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Heavy Metal-Polluted Aerosols Collected at a Rural Site, Northwest China

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ABSTRACT: Daily samples of aerosol (*n*=27) were collected from September 21st to October 4th, 2013 in Fukang (44.17°N, 88.45°E, 475 m a.s.l.), Xinjiang, Northwest China. The enrichment factors (EFc) of selected 49 elements showed that the aerosols had extremely high concentrations of heavy metals, probably indicating their anthropogenic origins. Morphology of individual aerosol particles was determined by scanning electron microscopy and energy-dispersive X-ray microanalysis. Based on morphology and elemental composition, the particles were clustered into three dominant types: (I) crustal originated particles: Si/Al-rich particles (36%) and Si/Fe-rich particles (24%); (II) mixed source particles; and (III) pollution derived particles: Pb-rich particles (10%). The backward trajectories were calculated using the HYSPLIT model, and the results indicated the different anthropogenic sources for heavy metals in Fukang aerosols. Air mass from north was identified as the most polluted source when compared to south and west.

KEY WORDS: heavy metal pollution, aerosols, Central Asia.

0 INTRODUCTION

Atmospheric pollutions in China especially metropolises have become increasingly heavier during the last decades with the rapid development of industrialization and urbanization (Tan et al., 2013). A great deal of relative research has focused on the middle and east parts of China (Huang et al., 2014; Jiang et al., 2014; Shen et al., 2014; Wei et al., 2014; Yin et al., 2014; Lu C et al., 2013; Xiao et al., 2013; Cheng and Hu, 2010; Lu H et al., 2008; Shi et al., 2008; Hao et al., 2007; Wang et al., 2007; Wong et al., 2002). While those in western China, especially rural areas, have received little attention because of the undeveloped economy and relatively lower pollutant emissions as well as a lack of field measurements (Li et al., 2014; Wang et al., 2011; Guo et al., 2004; Borbon et al., 2002; Sexton and Westberg, 1984).

However, previous studies showed that long-range transport under westerly wind, such as Asian dust events and biomass burning, could be contributing to the concentrations of aerosol pollutants in eastern China (Fang et al., 1999). Jaffe et al. (1999) suggested the surface emissions could be lifted into the free troposphere over Asia and then transported to North America in ~6 days. Regional air pollution study of a rural site in western China therefore will be a necessary supplement to the global atmosphere quality research.

In this article, we focused on the trace metals pollution of rural ambient in Fukang Station (FK), Xinjiang Uygur Autonomous Region, Northwest China (Fig. 1). Ion chemistry and individual particle analysis of atmospheric aerosols near this station has been reported by Zhao et al. (2011). However, previous work concerned only on inorganic ion chemistry properties, there is no data reported to reflect the heavy metals on the atmosphere over this region, and only using regular inorganic ions to deduce the anthropogenic pollution is insufficient. Trace metals analyses in this work will reflect the air conditions in this region more directly and effectively.

The overall purpose of this research was attempted to answer the following three questions: ① Does FK under clean or polluted atmospheric conditions? ② Are these pollution sporadic cases or daily events? ③ Where does pollution come from? Whether the different pollution sources carry the same element? To answer the first question, the general concentration characteristics of trace elements in FK aerosols were examined, followed by the comparison with other places. Then, enrichment factor analysis using aluminum as a reference element was determined combined with morphology results to answer the second question. Finally, the daily variations of pollutant percentage and individual heavy metals concentrations as well as backward trajectories were calculated in order

