

Chemical characteristics of aerosol particles (PM_{2.5}) at a site of Horqin Sand-land in northeast China

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Abstract: The objective of this study was to characterize the mass concentration and chemical composition of aerosol particles (PM_{2.5}) collected at Tongliao (Inner Mongolia Autonomous Region, China), a site in Horqin Sand-land in northeast China. During spring 2005, the mass concentration for PM_{2.5} was $(126 \pm 71) \mu\text{g}/\text{m}^3$ in average. Five dust storm events were monitored with higher concentration of $(255 \pm 77) \mu\text{g}/\text{m}^3$ in average than the non dusty days of $(106 \pm 44) \mu\text{g}/\text{m}^3$. Concentrations for 20 elements were obtained by the PIXE method. Mass concentrations of Al, Mg, Si, K, Ca, Ti, Mn, and V, which increased with the PM_{2.5} concentration, were higher than the pollution elements (S, Cl, Zn, Ar, Se, Br, and Pb). Enrichment factor relative to crust material was also calculated, which showed dust trace elements were mainly from earth upper crust and pollution elements were dominated the anthropogenic aerosols. The Si/Al, Ca/Al, and Fe/Al ratios in PM_{2.5} samples at Tongliao were 4.07, 0.94, and 0.82, respectively, which were remarkably different with those on other source regions, such as "Western desert source region", "North desert source region" and central Asia source. Air mass back-trajectory analysis identified three kinds of general pathways were associated with the aerosol particle transport to Tongliao, but have the similar elemental ratios, implying that elemental signatures for dust aerosol from Horqin Sand-land were different with other regions.

Keywords: dust aerosol; mass concentration; elemental composition; transport pathway

Introduction

Soil dust derived from the wind erosion process in arid and semi-arid region is an important factor on the climate forcing (Tegen *et al.*, 1996; Sokolik and Toon, 1996). Desert regions in East Asia were considered as the major sources for Asian dust according to the rain-dust records in Chinese historical documents and present day observations (Zhang, 1984; Merrill *et al.*, 1989, 1994; Zhang *et al.*, 2003a). Mineral aerosol particles directly affect the Earth's radiative balance by scattering and absorbing incoming solar and outgoing long-wave energy of terrestrial radiation, may modify the cloud properties by acting as cloud condensation nuclei (Charlson *et al.*, 1992), and may influence on the tropospheric chemistry by serving as media for various heterogeneous reactions of different chemical species (Franket *et al.*, 1996). The long distance transport and deposition of eolian dust can also play an important role in biogeochemical cycles in the land and oceans (Zhuang *et al.*, 2001; Young *et al.*, 1991), and high aerosol concentration in spring also affect the urban and rural air quality and human health (Harrison and Yin, 2000; Ren *et al.*, 2003; Wang *et al.*, 2002; Zhang *et al.*, 2003, 2005). Unfortunately, a more adequate assessment of regional forcing by dust is hardly possible because of our poor knowledge of the dust chemical, physical and optical characteristics and

their temporal and spatial variation.

Recently, accompanying with the international Asia Pacific Regional Aerosol Characterization Experiment (ACE-Asia) ongoing, many studies have shown the physical and chemical properties of eolian dust from the arid and semi-arid source regions in Northern China and their characterization of spatial and temporal variation in remote marine regions and beyond (Zhuang *et al.*, 2001; Alfaro *et al.*, 2003; Iwasaka *et al.*, 2003; Zhang *et al.*, 2003a; Arimoto *et al.*, 2004; Xuan *et al.*, 2004; Cao *et al.*, 2005; Zhang *et al.*, 2005).

Horqin Sand-land, located in the east of the Inner Mongolia Autonomous Region of China, 350 mm of the mean annual precipitation, was proved to be one of important source region by model simulations of Asian dust emissions in Gobi and desert regions in northern China (Zhang *et al.*, 2003b). Horqin Sand-land, covering a land area of about 50600 km², is also the biggest one among the four largest sand-lands in northern China (including Mu Us Sand-land, Hunshan Lake Sand-land, Horqin Sand-land, and Hulun Beer Sand-land). Asian dust, originating from Horqin Sand-land and other desert regions, are transported from Asia to the North Pacific and beyond by northerly or westerly wind and can easily influence airborne aerosol loading over downwind regions such as north China, especially Beijing (Zhang *et al.*, 2005),

Korea and Japan, and so on. However, few research studies focused on the chemical characteristics of soil dust from Horqin Sand-land. The studies were designed to investigate the mass concentration and characterization of elemental composition of fine size aerosol particles(PM_{2.5}) at Horqin Sand-land (Tongliao station).

