



Characteristics of major elements and heavy metals in atmospheric dust in Beijing, China



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ABSTRACT

Atmospheric dust is directly related to air quality and human health in urban areas. In this study, monthly atmospheric dust of fifteen major elements and heavy metals was measured from March 2008 to February 2009 in Beijing, China, in an effort to determine both natural and anthropogenic sources. The results showed average concentrations of Ca, Pb, Zn, and Cu in atmospheric dust were higher than those of topsoil. Cluster analysis suggested that the elements be divided into three groups: Ca, natural, and anthropogenic elements. Seasonal variations of the elements in dust showed that Ca and the anthropogenic elements of Cu, Zn, Pb were relatively higher in summer and autumn than in winter and spring due to the absence of dusty weather. Enrichment factors (EF) indicated that most of the dust was enriched in Ca, Zn, Pb, Mg, Cu, with EF values > 5; EF values were higher in summer and autumn months than in other months. The atmospheric dust in Beijing is affected by both natural and anthropogenic sources, which have different sources in different seasons. In spring and winter, atmospheric dust arises mainly from natural sources of dusty weather, while the atmospheric dust in summer and autumn was dominated by anthropogenic sources due to the lack of dusty weather.

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1. Introduction

Atmospheric dust is one of the important parts of the global biogeochemical cycle (Lawrence and Neff, 2009). Atmospheric dust is the medium that brings several elements to ecosystems (Farmer, 1993; Kennedy et al., 1998; Soderberg and Compton, 2007). In some cases, alkaline atmospheric particulates have played an important role in neutralizing the acidity of rainwater (Xu and Han, 2009; Han et al., 2011; Wang and Han, 2011). The raising, transportation, and deposition of dust are mainly from natural sources, whereas anthropogenic inputs (i.e., emissions) are released into the air and mixed with natural sources (Lawrence and Neff, 2009). Especially in urban areas, atmospheric dust is easily polluted by anthropogenic inputs, which are typically heavy metals produced by a series of human activities (e.g., vehicle emissions, industrial discharges, domestic heating, waste incineration) (Chen et al., 2005a; Chen et al., 2005b; Onder and Dursun, 2006; Valiulis et al., 2002). The heavy metals of concern for researchers in urban areas include Cd, Ni, Cu, Zn, Cr, Pb, and Hg (Guney et al., 2010; Kumar et al., 2010; Poikolainen et al., 2004; Zheng et al., 2010). Previous studies have shown that urban soils and urban road dusts are significantly impacted by heavy metals derived from anthropogenic activities (Christoforidis and Stamatis, 2009; Wei and Yang, 2010); these metals

are primarily derived from atmospheric deposition of pollutants (Hovmand et al., 2008; Zheng et al., 2010).

As the capital of China, Beijing has been troubled by air pollution not only from industrial production and automobile exhaust emissions, but from frequent sandstorms in the spring (Tang et al., 2013). In PM_{2.5}, PM₁₀, and TSP samples from Beijing, different elements showed different size distributions in foggy/hazy weather (Zhang et al., 2010). Aerosol monitoring in Beijing showed that crustal elements did not increase from 2001 to 2006; however, anthropogenic sources of trace metals (Cu, Zn, As, Cd, and Pb) did increase (Okuda et al., 2008). Sun et al. (2005) found that dust storms not only delivered large amounts of mineral elements but also carried significant quantities of pollutants to Beijing. Okuda et al. (2004) reported that trace metal concentrations in TSP in Beijing were much higher than those in TSP in the centre of Tokyo, Japan. A chemical mass balance (CMB) receptor model indicated that the primary sources of aerosols in Beijing were soil, dust, and coal combustion, while the contribution of vehicle exhaust tended to increase.

In this study, atmospheric dust samples were collected monthly over the period from March 2008 to February 2009 from an urban site in Beijing, and the chemical characteristics of the dust were analysed. The concentrations of six major elements and nine heavy metals in atmospheric dust were targeted as indicators of pollution conditions. Seasonal variations of the elemental contents and the enrichment factors were taken into consideration, and both natural and anthropogenic impacts on atmospheric dust were examined, in an effort to improve understanding of the air quality in Beijing.