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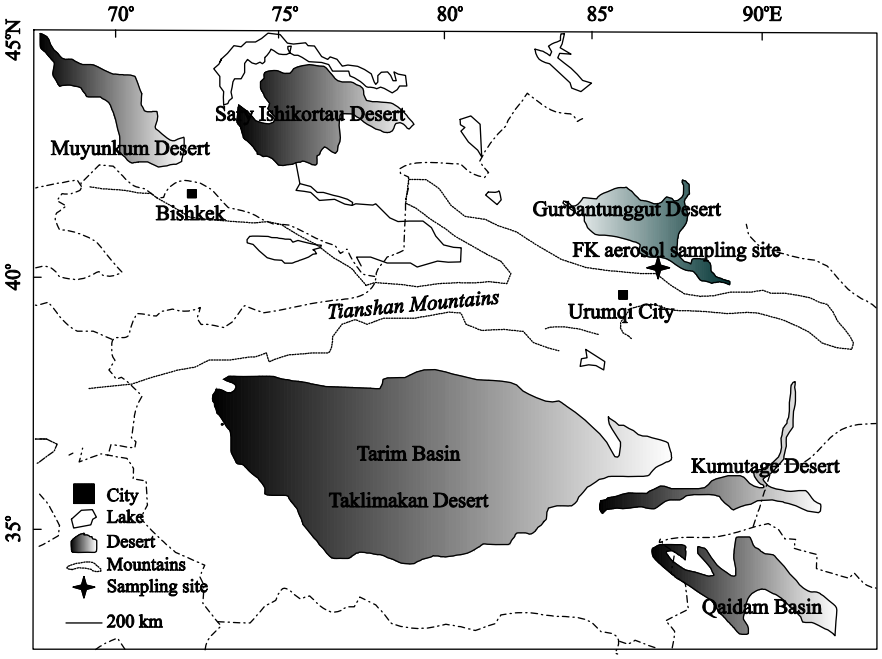

Figure 1. Location of the sampling site in Fukang Station and surrounding geographic environment.

to identify emission sources and feature of different pollution sources in FK.

We expect that our results could serve as a fundamental data set for the health risk assessment in the future and can be useful for making pollution control strategies in the FK as well as rural sites of Northwest China. Moreover, we were trying to relate the heavy metal pollution in ambient air to other environmental medium, such as snow and ice core in the Tianshan Mountains region.

1 METHODS

1.1 Site Description and Sampling

The aerosol samples were consecutively collected from September 21st to October 4th, 2013 at the Fukang Station of Desert Ecology, Chinese Academy of Sciences (44.17ºN, 88.45ºE, 475 m a.s.l.) (Fig. 1).

The FK Station located on the north foot of the eastern Tianshan Mountains, controlled by high atmospheric pressure from Mongolia all the year round with westerly jet stream prevails across this area. The sampling site is approximately 18 km north from central Fukang City, 63 km from Mt. Bogda and 72 km northeast and commonly downwind from Urumqi, which is the capital city with one million people and the industrial center of the Xinjiang Uygur Autonomous Region.

Aerosol samples were recovered on ZEFLUOR filters (2.0-μm pore size, 47 mm, Pall Life Sciences) using a 12-V diaphragm pump powered by solar cells, avoiding the contamination from the power generator. Filters were loaded in the field and mounted face down about 2 m above the ground surface. The sampling period for each day and night aerosol sample was 8–10 h, respectively. Twenty-seven aerosol samples were collected. The air volume was converted into standard conditions according to the local ambient conditions. After sampling, the filters were removed from the filter holder and placed into clean airtight plastic containers stored at 4 ºC before analysis after sampling.

1.2 Chemical Analysis

The collected aerosol filters were placed in a Teflon high pressure vessel for digestion using 2 mL of mixed solution (1.5 mL concentrated HNO_3 and 0.5 mL concentrated HF). The vessels were treated in ultrasonic bath for 25 min in the second step. Subsequently, the samples were digested in an oven for 4 h at 190 ºC. After cooling, the solutions were heated at 150 ºC, and another 0.5 mL HNO_3 was added into the residue, and further heated on a hot plate at 170 ºC for 4 h. This procedure for digestion was repeated twice. In each digestion batch (27 samples), a reagent blank was also used to check the sample handling processes. A total of 68 elements were measured by inductively coupled plasma-mass spectrometry (ICP-SFMS, Element, Bremen, Germany) in a 1 000-class clean room in the State Key Laboratory of Cryospheric Sciences, Cold and Arid Regions Environmental and Engineering Research Institute, Chinese Academy of Sciences, Lanzhou.

Elemental concentrations were quantified using external calibration standards (AccuTrace Reference Standard). A check standard was analyzed after the initial calibration and after every 10 samples. The method detection limits were listed as follow (Table S1). For precision, the corresponding RSD values of all element concentrations measured in the reference material were less than 5%. The final concentrations were corrected with reagent and filter blanks.

