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A mechanism for the increase of pollution elements in dust storms in Beijing

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Abstract

It is well known that springtime dust storms in northern China are created by strong winds associated with cold fronts. One unexpected aspect of the dust, observed recently, is that it contains high concentrations of pollution elements. It is believed that dust particles were mixed well with pollution substances during the transport, thus the pollution elements are thought to be pollution derived. A careful examination of concentrations of elements, pollution gases, and meteorology during March 2001 and March 2002 in Beijing showed that the dust itself was an important source for some pollution elements. A more important by-product was the finding that dust storms in Beijing typically contain four stages: (1) accumulation of pollutants in the atmosphere of Beijing; (2) clear-out of pollutants; (3) addition of pure dust; and (4) clear-out of the dust. The pure dust can appear before the accumulated pollutants are cleared out, which mixes the two and makes it appear that the pollutants came from it. Pollution gases and PM_{10} also reveal the same four stages. When PM10 varies inversely with the pollution gases, as it does during dust storms in Beijing and occasionally in Shanghai, it includes transported dust. This pattern makes it possible to study dust storms by examining the longer records of pollution gases and PM10 that are now being generated by municipal environmental bureaus throughout China.

Keywords: Dust storm; Pollution element; Beijing aerosol; Source; Air mass

1. Introduction

Dust storms are frequent in northern China during spring, and typically invade Beijing in March and April (Liu et al., 1981; Gao et al., 2002). Scientists have been studying them since the 1970s with chemical, physical, and meteorological techniques. These efforts have led to a good understanding of the effect of Asian dust on the global biogeochemical cycle (Duce et al., 1983; Zhuang et al., 1992). They are generated over dry/semi-dry areas (including deserts) by strong winds near cold fronts (Gao et al., 2000; Husar et al., 2001) and are transported to eastern and southeastern China (Zhuang et al., 2001; Zhang et al., 2000; Lin, 2001; Murayama et al., 2001), to Korea (Yi et al., 2001; Chun et al., 2001), to Japan (Ma et al., 2001; Murayama et al., 2001, Uematsu et al., 2002), and sometimes even to North America (Perry et al., 1999; Tratt et al., 2001; Husar et al., 2001). The dust clouds are transported in layers, particularly when longer distances are involved (Tratt et al., 2001; Murayama et al., 2001; Husar et al., 2001). As the dust storms reach Beijing, their coarse particles often peak hours before their fine particles do (Zhang et al., 2000). Together with the dust come strong winds, cold and dry air, and high concentrations of particulate pollutants (Zhuang et al., 2001).

We were puzzled about the relations between the dust and the pollutants, particularly in cases where high concentrations of certain particulate pollutants

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appeared with the highest dust. Had the pollutants come from their usual sources, or had they somehow been made airborne along with the dust, either as natural enrichments in dust or as previously deposited pollutants resuspended with local dust in Beijing? We tried to resolve this problem by carefully examining the elemental data from a series of aerosol samples taken before, during, and after major dust storms in Beijing in March of 2001 and 2002. We supplemented those data with pollution gases and local meteorological variables.

This work has placed Chinese dust storms into a more general meteorological picture. It is now clear that as the air masses over Beijing alternate between northern and southern, much as they do at other midlatitude sites, occasional strong winds behind the cold fronts raise significant quantities of dust that are then carried southeastward as dust storms.

2. Sampling and analysis

We collected daily TSP (particle diameter smaller than 100 μm) samples between June 2000 and June 2001, and more samples on days of dust storms. On 20 and 21 March 2002, a strong dust storm passed Beijing. We took several TSP samples before, during, and after it, all from the roof of the Science and Technology Building of Beijing Normal University, 12 stories above ground (~40 m). The samples were taken on a medium-volume aerosol sampler (model (TSP/PM10/PM2.5)-2) made by Beijing Geological Instrument-Dickel Co., Ltd., with 90-mm Whatman 41 cellulose filters at flow rates of $100\,\mathrm{l\,min^{-1}}$. Because the aerosol concentrations were very high and these filters clog rapidly, most samples had to be limited to 2–4 h.

