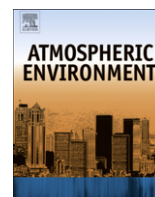




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Mixing and transformation of Asian dust with pollution in the two dust storms over the northern China in 2006

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ABSTRACT

To study the mixing and transformation of Asian dust with pollution in the two dust storms over the northern China in 2006, both TSP and PM_{2.5} samples were collected at three sites of northern China in addition to the dry deposition samples collected in an episode in Beijing. 23 elements, 15 ions, and 16 PAHs in each sample were analyzed. The two dust storms in northern China were observed in April 8–10 (DS1) and April 16–18 (DS2). Compared to DS2, DS1 was weaker and more polluted with stronger mixing between crustal and pollutant aerosols during their long-range transport. The concentrations of pollution species, e.g. pollution elements, ions, and PAHs were higher in DS1 than that in DS2, while the crustal species showed adverse variation. The correlation between chemical species and Al and between PAH(4) and PAH(5,6) further confirmed the stronger chemical transformation and aerosol mixing in DS1 than that in DS2. Back trajectory and chemical analysis revealed that in DS1 the air masses at Beijing were mostly from southern or southwestern direction at lower altitude with much more pollution, while in DS2 the air masses were mostly from the northwestern and northern direction with dust mainly, which explained why there was a stronger mixing of dust with pollution aerosol in DS1 than that in DS2 over Beijing.

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1. Introduction

Dust storm, blowing dust, and floating dust are clarified according to the horizontal visibility degraded by the dust and sand. Dust storm not only deteriorates the air quality and ecological system and accelerates the desertification in source area and pathways, but also affect the local climate by decreasing the visibility and influencing the radiation balance of the atmosphere, which may cause extensive economic loss (Wang et al., 2005a,b). Asian dust, starting from Gobi deserts in Mongolia and northern China and Taklimakan desert in western China (Sun et al., 2005), spreads rapidly over China, Korea (Chung et al., 2003), Japan (Nishikawa et al., 1991), and even to the west coast of North America (Husar et al., 2001; Perry et al., 1999). The crustal dust can mix with pollution aerosol on the pathway and change the chemical composition of the aerosols during its long-range transport. Furthermore, different transport pathway could result in different characteristics of the aerosols (Sun et al., 2005).

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Northern China located in the eastern Asian monsoon region, where arid and semiarid climate dominate, is one of the four regions of dust storm frequently occurred in the world for the past million years (Yan, 1993). The total area of Gobi desert and desertification land is approximately 1.653 million square kilometers (Wang and Zhu, 2001). Wang et al. (2005a,b) concluded that dust storm events gradually increased since 1998 based on the study on the frequencies of dust storm events during 1954–2001. A super dust storm attacked Beijing on March 20, 2002, which was the greatest one ever recorded until then (Sun et al., 2004). Although no such a big dust storm occurred in the next 3 years, another super dust storm attacked Beijing on April 17, 2006, which would verify the conclusion that the dust storm events happened in northern China with a cycle of 3–4 year (Wang et al., 2005a,b). The previous studies in our group (Zhuang et al., 2001; Guo et al., 2004; Wang et al., 2005a,b; Sun et al., 2005) revealed that dust storm not only delivered large amounts of crustal elements but also carried significant quantities of pollutants. In addition, four stages in the dust episode were observed and both crustal and pollutant species increased especially in PM_{2.5} during the dust storm, indicating that the dust storm was also a “pollutant”

storm (Guo et al., 2004; Sun et al., 2004, 2005). The dust particles could mix well with pollution aerosols, including sulfate, nitrate (Sun et al., 2005; Shen et al., 2007), the pollutant elements, such as Se, Ni, Pb, Br, and Cu (Zhang et al., 2005), sea salt particles (Fan et al., 1996), black carbon (Kim et al., 2004), and volatile organic compounds (Cheng et al., 2006) during the long-range transport. Hou et al. (2006) also reported that dust storm could carry and transport much organic pollutants, such as polycyclic aromatic hydrocarbons (PAH) in dust aerosols. However, Zhang et al. (2005) argued that Asian dust plume and polluted air masses were found not mixed with each other; instead, they were separated in two consecutive air parcels. To date there seems no concurrent measurements of inorganic and organic pollutants during dust storms. In this study both PM_{2.5} and TSP samples were collected at three sites of northern China in the spring dust season in 2006 with the measurement of inorganic and organic pollutants. In addition, the dry deposition samples were collected on the day of the super dust fall. This study would provide more evidences of mixing and transformation of Asian dust with pollution aerosols during the long-range transport.

2. Sampling and experiment

2.1. Sampling and mass concentration analysis

Aerosol samples of TSP and PM_{2.5} were synchronously collected at Yulin (YL) in Shaanxi province, Duolun (DL) in Inner Mongolia, and Beijing (BJ) in the spring of 2006 (Fig. 1), using medium-volume samplers manufactured by Beijing Geological Instrument–Dickel Co., Ltd. (model: TSP/PM₁₀/PM_{2.5}–2; flow rate: 77.59 L min^{−1}). The detailed descriptions of these sampling sites were given in Sun et al. (2006). All the aerosol samples were collected with Whatman® 41 filters and quartz fiber filters (Whatman Inc., Maidstone, UK) for inorganic and organic measurement, respectively. The filters were weighed before and after sampling using an analytical balance (Sartorius 2004 MP, with a reading precision of 10 µg) after keeping them under constant temperature (20 ± 1 °C) and humidity (40 ± 1%) for 48 h. The dry deposition samples were collected in a clean container in the morning of April 17, 2006, when the super dust fall attacked Beijing. The dry deposition samples were gridded into three sizes: <30, 30–74, and >74 µm, by filtrating through 500 and 300 sieve pores.

2.2. Chemical analysis

2.2.1. Element analysis

Total 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) in the aerosol and dry

deposition samples were analyzed by inductively coupled plasma spectroscopy and atomic emission spectroscopy (ICP-AES) (Model: ULTIMA, JOBIN-YVON Company, French). The detailed analytical procedure has been described in Zhuang et al. (2001).

2.2.2. Ion analysis

Eleven inorganic ions (SO₄^{2−}, NO₃[−], F[−], Cl[−], NO₂[−], PO₄^{3−}, NH₄⁺, Na⁺, K⁺, Ca²⁺, Mg²⁺) 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 filter samples were extracted using ultrasonic cleaner (KQ-50B), and then filtered with 0.45 µm micro-porous membrane (*d* = 25 mm, Attached factory of Beijing chemical school) and injected to IC system by polypropylene injector. The details were given elsewhere (Yuan et al., 2003).