Individual particles were analyzed using a field emission scanning electron microscope (JSM-6701F) equipped with an energy-dispersive X-ray spectrometer (EDX/EDS). Section of each filter was cut and mounted onto the electron microprobe stub, and coated with a thin gold film for a higher quality secondary electron image. Operating conditions were: accelerating voltage=5–10 kV; spectral acquisition times=60 s. Noran System software for energy-dispersive microanalysis was used for

the quantitative analysis of individual particles.

2 RESULTS

2.1 General Characteristics of Trace Elements

Table S1 describes the average concentrations (μg/m^3) of selected 49 elements at the FK site during the observation period. Based on the standard periodic table and chemical properties, the elements analyzed can be grouped into four categories: (1) alkali metals and alkali earth metals: Na, K, Rb, Cs, Be, Mg, Ca, Sr, Ba, and Al; (2) transition metals: Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, and Zn; (3) lanthanoids: La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu; (4) heavy elements: As, Sb, Se, Ag, Cd, Hg, Sn, Pb, Tl, Zr, Hf, Ta, W, Th, and U.

According to the statistical summary in Table S1, the daily average concentration of selected 49 elements ranged from 0.000 5 to 20.87 μg/m^3. The lanthanoids was found to be the lowest with a value of 0.015 μg/m^3, accounting for 0.4% of the total metal concentrations in the air mass, while alkali metals and alkali earth metals showed the highest concentration ranged from 0.000 5 to 20.866 1 μg/m^3, constituted about 78.4% of the measured concentrations. The transition metals and heavy metals accounted for about 11.1% and 10.1% of the total concentration, respectively.

Overall, the average concentrations of alkali metals, alkali earth metals and geogenic elements (e.g., Fe) were 2–4 orders of magnitude higher than the transition metals, heavy elements and lanthanoids.

2.2 Morphology Result

The final dataset included around 1 500 particles also analyzed on 12 filters. The particles were grouped into three dominant types according to their morphology and EDX signal: Si/Al-rich particles (abundance 36%), Si/Fe-rich particles (abundance 24%) and Pb-rich particles (abundance 10%) (Fig. 2).

