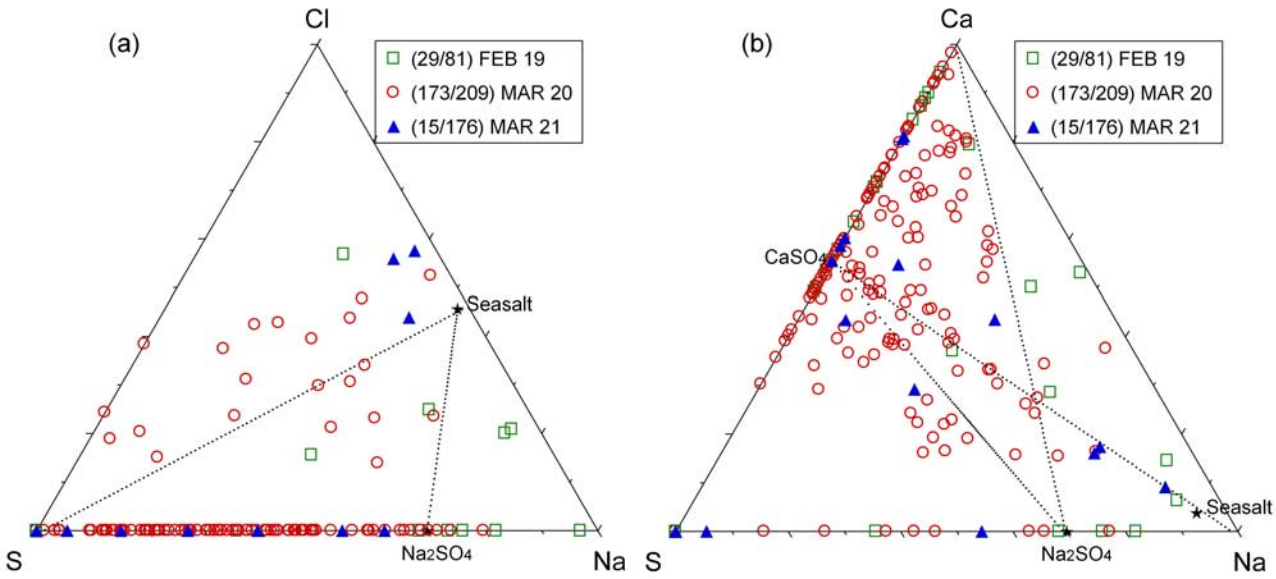

Figure 5. Weight ratios of (a) Cl:Na:S and (b) Ca:Na:S in S-containing mineral particles. The stars show the positions of fresh sea salt particles, Na_2SO_4, and $CaSO_4$ identified by the scanning electron microscope energy dispersive X-ray spectrometer under the same analysis conditions. In Figure 5a the dotted line of sea salt-Na_2SO_4 shows the positions of Na:S:Cl in sea salt when the ratio is presumably changed only by the reaction of $2NaCl + H_2SO_4 \rightarrow Na_2SO_4 + 2HCl(g)$, and ratios of Na/Cl on the dotted line of sea salt-S are the same as that in fresh sea salt. In Figure 5b, ratios of Na/S on the dotted line of Ca-Na_2SO_4 are the same as that of Na_2SO_4, ratios of Ca/S on the dotted line of Na-$CaSO_4$ are the same as that of $CaSO_4$, and the dotted line of Na_2SO_4-$CaSO_4$ shows that S exists as either Na_2SO_4 or $CaSO_4$. Data in the parentheses are (number of S-containing mineral particles)/(number of detected mineral particles) in each case.

salt, the sea salt should have combined with the anthropogenic mineral particles and Cl originally contained in the sea salt must have been completely displaced. S contents actually could compensate Cl loss in particles of (S:Na) > $(S:Na)_{Na2SO4}$ in Figure 5a if Na was totally from sea salt. Weather records showed that it was fog at Qingdao, and the relative humidity was very large (85% in Table 1) on 20 March, which favored heterogeneous conversions of S from SO_2 to sulfate on particles surface. However, it is questionable if displacing reactions could be so efficient as to have caused the complete loss of Cl before or after the sea salt combined with the mineral particles because the prefrontal air met marine air masses only in the latest several hours (Figure 2).

[12] Another possibility is that Na had Cl-free sources. From comparisons of Figure 5a and Figure 5b, it can be found that most Cl-free S-containing particles contained Ca. S content could be consumed by Ca and/or Na in the forms of $CaSO_4$ and Na_2SO_4 in most S-containing particles (particles in the Quadrangle Ca-$CaSO_4$-Na_2SO_4-Na in Figure 5b) and only a small number of the particles contained extra S that could not be compensated by Ca and Na in the above forms (particles in the Triangle S-$CaSO_4$-Na_2SO_4 in Figure 5b). Furthermore, a large number of the particles contained extra Ca even assuming that all S in the particles was in the form of $CaSO_4$ (particles in the Triangle Ca-$CaSO_4$-Na in Figure 5b). It is hard to believe that S deposited onto the particles choosing NaCl to react to have led to the complete loss of Cl in the particles while ignoring the existence of Ca. So the reasonable interpretation should be that Na had Cl-free sources in east China. The Na might be from surface soil or coal burning because it appeared mainly in particles abundant in mineral elements. Unfortunately, we are unable to identify the sources by the present data and further studies are necessary.