2.2.3. PAHs analysis

PAHs were extracted and analyzed by high performance liquid chromatography (HPLC) (Water® 2690 Separations Module, Water® 474 Scanning Fluorescence Detector and Symmetry® C18 3.9 mm × 150 mm Φ5 µm column) with the method of U.S. EPA (1998). 16 PAHs, including Naphthalene (Nap), Acenaphthylene (Acy), Fluorene (Flu), Acenaphthene (Ace), Phenanthrene (Phen), Anthracene (Anthr), Fluoranthene (Fluor), Pyrene (Pyr), Chrysene (Chy), Benzo[a]anthracene (B[a]A), Benzo[b]fluoranthene (B[b]F), Benzo[k]fluoranthene (B[k]F), Benzo[a]pyrene (B[a]P), Dibenzo[a,h]anthracene (DBA), Indeno[1,2,3-cd]pyrene (IND), and Benzo[ghi]perylene (B[ghi]P), were measured. The details were described in Zhang et al. (2007).

2.2.4. The meteorology analysis

The meteorological data, including temperature, dew point, wind speed, relative humidity, and atmospheric pressure, were obtained from the website (<http://www.wunderground.com>).

3. Results and discussion

3.1. General description of the dust storm in 2006

Two dust storm events occurred over the northern China in the spring of 2006, one lasted from April 5–10 (DS1), and the other from April 16–18 (DS2). Specifically, the two dust storms, DS1 and DS2 spread over the three sampling sites DL, YL, and BJ, where we were collecting samples. DS1 reached DL, YL and BJ during April

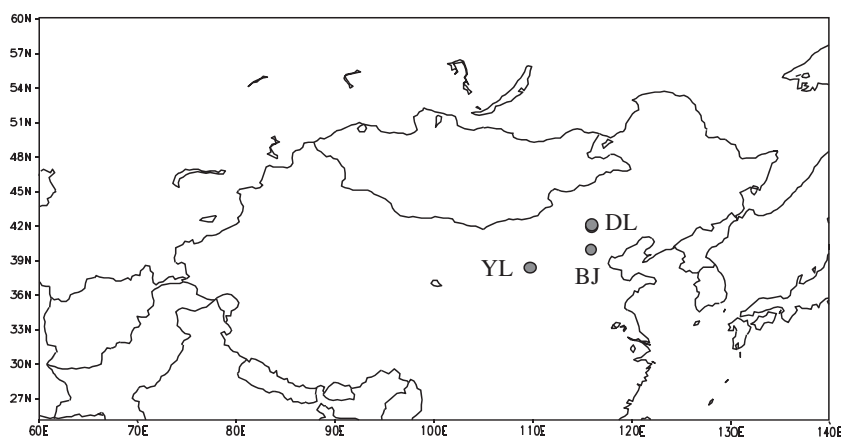


Fig. 1. Map of the sampling sites (BJ: Beijing, DL: Duolun, YL: Yulin).

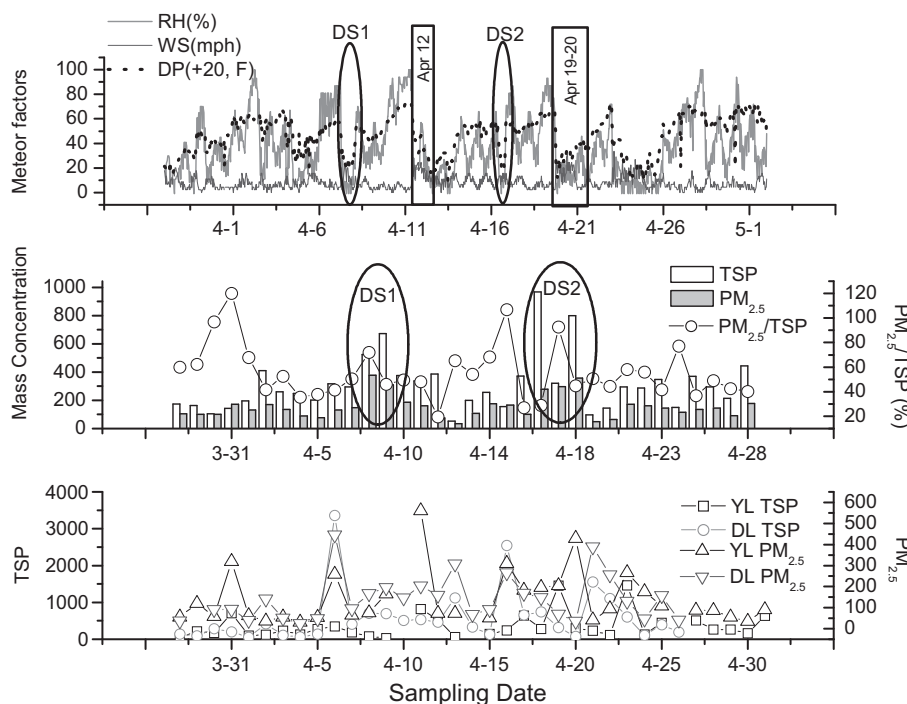


Fig. 2. Mass concentration of $PM_{2.5}$, TSP in BJ, DL, YL, and related meteorological information (RH, DP, WS) in Beijing during the sampling period.

5–6, April 6 and April 8–10, respectively. DS2 reached the above three sites during April 16, April 16–17 and April 16–18, respectively. It must note that in YL, besides the two dust storms, there were more dust storm in spring 2006, which, however, did not spread over northern China and were not mentioned in this study.

According to the weather report of Monitoring and Warning system of China Meteorological Administration (CMA) (<http://www.cma.gov.cn>), DS1 was mainly induced by the Mongolia cyclone and cold front from the northwest of China. DS2 originated from the northern and northwestern China, and reached Beijing at

night, and it was found outside a “yellow” world in the morning of April 17, 2006. DS2 was the strongest dust storm since 2003 and much stronger and more widespread than DS1. The visibility even dropped to less than 3 km at 8:00 of April 17. The dust storm brought ~300,000 tons of dust to Beijing. During the next two days, the dust kept moving southeastward and influenced a wide range of more than 300 thousand square kilometers in China.