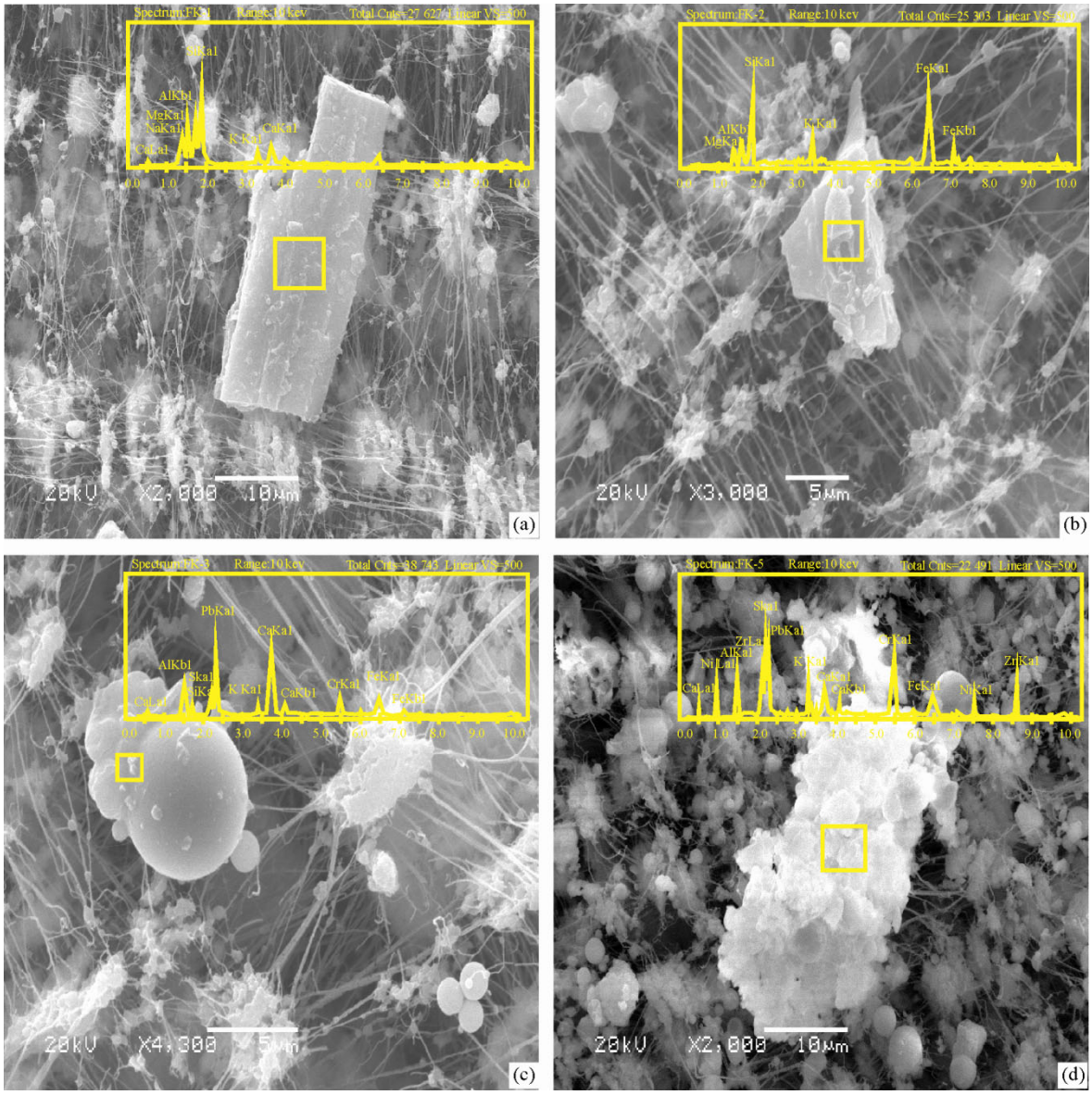

Figure 2. SEM images of typical aerosol particles. (a) Si/Al-rich particles; (b) Si/Fe-rich particles; (c) Pb-rich particles; (d) particles containing soot with varying heavy metals and crustal elements.

3 DISCUSSION

3.1 Comparison of Trace Element Concentration with Other Data

To further compare the elements concentrations in FK aerosols, data from other cities have also been included in Table S2. It is clear that concentrations of different elements in FK were generally comparable with the Urumqi and Beijing (two heavy polluted cities in China), except for As, Se, Sb and Cd. The concentration of As, Se, Sb and Cd in FK is distinctly higher than those in Beijing, indicating that FK receives even more anthropogenic influence since the dominant sources of As, Se, Sb and Cd are closely related to human activities (Ayrault et al., 2010; Pey et al., 2010).

In comparison with south of the city of Almaty, a remote site (43.04ºN, 76.58ºE, 2 760 m a.s.l.) representing the regional background condition in Tianshan Mountains (Hoornaert et al., 2004), the averaged concentrations of elements in FK were significantly higher, from 10 fold for Cr to 323 fold for Zn.

Our mean concentration values measured in FK were even higher than previous reliable data obtained from Tibetan Plateau, despite difference in time periods investigated. For example, arsenic concentrations in HK were 247 to 11 106 times higher than that in Lhasa and Nam Co, respectively (Table S2).

The average concentration of total detected elements in FK was 74.1 μg/m^3, higher than the China's Ambient Air Quality Standard (GB 3095-2012) (MEP and GAQS, 2012) for cities (limit of 50 μg/m^3).

The results indicated that there might be influences of anthropogenic activities on the atmospheric environment in the FK. Considering that alkali metals, alkali earth metals and other main geogenic elements (e.g., Fe) in crust/soil/street dust are also several orders than trace metals, the absolute concentration is not an ideal measure for the differentiation between natural and anthropogenic sources. Instead, enrichment factor analysis is discussed below.