1 Method

1.1 Aerosol sampling

Tongliao station (43° 36'N, 122° 16'E, 178 m above sea level) is located in the centre of the Horqin Sand-land, 5 km from Tongliao downtown (Inner Mongolia Autonomous Region, China). There were no major industrial activities surrounding the sampling site. Sampling site is on the rooftop of the building of Tongliao Meteorological Agency, at 12 m above the ground level. Aerosol samples were obtained with a mini-volume sampler (Airmetrics, Springfield, Oregon, USA) operating with a flow rates of 5 L/min on 46.2 mm diameter Teflon membrane filters (PM_{2.5} Air Monitoring PTFE Filters, Whatman). This kind of sampler is convenient for field observation for that it is equipped with a storage battery to avoid power failure during serious weather condition (such as high wind speed during dust storm). Particles with aerodynamic diameter $\leq 2.5 \mu\text{m}$ (PM_{2.5}) were collected daily from 3 March to 31 May 2005. Each sample had a 24-h sampling period from 10:00 in the morning to 10:00 next day morning. A total of 90 aerosol samples were collected in spring 2005(90 d) at Tongliao station. During the sampling period, meteorological data, including ambient temperature, ambient pressure, ambient relative humidity(RH), wind speed, and wind direction, were also recorded four times (at 02:00, 08:00, 14:00, and 20:00 Beijing time) in each sampling day at the Tongliao site by Tongliao Meteorological Agency.

1.2 Mass and elemental analysis

The aerosol mass were determined gravimetrically using a Sartorius MC5 electronic microbalance with a $\pm 1 \mu\text{g}$ sensitivity (Sartorius, Göttingen, Germany). Before weighted, Teflon membrane filters were equilibrated for 24 h at a constant temperature between 20 and 23°C and relative humidity between 35% and 45%. Each filter was weighted at least three times before and after sampling by a 24-h equilibration. The mean net mass for each filter was obtained by the average post-weight data subtracting the pre-weight. The precision of the weighing were $<10 \mu\text{g}$ for blank filters and $<20 \mu\text{g}$ for filter samples.

The elemental concentration of PM_{2.5} samples were analyzed directly by a proton induced X-ray emission (PIXE) method using the 2.5 MeV protons with a 50 nA beam current produced by the twin 1.7 MV beams tandem accelerator at the Institute of Low

Energy Nuclear Physics, Beijing Normal University (Zhang, 1993; Zhu and Wang, 1998). All concentrations were corrected for backgrounds from blank filters. 20 elements concentration for each sample were determined, which are Mg, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br and Pb. This PIXE method has been widely used to study the chemical components of aerosols particles in China in recent years (Zhang *et al.*, 2003a, 2001, 2005; Arimoto *et al.*, 2004; Cao *et al.*, 2005).

2 Results and discussion

2.1 Temporal variations of PM_{2.5} mass concentration

Daily PM_{2.5} mass concentration and observational meteorological data from 2 March to 31 May at Tongliao site are shown in Fig.1. The average mass concentration of PM_{2.5} at Tongliao station was $(126 \pm 71) \mu\text{g}/\text{m}^3$ during the whole spring 2005. During the field sampling period, five dust storm events (DS) were observed at Tongliao station on 6 March (DS1), 5—7 April (DS2), 13—14 April (DS3), 27—30 April and 1st May (DS4), and 30 May (DS5), corresponding with PM_{2.5} loading of 276, 299, 287, 222, and 203 $\mu\text{g}/\text{m}^3$, respectively, higher than the spring mean value (Table 1). The average mass concentration of these five dust storm events was $(255 \pm 77) \mu\text{g}/\text{m}^3$. The highest PM_{2.5} mass concentration was 399 $\mu\text{g}/\text{m}^3$ on 13 April, which was almost 4 times than the daily average mass concentration during non dust storm period (NDS) of $(106 \pm 44) \mu\text{g}/\text{m}^3$. Previous studies of PM_{2.5} mass concentration during Asian dust storm in other regions can compare with our measurement. Continuous monitoring of airborne particles at Chongwan-chongju, in Korea, showed that the average maximum concentration of strong dust storms in 2002 was 166.9 $\mu\text{g}/\text{m}^3$ for PM_{2.5} (Chung *et al.*, 2003). High PM_{2.5} concentration was also measured during a heavy dust storm event over a site near Asian dust source regions, Xi'an on 14 April 2002, which showed PM_{2.5} concentration was 740 $\mu\text{g}/\text{m}^3$, 3 times more than daily average concentration during NDS condition of 200 $\mu\text{g}/\text{m}^3$ (Cao *et al.*, 2005). Most of the dust storm events at Tongliao were associated with high wind speed (Table 1), which exceeding spring-time average value of 3.9 m/s. For example, observed the wind speeds of DS1 (on 6 March) were 6.4 m/s at 08:00, 8.8 m/s at 14:00, and 6.4 m/s at 20:00. Low relative humid(ranging from 29% to 41%) and low ambient pressure were also observed during most DS events (Fig.1). It implies that the occurrences of dust storm at Horqin Sand-land are commonly associated with dry, low pressure and high wind speed conditions. This phenomenon was also observed at ZBT station (Yulin, Shannxi Province, China) during the dust storm happened (Zhang *et al.*, 2003a). Table