Dust events were generally characterized of cold fronts with relative dry conditions, i.e. low relative humidity, and often with high wind speed (Wang et al., 2005a,b). Fig. 2 showed the

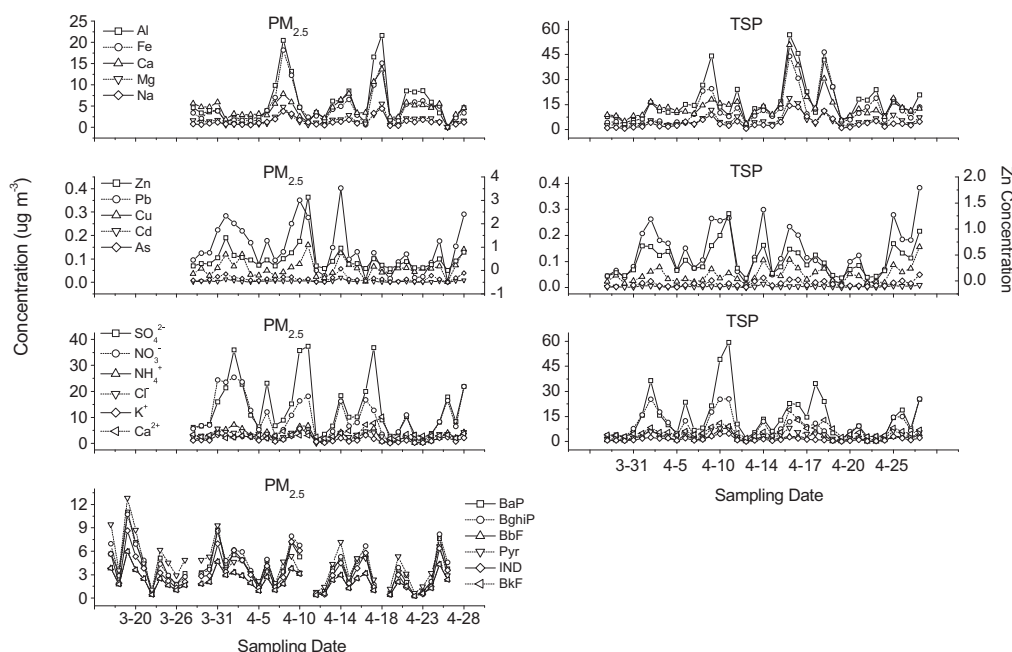


Fig. 3. The variation of chemical species in $PM_{2.5}$ and TSP during sampling time in Beijing.

temporal variations of $PM_{2.5}$ and TSP at the three sampling sites, along with the meteorological conditions in Beijing during the sampling period. Dust events were highlighted in the figure. Distinct weather conditions were observed between the two dust events and the non-dust days. The relative humidity (RH) and dew point (DP) decreased abruptly as the dust storm came and then increased again after the passage of the storm, while the wind speed (WS) showed an inverse trend. The concentrations of TSP and $PM_{2.5}$ in Beijing reached a peak of 672.1 and 377.5 $\mu\text{g}/\text{m}^3$ on April 8–9 in DS1, which was ~ 13 and 11 times of that on April 12th after DS1 (52.5 and 32.3 $\mu\text{g}/\text{m}^3$ in TSP and $PM_{2.5}$), respectively, and 966.9 and 358.1 $\mu\text{g}/\text{m}^3$ on April 17–18 in DS2, which was ~ 10 and 7 times of that on April 19 after DS1 (97.0 and 49.2 $\mu\text{g}/\text{m}^3$ in TSP and $PM_{2.5}$), respectively. In DL, TSP and $PM_{2.5}$ concentrations reached as high as 3361.6 and 444.9 $\mu\text{g}/\text{m}^3$ in DS1 at the peak day on April 6 and increased 26 and 5 times of the lowest concentration after DS1 (130.5 and 90.5 $\mu\text{g}/\text{m}^3$ in TSP and $PM_{2.5}$), respectively, while reached 2547.7 and 265.3 $\mu\text{g}/\text{m}^3$ in DS2 at the peak day on April 16 and to be 34 and 7.4 times of the

lowest concentration after DS2 (74.2 and 35.7 $\mu\text{g}/\text{m}^3$ in TSP and $PM_{2.5}$). In YL, TSP and $PM_{2.5}$ reached 347.4 and 258.7 $\mu\text{g}/\text{m}^3$ on April 6, respectively, which was ~ 5.6 and 4.9 times of that day after DS1 (62.5 and 53.1 $\mu\text{g}/\text{m}^3$ in TSP and $PM_{2.5}$), respectively, and on April 16–17 TSP and $PM_{2.5}$ reached as high as 639.8 and 310.7 $\mu\text{g}/\text{m}^3$, which was 5.5 and 3.3 times of that day after DS2 (116.9 and 94.7 $\mu\text{g}/\text{m}^3$ in TSP and $PM_{2.5}$). As shown in Fig. 2(a), in Beijing RH and DP decreased to be $<10\%$ and $<20^\circ\text{F}$ at the peak of the two dust storms, while the WS showed highest values of 22.4 and 15.7 mph in DS1 and DS2, respectively, when the dust storm firstly attacked. After the peak, the aerosol mass concentrations in DS1 decreased to 52.5 and 32.3 $\mu\text{g}/\text{m}^3$ of TSP and $PM_{2.5}$, while in DS2 decreased sharply to 97.0 and 49.2 $\mu\text{g}/\text{m}^3$, respectively; and the RH/DP increased up to 94%/52°F in DS1 and 81%/36°F in DS2, while the WS decreased to be 4.5 mph in both DS1 and DS2. Also, $PM_{2.5}$ reached the highest concentration prior to TSP in Beijing, which may be due to the pollutants pushed by cold front and reached Beijing first (Chuang et al., 2008), while in DS2 TSP reached the highest concentration prior to $PM_{2.5}$.

