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# Mixing of dust with pollution on the transport path of Asian dust – Revealed from the aerosol over Yulin, the north edge of Loess Plateau

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## ABSTRACT

Both PM<sub>2.5</sub> and TSP were monitored in the spring from 2006 to 2008 in an intensive ground monitoring network of five sites (Tazhong, Yulin, Duolun, Beijing, and Shanghai) along the pathway of Asian dust storm across China to investigate the mixing of dust with pollution on the pathway of the long-range transport of Asian dust. Mineral was found to be the most loading component of aerosols both in dust event days and non-dust days. The concentrations of those pollution elements, As, Cd, Pb, Zn, and S in aerosol were much higher than their mean abundances in the crust even in dust event days. The high concentration of SO<sub>4</sub><sup>2−</sup> could be from both sources: one from the transformation of the local emitted SO<sub>2</sub> and the other from the sulfate that existed in primary dust, which was transported to Yulin. Na<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup> were mainly from the crustal source, while NO<sub>3</sub><sup>−</sup> and NH<sub>4</sub><sup>+</sup> were from the local pollution sources. The mixing of dust with pollution aerosol over Yulin in dust event day was found to be ubiquitous, and the mixing extent could be expressed by the ratio of NO<sub>3</sub><sup>−</sup>/Al in dust aerosol. The ratio of Ca/Al was used as a tracer to study the dust source. The comparison of the ratios of Ca/Al together with back trajectory analysis indicated that the sources of the dust aerosol that invaded Yulin could be from the northwestern desert in China and Mongolia Gobi.

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## 1. Introduction

Asian dust can be transported to the Northern Pacific, even to North America (Duce et al., 1980, 1991). Asian dust and its long-range transport can impact on the air quality and human health of those downstream densely populated areas (Cao et al., 2005; Meng and Bin, 2007; Zhang et al., 2005) and play an important role in biogeochemical cycles (Zhuang et al., 1992). The dust can also impact on the climate directly by influencing the absorption and scattering of solar radiation (Kim et al., 2006; Du et al., 2008) and indirectly by acting as cloud condensation nuclei (Arimoto, 2001). During the transport, dust aerosol can mix with anthropogenic pollutant (Zhang et al., 1994; Dentener et al., 1996; Song and Carmichael, 2001; Zhang and Iwasaka, 2001). The composition and morphology of dust particles can be changed by its reactions with acid gases like SO<sub>2</sub>, NO<sub>x</sub>, and HCl (Chou et al., 2008), and particulate sulfate/nitrate can be enhanced (Zhang et al., 2000; Usher et al., 2002). This interaction depends in part on the size distribution and chemical composition of dust particles (Quan and Zhang, 2008; Jae-In and Soon-Ung, 2008; Shi et al., 2008) and may alter the nitrogen and sulfur cycles and acid/base balance (Parungo et al., 1996; Iwasaka et al., 2003; Chung et al., 2003).

The Loess Plateau in China, located on the transport pathway of Asian dust from the northern and northwestern China to the Northern Pacific (Zhang et al., 2008), is one of the sources of Asian dust (Zhuang et al., 2001; Zhang et al., 2008). It is also the depositional place of dust from both the Taklimakan desert and Mongolia Gobi (Liu et al., 1997; Zhang et al., 1993; Sun et al., 2001). The dust deposited in the Loess Plateau has been shown to be from Tarim Basin (Liu et al., 1997). However, other studies show that the Gobi desert in North China and Mongolia is one of the main sources of dust in the Loess Plateau (Sun et al., 2001). Yulin (YL), located at the north edge of the Loess Plateau, is a good representative place in the study of dust aerosol over the Loess Plateau (Wang et al., 2005; Cao et al., 2008). During the Campaign of Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia), the chemical, physical, and radiative characteristics of aerosol over YL were studied and the aerosol was shown to be polluted because of the enrichment of mines and industry pollution (Zhang and Iwasaka, 2001; Alfaro et al., 2003; Zhang et al., 2003; Xu et al., 2004; Arimoto et al., 2006). However, the mixing extent of mineral aerosol with pollution and the sources of dust that invaded YL have not been well studied. In this study, both PM<sub>2.5</sub> and TSP were monitored in the spring from 2006 to 2008 in an intensive ground monitoring network of five sites (Tazhong, Yulin, Duolun, Beijing, and Shanghai) along the pathway of the dust storm across China. YL, as the representative of Loess Plateau and the pathway of Asian dust, was the

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focus of this study to investigate the mixing of dust aerosol with pollution on the pathway of the long-range transport of Asian dust.

## 2. Experiments

### 2.1. Sampling

Aerosol samples of PM<sub>2.5</sub> and TSP were collected in an intensive ground monitoring network of five sites (Tazhong (TZ), Yulin (YL), Duolun (DL), Beijing (BJ), and Shanghai (SH)) along the pathway of dust storm across China (Fig. 1) in the spring of 2006, 2007, and 2008 to monitor synchronously the dust episodes. The samplers were set up at a place about 10 m above the ground. Aerosol samples were collected for 24 h (normally from 8:00 a.m. to 8:00 a.m. the next day; in the dust storm episode it could be 1–3 h for a sample depending on the intensity of the dust storm) on Whatman 41 filters (Whatman Inc., Maidstone, UK) by a medium-volume sampler (model: (TSP/PM<sub>10</sub>/PM<sub>2.5</sub>)<sup>-2</sup>, flow rate: 77.59 l min<sup>-1</sup>). The samples were put in polyethylene plastic bags immediately after sampling and then reserved in a refrigerator. All of the filters were weighed before and after sampling with an analytical balance (Sartorius BT 25s, reading precision 10 µg) after stabilizing under constant temperature (20 ± 1 °C) and humidity (40 ± 2%) for over 24 h. All the procedures were strictly quality controlled to avoid any possible contamination of the samples.

### 2.2. Chemical analysis

#### 2.2.1. Ion analysis

One-fourth of each sample and the blank filter were extracted ultrasonically by 10 ml deionized water (18 MΩ cm<sup>-1</sup>). Eleven inorganic ions (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>) and four organic acids (formic, acetic, oxalic, and methylsulphonic acid (MSA)) were analyzed by Ion Chromatography (IC; Dionex ICS 3000, USA) which consists of a separation column (Dionex Ionpac AS 11 for Anion, Dionex IonPac CS 12A for Cation), a guard column (Dionex Ionpac AG 11 for Anion, Dionex IonPac AG 12A for Cation), a self-regenerating suppressed conductivity detector (Dionex Ionpac ED50), and a gradient pump (Dionex Ionpac GP50). The detail procedures were given elsewhere (Yuan et al., 2003).