1 shows the dust storm data for spring 2005, including PM_{2.5} mass loadings, trace elemental concentrations, and meteorological data. Compared with the dust storm events, NDS

Table 1 Summary statistics for the concentration of PM_{2.5}, dust elements, and meteorological data during dust storms in spring 2005 at Tongliao

Dust storm event	Concentration, μg/m ³									Pre,	RH,	WD,
	PM _{2.5}	Si	Al	Mg	K	Ca	Ti	Mn	Fe	Pa	%	m/s
DS1(6 March)	276.0	40.1	10.3	2.5	4.0	6.8	0.67	0.22	7.3	99070	26	7.6
DS2(5—7 April)	299.1	48.0	12.0	1.7	5.7	9.8	0.75	0.24	9.2	98028	32	5.2
DS3(13—14 April)	287.3	45.6	11.3	1.4	4.4	8.0	0.72	0.18	8.3	98871	19	5.7
DS4(27—30 April,1 May)	222.4	44.0	10.9	2.8	4.2	7.8	0.74	0.21	8.2	97778	26	6.4
DS5(30 May)	203.0	50.9	10.9	0.7	6.2	10.2	0.98	0.31	11	97940	41	4.9

Notes: DS. dust storm events; Pre. pressure; RH. relative humidity; WD. wind speed

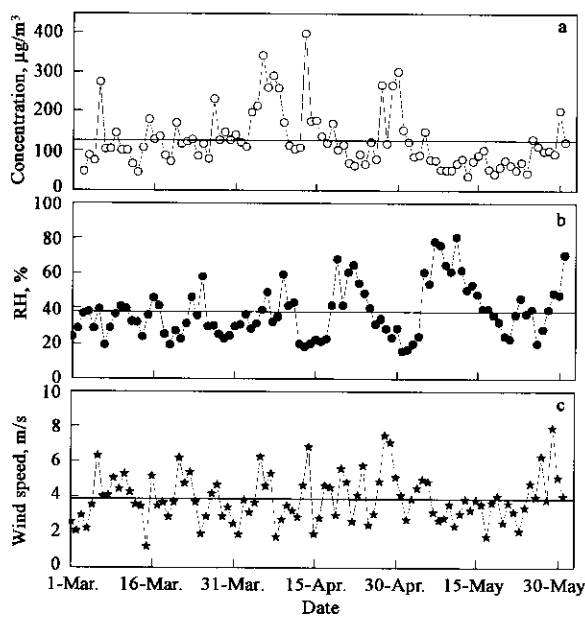


Fig.1 Temporal variations of PM_{2.5}(a), relative humidity(b), and wind speed(c) at Tongliao during 2 March to 31 May

conditions with calm or weak wind had a background aerosol loading of 106 μg/m³ in the whole spring, which exceeded the American EPA's new PM_{2.5} air quality standard of 24 h mean value of 65 μg/m³ and year mean value of 15 μg/m³. This high background loading at Horqin Sand-land implies that fine size particles pollution was very serious in north China, even if in the small or middle cities near the desert regions, which the industry was not developed.

The mass concentration of soil dust can be estimated by summing the concentration of several elements predominantly associated with soil, plus oxygen assuming the compounds involved are the most common oxides (Malm *et al.*, 1994). The following equation can be used to calculate the dust concentration by elemental concentrations:

$$C_{\text{soil}} = 2.2C_{\text{Al}} + 2.49C_{\text{Si}} + 1.63C_{\text{Ca}} + 2.42C_{\text{Fe}} + 1.94C_{\text{Ti}}$$

(1)

where C_{soil} is the calculated soil dust concentration,

and C_{Al} , C_{Si} , C_{Ca} , C_{Fe} , and C_{Ti} represent the elemental concentration of Al, Si, Ca, Fe, and Ti in PM_{2.5} samples. The average soil dust PM_{2.5} mass concentration were 59 μg/m³ during the whole spring, 174 μg/m³ during DS period, and 41 μg/m³ on non-dusty days. For the five dust storm events, calculated of the dust concentrations for PM_{2.5} were 153, 186, 173, 168, and 197 μg/m³, respectively.

It was particularly valuable to assess the contributions of Asian dust (geological materials) emitted by the wind erosion process to the aerosol particles at Horqin Sand-land. It showed that geological mineral dust accounted for 54% of PM_{2.5} samples. The residual about 46% of PM_{2.5} was assumed to be pollution aerosols and unidentified materials. Our result reflected the significant contribution of anthropogenic sources from the remote upwind regions or south of Tongliao site (Tongliao City) to the aerosol mass concentration. Previous studies on the Asian dust also found the phenomenon that pollution-derived aerosols often mixed with the natural soil dust, even if at Asian dust source regions of Chinese desert areas(Arimoto *et al.*, 2004; Alfaro *et al.*, 2003; Prospero *et al.*, 2003; Zhang *et al.*, 2003a). Chemical analysis of PM_{2.5} samples also confirmed that soil-derived dust mixes with anthropogenic aerosol at Tongliao station, and these data will be discussed later.