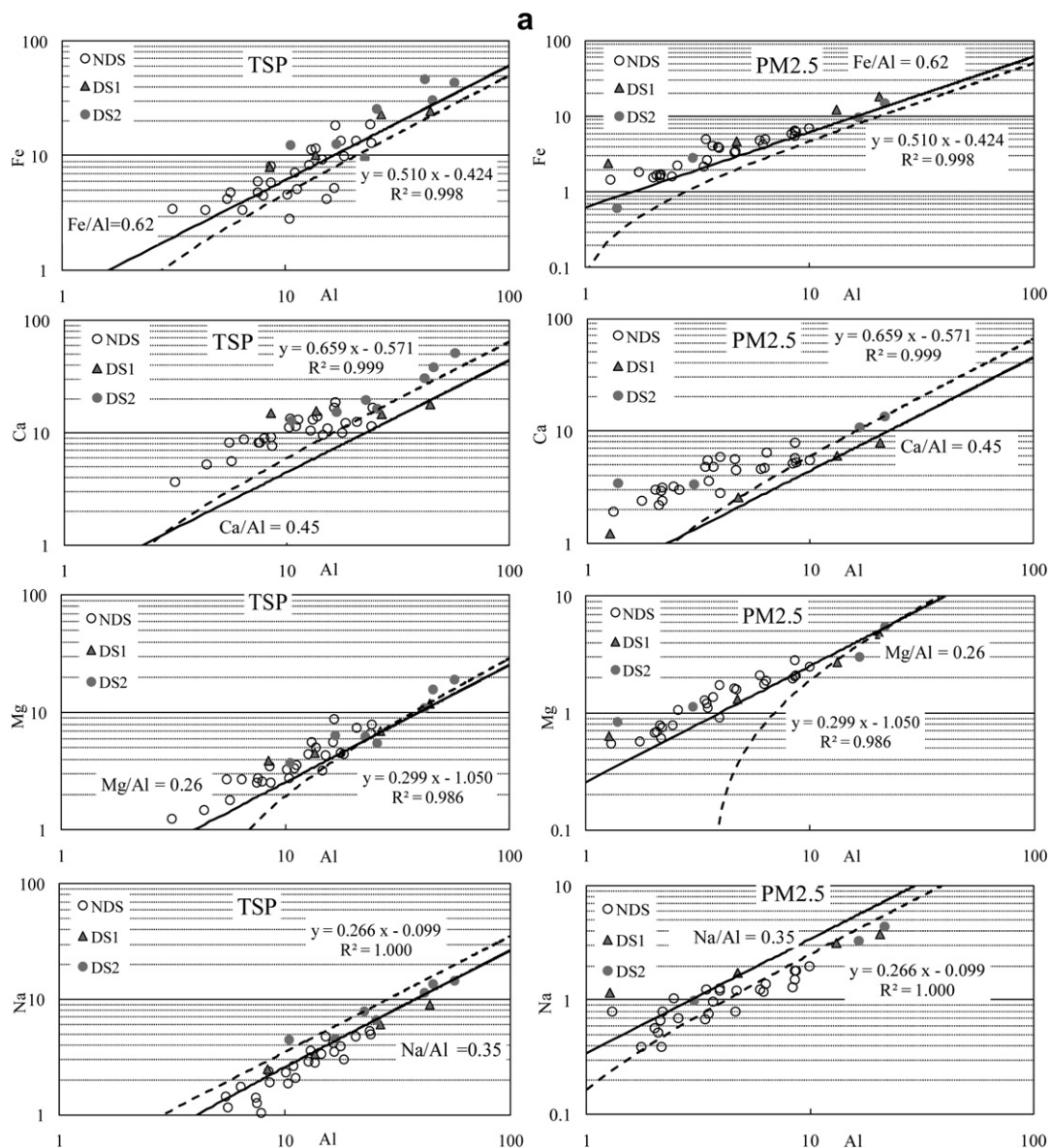


Fig. 4. X versus Al in TSP and $PM_{2.5}$ samples collected from March 28 to April 30 in 2006 in Beijing. (a) Fe, Ca, Mg, Na; (b) Cu, Zn, As, Pb, Cd; (c) SO_4^{2-} , NO_3^- , NH_4^+ , Ca^{2+} , K^+ .

3.2. Aerosol mixing and transformation in DS1 and DS2

3.2.1. Aerosol mixing in DS1 and DS2

3.2.1.1. Evidence of mixing from the chemical species. Fig. 3 presented the temporal variation of the major chemical species during the sampling period in Beijing. For those crustal elements, Al, Ca, Fe, Mg, and Na, the highest concentration in TSP were 44.2, 18.1, 24.6, 11.9, and 8.9 $\mu\text{g}/\text{m}^3$ on April 9 in DS1, and 56.8, 50.8, 43.9, 18.9, and 14.5 $\mu\text{g}/\text{m}^3$ on April 17 in DS2, which were 2~6 times higher than the average of 12.1, 10.7, 7.9, 4.0, 2.7 $\mu\text{g}/\text{m}^3$ in non-dust storm day (NDS). The highest concentration of Al, Ca, Fe, Mg, and Na in $\text{PM}_{2.5}$

reach the peaks of 20.5, 7.8, 18.2, 4.8, and 3.8 $\mu\text{g}/\text{m}^3$ on April 9 in DS1, and 21.6, 13.6, 15.1, 5.6, and 4.4 $\mu\text{g}/\text{m}^3$ on April 18 in DS2, respectively, which were also 2~5 times higher than those on average in NDS. Furthermore, in $\text{PM}_{2.5}$ almost all the crustal elements reached their peak values, just one day after the peak of those in TSP, which might be due to the re-suspension of the dust and the mixing of the crustal elements with the pollution species (Guo et al., 2004; Sun et al., 2004, 2005; Wang et al., 2005a,b). In Fig. 3, it is obvious that the concentrations of the crustal elements in TSP in DS2 were higher than those in DS1, indicating the crustal part contributed more to the total mass of DS2 than DS1. The dust