The samples were dissolved at 170°C for 4 h in 3 ml of concentrated nitric acid, 1 ml of concentrated perchloric acid, and 1 ml of concentrated hydrofluoric acid, then cooled and oven-dried. One milliliter of concentrated HNO_3 was added to each, and the sample was diluted to 10 ml with deionized water. The samples were then analyzed by ICP-AES (Model ULTIMA, JOBIN-YVON Company, France). Seventeen elements were measured. This paper limits itself to the variations of S, Cu, Zn, and Al.

We collected hourly meteorological data for March–May 2002 from Weather Underground (http://www.wunderground.com/).

As of July 2000, each city in China is required to publish an Air Pollution Index (API, API = 100 corresponds to Chinese Air quality standard II) measured for the urban region each day. The observations comprise NO_x, SO_2, and TSP (in some cities, PM10), and in some cases CO and O_3. API data for SO_2 and PM10 were collected for Beijing and Shanghai and converted to concentrations so as to examine their relations with dust storm.

3. Results and discussion

3.1. Stages of dust storms revealed by elemental data

3.1.1. Stages in the super dust storm of March 2002

On 20 and 21 March 2002, an unusually strong dust storm invaded Beijing, the strongest one in a decade. Its TSP reached $11{,}000\,\mu\mathrm{g\,m^{-3}}$. The sky was covered by reddish dust, 3×10^{10} g of which settled onto Beijing (Xinhua News Agency, 22 March 2002). The National Environmental Protection Bureau of China reported that this was a floating-dust event transported from the west at a relatively high altitude.

Al was chosen as the indicator element for the crustal dust. Its concentration increased sharply from $\sim20\,\mu\mathrm{g\,m^{-3}}$ on 19 March (point 19 in Fig. 1) to $570\,\mu\mathrm{g\,m^{-3}}$ in the first sample of 20 March (point 20^{a}), the highest concentration we have seen in Beijing. This marked the abrupt arrival of the dust cloud. The concentration dropped to $70\,\mu\mathrm{g\,m^{-3}}$ in the evening (points 20^{d}, 20^{e}) and then rose to $180\,\mu\mathrm{g\,m^{-3}}$ in the last sample of 20 March (point 20^{f}) as the wind speed dropped to its lowest value of the day (Section 3.3.2). We are not sure what made the Al rise with decreasing wind speed during the night, but we saw the same pattern the next day, when Al rose from $30\,\mu\mathrm{g\,m^{-3}}$ during the day (points $21^{a\text{–}d}$) to $60\,\mu\mathrm{g\,m^{-3}}$ at night (point 21^{e}). By the end of the 21st, the dew point was beginning to increase, which showed that the new air mass was settling in (Section 3.3.2). On 22 March (point 22^{a}), Al began low ($6\,\mu\mathrm{g\,m^{-3}}$) and increased to $30\,\mu\mathrm{g\,m^{-3}}$ (points $22^{c,d}$). From 23 to 25 March (points 23–25), Al remained around $30\,\mu\mathrm{g\,m^{-3}}$. The dust event was considered to be over on 21 March, and 22–25 March were considered to be "normal" spring days in Beijing.

The rise of the "crustal" element Al after the dust storm shows that it also has pollution sources. To discriminate between the types of Al (and of inorganic aerosol) we use the ratios of pollution elements to Al (X/Al). When these ratios are high, the elements involved are mostly pollution derived; when they are low and close to crustal ratios, the elements (and probably much of the inorganic aerosol) are crustal. We used Cu, Zn, and S as indicators of pollution. Their concentrations are plotted against Al in the three parts of Fig. 1. Ratios of the elements to Al in Inner Mongolia loess are shown there as well.