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2. Materials and methods

2.1. Study area

Beijing is located in the northern region of the North China Plain and in southern Inner Mongolia (Fig. 1). The climate of Beijing is typically semiarid, with an annual precipitation of 300–800 mm. Beijing and the surrounding areas are located on a plain at elevations of 20–60 m. The Yanghe River valley extends from the Zhangjiakou Highlands to the Beijing plain. Most of the Beijing plain is covered by quaternary loess (An et al., 1991; Liu and Ding, 1998; Xiong et al., 2001). The Yanshan mountains around Beijing consist mainly of igneous rocks, most of which are granite (Yang et al., 2008); limestone and dolomite occur locally at some outcrops.

The population of Beijing is more than 16 million, and there are approximately 3 million motor vehicles in the city. In the years of 2007 and 2008, the total energy consumption of Beijing was 63.27 and 65.70 billion tons of standard coal equivalent (SEC), which was mainly from coal, petrol, and natural gas. Due to the use of such large amounts of fossil fuel and the corresponding emission of a large number of air particulates and pollutants, there is significant pressure on the local atmospheric environment.

2.2. Sampling methods

A typical urban background site was selected as the sampling site; for this study we used the roof of a five storey office building (height of 20 m) within the Institute of Geology and Geophysics at the Chinese Academy of Sciences (N 39°58'55", E 116°22'63") (Fig. 1). This site is located 2 km to the northwest of Beijing National Stadium. Dust samples were collected in duplicate using wet method procedures; the collectors were made of polypropylene (Ganor et al., 2003) with a size of $\Phi 30$ cm \times h40 cm. The collectors were fixed to steel shelves and mounted on 3 m tall field platforms. To depress algal and microbial growth, a collection medium consisting of 700 ml of purified glycol solution (20%) was added to each collector.

The collectors were checked routinely, and glycol solution was added as necessary to keep the bottom of the sampling containers submerged. The collectors were kept covered during rainy periods, but were exposed at all other times. Dust samples were gathered monthly. First, the solid/liquid mixture in the collector was emptied into a clean glass container; ultra-clean water and a small brush were used to remove any dust particles adhering to the collector wall. The samples were then transported to the lab for subsequent processing. After removing impurities, such as leaves and insects, the mixture was dried in an evaporating dish at 80 °C.

To better distinguish between natural and anthropogenic sources of atmospheric dust, two topsoil samples from Changping and Miyun, near Beijing city centre, were collected and tested.

The topsoil samples were a mixture of common soil types taken from the area surrounding the sampling site using methods of quartering; these soil types include cultivated land, woodland, and wasteland. The sampling depth was 0–10 cm. Both of the two topsoil samples had extensive coverage of local Quaternary loess, which was used as the background value for atmospheric dust in this study.

2.3. Analytical methods

Dust and soil samples were ground into a powder in an agate mortar, sieved (mesh size 75 μ m), and dried in an oven at 105 °C for 3 h. The methods used to digest the sample powders have been previously described (Yang et al., 2007; Roy and Smykatz-Kloss, 2007). In brief:

- 1) 100 mg of sample powder was digested with 1 ml HF and 2 ml HNO₃ in a PFA sample jar (Saville, US) at 140 °C for 7 days.
- 2) Samples were resuspended in 1 ml HF and 2 ml HNO₃ using the same treatment in step 1. This was repeated until the solution became clear.
- 3) After the samples were completely digested, 2 ml HNO₃ (1:1) was added two times to break up fluorine compounds. Then, samples were dried and vaporized on a hot plate.