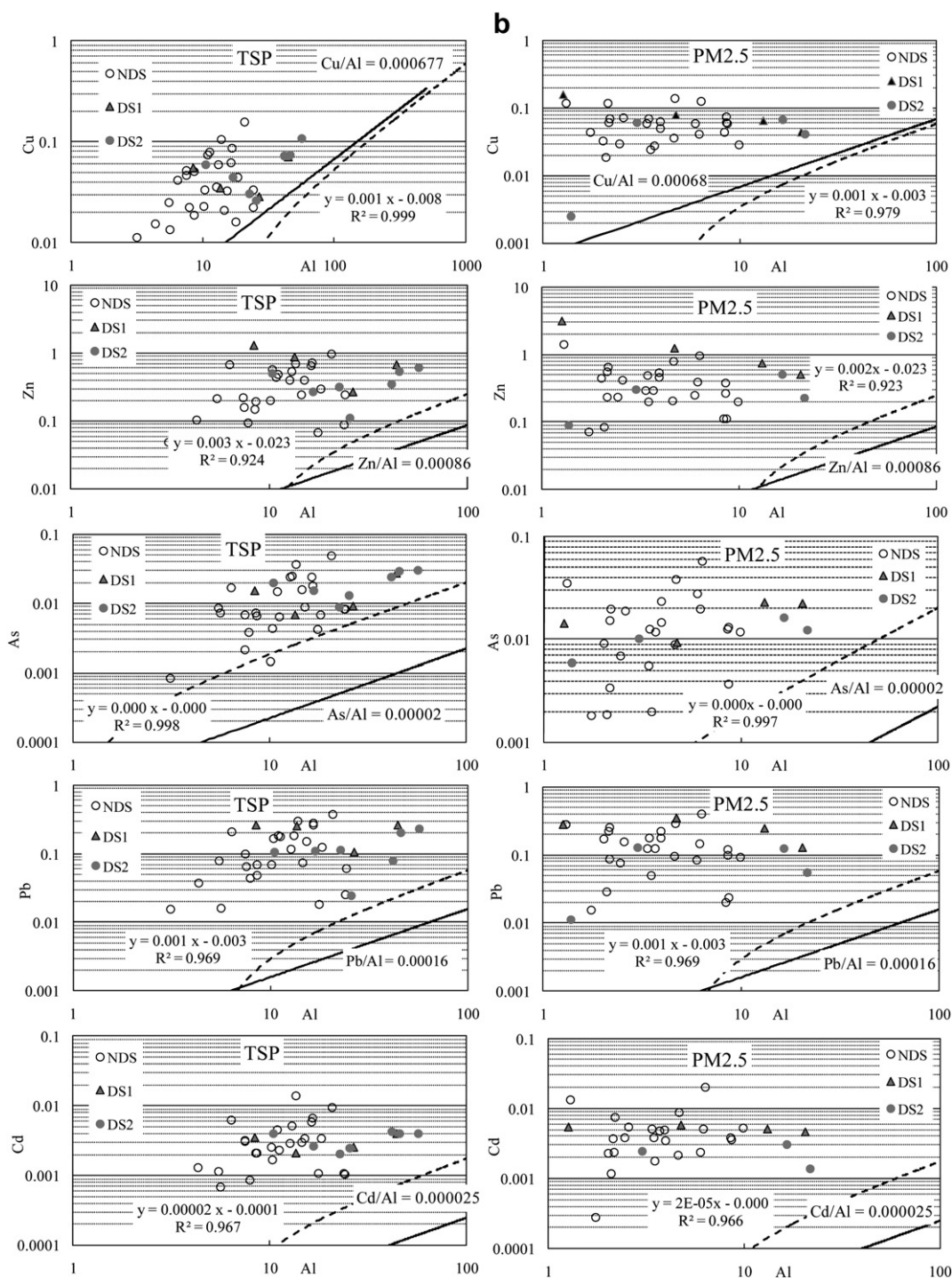


Fig. 4. (continued).

storm not only delivers large amounts of crustal aerosols but also carries significant quantities of pollution aerosols (Sun et al., 2004, 2005). In DS1, the highest concentrations of those pollution elements, As, Zn, Cu, Cd, and Pb, in PM_{2.5} were 0.023, 3.13, 0.16, 0.006, and 0.35 $\mu\text{g}/\text{m}^3$, respectively, on April 10 in DS1, which were 2~4 times (12 times for Zn) higher than their averages of 0.0089, 0.27, 0.046, 0.0028, and 0.093 $\mu\text{g}/\text{m}^3$ in NDS, respectively. In TSP, they were 0.027, 1.29, 0.071, 0.004, and 0.27 $\mu\text{g}/\text{m}^3$ on April 10 in

DS1, which were 2~5 times higher than their averages of 0.008, 0.25, 0.036, 0.002, and 0.088 $\mu\text{g}/\text{m}^3$ in NDS, respectively. In DS2, there were no significant variations in pollution elements between DS and NDS in both TSP and PM_{2.5}, as shown in Figs. 3 and 4(a). This indicated that the mixing between crustal and pollution aerosols was stronger in DS1 than that in DS2.

The correlations between elements/ions and Al in TSP/PM_{2.5} samples in Beijing were shown in Fig. 4(a)–(c). The major water-