Consider first the data for Cu in Fig. 1a. Its concentration began near $0.1\,\mu\mathrm{g\,m^{-3}}$ on 19 March (point 19). Its crustal contribution at the time, $0.007\,\mu\mathrm{g\,m^{-3}}$ (calculated from rock, Turekian, 1971), meant that more than 99% of the Cu was from

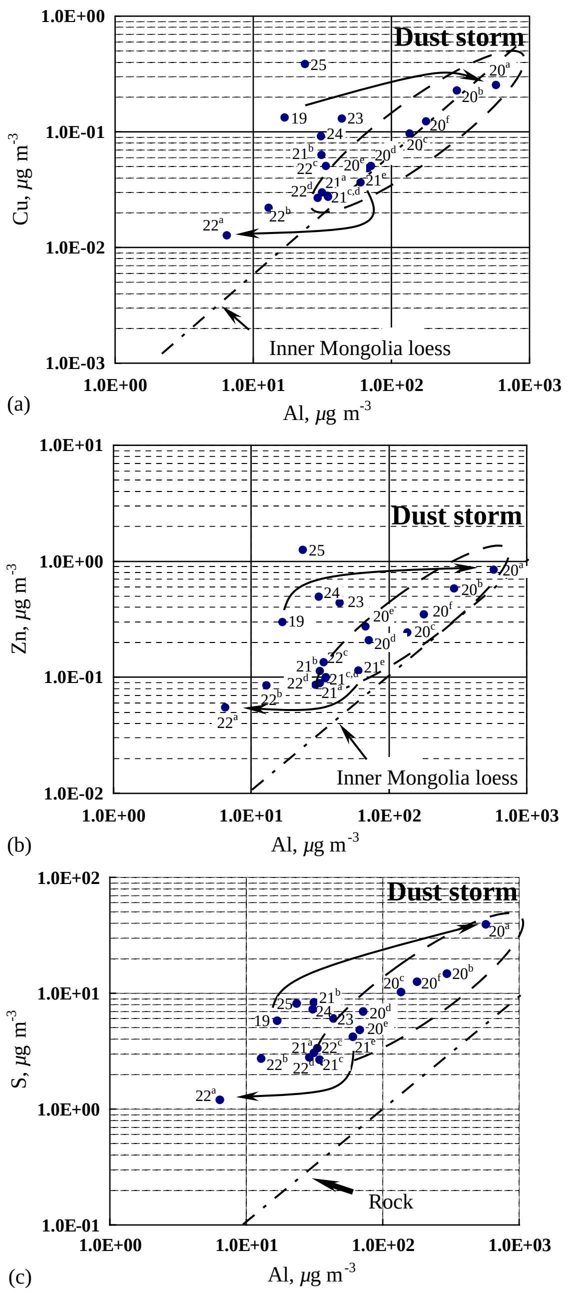

Fig. 1. X vs. Al in TSP samples collected during March 2002. The numbers in the figure illustrate the sampling time. For example, 20^b means the second sample collected on 20 March 2002.

pollution. On 20 March, Cu increased to 0.25 µg m^{-3} as Al rose to 570 µg m^{-3} (point 20^a). The increased crustal component, 0.23 µg m^{-3}, meant that most of the increased Cu was crustal. The Cu/Al ratio had decreased by 3-fold to 4.4×10^{-4}, essentially the same as its nominal crustal ratio of 5.6×10^{-4} for Inner Mongolia loess and 6.4×10^{-4} for Minqin dust (Liu et al., 2002). The replacement of pollution Cu by crustal Cu showed that pollution aerosol had been replaced by crustal aerosol. For all the samples taken on 20 and 21 March, Cu varied closely enough with Al to keep the Cu/Al ratio within a factor of two of its crustal ratio (Fig. 1a) and to confirm the crustal origin of both elements and probably the great mass of inorganic aerosol. As the dust storm ended on 22 March, Cu/Al climbed above the crustal ratio and remained high. This brought Cu and Al back to their normal pollution proportions for spring in Beijing.

Zn varied similarly to Cu (Fig. 1b). On 19 March, its concentration was 0.3 µg m^{-3}, with a crustal component of 0.01 µg m^{-3} (point 19). The Zn was almost completely from pollution sources, and its ratio to Al was 30 times higher than its nominal crustal ratio of 1.0×10^{-3} for Inner Mongolia loess and 6.6×10^{-4} for Minqin dust (Liu et al., 2002). As the dust storm arrived on 20 March, Zn rose to 0.85 µg m^{-3} but the Zn/Al ratio dropped to 1.5×10^{-3} (point 20^a), only 1.5 times greater than the nominal crustal ratio and probably close to the actual ratio. This was the same progression as for Cu: replacement of pollution Zn by crustal Zn and pollution aerosol by crustal aerosol. The Zn/Al ratio remained within factors of 1–4 of its nominal crustal ratio throughout this dust event, reinforcing the purely crustal origin of the dust. Zn/Al climbed on the 22nd and remained high, indicating the return of normal spring aerosol.