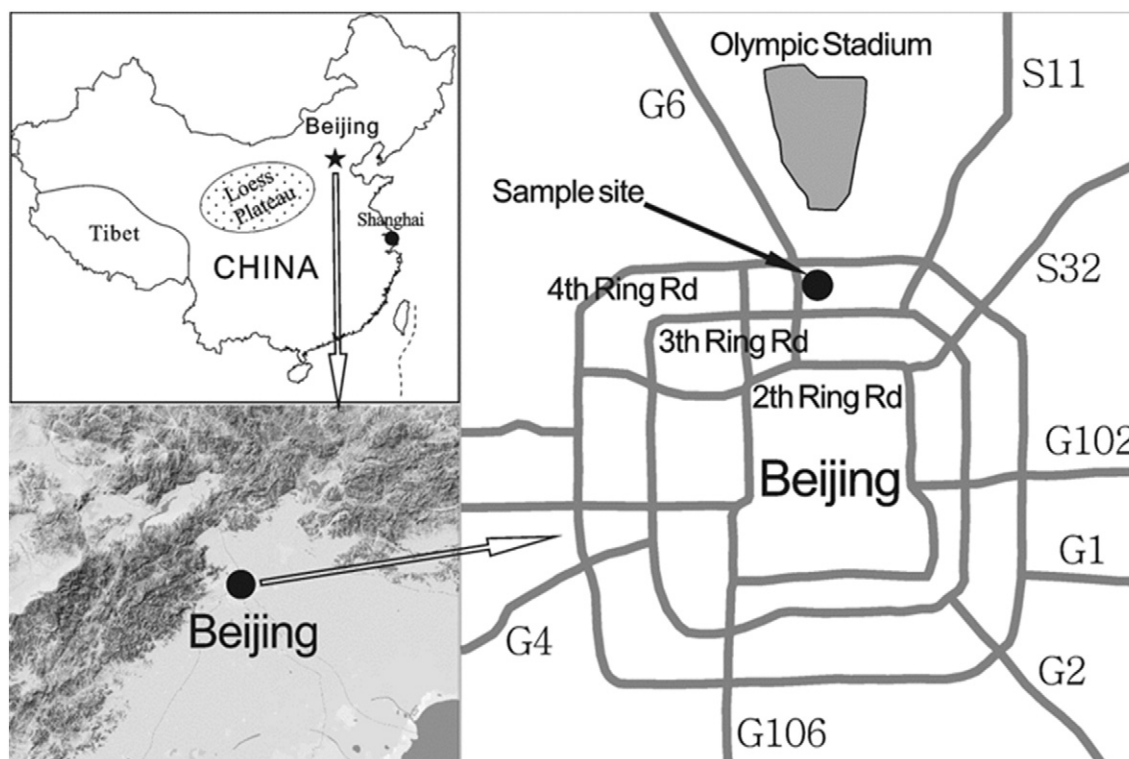


Fig. 1. Map of study area and sample site.

4) Finally, the remaining digest was dissolved in a 100-ml volumetric flask using 3% HNO₃. The digestion processes was performed in the Ultra-Clean Lab of the Institute of Geochemistry, Chinese Academy of Sciences.

The major elements (Na, Mg, Al, K, Ca, and Fe) concentrations of the digested solutions were analysed using an Vista MPX ICP-OES. Heavy metals (Sc, V, Cr, Co, Ni, Cu, Zn, Ba, and Pb) were tested by an Agilent 7500a ICP-MS at the State Key Laboratory of Geological Processes and Mineral Resources at the China University of Geosciences in Wuhan. Testing error was $\pm 6\%$ for Fe and $\pm 3\%$ for the other major elements. Analytical precision was estimated to be $<5\%$ for the heavy metals, based on duplicate analyses of samples and standards. Quality control was performed using the national standard reference material of China (GBW07427).

3. Results and discussion

3.1. Elements cluster analysis and correlation

The concentrations of major elements and heavy metals in atmospheric dust and topsoil samples of Beijing city are shown in Table 1. Main elements and heavy metals concentration of the dust and the topsoil were different, and can be ranked by abundance as follows:

Dust: Ca > Al > Fe > Mg > K > Na > Ba > Zn > Pb > Cu > Cr > V > Ni > Co > Sc.

Topsoil: Al > Fe > K > Na > Ca > Mg > Ba > V > Zn > Cu > Cr > Ni > Pb > Co > Sc.

The atmospheric dust in Beijing has higher average Ca and Mg content compared with topsoil, whereas Fe content in dust is lower than that of the topsoil; other major elements in dust are similar to topsoil. For heavy metals, the average concentration of Pb, Zn, and Cu in dust were higher than those in topsoil.

To determine whether elements are derived from natural or anthropogenic sources, concentration data of the dust was analysed using the hierarchical cluster method. The results (Fig. 2) show that the elements can be divided into three groups: Ca comprises group I; Zn, Pb, Cu, and Ni form group II; and V, Sc, Al, Fe, Co, K, Cr, Mg, Na, and Ba form group III. This cluster pattern was similar to the results of the elements in the urban soil of Beijing (Wang et al., 2012). In urban areas, cement was widely used for buildings and roads, which can be the major source of Ca in dust; thus, Ca exhibits distinctive geochemical behaviours. Group II is comprised of typical heavy metals derived from anthropogenic sources; these elements are generally from emissions industrial emissions and vehicles (Azimi et al., 2003; Schleicher et al., 2011). In urban soils of Beijing, Zn, Pb, and Cu were mainly affected by anthropogenic