2.2 Elemental composition of PM_{2.5}

A major objective of our studies was to characterize the chemical composition of aerosol dust particles at Tongliao station. We calculated the arithmetic mean concentrations of 20 elements, which were shown in Table 2. Chemical results showed that high elemental concentrations were mainly for the eight typical dust elements, which are Al, Si, Mg, Ca, Fe, K, Mn, and Ti. The mean concentrations for eight dust elements were 3.8 ± 3.6, 15.2 ± 14.3, 1.3 ± 1.3, 3.1 ± 2.6, 3.0 ± 2.8, 2.1 ± 1.5, 0.1 ± 0.1, and 0.2 ± 0.2 μg/m³, respectively. Most elements content of PM_{2.5} at Tongliao were similar with the chemical results for the

same size fraction samples collected at ZBT, an ACE-Asia ground station (Arimoto *et al.*, 2004), in which the mean elemental concentrations for Al, Si, Mg, Ca, Fe, K, and Mn were 4.3, 8.8, 1.5, 5.2, 2.5, 1.5, and 0.1 $\mu\text{g}/\text{m}^3$, respectively. However, the important difference of Si concentration should be noticed. For instance, Si concentration at Tongliao was almost two times than that at ZBT, showing dust at Horqin Sand-land was characterized by a high Si concentration. During dust storm events, most element concentrations were higher than the spring mean values (Table 1). For example, mass concentrations for Si, Al, Ti and Fe during dust storm episodes were 4 times, Ca, Mg, Mn, and K were 2 or 3 times, more than those of spring mean concentrations. The calculation of enrichment factor relative to earth upper crust can compare the elemental composition between aerosol samples and crust materials (Taylor and Melennand, 1995). This method was often used to identify the elements origin from natural source or anthropogenic source. The enrichment factors (EF) for 20 elements were calculated as follows:

$$EF_{\text{crust}} = (C_{\text{element}}/C_{\text{reference}})_{\text{air}} / (C_{\text{element}}/C_{\text{reference}})_{\text{crust}} \quad (2)$$

where C_{element} is the concentration of any elements, $C_{\text{reference}}$ is the concentration of reference element. Typically, Al, Fe, and Si were often chosen as the reference element. The EF_{crust} results of 20 elements are shown in Table 2. The values of EF_{crust} for Al, Mg, Si, K, Ca, Ti, Mn, and V were in the same range of 1 to 5 in both the DS and NDS samples. Therefore, these nine elements are mainly from crust origin.

The relationship between Mg, Si, Fe, Ca, K, Mn, Ti and Al in Tongliao $\text{PM}_{2.5}$ samples were plotted in Fig.2. The correlation coefficient (r) for the dust trace elements versus Al ranged from 0.86 to 1.0. These high correlations also verify the crystal origin of these nine dust elements. The ratios of 20 elements to Al were calculated and shown in Table 2. Si, Al, Ca and Fe are four major elements in mineral dust, and the ratio of Si/Al, Ca/Al and Fe/Al will be described detailed in the following section.

The mean value of Si/Al ratio in $\text{PM}_{2.5}$ at Tongliao was 4.07 of total samples, and there was no significant difference between DS and NDS condition, indicating the major source for Si was dust aerosol. These Si/Al ratios were consistent with average

Table 2 Concentration and enrichment factor (EF) for 20 elements and relationships between dust elements and Al in $\text{PM}_{2.5}$ aerosol particles