The progression for S (Fig. 1c) was much the same as for Cu and Zn. It began on 19 March by being mostly pollution derived, with a high S/Al (point 19). Its concentration rose as the dust storm arrived on the 20th, but its ratio to Al dropped to the lowest value we have ever seen for Beijing. The ratio remained low throughout the dust event, while the concentrations rose and fell almost linearly with Al. After the dust storm, S/Al climbed back to its normal value and remained there. Because the S/Al ratio did not decline all the way to its nominal crustal value, it is likely that some of the S in the dust storm was crustal. Some of this might be from sulfates in the long-range transported soil, and some from uptake of SO_2 by alkaline crustal particles during transport to Beijing (the absorption mechanism suggested by Andreae et al., 1986). More work needs to be done to determine the real source of the "extra" S in dust storms.

Collectively, these three pollution elements revealed the replacement of pollution aerosol by crustal aerosol during the dust storm, the clear-out of the crustal aerosol as the storm passed, and the subsequent build-up of pollution aerosol. The elements also showed that a dust storm could increase the concentration of "pollution" elements by replacing them with unusually large crustal components. The elements also showed that at least during a super dust storm, as we have seen on 20 March 2002, the dust is mostly crustal material. But this leaves unanswered the question of whether this pure crustal dust is something special for the super dust storms. Where do the pollution elements come from in medium dust storms, when their ratios to Al remain above the crustal ratios? We addressed this problem by examining a longer set of samples taken in March 2001. Instead of resolving the problem, however, we found the four stages of dust storms that are the focus of this article.

3.1.2. Stages in weaker dust storms of March 2001

We examined the TSP samples of 14–26 March 2001, again using Al as the indicator of crustal aerosol and the ratios of pollution elements to Al as the indicator of the origin of the pollution elements. The higher the X/Al ratio, the more of element X (and probably Al) comes from pollution.

The concentrations of Al showed that there were two dust events during this period, and the four X/Al ratios revealed four "stages": (1) accumulation of pollutants; (2) clear-out of pollutants; (3) arrival of dust; and (4) clear-out of dust (Cu/Al ratio is shown as an example in Fig. 2). From 14 to 16 March, the X/Al ratios increased slowly and indicated that pollutants were accumulating (stage 1). On 17 March, all the X/Al ratios except for S began to decrease (S/Al began one day later), and continued until they reached minimum values on the 20th. Because Al did not change much during these days, the decrease in X/Al ratios meant that the concentrations of the pollution elements were decreasing, too. This showed that the pollutants were being cleared out (stage 2). The sample taken from 12:00 to 14:00 on 21 March showed an increase of pollutants and a small increase of crustal elements. We are not sure whether these pollutants were transported or accumulated locally in stagnant air. The real dust storm arrived around 15:00, with crustal concentrations increasing sharply and the X/Al ratios dropping to near-crustal values (stage 3). By 22 March, the dust storm had ended (stage 4). Although the crustal elements were still high, they remained within "normal" spring concentrations for Beijing, so that 22 March was not considered a dust-storm day. The pollutants started to climb again on the 23rd, indicating a possible new stage 1. The noontime sample on the 24th resembled that of the 23rd, and showed no indication of a dust storm. Around 15:00, however, a cloud of dust arrived. Al increased sharply, and the X/Al ratios dropped (stages 2 and 3). This dust storm was not as strong as the one on the 21st, and did not last as long. It ended early in the evening of the 24th (stage 4). Then the cycle progressed to the next stage 1.