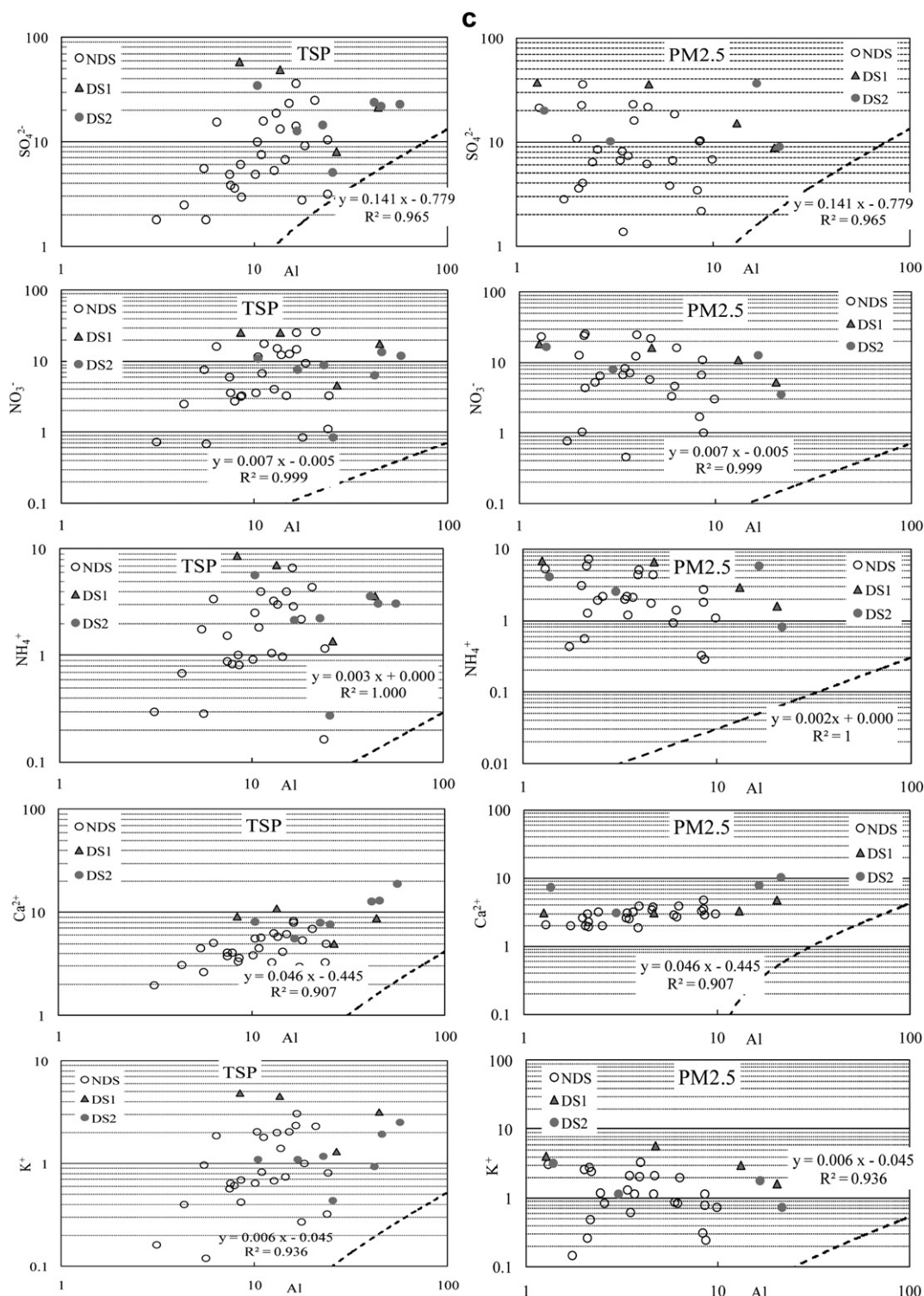


Fig. 4. (continued).

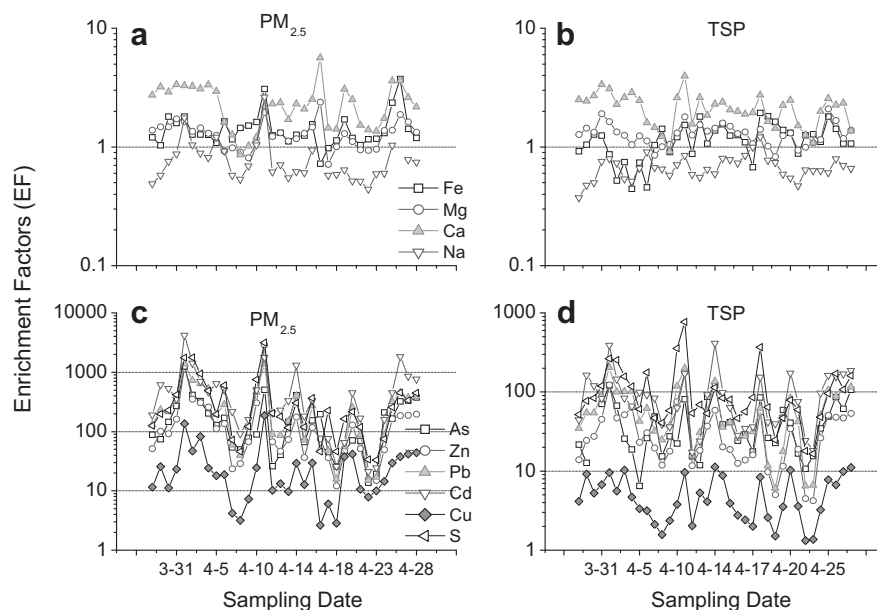


Fig. 5. The enrichment factors (EF) of the elements in $PM_{2.5}$ and TSP during the sampling time in Beijing (a) Fe, Mg, Ca, Na in $PM_{2.5}$; (b) Fe, Mg, Ca, Na in TSP; (c) As, Zn, Pb, Cd, Cu, S in $PM_{2.5}$; (d) As, Zn, Pb, Cd, Cu, S in TSP.

soluble ions, NH_4^+ , Ca^{2+} , Mg^{2+} , K^+ , SO_4^{2-} , NO_3^- , F^- , and Cl^- , could be classified into three groups according to their source. Ca^{2+} , Mg^{2+} , Na^+ were classified as the “crustal”, NO_3^- , NH_4^+ , F^- as the “pollution”, and SO_4^{2-} , Cl^- , K^+ as the crustal/pollution mixed group. Taking Ca^{2+} as the representative of the “crustal” group, it showed better correlation with Al in DS2 ($R = 0.93$ in TSP and 0.76 in $PM_{2.5}$), compared with those in NDS ($R = 0.45$ in TSP and 0.51 in $PM_{2.5}$). In DS2 the correlation of Ca^{2+} with Al in TSP ($R = 0.93$) was better than that in $PM_{2.5}$ ($R = 0.76$), while in DS1 it in $PM_{2.5}$ ($R = 0.88$) was better than that in TSP ($R = -0.33$). These results would indicate that compared to DS1, DS2 could be seen as more “crustal” storm with more in coarse part, and the mixing between crustal and pollutant aerosol could occur more in fine particle than that in coarse one (see Fig. 4(c)).

Taking SO_4^{2-} , NO_3^- as the representatives of the mixed and pollution group, they showed higher concentrations in dust storms compared with those in NDS, as shown in Fig. 3. Although DS2 was stronger than DS1, the concentrations of SO_4^{2-} , NO_3^- and NH_4^+ in DS1 were higher than those in DS2. This might be due to the overlap of the two stages of “clear-out pollutants” and “addition of pure dust” (Guo et al., 2004) in DS1. Furthermore, the weaker the dust storm was, the deeper extent of the mixing between crustal and pollutant aerosols (Wang et al., 2005a,b), which might explain that the relative weaker DS1 was associated with more pollutants in it, as compared to DS2.

16 PAHs that are the priority pollutants publicized by U.S. EPA were measured in $PM_{2.5}$. As the PAH compounds with 2 and 3 rings (NaP, Acy, Ace, Flu, Phen and Anthr) are of low molecular weight and high volatility, only those PAHs with 4–6 rings were discussed in this study. The semi-volatile PAHs with 4 rings are Fluor, Pyr, Chry, and B[a]A, and the non-volatile PAHs with 5 and 6 rings are B[b]F, B[k]F, B[a]P, DBA, IND, and B[ghi]P. No significant difference was found between DS and NDS for the PAH species mentioned above, suggesting that PAHs were more from local emissions, compared with those from the transport by dust events (see Fig. 6).

The highest concentrations of PAHs were observed on April 9 in contrast to the highest crustal and anthropogenic inorganic species on April 8, and April 10, respectively. This might verify again that the stages of “addition of pure dust” on April 8 ahead of time, “accumulation of local pollutants” on April 9, “clear-out pollutants” and “mixing of local pollution” on April 10 was overlapped together in

a weaker dust storm. This could partially explain why there was more pollution mixed with crustal in DS1 than DS2. However, in DS2 the highest concentration of PAHs was on April 17 (20.1 ng/m^3 , 29.2 ng/m^3 for the sum of 4 rings, PAH(4), and sum of 5/6 rings, PAH(5,6), respectively), and decreased significantly on April 18 (7.2 and 8.2 ng/m^3 for PAH(4) and PAH(5,6), respectively), which indicated the two stages of “accumulation of local pollutants” on April 17 and “clear-out local pollutants” on April 18. Furthermore, the highest concentration of crustal aerosol in later April 18 suggested the stage of “addition of pure dust”, which could partially explain the crustal characteristics of DS2. This might again verify that the different mixing extent between crustal and pollutant aerosols in the two dust storms, i.e. the well mixing in DS1 and incomplete mixing in DS2.