#### 2.2.2. Element analysis

Half of each sample filter was digested at 170 °C for 4 h in a high-pressure Teflon digestion vessel with 3 ml concentrated HNO<sub>3</sub>, 1 ml concentrated HClO<sub>4</sub>, and 1 ml concentrated HF. After cooling, the solutions were dried, and then diluted to 10 ml with deionized water (18 MΩ cm<sup>-1</sup>). Total 19 elements (Al, Fe, Mn, Mg, Ti, Sc, Na, Sr, Ca, Co, Cr, Ni, Cu, Pb, Zn, Cd, V, S, and As) were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES; Model: ULTIMA, JOBIN-YVON Company, France). The detailed analytical procedures were given elsewhere (Zhuang et al., 2001, 2003; Sun et al., 2004a, b).

#### 2.2.3. Meteorological data

The meteorological data, including wind speed, wind direction, etc., was downloaded from <http://www.wunderground.com>. The information of remote detection from satellites was downloaded from <http://www.duststorm.com.cn>.

## 3. Results and discussion

### 3.1. The concentration of TSP and PM<sub>2.5</sub> during dust event and non-dust days

During the sampling time there were 21 dust event days (DS), March 31st–April 1st, April 5th–6th, April 11th–12th, April 17th–19th, April 23rd, April 25th, April 27th–28th, April 20th–May 1st in 2006, March 30th–April 1st, April 15th, April 19th in 2007, April 1st in 2008 and 61 non-dust days (NDS) according to the meteorological data and the remote sensing from satellites. In NDS, the daily concentration of TSP ranged from 25.0 to 369.1 µg/m<sup>3</sup>. The average concentration was 160.3 µg/m<sup>3</sup>, higher than the health-based national air quality standard of 120 µg/m<sup>3</sup>. The average concentration of PM<sub>2.5</sub> in NDS was 56.7 µg/m<sup>3</sup> (ranging from 7.1 to 218.9 µg/m<sup>3</sup>), much higher than USA EPA's standard of 35 µg/m<sup>3</sup>. Compared to those seriously air-polluted cities, Urumqi (Li et al., 2008), Beijing, Shanghai (Wang et al., 2007), and Lanzhou (Chu et al., 2008), the aerosol concentration over YL in NDS was not so high, however, it increased greatly in DS. The average TSP concentration in DS was 591.0 µg/m<sup>3</sup> (ranging from 104.3 to 1464.3 µg/m<sup>3</sup>), 3.7 times of that in NDS, while PM<sub>2.5</sub> was 159.5 µg/m<sup>3</sup> (ranging from 35.5 to 561.0 µg/m<sup>3</sup>), 2.8 times of that in NDS. The TSP concentrations in DS over YL were comparable to those in dust

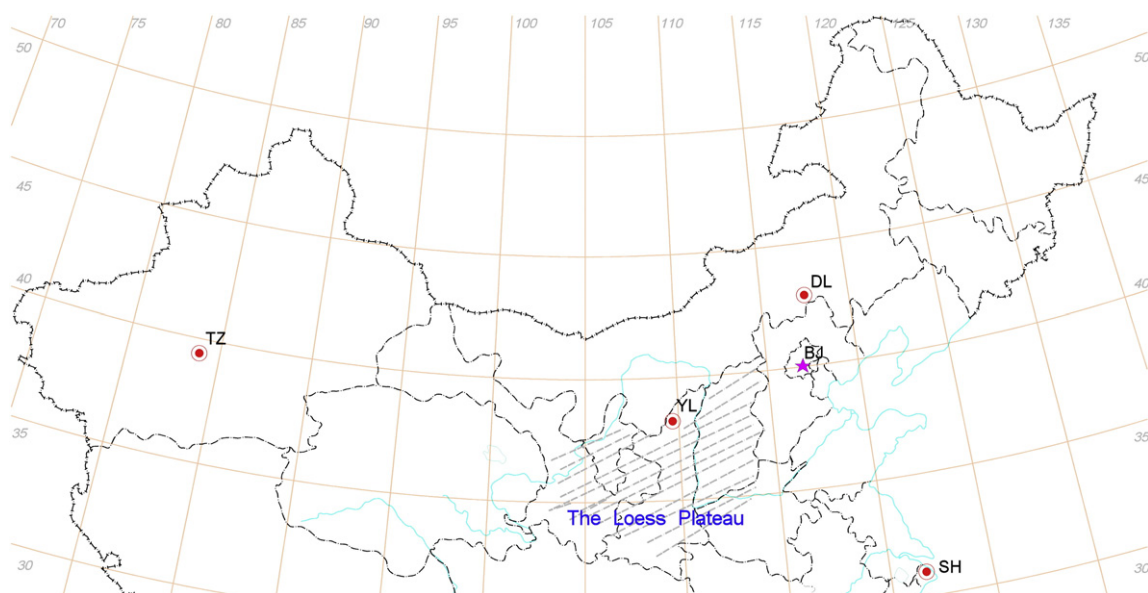


Fig. 1. Map of the sampling sites and the Loess Plateau, China.

storms ( $317\text{--}1000\text{ }\mu\text{g}/\text{m}^3$ ) over Dunhuang, a place nearby the dust source, and those in dust storms ( $260\text{--}1000\text{ }\mu\text{g}/\text{m}^3$ ) over Zhengbaitai in YL (Zhang et al., 2003; Duvall et al., 2008). The high concentrations of  $\text{PM}_{2.5}$  and TSP indicated that there has been serious air pollution over YL.