3.1.3. Summary of the elemental stages

The data from both 2001 and 2002 clearly reveal four stages to the cycle of dust storms. The first two stages can be separated from the last two or can overlap. When they are separate, the dust storm can be "pure," that is, with "pollution" elements being high in concentration but purely crustal in origin (crustal ratios to Al). When the stages overlap, especially stages 2 and 3, the X/Al ratios can be high at the same time that Al is high. Although we are not sure how much of a role resuspension plays in weaker dust events in Beijing, overlapping stages seem to be one of the most important ways of getting high pollution in a dust storm. As we know, dust storms are activated by strong winds associated with cold fronts and transported to Beijing via different pathways (Gao et al., 2002). Since there is less population in the areas north and northwest of Beijing, air masses from that direction are usually less polluted. When a dust cloud is injected into that air mass, which then replaces the previous, polluted air mass in Beijing, the pollutants are effectively cleared out. If the dust cloud follows immediately after the front, or even precedes it by falling from the colder air mass above, it will arrive in Beijing before the pollutants are cleared out. This overlaps stages 2 and 3 and obscures the origin of the pollutants unless they have been sampled beforehand and the meteorology has been

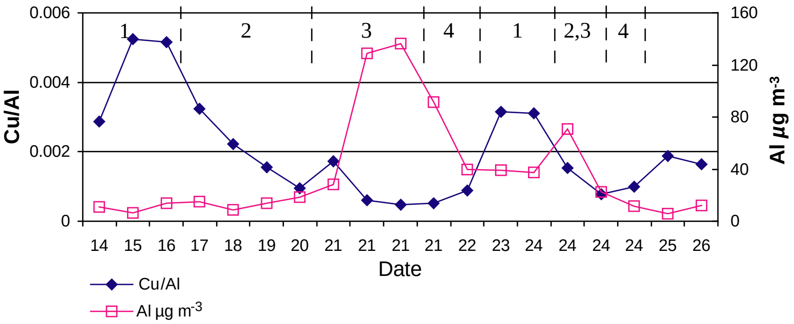

Fig. 2. Cu/Al and Al concentration in TSP samples collected during March 2001.

carefully followed. If the dust cloud is far enough behind the front, the pollutants can be cleared out before the dust arrives, leading to a well-defined four-stage dust storm.

3.2. Stages of dust storms revealed by pollution gases and PM10

We collected daily average concentrations of SO_2, CO, NO_2, O_3, and PM10 for Beijing from the web site of the Beijing Environmental Protection Bureau (http://www.bjepb.gov.cn), and SO_2, NO_2, and PM10 for Shanghai from the web site of the Shanghai Environmental Protection Bureau (http://www.envir.online.sh.cn). Here we use SO_2 as the indicator of gaseous pollution.

As shown in Fig. 3a, SO_2 and PM10 are positively correlated in winter in Beijing, which means that PM10 has the same general sources as SO_2 (pollution). As the air in Beijing alternates between clean northern air and polluted southern air, concentrations of SO_2 and PM10 rise and fall together. Fig. 3b shows that the same relation holds for SO_2 and PM10 in winter in Shanghai, but with the air mass alternating between clean air from the sea and polluted air from the land. The situation is different in springtime in Beijing, however (Fig. 3c)—both inverse and direct relations between SO_2 and PM10 are seen. This is the season of dust storms. The northern air mass becomes loaded with dust as it is injected by fronts over the deserts north and northwest of Beijing. When cold, dry, dusty air comes to Beijing from the north, the pollution is cleared out and dust is added to the air mass. As the wind speeds of this air mass decrease, the dust gets cleared out and pollution begins to accumulate. Consequently, PM10 varies inversely with SO_2 when dust storms come. In 2002 in Beijing, dust storms were reported on 15–16 March, 20–21 March, 6–8 April, 11–12 April, and 15–16 April. The pollution gases and PM10 correlated inversely throughout these events (Fig. 3c), agreeing with the informal assessment of dust storms. They correlated directly, however, during the other (non-dust) days, and thereby confirmed that that PM10 was pollution derived then. The big decrease of pollution gases on the days of dust storms shows that the cleaner air mass has replaced the dirtier mass. The pollution gases typically remained low