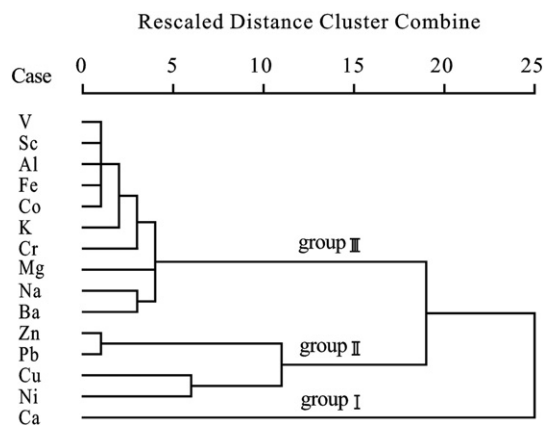


Fig. 2. Results of hierarchical cluster analysis (dendrogram) of the major elements and heavy metal concentrations of atmospheric dust in Beijing ($n = 12$).

sources (Xia et al., 2011). Most elements of group III have similar levels in dust and topsoil. These elements are generally considered to be minimally influenced by human activities and are mainly derived from natural sources; therefore, it makes sense that they are similar to local background values. Ca was negatively correlated with the other elements according to Pearson's coefficient (Table 2); the correlation was stronger between Ca and the elements of group III ($-0.32 > r > -0.77$, $n = 12$), indicating that Ca and group III are complementary as the main source of the dust. This may be related to monsoons; in the winter and spring the northwest wind dominates. Although windy weather can produce more dust from concrete buildings and roads, a large number of EAD inputs made atmospheric dust more prone to display the natural characteristics of group III. While the southeast monsoon brings less EAD in summer and autumn, the dust from concrete buildings and roads is rich in calcium. Elements of group III were significantly positively correlated in couples ($0.61 < r < 0.99$, $n = 12$), which suggests that they are derived from the same sources, likely natural. In group II, Zn and Pb were strongly positive correlated ($r = 0.93$, $n = 12$); they were also positively correlated to Ni, but Cu had a weak correlation to the other elements of group II, indicating that elements of group II were produced by different types of human activities.

3.2. Seasonal variation

Both main element and heavy metal concentrations in atmospheric dust varied seasonal. In the summer of 2008 (Beijing Olympic Games), the concentration of Ca in dust was relatively higher than in other seasons, whereas contents of other major elements were relatively lower

Table 1
Concentrations of six major elements ($\text{g}\cdot\text{kg}^{-1}$) and nine heavy metals ($\text{mg}\cdot\text{kg}^{-1}$) in atmospheric dust of Beijing.

Sample	Na	Mg	Al	K	Ca	Fe	Sc	V	Cr	Co	Ni	Cu	Zn	Ba	Pb
Mar	14.7	16.8	51.4	15.9	58.3	28.9	8.80	69.5	71.5	12.2	34.9	81.6	389	581	62.7
Apr	11.9	18.4	48.1	15.6	76.9	27.6	8.23	66.4	70.1	11.4	31.9	70.0	409	547	87.2
May	12.3	15.8	53.7	16.4	55.0	29.9	9.41	72.6	69.0	11.8	31.8	52.5	269	578	63.0
Jun	10.0	14.7	58.6	15.1	30.6	35.4	9.04	76.3	94.5	12.8	48.1	110	784	507	259
Jul	7.92	12.9	31.2	9.40	77.7	22.2	5.11	46.6	68.1	9.27	36.2	105	549	494	168
Aug	6.54	8.65	34.0	9.38	50.0	19.7	4.93	42.9	60.4	8.22	44.0	94.1	612	347	241
Sep	6.54	8.79	16.8	8.25	106	11.4	2.82	25.7	36.0	6.29	30.6	78.2	374	330	121
Oct	10.4	17.0	44.0	15.0	72.8	27.8	7.53	59.3	74.2	11.2	35.9	75.3	369	583	106
Nov	12.1	17.5	51.7	13.9	69.2	29.2	8.32	67.6	76.4	12.3	37.9	98.1	302	580	68.7
Dec	11.8	15.8	48.4	13.6	59.8	28.3	8.07	62.6	74.6	13.8	36.8	96.6	302	620	72.7
Jan	15.0	16.6	58.6	16.9	51.9	33.4	9.86	77.3	80.7	14.7	41.2	143	216	999	66.8
Feb	15.5	16.4	51.3	15.4	59.0	34.2	8.94	74.1	89.4	13.5	41.9	172	325	953	109
dust-mean	11.2	14.9	45.7	13.7	63.9	27.3	7.59	61.7	72.1	11.5	37.6	98.0	408	593	119
soil-1	15.1	7.39	38.4	18.3	10.6	31.4	7.02	83.8	61.0	10.6	25.7	20.7	67.0	441	25.1
soil-2	18.4	8.40	53.3	21.4	9.64	44.1	9.41	103	58.6	16.9	24.0	66.6	72.8	588	19.6
soil-mean	16.7	7.89	45.9	19.9	10.1	37.8	8.21	93.6	59.8	13.7	24.9	43.6	69.9	514	22.3
blank	0.01	0.01	0.02	0.07	0.04	0.06	0.03	0.07	0.16	0.09	0.07	0.23	0.35	0.32	0.04