### 3.2. Aerosol composition

#### 3.2.1. Elemental composition

Table 1 showed the average daily mass concentrations and mass loading percentage, the ratio of the mass of each element to the mass of the aerosol, of 19 elements in aerosol in DS, and NDS. For comparison, the mean abundances in the crust of each element (Lida, 2005/2006) were also listed. Those elements, Al, Fe, Ca, Na, and Mg, which were usually thought to be dust-derived, had high concentrations both in DS and NDS, and the mass loadings of these five elements were higher than 1%. The concentrations and mass loadings of these five crustal elements increased greatly from NDS to DS, in both TSP and  $\text{PM}_{2.5}$ , and the mass loadings were more closed to their abundances in DS than in NDS (except Ca), indicating that these five elements were from the crustal source. The mass loading of Ca in aerosol was much higher than its mean abundance in the crust in both DS and NDS, showing that dust aerosol over YL had a high content of Ca, which was the feature of the dust aerosol over YL. Earlier studies had shown that Loess (Arimoto et al., 2004) and aerosol over YL (Arimoto et al., 2006) had a high percentage of Ca. Thus, the high content of Ca in dust aerosol could be used as a tracer to identify the source of dust aerosol.

The mass loadings of elements Ti, P, Mn, Sr, Cr, Co, V, and Ni were higher than 0.01% and lower than 1%. The concentrations and mass loadings of these 8 elements increased obviously from NDS to DS, showing that these elements were mainly from the crustal source. The sources of these crustal elements were further studied by enrichment factors (EFs) shown in Fig. 2, which were calculated by using Al as

reference. Elements, including Fe, Na, Ti, P, Mg, Co, V, Ni, Mn, Sr, Cr, and Ca, had EFs of  $<5$ , indicating that these elements were mostly from crustal dust. In NDS the mass loadings of most crustal elements were much lower than their mean crustal abundances, indicating that during NDS aerosol over YL contained more pollution components. Furthermore, even in DS the mass loadings of most crustal elements were lower than their abundances in the crust, indicating that those pollution components were significantly mixed with those mineral components in the dust aerosol over YL.

Compared to those crustal elements, the mass loadings of the pollution elements, Pb, Zn, Cd, S, and As, in aerosols mainly decreased from NDS to DS due to the dilution of the invaded dust in DS, with the high loadings of 0.043, 0.16, 0.00088, 0.058, and 2.35 for Pb, Zn, Cd, As and S, respectively, in  $\text{PM}_{2.5}$  and 0.027, 0.094, 0.00048, 0.0066, and 1.79% in TSP in NDS. However, even in dust event days their mass loadings were much higher than their mean abundances in the crust. In  $\text{PM}_{2.5}$ , Pb, Zn, Cd, As, and S in DS were 11.0, 21.1, 31.4, 106.3, and 35.0 times of their abundances in the crust, while in TSP were 4.7, 3.4, 11.0, 23.5, and 17.1. These results indicated that these elements mainly from pollution sources, e.g. As and S were mainly from coal and oil combustion used in heating and industry, while Pb was associated to traffic and oil combustion (Borbély-Kiss et al., 1998; Hien et al., 2001). Compared to other studies, the concentrations of these pollution elements were lower than those in the mega-cities in China (Fu et al., 2008; Zhuang et al., 2001), but much higher than those in Tibetan Plateau, which were 0.0018, 0.00004, and  $0.101\text{ }\mu\text{g}/\text{m}^3$  for Zn, As, and S, respectively (Cong et al., 2007), indicating further that in the dust aerosols over YL these pollution components were mixed with those mineral components.

The EFs of Pb, Zn, Cd, S, and As in  $\text{PM}_{2.5}$  in DS were greater than 10 and were even greater than 100 in NDS, indicating these elements were mainly from the pollution source. The EFs of these elements in NDS were much higher than that in DS, which further indicated that these elements were mostly from those local pollution sources in YL.

**Table 1**

The mean concentrations ( $\mu\text{g}/\text{m}^3$ ) and the mass loading percentage (%) of elements and water-soluble ions in DS and NDS.

	Abundance /(%)	Concentration ( $\text{PM}_{2.5}$ )		Percentage ( $\text{PM}_{2.5}$ )		Concentration (TSP)		Percentage (TSP)	
		DS	NDS	DS	NDS	DS	NDS	DS	NDS
Al	8.23	10.86	1.70	6.57	3.65	36.38	6.91	6.21	4.25
Fe	5.63	6.88	0.99	3.98	2.05	20.31	3.95	3.59	2.46
Ca	4.15	10.22	2.07	6.81	4.60	25.61	7.90	4.80	5.04
Na	2.36	2.41	0.48	1.49	1.04	10.45	1.79	1.79	1.10
Mg	2.33	3.26	0.59	2.05	1.28	9.84	2.07	1.74	1.28
Ti	0.565	0.68	0.10	0.41	0.22	2.53	0.43	0.43	0.26
P	0.105	0.17	0.03	0.10	0.06	0.54	0.12	0.09	0.07
Mn	0.095	0.18	0.03	0.11	0.08	0.48	0.11	0.08	0.07
Sr	0.037	0.08	0.02	0.05	0.03	0.21	0.06	0.04	0.04
Cr	0.0102	0.03	0.01	0.02	0.02	0.06	0.01	0.01	0.01
Co	0.0025	0.01	0.00	0.00	0.00	0.01	0.00	0.00	0.00
V	0.012	0.02	0.00	0.01	0.01	0.06	0.01	0.01	0.01
Ni	0.0084	0.01	0.00	0.01	0.01	0.03	0.00	0.00	0.00
Cu	0.006	0.01	0.01	0.01	0.02	0.03	0.01	0.01	0.01
Pb	0.0014	0.02	0.02	0.02	0.05	0.04	0.05	0.01	0.03
Zn	0.007	0.21	0.09	0.15	0.16	0.12	0.15	0.02	0.10
Cd	0.000015	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
As	0.00018	0.02	0.02	0.02	0.06	0.02	0.01	0.00	0.01
S	0.035	1.75	1.29	1.22	2.35	3.27	2.80	0.60	1.79
$\text{SO}_4^{2-}$		5.00	5.02	3.53	8.96	10.93	9.09	2.03	5.76
$\text{NO}_3^-$		1.05	1.64	0.81	3.33	1.45	3.86	0.30	2.51
$\text{Cl}^-$		1.05	0.99	0.78	2.06	2.29	1.43	0.41	1.00
$\text{NO}_2^-$		0.23	0.35	0.17	0.79	0.31	0.47	0.06	0.30
$\text{F}^-$		0.14	0.16	0.11	0.36	0.56	0.53	0.10	0.44
$\text{Ca}^{2+}$		3.99	2.70	3.23	6.01	6.43	6.37	1.30	4.38
$\text{NH}_4^+$		0.52	2.28	0.45	4.08	0.54	3.34	0.12	2.20
$\text{Na}^+$		1.40	0.62	1.02	1.41	3.71	1.14	0.67	0.76
$\text{K}^+$		0.31	0.37	0.23	0.78	0.50	0.59	0.09	0.40
$\text{Mg}^{2+}$		0.32	0.26	0.25	0.60	0.60	0.48	0.12	0.33