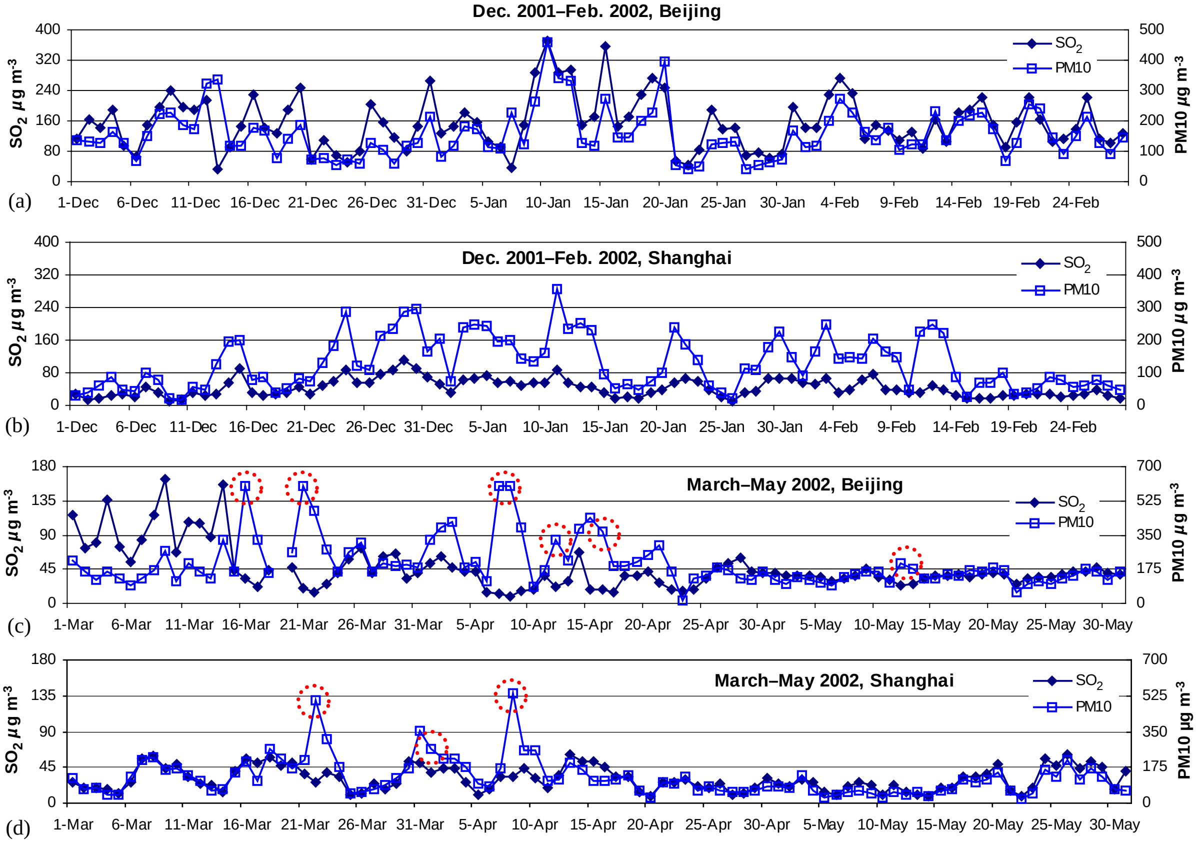

Fig. 3. SO_2 and PM10 concentrations in Beijing and Shanghai in winter (a and b) and spring (c and d), circles indicate reported dust storms.

throughout the dust event, but PM10 peaked on the second day. Thus the "clean" air mass contained considerable dust, which makes "clean" mean lack of pollution aerosol. After the dust event, the pollution gases began to climb and PM10 dropped. When the new air mass does not contain extra dust, PM10 correlates positively with the pollution gases and so is pollution derived. There are far fewer dust events in Shanghai in spring (Fig. 3d), where SO_2 and PM10 remain positively correlated most of the time.