Table 2
Pearson correlation coefficient between fifteen elements of atmospheric dust in Beijing (n = 12).

	Na	Mg	Al	K	Ca	Fe	Sc	V	Cr	Co	Ni	Cu	Zn	Ba	Pb
Na	1.00														
Mg	0.81**	1.00													
Al	0.79**	0.79**	1.00												
K	0.87**	0.87**	0.92**	1.00											
Ca	-0.36	-0.25	-0.76**	-0.52	1.00										
Fe	0.79**	0.78**	0.96**	0.88**	-0.74**	1.00									
Sc	0.87**	0.85**	0.98**	0.96**	-0.66*	0.95**	1.00								
V	0.84**	0.83**	0.99**	0.94**	-0.70*	0.98**	0.99**	1.00							
Cr	0.63*	0.66*	0.86**	0.71*	-0.77**	0.95**	0.82**	0.88**	1.00						
Co	0.86**	0.81**	0.93**	0.86**	-0.62*	0.93**	0.94**	0.93**	0.85**	1.00					
Ni	0.05	-0.09	0.39	0.09	-0.76**	0.47	0.25	0.34	0.64*	0.34	1.00				
Cu	0.41	0.08	0.26	0.13	-0.31	0.42	0.24	0.31	0.53	0.44	0.64*	1.00			
Zn	-0.57	-0.43	-0.16	-0.37	-0.34	-0.09	-0.29	-0.20	0.12	-0.34	0.55	-0.03	1.00		
Ba	0.85**	0.61*	0.65*	0.68*	-0.33	0.72**	0.71*	0.71*	0.64*	0.81**	0.25	0.72**	-0.54	1.00	
Pb	-0.64*	-0.61*	-0.25	-0.48	-0.33	-0.17	-0.39	-0.29	0.07	-0.39	0.66*	0.13	0.93**	-0.46	1.00

** Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2-tailed).

than in other seasons. Both the maximum and minimum values appeared in September. The same pattern was observed in the heavy metals: Cu, Zn, and Pb reached maximum levels in June. Heavy metal contents of roadside soils in Beijing showed that these elements were mainly influenced by traffic (Chen et al., 2010). The remaining heavy metals reached minimal content levels in September. The seasonal variations in major element and heavy metal concentrations of atmospheric dust might be jointly influenced by the monsoon, the climate, and the government policies for the 2008 Beijing Olympic Games. Beijing has been troubled by East Asia Dust (EAD) for a long time; according to previous studies, this dust was mainly from the sandy lands of northern China (Sun et al., 2005). Mass concentrations of most natural mineral elements of particles increased during dusty weather (Zhang et al.,

2009). Almost all of the dusty weather occurred in winter and spring (Guo et al., 2004; Qian et al., 2002); in these two seasons, the atmospheric dust of Beijing was dominated by EAD, which had a higher concentration of elements from group III. Anthropogenic elements of group II were depleted in the dust during these seasons.