3.2.1.2. Evidence of mixing from the correlation and EF analysis.

Enrichment factors (EFs) of major elements in aerosols were calculated using Al as a reference element, X is the interest element ($EF = (X/Al)_{\text{aerosol}} / (X/Al)_{\text{crust}}$) (Fig. 5). EFs of crustal elements (Fe, Ca, Mg, Na) in TSP approached 1 in DS2 especially on April 17, which clearly indicated the crustal origin of aerosols in DS2 (Fig. 5(a) and (b)). In DS1, high EFs of Fe, Ca, Mg, Na were observed on April 10, indicating there was somewhat anthropogenic origin in addition to the crust sources.

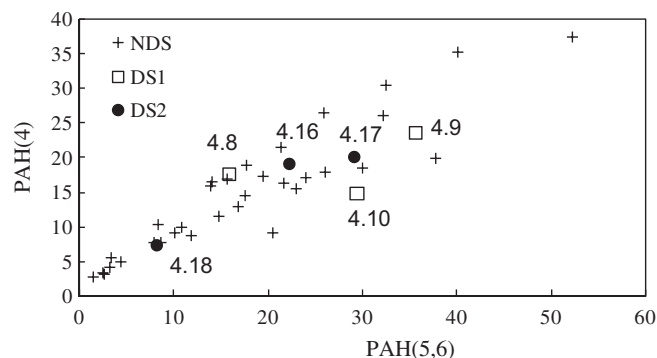


Fig. 6. The ratios of PAH with four rings of PAH (PAH (4)) to 5 and 6 rings (PAH (5, 6)) in Beijing. The number refers the sampling date of the two dust storms.

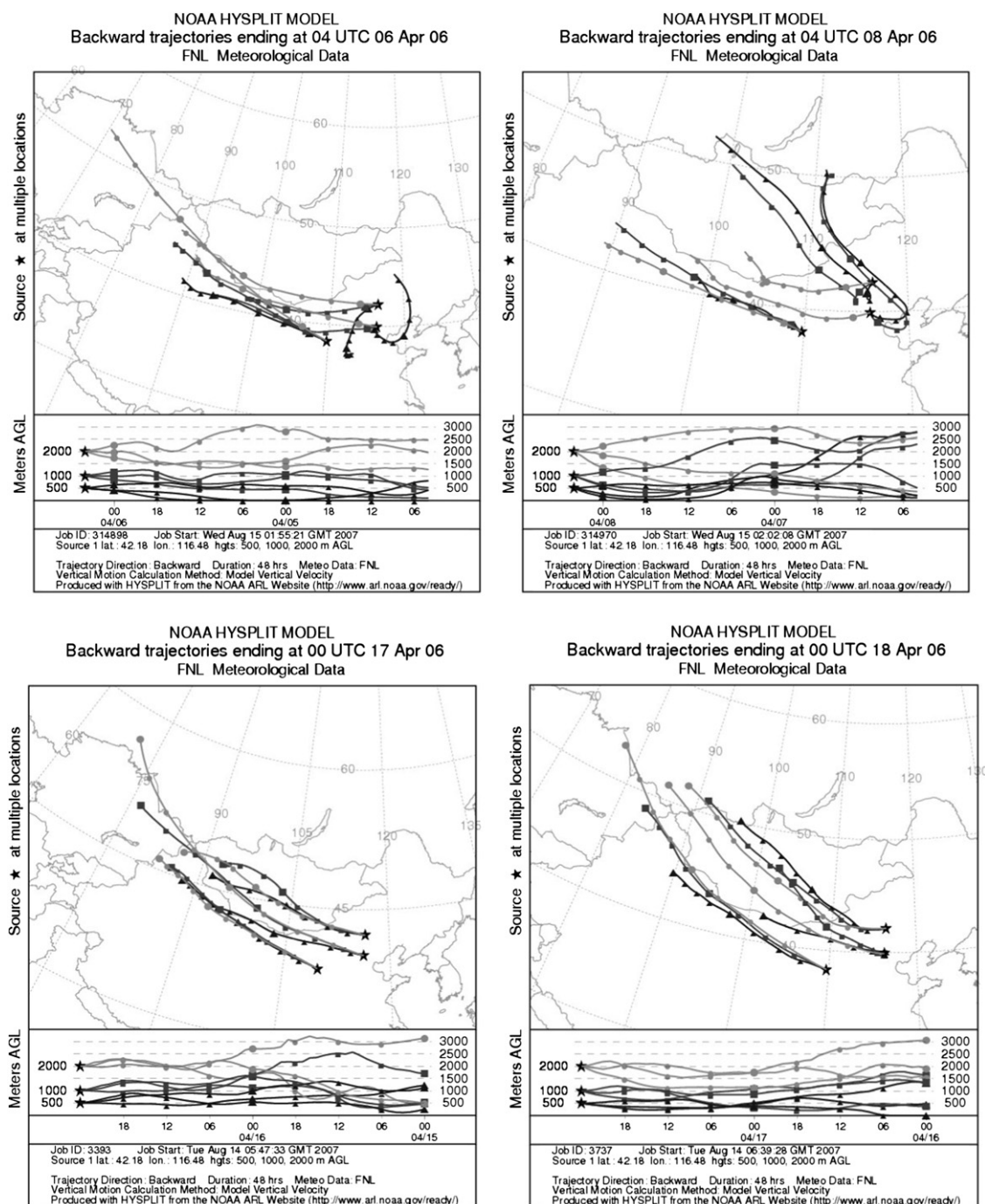


Fig. 7. 48-h backward trajectory from Beijing (BJ), Duolun (DL) and Yunlin (YL) for DS1 (04 UTC 6 April, 04 UTC 8 April) and DS2 (00 UTC 17 April).