PM10 here works the same way that Al does in aerosol data, and the pollution gases work the same as X/Al ratios. A closer look at the variations of PM10 and SO_2 for March–April 2002 reveals the same four stages (accumulation and clear-out of pollutants, arrival and clean-out of dust) that the elements in aerosol do. Both SO_2 and PM10 increased from 30 March to 2 April. This signaled the build-up of pollutants (stage 1). From 3 April to 6 April, both SO_2 and PM10 concentrations dropped gradually (stage 2, the clear-out of pollutants). On 7 April, SO_2 stayed low while PM10 concentration jumped to 600 $\mu g\,m^{-3}$, which meant that PM10 then had a different (crustal) source (stage 3, the arrival of dust). On 9 April, PM10 dropped below 100 $\mu g\,m^{-3}$ (stage 4, the clear-out of dust). Then another cycle began. This time, however, stages 2 and 3 overlapped rather than being separated. After stage 1 the pollutants were accumulating in the air mass, but without enough time for them to be cleared out before the dust arrived. (High PM10 with low SO_2 arrived directly after high PM10 and high SO_2.) These four stages are caused by air-mass movements. When the dust cloud is far enough behind the cold front to allow time for the pollutants to be cleared out, stages 2 and 3 are separated. When the dust cloud follows right behind the cold front and there is not enough time for the pollutants to be cleared out beforehand, stage 3 overlaps stage 2. Thus the relationships between PM10 and pollution gases reveal the movements of air masses and the properties of dust storms.

3.3. Meteorological aspects of stages of dust storms

3.3.1. The super dust storm

The hourly meteorological data from 19 to 25 March 2002 at Beijing showed that a dry cold front passed during the dust event (Fig. 4). Before the dry air mass arrived on the 20th, the dew point was high and the wind speed was low—typical spring weather for Beijing. When the front arrived, the dew point dropped sharply and the wind strengthened—a typical strong springtime cold front. Together with this front came the huge dust storm. The aerosol data show, however, that the most-concentrated dust appeared before the dry air and strong winds arrived. When the winds strengthened, the aerosol fell instead of rose. This means that the dust in this event was mostly from transport rather than local resuspension. We call this quiet, dusty period before the strong winds the calm stage of the dust storm. One possible explanation for this phenomenon is: when the dust was kicked up back in the source area by the low-pressure system, it was lifted high and transported far downwind. It was transported faster and arrived at Beijing before the surface dust and wind because the winds aloft were stronger than at the ground. The coarsest particles aloft fell into the calm, humid air mass below and created the initial quiet, dusty period. After the new air mass settled in, the dew point started to rise and local pollution started to accumulate.

3.3.2. Dust storms of March and April 2002

The hourly variations of temperature, dew point, and wind speed for March and April 2002 at Beijing (Fig. 5) revealed the more general comings and goings of air masses. The basic air masses in question can be simplified to a northern one that is relatively cold, dry,

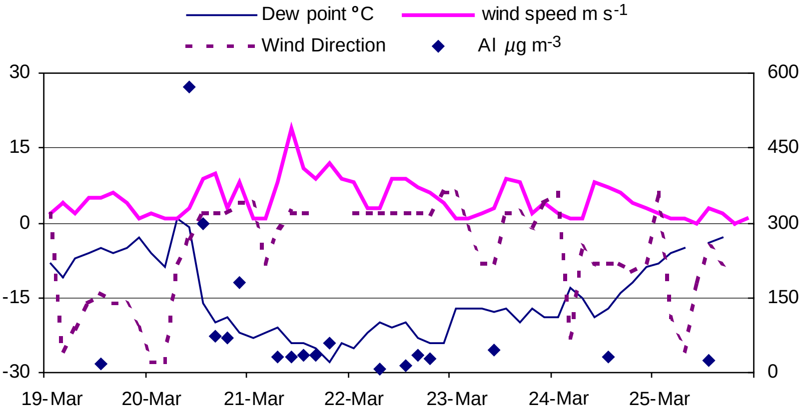

Fig. 4. Meteorological data and Al concentration for TSP samples of 19–25 March 2002. Wind speed and dew point left axis, Al concentration and wind direction right axis.