[13] To investigate the origin of Ca, its weight ratios were compared with those of S and mineral elements of Al, Si, and Fe in Ca-containing particles. Al, Si and Fe were chosen as indicators of crustal contributions. This was because the number fractions of Al- and Fe-containing mineral particles in the prefrontal, postfrontal and local mineral particles were approximately same besides Si (Figure 4). Figure 6 shows (Al + Si + Fe):S:Ca in weight and the relative weight ratios of Al + Si + Fi, S and Ca in detected Ca-containing particles, where Al + Si + Fi is the sum of relative weight ratios of Al, Si and Fe in an individual particle. It should be noted that only Ca-containing particles, most of which were mineral particles, appeared in Figure 6 and the number fraction of Ca-containing mineral particles in the postfrontal air were much less than that in the prefrontal air (Figure 4). In contrast to that postfrontal Ca-containing particles rarely contained S, most prefrontal particles and many local particles were S carriers (Figure 6). (Al + Si + Fe):S:Ca and the relative ratios of Al + Si + Fi, S and Ca in the prefrontal ones were similar to those of local particles except that more prefrontal particles contained S. Further, Ca and S were more likely in the form of $CaSO_4$ with the increase of Ca weight ratios in a number of the particles (particles along or close to the line $CaSO_4$-(Al + Si + Fe) in Figure 6a). These results indicate that Ca in the prefrontal particles was closely related to S and significantly contributed from noncrustal origins. Several recent measurements of particles in urban atmosphere in China revealed possible large emissions of anthropogenic Ca from construction sites where gypsum was well applied [*He et al.*, 2001; *Zhang et al.*, 2002] and analyses of winter PM_{10} samples of Shanghai (a megacity in about 500 km south of Qingdao) revealed the significant contributions of anthropogenic S and Ca [*Zheng et al.*, 2004]. Therefore the abundance of Ca in the prefrontal particles also indicates that the prefrontal air contained significant anthropogenic pollutants. Consequently, the remarkable difference of Ca content in prefrontal and postfrontal particles further sup-

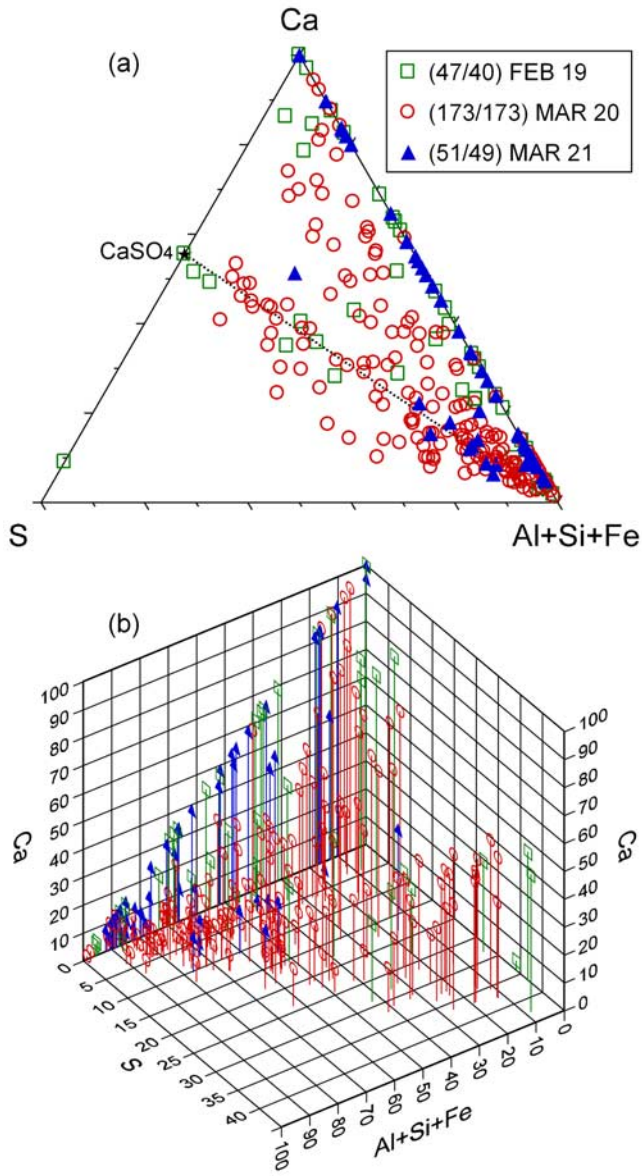

Figure 6. (a) Weight ratios of Ca:(Al + Si + Fe):S in Ca-containing particles and (b) relative weight ratios of Ca, Al + Si + Fe, and S among the detected elements in the particles. In Figure 6a the star shows the position of $CaSO_4$, and ratios of Ca/S on the dotted line of (Al + Si + Fe)-$CaSO_4$ are the same as that of $CaSO_4$. Data in the parentheses are (number of Ca-containing particles)/(number of Ca-containing mineral particles).