Total samples <i>n</i> =9	Concentration, ng/m ³		<i>EF</i> _{crust}		Ratios to Al		
	Mean	<i>SD</i>	Mean	<i>SD</i>	Total samples	DS (<i>n</i> =12)	NDS (<i>n</i> =78)
$\text{PM}_{2.5}$	1.3×10^3	7.1×10^4					
Al	3.8×10^3	3.6×10^3	0.8	0.2			
Mg	1.3×10^3	1.4×10^3	1.5	1.8	0.50	0.19	0.55
Si	15.2×10^3	14.8×10^3	0.9	0.1	4.07	4.06	4.07
P	1.7×10^2	2.1×10^2			0.046	0.046	0.046
S	2.4×10^3	2.3×10^3	239.4	250.0	1.11	0.33	1.23
Cl	1.1×10^3	1.0×10^3	249.6	295.6	0.55	0.11	0.61
K	2.1×10^3	1.5×10^3	1.7	0.9	0.76	0.44	0.81
Ca	3.1×10^3	2.7×10^3	1.6	0.3	0.94	0.76	0.97
Ti	2.5×10^2	2.5×10^2	0.9	0.2	0.063	0.067	0.063
V	2.2×10^1	2.4×10^1	3.4	3.2	0.007	0.005	0.008
Cr	3.7×10^1	4.1×10^1	9.4	12.8	0.015	0.006	0.017
Mn	1.0×10^2	6.9×10^1	2.4	1.7	0.039	0.020	0.042
Fe	3.0×10^3	2.8×10^3	1.0	0.0	0.82	0.78	0.83
Ni	1.9×10^1	1.9×10^1	5.9	5.6	0.007	0.003	0.008
Cu	6.7×10^1	5.4×10^1	26.4	17.4	0.024	0.013	0.026
Zn	6.5×10^2	5.9×10^2	283.1	360.9	0.34	0.090	0.38
As	6.5×10^1	4.9×10^1	1036.9	1266.6	0.030	0.008	0.034
Se	6.4×10^1	5.0×10^1	37815.0	46000.4	0.031	0.008	0.034
Br	9.3×10^2	6.5×10^1	1079.8	1180.3	0.043	0.011	0.048
Pb	1.2×10^2	9.5×10^1	263.5	295.1	0.059	0.017	0.066

Notes: *SD*. standard deviation; *n*. sample number

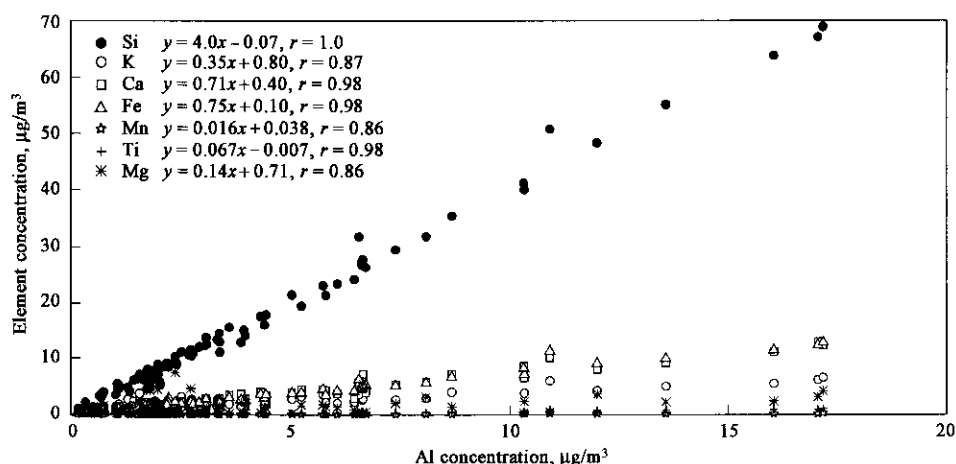


Fig.2 Relationship between dust elements and Al in PM_{2.5} at Tongliao site

composition of Si/Al of global upper crust or sedimentary rocks of 4.04 (Pye, 1987). In contrast, the Si/Al ratio in PM₉ and PM_{2.5} at ZBT, which represents the “Northern desert source region” (Zhang *et al.*, 2003a), were 2.8, and 1.9, respectively (Arimoto *et al.*, 2004; Alfaro *et al.*, 2003). So the dust aerosol at Horqin Sand-land characterized by a high Si/Al ratio distinguished from “Northern desert source regions”. Other observations also showed the Si/Al ratio of eolian dusts from Sahelian region ranges from 3.18 to 3.6, and soil dusts from the Tadzhikistan (central Asia) had a ratio of 3.06 (Gomes and Gillette, 1993), while for the dusts from northern Morocco varied from 2.67 to 2.87 (Bergametti *et al.*, 1989). So dust aerosol at Tongliao showed a high Si/Al ratio, implying that elemental signature of dust from Horqin Sand-land is different with other source regions.

The ratio of Ca/Al in PM_{2.5} samples at Tongliao was 0.94 in average, which compared quit well to a Ca/Al ratio approximately 1 of PM_{2.5} and TSP samples collected at ZBT (Arimoto *et al.*, 2004; Zhang *et al.*, 2003a). But the difference of Ca/Al ratio between DS events and non dusty days should be noticed (Table 2). Ca/Al ratio during NDS was larger than in DS. This abundance of Ca during NDS revealed the presence of local pollution aerosol particles that were richer in Ca than desert mineral dusts. For example, cement particles can be released by the city construction sites. This Ca rich phenomenon during non dusty days was also observed at ZBT by Alfaro *et al.* (2003), which showed the Ca/Al ratio were 0.79 for DS samples and 1.14 for NDS samples. The Ca/Al ratio in dust samples at Horqin Sand-land was remarkable different with those at “Western desert source region”, which showed a high Ca/Al ratio of 1.7 (Zhang *et al.*, 2003a). Therefore, elemental signature of soil dust from Horqin Sand-land was different with “Western desert source region”.