According to Han et al. (2005), contributions from outside Beijing in spring and winter are higher than those in summer for TSP, which was clearly related to various meteorological factors. In summer and autumn, when dusty weather has minimal impact on Beijing, the prevailing wind direction was southwest, and atmospheric dust from the EAD and the topsoil of surrounding area were minimal. During this time, cement buildings, industry, and vehicles became the main sources of dust; thus, the contents of Ca and elements of group II were relatively

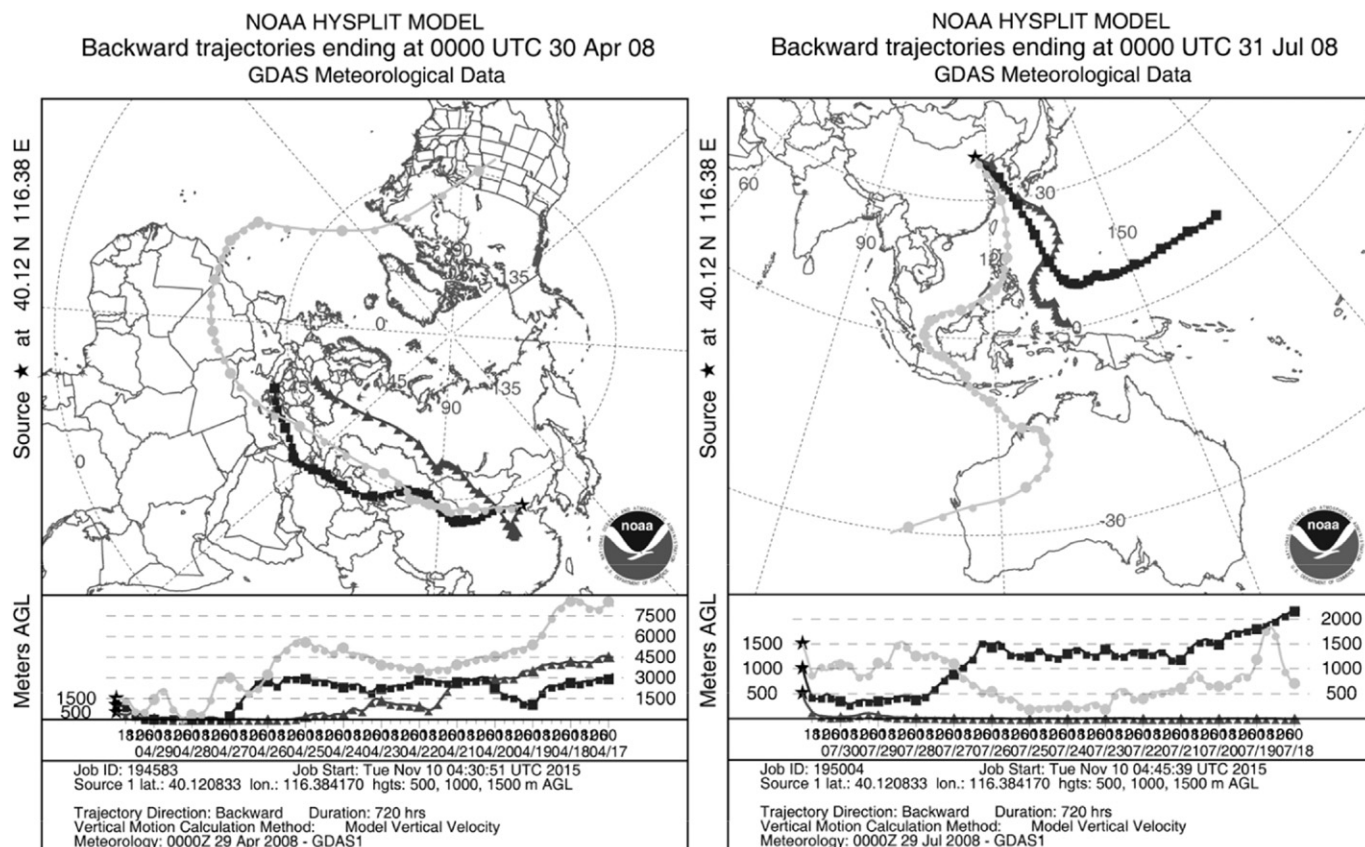


Fig. 3. 30-days backward trajectories at 30 Apr. 08 and 31 Jul. 08 in Beijing with heights 500 m, 1000 m and 1500 m.