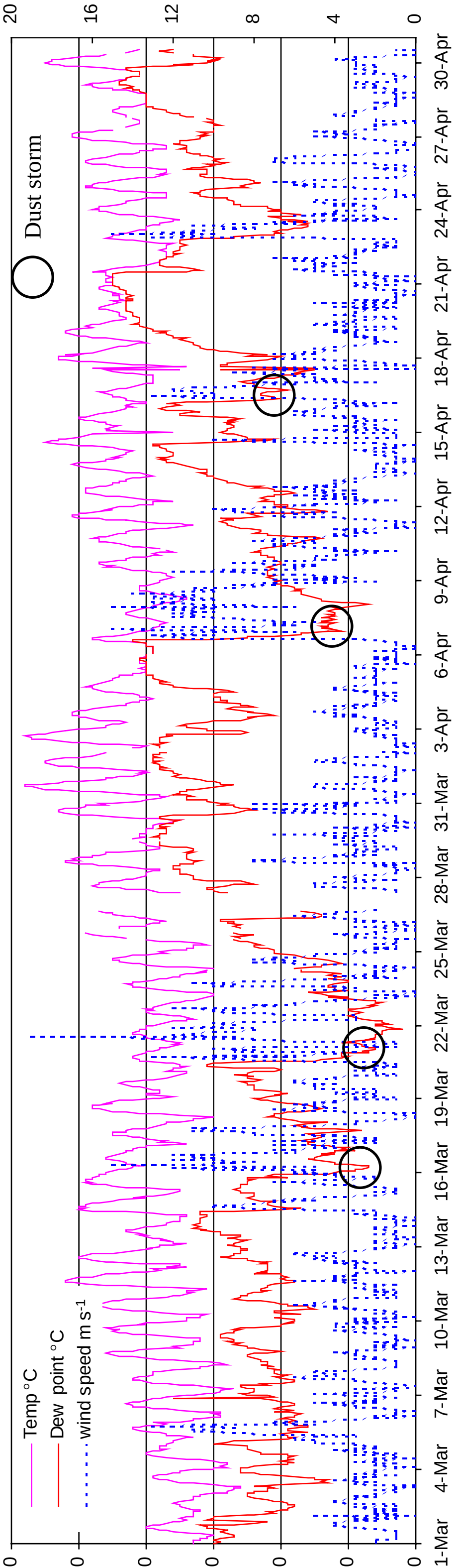

Fig. 5. Temperature, dew point, and wind speed for March and April 2002. Temperature and dew point left axis, wind speed right axis.

and clean (except when it contains dust) and a southern one that is warmer, more humid, and more polluted. As a northern air mass arrives in Beijing and replaces the southern one, the concentrations of pollution elements drop. If the air is enriched in dust, the concentrations of crustal elements rise; otherwise they drop, too. (Note in Fig. 5 that dust storms come with four of the five strongest drops in dew point.) Pollutants begin to accumulate as soon as the new air mass settles in. This sequence matches the variations of elements in dust storms, pollution gases, and PM10 very well.

4. Conclusions

PM10/SO$_2$, elemental ratios in aerosol, and meteorology have independently revealed four stages for dust storms, which represent the alternation of air masses: (1) accumulation of pollutants; (2) clear-out of pollutants; (3) arrival of dust; and (4) clear-out of dust. The first two stages can be separate from the last two or can overlap them. When they are separate, the dust storm is "clean," that is, with "pollution" elements high in concentration but purely crustal in origin (as shown by crustal ratios to Al). When the stages overlap, especially 2 and 3, the dust storm contains more pollution, which manifests itself in X/Al ratios that are high at the same time that Al is high. Although we are not sure how much of a role resuspension plays in this case, we are sure that overlapping of stages is one of the most important ways of getting high pollution concentrations in a dust storm.

The variations of PM10 and pollution gases (SO$_2$ here) also proved to be useful. The pollution gases serve as general indicators of anthropogenic aerosol (including pollution-derived "crustal" elements) being accumulated or diluted in the air mass, while PM10 indicates genuine crustal aerosol when it varies indirectly with the pollution gases, and anthropogenic aerosol when it varies inversely with them. Together, these indicators revealed the same four stages as the elements had. This result is especially important because each city in China has been required since July 2000 to publish an Air Pollution Index (API) measured daily, and they are now typically observing NO$_x$, SO$_2$, and TSP (PM10 in some cities), and sometimes CO and ozone. Studying this record will improve our understanding of the generation and transport of dust storms across China. Accompanied by meteorological data, we will get a clearer picture of dust storms in northern China.

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