Special attention should be paid to Ca (referred water-soluble (WS) Ca²⁺) materials, such as Calcite,

for which were involved in troposphere atmospheric chemistry reactions. Many investigators have suggested that some of the sulfate particles (like Gypsum) formed in the atmosphere through homogeneous and heterogeneous reactions. For example, Ca-rich mineral (Calcite) reacted with pollution aerosols such as sulphur oxide and formed gypsum (Zhang and Iwasaka, 1999). Arimoto *et al.* (2004) observed the important variations of the Ca/Al ratio and the ratio of SO₄²⁻ to WS Ca²⁺ between a Chinese desert region site of ZBT and a downwind region station of Gosan in Korea. Ca/Al ratio and SO₄²⁻ to WS Ca²⁺ ratio at ZBT were higher than those observed at Gosan, and the possible reason for these difference were mainly due to gas-particle conversion, size selective fractionation, or aerosol types mixing. Variation of element ratios (such as Ca/Al) during long distance transport process reflects changes of mineralogical composition of aerosol particles. The variation of mineralogical composition can influence on the optical properties (extinction coefficient, single scattering albedo, and asymmetry parameters) of dust aerosols, for which are determined by the relative abundance of various minerals and how the minerals mixed together (Sokolik and Toon, 1999).

We show the Fe/Al ratio in PM_{2.5} samples investigated at Tongliao was 0.8 in average, and no significant difference was observed between DS and NDS, similar with Si/Al ratio. This Fe/Al ratio was different with that of 0.6 for PM_{2.5} and PM₉ samples at ZBT (Arimoto *et al.*, 2004; Alfaro *et al.*, 2003). In contrast, aerosol particles at central Asia (Tadzhikistan) had low Fe/Al ratio of 0.4 (Gomes and Gillette, 1993). The Fe/Al ratio seems to be a good signature on the regional scale allowing us to identify the dust aerosol source, for example to distinguish central Asian dust source region from East Asia dust source regions. The difference of Fe/Al ratio involved in the output flux of Fe from Asian source regions to the North Pacific Ocean and beyond, and will

influence on ocean biogeochemical cycles and regional/global climate changes(Jickells *et al.*, 2005).

In comparing with the soil dust trace elements, the EF_{crust} values for S, Cl, Zn, As, Se, Br, and Pb were much larger than 5, implying the important contributions of non crystal materials to these elements. S, Cl, Zn, and Br were taken as important pollution elements for their high concentrations in $PM_{2.5}$. The mean mass concentration of S in ambient $PM_{2.5}$ in spring 2005 at Tongliao was $2.4 \mu g/m^3$, which was in the same level with that monitored at other source regions in north China (Zhang *et al.*, 2003a). The temporal variations of mass concentration and EF_{crust} for S were plotted in Fig.3. It shows the highest concentration of S during spring 2005 was $12.6 \mu g/m^3$ on 5 April (DS2). In most cases, high concentration and EF for S occurred during dust storm events, and the mean concentration of S during DS ($3.6 \mu g/m^3$) was higher than the mean value of $2.2 \mu g/m^3$ on NDS, suggests dusty air contained higher level of pollution material than non dusty air. A recent study on a very strong dust storm at Beijing on March 2002 also reported this fact (Zhang *et al.*, 2005). Other pollution elements such as Cl, Zn, Ar, Se, Br, and Pb were similar with S, showed higher concentrations during DS than NDS condition.

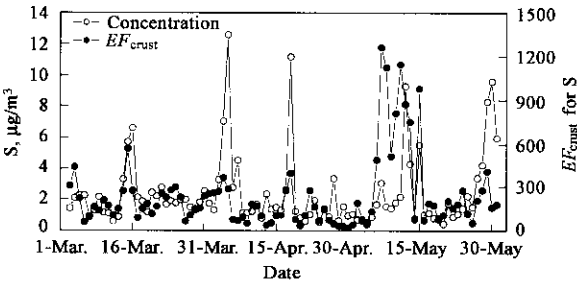


Fig.3 Temporal variations of mass concentration and EF_{crust} for S

2.3 Transport pathway of dust storms at Tongliao site

Five-day air mass back trajectories arriving at 1000 m above ground level (at 06 UTC) were calculated for Tongliao using the NOAA HYSPLIT 4 trajectory model to investigate the soil dust transport pathways. During the 12 dusty days of five dust storm events in spring 2005, three groups(A, B and C) of air mass trajectories that passed over Tongliao were identified. Trajectory group A was that the air masses passed from north to southeast through Gobi and desert regions Mongolia, and then moving southeast to Tongliao. Dusty days on 6 March, 13 April, 27 to 30 April, 1st May, and 30 May belong to this kind of pathway. The route of group B passed from the west to the east through the Gobi and desert areas of north Xinjiang Province, west and central part of Inner Mongolia Autonomous Region to Tongliao. This kind of transport pathway includes dusty days on 5—6

April. Another pathway of group C passed through the southeast Russia and then turned south through east boundary of Mongolia and northeast of Inner Mongolia to Tongliao. Dusty days on 7 April, 14 April were this kind of pathway. The ratios of dust elements to Al for the twelve dusty days were similar as shown in Table 1, although the air mass transport pathways are different. It demonstrated that elemental signatures of aerosol particles from Horqin Sand-land were different with other sources.