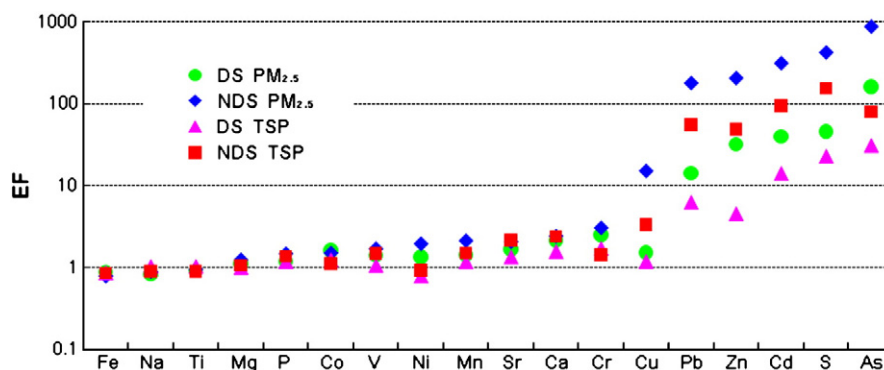


Fig. 2. Enrichment factors (EFs) of elements in PM<sub>2.5</sub> and TSP.

In DS, local pollution was diluted by the invaded mineral dust, resulting in the decrease of EFs of these elements. The comparison of the correlation coefficients between pollution elements and Al analyzed for all the samples collected in the whole year and for those samples collected only in NDS was used to further identify the sources of these pollution elements. The correlation analysis showed that Cd, Pb, As, and Zn were not correlated with Al in DS and NDS, implicating that Cd, Pb, As, and Zn were mainly from the local pollution source or from local dust, in which they were previously deposited (Cao et al., 2008). Unlike these four elements, the correlation coefficients of S with Al were 0.96 and 0.58 in 2006 and 2007, respectively, for PM<sub>2.5</sub> aerosols collected in the whole year, while for TSP were 0.73 and 0.57, indicating that S in the dust aerosol over YL was partially from the mineral dust.

### 3.2.2. Water-soluble ions

Table 1 showed the average daily mass concentrations and mass loading percentage, the ratio of the mass of each ion to the mass of the aerosol, of those major water-soluble ions, in DS and NDS. The concentrations of Ca<sup>2+</sup>, Na<sup>+</sup>, and Mg<sup>2+</sup>, increased from NDS to DS. The concentrations of Ca<sup>2+</sup>, Na<sup>+</sup>, and Mg<sup>2+</sup> in PM<sub>2.5</sub> in DS were 1.5, 2.3, and 1.2 times of that in NDS, while in TSP were 1.0, 3.3, and 1.3 times. Besides, Ca<sup>2+</sup>, Na<sup>+</sup>, and Mg<sup>2+</sup> highly correlated with Al, with correlation coefficients of 0.56–0.89 for Na<sup>+</sup>, 0.47–0.94 for Mg<sup>2+</sup>, and 0.57–0.97 for Ca<sup>2+</sup>. These results suggested that these three ions were mainly from the crustal source (Wang et al., 2006). Na<sup>+</sup> in the aerosol over YL was greater than that in those areas nearby desert (Shen et al., 2007), which might be owing to the fact that there were a number of Salinas around YL (Cao et al., 2008).

SO<sub>4</sub><sup>2−</sup> was the most abundant among all of those ions in both PM<sub>2.5</sub> and TSP. Like the crustal components, the concentration of SO<sub>4</sub><sup>2−</sup> increased from NDS to DS. Also, SO<sub>4</sub><sup>2−</sup> was highly correlated with Al during DS (the correlation coefficient was 0.55–0.94 for all samples in each year), but not correlated with Al if only for NDS. These results indicated that SO<sub>4</sub><sup>2−</sup> was partially from the “crustal” source, i.e. from the emitted mineral dust, and partially from the anthropogenic pollution source, the transformation of SO<sub>2</sub> into SO<sub>4</sub><sup>2−</sup> in the atmosphere. However, even in strong DS samples, the ratio of S in SO<sub>4</sub><sup>2−</sup> to the total S in the aerosol was 0.69 to 1.00, which meant that most of S in the invaded dust was in the form of SO<sub>4</sub><sup>2−</sup>. Our recent study indicated that the dust aerosol from Taklimakan contained most of the sulfur in the form of sulfate, which was from the emitted primary dust in the desert (unpublished data). The high concentration of SO<sub>4</sub><sup>2−</sup> in dust aerosols over YL could be from two sources: one from the absorption of gaseous SO<sub>2</sub>/H<sub>2</sub>SO<sub>4</sub> and the consequent transformation to sulfate on the surface of the dust aerosol (Arimoto et al., 2004); the other was from the “crustal” source as mentioned earlier (Wang et al., 2006; Xu et al., 2004). Cl<sup>−</sup>, NO<sub>2</sub><sup>−</sup>, F<sup>−</sup>, and K<sup>+</sup> correlated with Al in DS (the correlation coefficient was 0.57–0.90 for Cl<sup>−</sup>, 0.46–0.52 for NO<sub>2</sub><sup>−</sup>, 0.49–0.78 for F<sup>−</sup>, and 0.41–0.99 for K<sup>+</sup> for all samples in each year) but not

correlated with Al in NDS, indicating that these ions were from both pollution and crustal sources. As for NO<sub>3</sub><sup>−</sup> and NH<sub>4</sub><sup>+</sup>, both concentrations and mass loadings in DS were much lower than that in NDS, which might be due to the dilution of invaded dust in dust days, and these two ions were not correlated with Al in both PM<sub>2.5</sub> and TSP, indicating that these two ions were mainly from the anthropogenic pollution source.