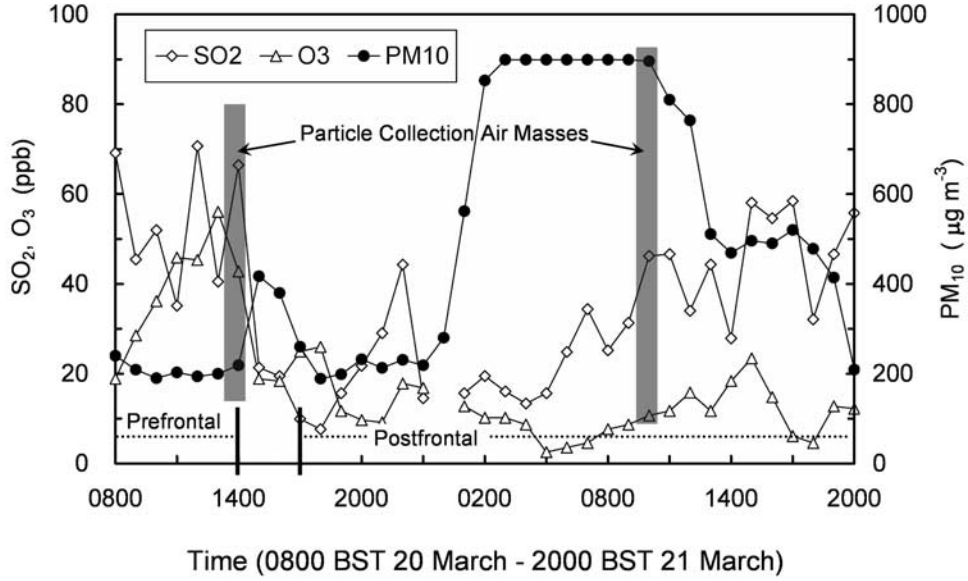

Figure 7. Variations of PM_{10}, SO_2, and O_3 concentrations recorded at the Sifang Air Quality Monitoring station from 800 BST on 20 March to 2000 BST on 21 March 2000 (BST is UTC plus 8 hours). The station is located about 11 km west from the particle collection site. The cold frontal passed the station between 1400 and 1700 BST (see temperature records in Figure 1). Periods when prefrontal air and postfrontal air passed the station are shown by straight dotted lines. The air masses in which particles were collected are marked by shaded bars, which were estimated as in Figure 1.

ports that the prefrontal air masses had been heavily polluted by anthropogenic emissions while the postfrontal dust plume was not.

[14] A speculative interpretation for the prefrontal mineral particles is that they were construction and road dust, fly ashes and/or their mixture from local and regional emissions. They were emitted into the prefrontal air parcel as the air moved northward from south to Qingdao and probably had experienced mutual interactions and modifications before arriving at there. East China is a region with huge populations, large numbers of construction sites and vigorous industrial activities. Coal is the major energy source there, and large amounts of fly ashes and SO_2 are being emitted from power plants and heating systems [*International Energy Agency*, 1999]. Note that major elemental components of fly ashes from coal burning are similar to surface soil and dust particles. Soot particles from coal burning are usually aggregates of fragments and chains of small spheres or their mixture. Such particles were frequently detected in the prefrontal particles and examples could be found in Figure 3a. The differences of number fractions of carbon-containing mineral particles in the prefrontal and postfrontal particles (Figure 4) also support this idea. In addition, we did find Se, Pb, Te, As and Rh of trace metals for pollutants in a few prefrontal particles while these elements were rarely found among the postfrontal particles.