higher compared to other seasons. Backward trajectory results also showed the seasonal difference of atmospheric circulation (Fig. 3). In spring (April) 2008, the air mass influencing Beijing was mainly passing through the Gobi desert of west China, which was the source of EAD. Meanwhile, in summer (July) 2008, the air mass was mainly coming from the southeast China Sea, where there was less dust. Thus, the atmospheric dust of Beijing might be influenced by the sources of the predominant air mass. Ca/Al proved to be a good signature for tracing different dust origin areas due to differences in Ca content among source regions (Sun et al., 2005; Wang et al., 2011; Yuan et al., 2008). Pb was a typical anthropogenic heavy metal mainly derived from fossil fuels (De Vleeschouwer et al., 2007; Simonetti et al., 2003). Together with Ca, Pb could be used to distinguish different anthropogenic sources of dust in different seasons. Fig. 4 shows that the Ca and Pb characteristics of atmospheric dust in Beijing were different from local topsoil. The dust was also divided into two groups: dust-A (Jun. to Sep.) was more dispersed and enriched in Pb than dust-B (the rest of the season). Most dust and spring aerosols of Beijing had higher Ca/Al values than the spring aerosols of Hunshan Dake (Chen et al., 2005a; Chen et al., 2005b), indicating that Ca was enriched in the air of urban areas due to cement use. Both Beijing and Hunshan Dake aerosols had significantly higher Pb/Al (> 1000) than the dust of Beijing, suggesting that anthropogenic heavy metals such as Pb were enriched in fine particulates (Zhao et al., 2010).

3.3. Abundance and enrichment factor

Abundance of major elements and heavy metals was normalized by the average topsoil values of Beijing in this study (Fig. 5). Most elements had a topsoil-normalized value which approached 1 and ranged from 0.3 to 3, with the exception of Ca, Cu, Zn, and Pb. Abundance values of Ca in dust were between 3.02 and 10.44, indicating that Ca was enriched in all the dust sampled. Cu, Zn, and Pb showed abundance of varying degrees, which was more obvious in summer and winter. To better understand the natural and anthropogenic sources of the dust in Beijing, enrichment factor (EF) was used to estimate the contribution rate of anthropogenic sources to dust. EF values were the concentration ratios of target elements in the dust samples normalized to corresponding local topsoil, which were calculated by the following expression, $EF_d = (C_{ed}/CAI_d)/(C_{es}/CAI_s)$, where C_{ed} and CAI_d are concentrations of the element x and Al in dusts, and C_{es} and CAI_s are those in the local topsoil. If the EF values are near 1, local sources are predominant. Generally, a value > 5 indicates that non-local or anthropogenic sources were substantial. Calculated EF values showed most of the dust was enriched in Ca, Zn, and Pb (Fig. 6). Mg, Cu, and Ni were slightly enriched, with EF

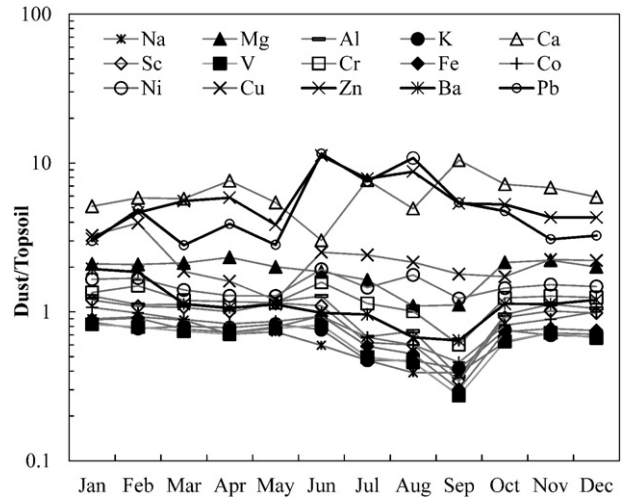


Fig. 5. Abundances of fifteen elements in atmospheric dust normalized to local topsoil in Beijing.

values between 1 and 5, while the rest of elements showed no enrichment characteristics. Higher EF values were closely related to human activities. Seasonal variations in EF values showed that all the elements were enriched in summer and autumn (Jun. to Sep., Fig. 7), which can be attributed to the absence of dusty weather and the fine vegetation coverage in this period. This is also explained by the different sources of atmospheric dust in Beijing that change with the season.

4. Conclusions

The major elements and heavy metals in the atmospheric dust of Beijing can be classified into three groups according to cluster analysis. Ca is a separate group, mainly due to the influence of cement buildings and roads on the atmospheric dust. Zn, Pb, Cu, and Ni can be classified as anthropogenic group. The remaining elements are classified as natural group.