### 3.2.3. Major components of aerosol

In order to study the major components of the dust aerosol over YL, the concentration of minerals was calculated based on the formula: Conc. = [Al]/0.08, as 8% is the mean abundance of the Al in the crust. The average mass contributions of minerals to PM<sub>2.5</sub> were 82.2% and 44.7% in DS and NDS, respectively, and to TSP were 84.1% and 60.3%, indicating that minerals were the most loading component in aerosols over YL in both DS and NDS. The contributions of minerals were lower than unity even in DS, indicating that there was a lot of pollution aerosol that mixed to the mineral in both NDS and DS. YL is surrounded by a number of big coal mines, and the coal mining, the industry emission, and the domestic heating would contribute much pollution aerosol there. The decrease of the contribution of minerals to the total mass of the dust aerosol from DS to NDS over YL indicated that more pollutants from the local anthropogenic sources in NDS were mixed with the dust than that in DS.

### 3.3. Mixing of mineral dust with pollution aerosol at YL

#### 3.3.1. Evidence from the composition of aerosols

As mentioned earlier, during the DS the dust aerosols over YL contained a lot of pollutants. As shown in Table 1, the mass loadings of pollution elements in aerosol were much higher than their mean abundance in the crust even in DS, implicating the mixing of mineral dust with pollution aerosol in DS. The contributions of crustal components to the total mass of the dust aerosol increased in DS, while of the pollution components decreased in DS, clearly showing that the mixing between mineral aerosol and pollution during dust events was ubiquitous.

#### 3.3.2. Evidence from the variation of the secondary aerosols

SO<sub>4</sub><sup>2−</sup> and NO<sub>3</sub><sup>−</sup> were considered to be mainly from chemical transformation in the atmosphere, and their variations are good indicators for the mixing of mineral aerosols with pollution. The variations of SO<sub>4</sub><sup>2−</sup> and NO<sub>3</sub><sup>−</sup> concentrations in the entire study time, together with Al were shown in Fig. 3. As mentioned earlier, in the dust aerosol over YL SO<sub>4</sub><sup>2−</sup> was from both pollution and mineral dust, while NO<sub>3</sub><sup>−</sup> was mainly from local pollution. SO<sub>4</sub><sup>2−</sup> increased obviously with the increase of Al in DS. On March 30, 2007 the concentration of SO<sub>4</sub><sup>2−</sup> was up to 4.3 times of the average in NDS in TSP. These results implicated that SO<sub>4</sub><sup>2−</sup> could largely be brought by the invaded dust in

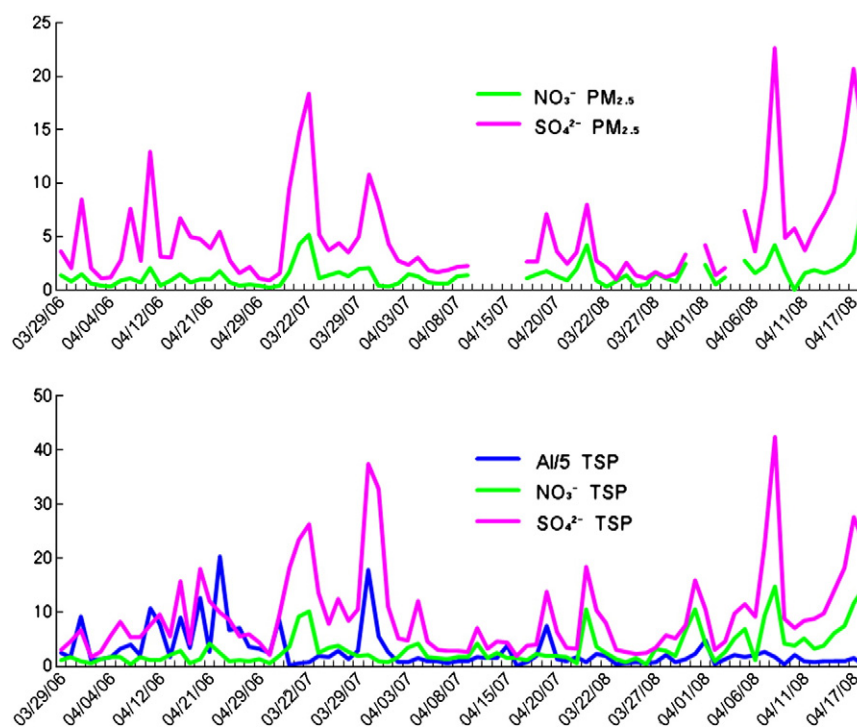


Fig. 3. Variations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and Al in the study time. (The missing data were due to a malfunction of the sampler.)

DS, i.e. from the primary dust, while in NDS,  $\text{SO}_4^{2-}$  mainly increased with the increase of  $\text{NO}_3^-$ , showing that  $\text{SO}_4^{2-}$  was from the pollution source in NDS, i.e. the interaction of  $\text{SO}_2$  with the alkaline component of the mineral dust like  $\text{CaCO}_3$ , as  $\text{NO}_3^-$  did (Wang et al., 2007). As there are many large coal mines located around YL, the use of coal to heating and cooking was the major sources of the local pollutants (Hou et al., 2006; Xu et al., 2004; Shi et al., 2008). Study showed that in winter  $\text{SO}_2$  from coal mine could contribute a large proportion to the aerosol in Xi'an, which is near YL (Zhang et al., 2002). The concentration of  $\text{NO}_3^-$  decreased in DS, which was due to the dilution of  $\text{NO}_3^-$  by the invaded dust. The average contributions of  $\text{NO}_3^-$  to the total mass in DS were 0.8% in  $\text{PM}_{2.5}$  and 0.3% in TSP, greater than that in the Taklimakan desert (0.5% in  $\text{PM}_{2.5}$  and 0.1% in TSP). This result also indicated that the interaction between acid gases and dust aerosol occurred in DS at YL.