3.2 Enrichment Factor Analysis

Trace elements in the atmosphere originate from natural sources such as rock and soil dust, sea-salt spray, volcanoes, wild forest fires and marine biogenic sources (Lee et al., 2008). On a global scale, however, the emissions of trace elements into the atmosphere from various anthropogenic sources are known to exceed those from natural sources (Nriagu et al., 1989). Crustal enrichment factors (EFc) is often calculated to identify source regions and evaluate the degree of anthropogenic influence (Hur Do et al., 2013; Öztürk et al., 2012; Lee et al., 2008; Cong et al., 2007; Duce et al., 1975) EFc is defined as the concentration ratio of a given element to that of Al (Al, Si, Ti, or Fe were commonly used as approximation of rock and soil dust because they are abundant in crustal material and are not significantly affected by contamination), normalized to the same concentration ratio characteristic of the upper continental crust. For example, the EFc for Cr is thus

$$EFc=(Cr/Al)_{aerosol}/(Cr/Al)_{crust}$$

where Cr represents the element of interest; Al is a reference element. Here, we have used the data for the upper continental crust given by Wedepohl (1995). The aerosol and crust subscripts refer to element in the aerosol samples and crustal material, respectively. An EFc value close to unity is considered to indicate a dominant input from rock and soil, while an EFc value greater than 10 suggests that the corresponding element originated mainly from other sources (Wu et al., 2009; Cong et al., 2007; Ferrari et al., 2001).

Figure 3 shows the mean values of crustal enrichment factors for the measured trace elements. The average EFc values are observed to be highly variable between elements, with the lowest value determined for Zr (0.18) and the highest determined for Se (37 137.60). This wide range of EFc values for each element is an indicator of the differences in crustal contributions from one group to another.

The mean values of EFc are relatively lower for alkali metals and alkali earth metals (all less than 10), with the following occurrence order: Mg<Al<Ba<Be<Sr<Na<K<Ca<Rb, showing that their compositions are close to that of the upper continental crust. The transition metals and lanthanoids have EFc values from 0.35 to 93.78, are considered moderately enriched. Heavy elements were found to be highly enriched (with EFc values greater than 100, ranged from 171.00 to 37 137.00) in the FK aerosol samples.

Our values are in broad agreement with the result from previous studies in Beijing. Yang et al. (2003) reported that EFc values of heavy elements such as Se, Sb, S, As and Pb in Beijing were significantly high, ranged from 121.1 to 10 622.71, while alkali metals, alkali earth metals and transition metals (e.g., Na, Mg, Ca, K, Al, V, Cr, and Mn) were also found below 10.

Based on the previous studies (Wu et al., 2009; Cong et al., 2007), elements can be classified into three types in this study: (I) Crustal originated metals (EFc<10): alkali metals and alkali earth metals are not enriched in FK and can be considered as soil originated metals. (II) Mixed source metals (10<EFc<100): it was possible to establish that transition metals and lanthanoids are influenced both from crustal and anthropogenic sources according to their enriched EFc values (Lee et al., 2008; Hong et al., 2004; Barbante et al., 2003). For example, for Sc, Ti, V, Cr, Mn, Fe, La, Gd and Yb are not enriched with EFc values below 5, can be attributed to crustal originated metals. While high EFc for Co, Ni, Cu, Zn and the remaining lanthanoids elements can be attributed to the presence of these metals in local soil in unusually high concentrations (Koçak et al., 2004; Güllü et al., 1998; Kubilay and Saydam, 1995), while non-ferrous metal production, combustion of petroleum products and waste incineration could also provide the major source of Ni, Cu, Zn and V (Wu et al., 2009; Pacyna and Pacyna, 2001). In addition, differences in reference and local soil composition might lead to slightly <10 EFc for this second group of elements (Öztürk et al., 2012). (III) Pollution derived metals (EFc>100): heavy elements in FK aerosols showed highly enriched with EFc order of Se>Tl>Hg>Sb>Sn>As>W>Ag>Ta>U>Hf>Pb. Previous studies showed that As and Se are elements typically derived from coal combustion (Ayrault et al., 2010; Pey et al., 2010). Stibium, Sn, Zn, Pb and W are released to the atmosphere mainly by industrial emissions and the combustion of leaded and low-leaded gasoline (Öztürk et al., 2012; Wu et al., 2009).