3 Conclusions

At Tongliao, the mean concentration for $PM_{2.5}$ during spring 2005 was $(126 \pm 71) \mu g/m^3$. Five dust storm events were observed with higher concentration of $(255 \pm 77) \mu g/m^3$ than non dusty days mean concentration of $(106 \pm 44) \mu g/m^3$. Large amount of fine size aerosol particles output from Horqin Sand-land will effect the climatic changes and environmental quality in the East Asia-Pacific regions.

Soil dust was the major component of $PM_{2.5}$ at Horqin Sand-land. The EF_{crust} for Al, Mg, Si, K, Ca, Ti, Mn, and V were lower than 5, showing these dust trace elements are maily from earth crust source. The Si/Al, Ca/Al, and Fe/Al ratios in $PM_{2.5}$ samples at Tongliao were remarkable different with those on other source regions, such as “Western desert source region”, “North desert source region”, and central Asia source, suggesting elemental composition of dust aerosol at Tongliao were different with other regions. Compared with dust trace elements, S, Cl, Zn, As, Se, Br, and Pb, which the EF_{crust} much were larger than 5, were dominated the anthropogenic pollution aerosols. Mass concentrations of Al, Mg, Si, K, Ca, Ti, Mn, and V, which increased with the $PM_{2.5}$ loading, were higher than the concentrations of pollution elements, such as S, Cl, Zn, Ar, Se, Br, and Pb.

Three general air mass trajectories which passed over Tongliao were identified during DS in spring 2005. Whatever the air masses transport pathways, the similar ratios of elements to Al were obtained, further demonstrated the chemical characteristics of dust aerosol at Horqin Sand-land were different with other source regions.

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References:

Alfaro S C, Gomes L, Rajot J L *et al.*, 2003. Chemical and optical characterization of aerosols measured in spring 2002 at the ACE-Asia supersite, Zhenbeitai, China [J]. *J Geophys Res*, 108 (D23), 8641, doi:10.1029/2002JD003214.
Arimoto R, Zhang X Y, Huebert B J *et al.*, 2004. Chemical composition of atmospheric aerosols from Zhenbeitai, China, and Gosan,