### 3.3.3. Mixing extent

$\text{NO}_3^-$  is one of the major secondary species and is mainly from the oxidation of  $\text{NO}_x$ . The ratio of  $\text{NO}_3^-/\text{Al}$  and  $\text{SO}_4^{2-}/\text{Al}$  had been reported to be a possible index to show the variation degree of these species due to the chemical reactions (Wang et al., 2006). As shown

in Fig. 4, the variation of  $\text{NO}_3^-$  was similar to almost all of those pollutants derived from the local anthropogenic source, such as Pb, Cd, Zn, and  $\text{NH}_4^+$ , and  $\text{NO}_3^-$  could be used to be the representative of the local pollution. Al is a typical crustal component, thus, the ratio of  $\text{NO}_3^-/\text{Al}$  could be used to express the mixing extent of pollution aerosol with mineral dust in the dust aerosol. Table 2 showed the calculated ratios of  $\text{NO}_3^-/\text{Al}$  in both DS and NDS. For comparison the ratios of  $\text{NO}_3^-/\text{Al}$  in those aerosols collected in other sites in the same period as in YL were also listed in Table 2. The ratio in  $\text{PM}_{2.5}$  was much higher than those in TSP in both DS and NDS, i.e. the mixing extent in fine particles was greater than that in coarse particles, indicating that the pollution components were more in fine particles and the interaction processes were more important in fine particles than in the coarse particles. The ratios of  $\text{NO}_3^-/\text{Al}$  were much higher in NDS than in DS, especially in TSP, implying that pollutants were diluted by the invaded dust during dust events. During the dust period of March 30 to April 1 in 2007, the ratios of  $\text{NO}_3^-/\text{Al}$  were 0.35 in the mixing dust aerosol on March 29 before the coming of the invaded dust, 0.71 in the first dust aerosol sample on March 30, 0.07 in the second sample on March 30, 0.04 in the third on March 30, 0.07 in the dust aerosol on March 31, 0.09 in the dust

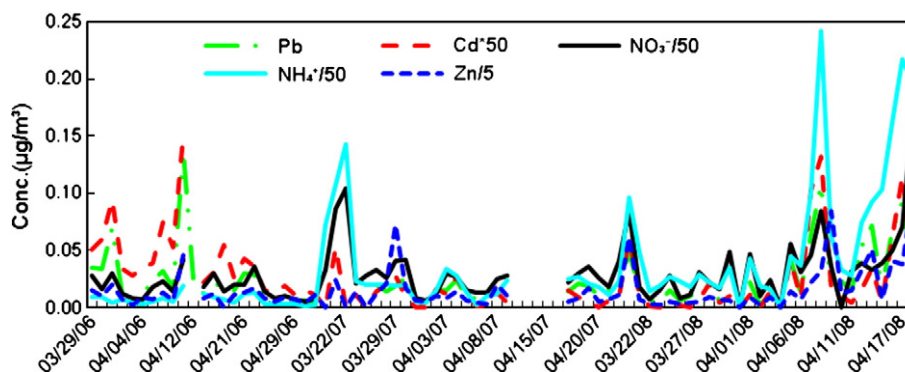


Fig. 4. Variations of the local pollution species Pb, Cd, Zn,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$ . (The missing data were due to a malfunction of the sampler.)

**Table 2**  
The ratios of  $\text{NO}_3^-/\text{Al}$  in aerosol at different sites.

Sites	Time	$\text{NO}_3^-/\text{Al}$ ( $\text{PM}_{2.5}$ )	$\text{NO}_3^-/\text{Al}$ (TSP)	Sites	Time	$\text{NO}_3^-/\text{Al}$ ( $\text{PM}_{2.5}$ )	$\text{NO}_3^-/\text{Al}$ (TSP)
YL, DS	2006	0.1	0.04	TZ, DS	2007	0.07	0.06
YL, DS	2007	0.19	0.05	TZ, DS	2008	0.16	0.08
YL, DS	2008	0.44	0.17	DL, NDS	2007	1.02	0.51
YL, NDS	2006	0.24	0.19	DL, NDS	2008	3.97	1.11
YL, NDS	2007	1.23	0.71	SH	2007	13.19	3.98
YL, NDS	2008	5.08	1.41	BJ	2007	7.63	1.9
DL, DS	2007	0.17	0.03				

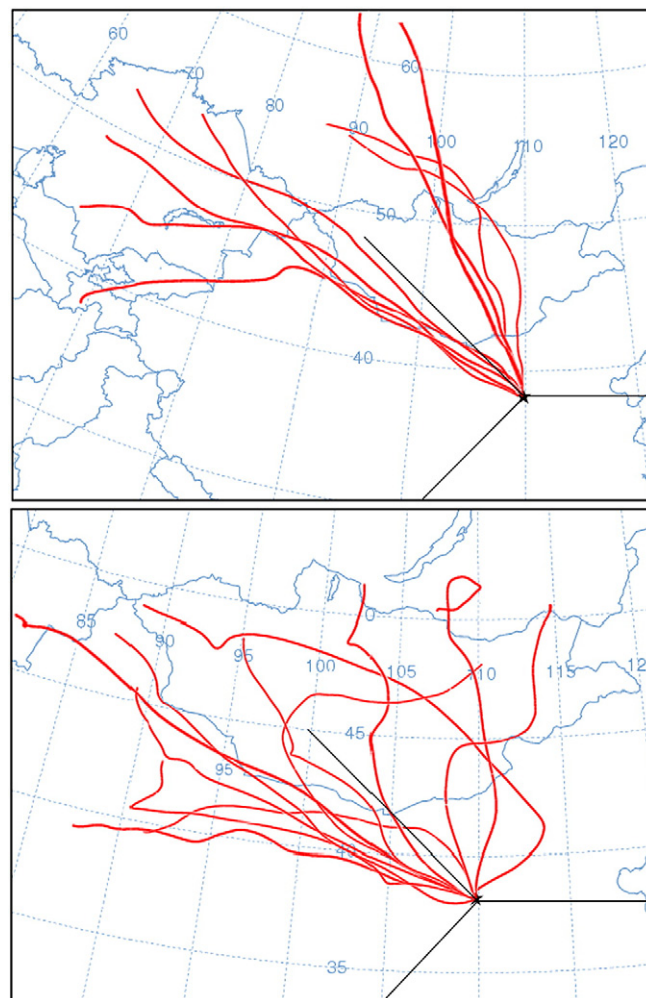
aerosol on April 1, and 0.74 in the dust aerosol on April 2. The ratio decreased gradually as the intensity of dust increased, and then increased with the mixing and interaction between dust and the pollution aerosol, showing that the mixing extent increased. The average ratios of  $\text{NO}_3^-/\text{Al}$  in NDS were 0.24, 1.23, and 5.08 in 2006, 2007, and 2008 in  $\text{PM}_{2.5}$  and 0.19, 0.71, and 1.41 in TSP, increasing sharply from 2006 to 2008. This was firstly due to the fact that the year of 2006 was the most dusty year and the year of 2008 had less dusty days among recent years. And also this was the indication of the increase of the pollution aerosol emitted from the local source from 2006 to 2008.