There is a negative correlation between Ca content and the other major elements. A negative correlation is also observed between the contents of Zn, Cu, and Pb and the remaining heavy metals. The concentrations of Ca, Zn, Cu, and Pb in atmospheric dust are relatively higher in summer and autumn than during other seasons. This is mainly because of the absence of dusty weather in summer and autumn in Beijing, when the atmospheric dust is mainly from anthropogenic sources.

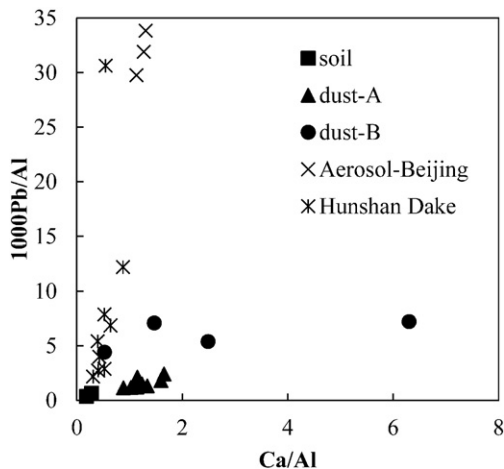


Fig. 4. Covariation of Ca/Al vs. 1000 Pb/Al of atmospheric dusts and local topsoil in Beijing, the aerosol of Beijing and Hunshan Dake in spring (Chen et al., 2005a; Chen et al., 2005b).

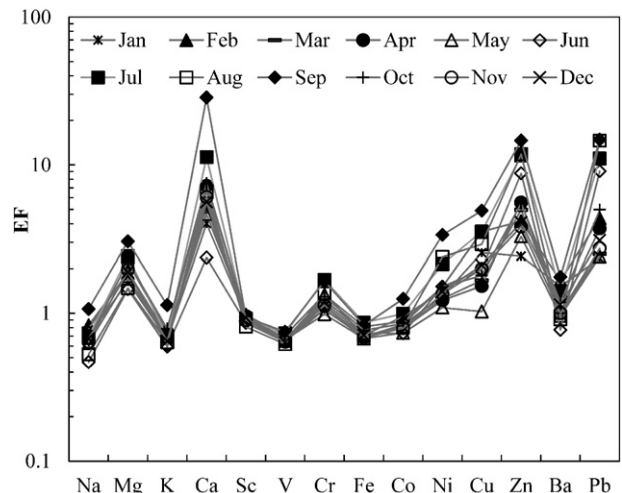


Fig. 6. Enrichment factors of atmospheric dust in Beijing.

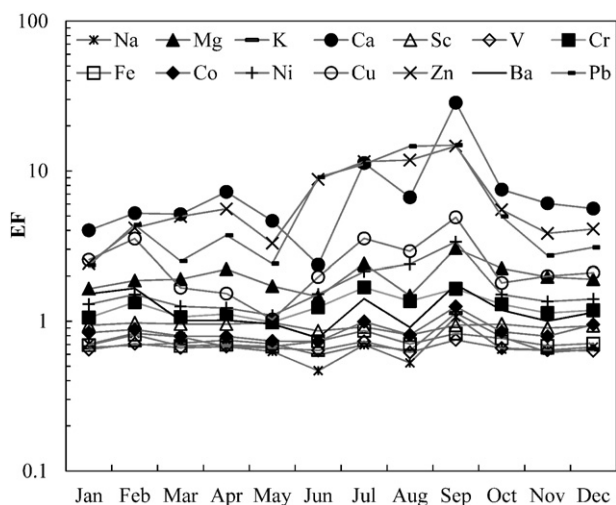


Fig. 7. Seasonal variations of EF values of atmospheric dust in Beijing.

Compared with the local topsoil, atmospheric dust in Beijing is relatively enriched in Ca, Cu, Zn, and Pb. Calculated EF values show that the enrichment characteristics are more obvious in the summer and autumn. The EF values of Ca, Cu, Zn, and Pb are indicative of significant anthropogenic sources. The primary source of atmospheric dust in Beijing is not the same in all seasons. The frequent dusty weather in spring and winter dominates the natural sources, while atmospheric dust is mainly influenced by anthropogenic sources in other seasons.

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