- South Korea, during ACE-Asia [J]. *J Geophys Res*, 109(D19), (S04), doi:10.1029/2003JD004323.
- Bergametti G, Gomes L, Coude-Gaussen G *et al.*, 1989. African dust observed over Canary Island: source-regions identification and transport pattern for some summer situation [J]. *J Geophys Res*, 94: 14855—14864.
- Cao J J, Lee S C, Zhang X Y *et al.*, 2005. Characterization of airborne carbonate over a site near Asian dust source regions during spring 2002 and its climatic and environmental significance [J]. *J Geophys Res*, 110, D03203, doi:10.1029/2004JD005244.
- Charlson R J, Schwartz S E, Hales J M *et al.*, 1992. Climate forcing by anthropogenic aerosols[J]. *Science*, 255: 423—430.
- Chung Y S, Kim H S, Dulama J *et al.*, 2003. On heavy dustfall observed with explosive sandstorms in Chongwon-Chongju, Korea in 2002 [J]. *Atmos Environ*, 37: 425—433.
- Frank J D, Gregory R C, Yang Z *et al.*, 1996. Role of mineral aerosol as a reactive surface in the global troposphere [J]. *J Geophys Res*, 101(D17): 22869—22889.
- Gomes L, Gillette D A, 1993. A comparison of characteristics of aerosol from dust storms in Central Asia with soil-derived dust from other regions[J]. *Atmos Environ*, 27A: 2539—2544.
- Harrison R M, Yin J, 2000. Particulate matter in the atmosphere: Which particle properties are important for its effects on health [J]. *Sci Total Environ*, 249: 85—101.
- Iwasaka Y, Shi G Y, Matsuki A *et al.*, 2003. Importance of dust particles in the free troposphere over the Taklamakan Desert: Electron microscopic experiments of particles collected with a balloonborne particle impactor at Dunhuang, China[J]. *J Geophys Res*, 108(D23), 8644, doi:10.1029/2002JD003270.
- Jickells T D, An Z S, Andersen K K *et al.*, 2005. Global iron connections—desert dust, ocean biogeochemistry and climate [J]. *Science*, 308: 67—71.
- Malm W C, Sisler J F, Huffman D *et al.*, 1994. Spatial and seasonal trends in particle concentration and optical extinction in the United States[J]. *J Geophys Res*, 99(D1): 1347—1370.
- Merrill J T, Uematsu M, Bleck R, 1989. Meteorological analysis of long range transport of mineral aerosol over the North Pacific [J]. *J Geophys Res*, 94: 8584—8598.
- Merrill J, Arnold E, Margaret L *et al.*, 1994. Mineralogy of aeolian dust reaching the North Pacific Ocean 2. Relationship of mineral assemblages to atmospheric transport patterns[J]. *J Geophys Res*, 99(D10): 21025—21032.
- Prospero J, Savoie D L, Arimoto R, 2003. Long-term record of nssulfate and nitrate in aerosols on Midway Island, 1981—2000: Evidence of increased (now decreasing?) anthropogenic emissions from Asia [J]. *J Geophys Res*, 108 (D1), 4019, doi: 10.1029/2001JD001524.
- Pye K, 1987. Aeolian dust and dust deposits[M]. Academic, San Diego, Calif.
- Ren Z H, Gao Q X, Su F Q *et al.*, 2003. The regional characteristics of the atmospheric environment and the impact of dust storm in Beijing[J]. *China Technology Science*, 5(2): 49—56.
- Sokolik I N, Toon O B, 1996. Direct radiative forcing by anthropogenic airborne mineral aerosols[J]. *Nature*, 380: 681—683.
- Sokolik I N, Toon O B, 1999. Incorporation of mineralogical composition into models of the radiative properties of mineral aerosol from UV to IR wavelengths [J]. *J Geophys Res*, 104(D8): 9423—9444.
- Taylor S R, McLennan S M, 1995. The geochemical evolution of the continental crust[J]. *Reviews of Geophysics*, 33: 241—265.
- Tegen I, Andrew A L, Fung I, 1996. The influence on climate forcing of mineral aerosols from disturbed soils[J]. *Nature*, 380: 419—422.
- Wang W, Yue X, Liu H J *et al.*, 2002. Study on pollution characteristics of aerosols during sand dust-storm weather in Beijing [J]. *Acta Scientiae Circumstantiae*, 22(4): 494—498.
- Xuan J, Sokolik I N, Hao J F *et al.*, 2004. Identification and characterization of sources of atmospheric mineral dust in East Asia[J]. *Atmos Environ*, 38: 6239—6252.
- Young R W, Carder K L, Betzer P R *et al.*, 1991. Atmospheric iron inputs and primary productivity: Phytoplankton responses in the North Pacific[J]. *Global Biogeochem Cycles*, 5: 119—134.
- Zhang D E, 1984. Synoptic-climatic studies of dust fall in China since the historic times[J]. *Scientia Sinica*, 27: 825—836.
- Zhang X Y, Arimoto R, An Z S *et al.*, 1993. Atmospheric trace elements over source regions for Chinese dust: concentrations, sources and atmospheric deposition on the Loess Plateau [J]. *Atmos Environ*, 27A(13): 2051—2067.
- Zhang D, Iwasaka Y, 1999. Nitrate and sulfate in individual Asian dust-storm particles in Beijing, China in spring of 1995 and 1996 [J]. *Atmos Environ*, 33: 3213—3223.
- Zhang R J, Xu X F, Han Z W, 2001. Inorganic chemical composition and source signature of PM_{2.5} in Beijing during ACE-Asia period [J]. *Chin Sci Bull*, 48(10): 1002—1005.
- Zhang X Y, Gong S L, Shen Z X *et al.*, 2003a. Characterization of soil dust aerosol in China and its transport and distribution during 2001 ACE-Asia: 1. Network observations[J]. *J Geophys Res*, 108 (D9), 4261, doi:10.1029/2002JD002632.
- Zhang X Y, Gong S L, Zhao T L *et al.*, 2003b. Sources of Asian dust and role of climate change versus desertification in Asian dust emission [J]. *Geophysical Research Letters*, 30, 2272. doi: 10.1029/2003GL018206.
- Zhang R J, Arimoto R, An J L *et al.*, 2005. Ground observation of an intense dust storm in Beijing in March 2002 [J]. *J Geophys Res*, 110(D18S06), doi:10.1029/2004JD004589.
- Zhu G H, Wang G F, 1998. Investigation of the particulate derived from indigenous zinc smelting using PIXE analytical technique [J]. *Nucl Instrum Methods Phys Res, Sect B*, 136: 966—969.
- Zhuang G S, Guo J H, Yuan H *et al.*, 2001. The compositions, sources, and size distribution of the dust storm from China in spring of 2000 and its impact on the global environment[J]. *Chin Sci Bull*, 46(11): 895—901.

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