As shown in Table 2, the ratio of  $\text{NO}_3^-/\text{Al}$  in YL was lower than that in Beijing and Shanghai, indicating that the air pollution in Beijing and Shanghai was more serious than in YL. However, the ratios of  $\text{NO}_3^-/\text{Al}$  in YL were higher than that in DL, located nearby deserts in Inner Mongolia, and that in Tongliao, located in a desert area (Shen et al., 2007). Also, the ratios of  $\text{NO}_3^-/\text{Al}$  in YL in both  $\text{PM}_{2.5}$  and TSP were higher than that in TZ, the center of the Taklimakan desert. These results indicated that the mixing extent of the pollution aerosol with mineral dust at YL was greater than that in those desert areas, such as DL, TZ, and Tongliao in China. It could be seen clearly that the mixing on the pathway of the long-range transport of Asian dust from west to east would make the dust aerosols much polluted.

### 3.4. Sources of the invaded dust

Fig. 5 showed a 3-day back trajectory of each dust day during the sampling time over Yulin. It could be seen that the dust that invaded YL from the long-range transport was from the direction of W–NW and N–EN in dust event days. Dust from the W–NW direction could be from the deserts in western/northwestern China or southwestern Mongolia, while dust from the N–EN direction could be from the deserts in middle Inner Mongolia in China or eastern Mongolia.

In addition to the back trajectory, several studies have shown that the ratios of elements, Ca, Mg, Fe, and Ti to Al could be used as tracers of dust sources (Sun et al., 2004a,b; Han et al., 2005; Shen et al., 2007). The ratios of these four crustal-driven elements to Al were calculated and the scatter plots of the ratios of Ca/Al, Fe/Al, Mg/Al, and Ti/Al against Al in TSP over YL were shown in Fig. 6. Obviously, the ratios of Fe/Al, Ti/Al, and Mg/Al varied slightly among DS and NDS, while the ratio of Ca/Al decreased obviously during DS. The decrease of Ca/Al from NDS to DS showed the significant influence of the invaded dust from the sources outside the city on dust aerosol over YL. Furthermore, as shown in Fig. 7, the ratio of Ca/Al tended to be high when wind was almost from the 90 to 225° direction of YL, where the Loess Plateau is located, while the ratio of Ca/Al tended to be low when wind was mostly from the north or northwest or northeast direction. Those aerosol samples collected on these days (March 21 in 2007, April 4, April 7, April 14, and April 16 in 2008), when 100% of the wind was from the 90 to 225° direction of YL, i.e. from the Loess Plateau, were the representative aerosol of the Loess Plateau. From the elemental analysis of the aerosols collected in these five days, the ratio of Ca/Al in local dust from the Loess Plateau was estimated to be 1.26 in  $\text{PM}_{2.5}$  and 1.28 in



**Fig. 5.** 3-day back trajectories of the dust events and the numbers of the aerosol samples from each dust event over Yulin.

TSP, which was comparable to that (1.22) in the fugitive dust in the Loess Plateau (Cao et al., 2008) and could be used as the representative value of the Loess Plateau. As shown in Fig. 5 the back trajectories of the dust that invaded YL from the long-range transport were mainly from the direction of W–NW and N–EN in DS. The ratio of Ca/Al in the dust from N–EN was lower than that from W–NW. Thus, the transported dust from sources outside YL could be identified as two sources with different characteristics: one from W–NW and the other from N–EN. The dust that invaded YL on April 11, 12, 19, and 23, May 1 in 2006, and April 1 and 15 in 2007 was mainly from N–EN based on the meteorological data and the back trajectory analysis, while the dust that invaded YL on March 31, April 1, 5, 6, 17, 18, 25, 27, 28, and 30, May 1 in 2006, March 30, 31, and 19 in 2007, and April 1 in 2008, was mainly from W–NW. The dust aerosol collected on April 11, 2006, when almost 100% of the wind was from N–EN, could be used as the representative of the dust from the N–EN source, and the ratio of Ca/Al in this dust aerosol was 0.50 in TSP and 0.61 in  $\text{PM}_{2.5}$ . This ratio was comparable to that of the dust collected from the Duolun, Hunshandake desert, which is part of Mongolia Gobi and located in N–EN of YL, for it was 0.47 and 0.61 in TSP and  $\text{PM}_{2.5}$  in non-dust days as measured in this study. Since Hunshandake desert is part of Mongolia Gobi, one of the two major sources of Asian dust, these results could further confirm that the long-range transported dust from N–EN was likely from Mongolia Gobi, which had a low ratio of Ca/Al. Like the dust aerosol collected on April 11, the dust aerosol on April 6, 2006 could be the representative of the dust from W–NW for almost 100% of the wind