[15] The above results proved that the prefrontal and postfrontal mineral particles were remarkably different. Since the particles were collected during two short periods in the prefrontal and postfrontal air when they passed the observational site, a remained question is where was the boundary between the air parcels containing the two kinds of mineral particles as the dust storm event passed Qingdao. In order to answer this question, we checked the continuous records of the concentrations of PM_{10}, sulfur dioxides (SO_2) and ozone (O_3), which were measured at Sifang Air Quality Monitoring Station of Qingdao Environment Agency. The monitoring station is located about 11 km west of the observational site and near the meteorological observatory. PM_{10} was measured by a GBAM-1020 monitor of Met One Instruments Incorporation, SO_2 by an API 100A Sulfur Dioxide Analyzer and O_3 by an API 400A Ozone Analyzer manufactured by Advanced Pollution Instrumentation Incorporation. Figure 7 shows the concentrations of PM_{10}, SO_2 and O_3 concentrations at the station when the dust storm event passed Qingdao areas. PM_{10} concentration was about 200 $\mu g\ m^{-3}$ in the prefrontal air and increased to about 400 $\mu g\ m^{-3}$ just before the arrival of the front, indicating that the air carried a large amount of particulate matters. This is consistent with the result that a large number of mineral particles were detected in the prefrontal particle samples. SO_2 and O_3 concentrations were also in high levels. Thus the whole prefrontal air contained not only mineral particles but also gaseous species such as SO_2, confirming that the air was heavily polluted by anthropogenic pollutants. With the arrival of the cold front, PM_{10} concentration rapidly increased up to about 900 $\mu g\ m^{-3}$ from 200 $\mu g\ m^{-3}$ while SO_2 and O_3 concentrations sharply decreased, suggesting that the cold front carried dust to arrive and the air masses with heavy anthropogenic pollutants were replaced by air masses with less anthropogenic pollutants. The arrival of the front eventually led to the increase of the concentration of particulate matters, which was due to the dust plume in the postfrontal air. This process was similar to that of the same dust event when it passed Beijing described by *J. Guo et al.* [2004]. These data revealed that the remarkable change of anthropogenic pollutant concentrations coincided with the movement of the cold front, suggesting that the cold front was the

boundary of the polluted and unpolluted air masses. This further indicates that the prefrontal and postfrontal mineral particles should be considered separately.

[16] The scenario of synoptic scale movements of the dust plume and polluted air masses should have been that, the dust plume moved eastward following the cold front while the polluted air masses moved northeastward before the front. This is normal in view of meteorology. The northeastward movement of the polluted air masses was due to the force of the cold front and the convergence of air from the south. This can be evidenced by the presence of a warm front in the north of the prefrontal air after 20 March when the prefrontal warm air met the cold air in the north (see Figure 1). With the NOAA HYSPLIT model, we have confirmed the above scenario at different stages when the cold front was approaching Qingdao.

5. Summary

[17] In the observed dust storm event, the so-called Asian dust particles from northwest China were in the postfrontal air and anthropogenic pollutants including significant mineral particles from east China were in the prefrontal air at the coastal area. The dust plume and the polluted air masses were separated by the cold front. Although this result is from one dust storm event, such phenomena is possibly a usual status of dust plumes and polluted air masses or frequently occur at the coastal areas of China with respect to structures of low-pressure systems. Recent studies of dust storm particles collected at the same site in postfrontal air of three dust events revealed that the particle compositions were always similar to the postfrontal mineral particles of this study and they rarely showed polluted symptoms [*Zhang et al.*, 2003]. We confirmed by trajectory analysis that the movements of postfrontal dust plumes and prefrontal air masses were similar to that shown in Figure 2 in most dust events in east Asia since 1998.

[18] An important implication of the above results is that Asian dust storm particles and anthropogenic pollutants leave the continent separately in different air parcels. *Matsumoto et al.* [2003] reported frequent time discrepancies between the arrival of dust particles and anthropogenic pollutants in northeastern Japan. This was also found in a surface observation at southwestern Japan [*Uematsu et al.*, 2002], aerial measurements over the East China Sea [*Hatakeyama et al.*, 2004] and an ACE-Asia ship cruise around southern Japan [*Bates et al.*, 2004]. In addition, the time discrepancies were testified by regional transport model simulations by *Matsumoto et al.* [2003] and *Bates et al.* [2004]. These results suggest that the separated status of dust plumes and polluted air masses could last more than several hours after they left the continent. Although more case data are necessary, it can be concluded from this study that the mixing of dust plumes and polluted air masses in east Asia should not be regarded as a homogeneous process. Mineral particles in prefrontal and postfrontal air of Asian dust events in the continental atmosphere should be treated separately in the investigation of their physical and chemical properties. It could be consequently expected that formation of pollutants such as particulate sulfate and nitrate in the continent atmosphere should be mainly enhanced by anthropogenic mineral particles rather than natural dust.

[19] In addition, the anthropogenic mineral particles in the prefrontal air were abundant in Ca. PM_{10} concentration in the air was in the range of $200 \sim 400\ \mu g\ m^{-3}$, which was a level comparable to weak dust events in China. If Ca were applied as the indicator of natural dust, the prefrontal mineral particles would have been attributed to Asian dust from nature, which is apparently incorrect in the dust event analyzed in this study. Therefore Ca is not always a good indicator of Asian dust. Weather conditions and other indexes should be considered.

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