Furthermore, as Fig. 5(c) and (d) showed, the EFs of As, Zn, Cu, Cd, Pb, and S in TSP increased from the averages of 34, 27, 5, 90, 52, and 81 in NDS to 81, 178, 10, 171, 198, and 768 on April 10 during DS1, but decreased to 31, 15, 3, 48, 24, and 72 during DS2, respectively. Similarly, the EFs of these elements in PM_{2.5} increased greatly from the averages of 125, 97, 21, 358, 181, and 230 in NDS to 506, 2873, 186, 1766, 1368, and 3143 on April 10 during DS1, but decreased to the averages of 76, 44, 6, 87, 57, and 115 during DS2, respectively. This again verified that DS1 was a “pollution storm” with well mixing between crustal and pollution aerosol, and DS2 was a “crustal” storm in less mixing extent with pollution aerosol than DS1.

As for the ratio of X/Al (X is the element interested) the pollution elements showed different characteristics in NDS, DS1 and DS2. As shown in Fig. 4(b), the ratio of X/Al in TSP was higher in NDS and DS1 than that in DS2. For example, the average ratios of Zn/Al and Pb/Al were from 0.030 and 0.011 in NDS increased to be 0.061 and 0.015 in DS1, and decreased to be 0.016 and 0.005 in DS2. Similarly, in PM_{2.5}, Zn/Al and Pb/Al were 0.146 and 0.047 in NDS, increased greatly to be 0.704 and 0.080 in DS1, and decreased to be 0.052 and 0.015 in DS2, respectively. This again verified the much deeper mixing of crustal with pollutant aerosol in DS1 than in DS2.

3.2.2. The chemical transformation of aerosols in the two dust storms

As shown in Fig. 4(c), sulfate, nitrate, and ammonium of those secondary aerosol ions in Beijing showed weak correlations with Al in NDS and DS2, as the correlation coefficients were 0.2–0.5, while in DS1 there were strong negative correlations of Al with sulfate, nitrate, and ammonium (the R were -0.98 , -1.0 , -0.98 in $PM_{2.5}$ and -0.77 , -0.50 , -0.74 in TSP, respectively). The negative correlations mean that the concentrations of these secondary soluble pollutants decrease with the increase of the crustal aerosol in DS1, which suggested a dilution effect of the pollution species by the dust storm. However, the geometric mean ratios of $[SO_4^{2-}]/Al$, $[NO_3^-]/Al$, and $[NH_4^+]/Al$ in Beijing were 1.39, 0.79, and 0.22 in DS1 and 0.62, 0.25, and 0.08 in DS2, respectively. The geometric mean ratios of $[SO_4^{2-}]/Al$, $[NO_3^-]/Al$, and $[NH_4^+]/Al$ were 0.21, 0.062 and 0.022 in DL and 0.23, 0.074, and 0.0301, respectively, in YL in DS2. These ratios were all greater than 0.044, <0.002 , and <0.002 , respectively, in the surface soil of Taklimakan Desert and 0.002, <0.002 , and <0.002 in the soil of Gobi Desert and Loess Plateau (Nishikawa et al., 1991). These ratios were also significantly higher than 0.12, 0.008, and 0.003 in the sample of the surface dry deposition in DS2. All of these results mentioned above indicated that there were much greater concentrations of those secondary aerosol ions in dust storm DS1 and DS2 than that from their original sources, and there must be other sources of the secondary species, other than the carrying or/and combining of the local dust, during the dust storm DS1 and DS2. Those secondary ion species might be from the reactions of SO_2 , NO_x , NH_3 , sulfuric acid, and nitric acid on the surface of dust particles, or the re-suspension on the pathway to be attached to the dust particles during the long-range transport (Sun et al., 2005), or the overlap of the two stages of “clear-out pollutants” of local pollution and “addition of pure dust” of dust storm (Guo et al., 2004). The ratios of $[SO_4^{2-}]/Al$, $[NO_3^-]/Al$, and $[NH_4^+]/Al$ in DS1 were much higher than that in DS2, indicating again that the mixing extent of dust with pollution aerosol was much deeper in DS1 than that in DS2. It should be noted that the interaction between dust and the precursors of those secondary pollution aerosol, i.e. the heterogeneous reactions on the surface of particles, increased the concentrations of aerosol several days after the peak of the dust storm in Beijing, as shown in Figs. 2 and 3, the weaker and finer DS1 contributed higher concentration of aerosols on April 14, several days after the peak of the dust storm. The finer the particles were, the higher the concentrations of aerosol were. The coarser and stronger DS2 might affect longer and stronger due to the friction and re-suspension of the coarse particles, as indicated with the high concentration of aerosol on April 28.

3.3. Source identification of the two dust storms

To identify the different source regions of the two dust storms during the sampling time, the air mass trajectories were applied using the HYSPLIT 4 model of the Air Resources Laboratory of NOAA (Draxler and Rolph, 2003). For every sampling day, a 48 h backward trajectory was computed at three sites of Beijing (BJ), Duolun (DL) and Yulin (YL) for DS1 (04 UTC 6 April, 04 UTC 8 April) and DS2 (00 UTC 17 April, 00 UTC 18 April), at 500 m, 1000 m and 2000 m above the starting point at ground level with FNL Meteorological data as input (Draxler and Rolph, 2003; <http://www.arl.noaa.gov/ready/hysplit4.html>). Based on the transport pathways of air masses in Fig. 7, two different kinds of transport modes were identified: polluted and non-polluted. It was obviously that DS1 was mostly from southeastern or southwestern direction at lower altitude but western direction at higher altitude. The southeastern and southwestern direction in lower altitude might carry more pollutants, while the western and northwestern direction in higher altitudes might contribute more

crust. Air masses from different directions probably went through a mixing process during the long-range transport before reach the BNU sampling site, which could explain why the mixing extent between crustal and pollutant aerosol in DS1 was so high. As in DS2, the air masses almost traveled from the same direction, the north-west/northern direction, at various altitudes, as inferred from the back trajectory. Thus, the mixing extent between crustal and pollutant aerosols in DS2 should be weaker than that in DS1.

4. Conclusion

Two different types of dust storms were observed: the weaker but more polluted DS1 and the stronger but less polluted DS2. Also, there was a deeper mixing between crustal and pollutant aerosols during their long-range transport in DS1 than in DS2. The correlation between chemical species and Al and between PAH(4) and PAH (5,6) further confirmed the stronger chemical transformation and aerosol mixing in DS1 than that in DS2. Back trajectory and chemical analysis revealed that in DS1 the air masses at Beijing were mostly from southern or southwestern direction at lower altitude with much more pollution, while in DS2 the air masses were mostly from the northwestern and northern direction with dust mainly, which explained why there was a stronger mixing of dust with pollution aerosol in DS1 than that in DS2 over Beijing.

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