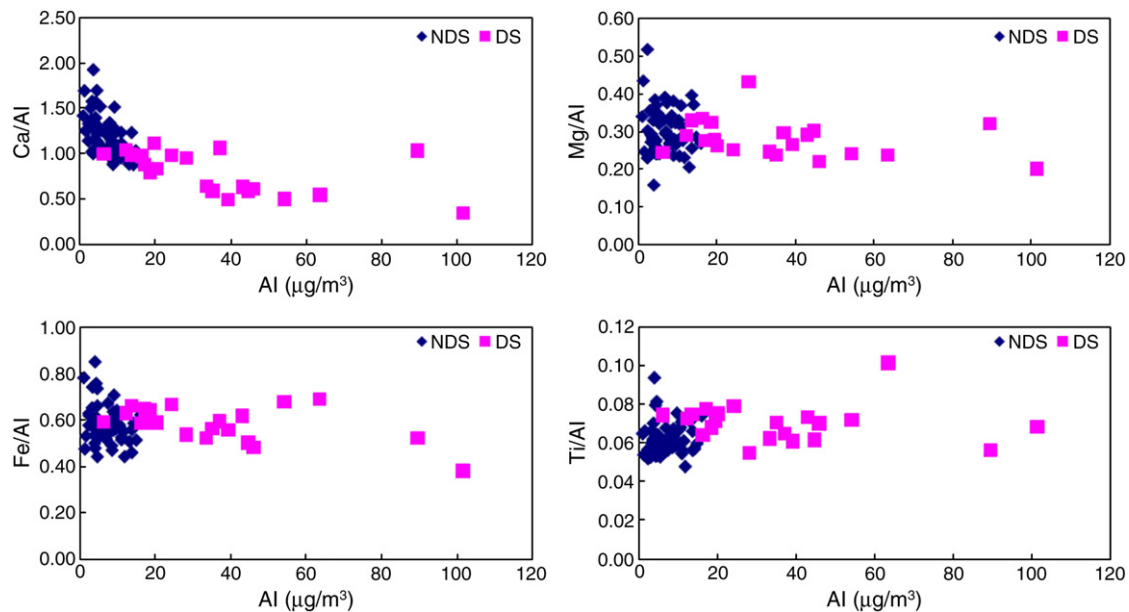


Fig. 6. Ratios of Ca/Al, Mg/Al, Fe/Al, and Ti/Al vs. Al in the aerosol at Yulin.

in this day was from W–NW, and the ratio of Ca/Al was 0.84 in TSP and 0.78 in PM<sub>2.5</sub>. This ratio was comparable to that (0.94) in the TSP collected from Dunhuang, Gansu province, which was located in W–NW of YL (Zhang et al., 2003). It must be noted that the ratio of Ca/Al in the dust aerosols collected in W–NW of YL was higher than that from N–EN. In this study, we also measured the dust aerosol collected from the Taklimakan desert, which is located far away in the direction of W–NW of YL, and the ratio of Ca/Al was 1.67 and 1.65 in TSP and PM<sub>2.5</sub>, respectively. The ratio of Ca/Al in the dust aerosol transported from W–NW of YL was just between the two numbers, the higher one from the Taklimakan desert and the lower one from Mongolia Gobi. This result was the evidence that the dust aerosol that invaded YL from the long-range transport could be the

mixture of the dust from the Taklimakan desert located on its far west direction and the dust from Mongolia Gobi located on its N–EN direction, so as we found in the back trajectory analysis.

#### 4. Conclusion

- (1) The average daily concentrations of PM<sub>2.5</sub> and TSP in dust days were 159.5 μg/m<sup>3</sup> and 591.0 μg/m<sup>3</sup>, respectively, with peak values of 561.0 μg/m<sup>3</sup> in PM<sub>2.5</sub> and 1464.3 μg/m<sup>3</sup> in TSP, indicating that in Yulin, an area near the desert, the air was seriously polluted.
- (2) The mineral dust was the most loading component in dust aerosols over Yulin in both dust event days and non-dust days.

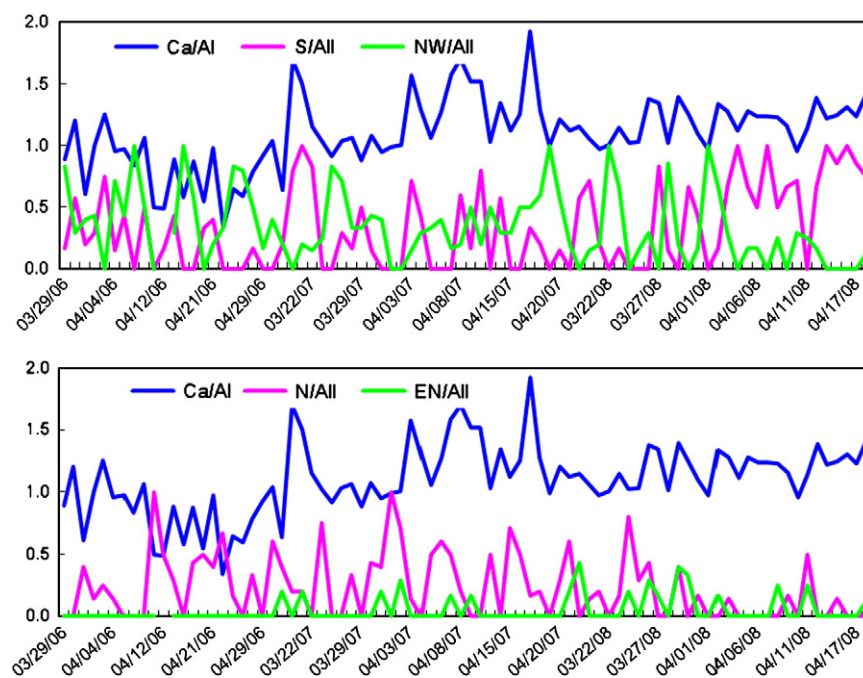


Fig. 7. Variation of the ratios of Ca/Al in TSP and the ratios of S/Al, NW/Al, N/Al, and EN/Al in the study time (S: represents the wind direction from the 90° to 225° section, where the Loess Plateau is located; NW: wind from the 270 to 315° section; N: wind from 315 to 22.5°; EN: wind from 22.5 to 67.5°, Al: represents the wind from all directions in a day).



The concentrations of those pollution elements, As, Cd, Pb, Zn, and S were much higher than their mean abundances in crust even in dust event days. Cd, Pb, As, and Zn were mainly from local pollution, while S was also greatly influenced by the long-range transported dust.

- (3) The high concentration of  $\text{SO}_4^{2-}$  could be from both sources: one from the transformation of the local emitted  $\text{SO}_2$  and the other from the sulfate that existed in primary dust, which was transported to Yulin.  $\text{NO}_3^-$  and  $\text{NH}_4^+$  were from local pollution source, while  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  were mainly from the crustal source.
- (4) The mixing of dust with pollution aerosols on the pathway of the long-range transport of Asian dust was ubiquitous, which would make the dust aerosols much polluted. The mixing extent could be expressed by the ratio of  $\text{NO}_3^-/\text{Al}$  in aerosol.
- (5) The ratio of  $\text{Ca}/\text{Al}$  was used as a tracer to study the dust source. The comparison of the ratio of  $\text{Ca}/\text{Al}$  together with back trajectory analysis indicated that the sources of the dust aerosol that invaded Yulin could be from the northwestern desert in China and Mongolia Gobi.

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