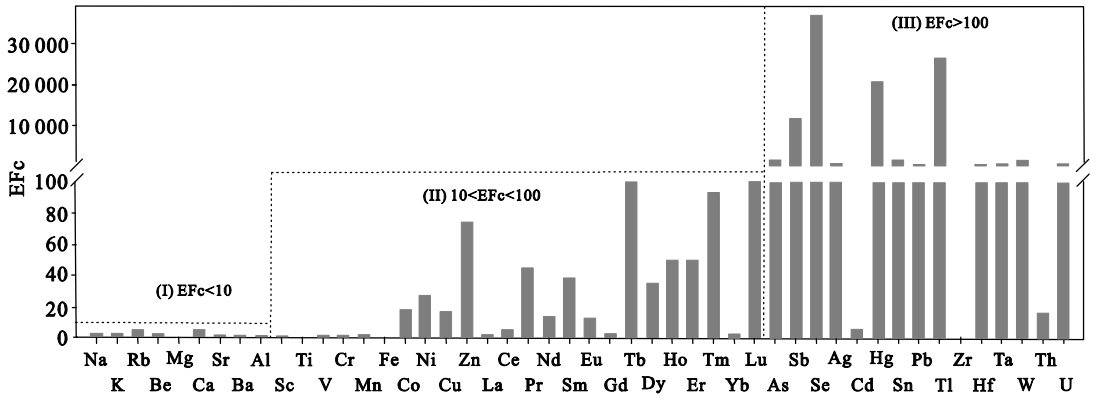

Figure 3. Enrichment factors (EFc) in FK aerosols, relative to Al as the reference element. (I) Crustal originated metals (EFc<10); (II) mixed source metals (10<EFc<100); (III) pollution derived metals (EFc>100).

3.3 Individual Particles Analysis

Figure 2 shows the typical individual particles loaded on the filters at different magnifications. According to the results of enrichment factor analysis above, three particle categories could be obtained based on the morphology and chemical characteristics of individual particles. The distinct characteristics and possible origins of each group are described in detail as follows.

Crustal originated particles: Si/Al-rich particles (abundance 36%). The alumosilicates are characterized by high contents of Si and Al with varying Na, Mg, K and Ca. Particles containing predominantly silicon are classified as silica (e.g., quartz). In our study, alumosilicates and silica were found to be the most abundant particle type with average abundance of 36%. From the point of view of mineralogy, alumosilicates usually include $Al_4[Si_4O_{10}](OH)_4$, $Al_4SiO_{20}nH_2O$, $Na[AlSi_3O_8]$, $K[AlSi_3O_8]$, $Ca[Al_2Si_2O_8]$ and $Ba[Al_2Si_2O_8]$ which are typical terrigenous minerals (Yang et al., 2015; Zhang et al., 2015). Therefore, a large fraction of the irregular shaped particles (Fig. 2a) identified as alumosilicates could be attributed to eolian dispersion of soil particles.

Crustal originated particles: Si/Fe-rich particles (abundance 24%). Particles consisting predominantly of Si and Fe account for 24% of the total aerosol abundance. The Si/Fe-rich particles are considered to be biotite ($K(Mg,Fe)_3[AlSi_3O_{10}(OH,F)_2]$) due to biotite generally associated with magnetite, is abundant in West China, especially in Xinjiang Uygur Autonomous Region. Besides soil dispersion, Fe-rich particle could also be produced by coal-fired boilers and metal industry. Fe-rich particles emanated from high-temperature furnace usually show spherical shape. However, all of Si/Fe-rich particles in this study have irregular (non-spherical) morphology (Fig. 2b), thus identified as natural origin.

Pollution derived particles: Pb-rich particles (abundance 10%). Minor amounts of Pb-rich particles, containing a small amount of S, Ca, Cr Al, Fe and Ca elements occurred in the aerosol samples (Fig. 2c). As analyzed above, Pb had the high absolute EFc value of 230.3, indicating a serious impact of anthropogenic activities. Important sources of atmospheric lead include emissions from automotive emissions, coal combustion, mining or smelting operations and waste incineration. Cheng and Hu (2010) suggested that Pb originates mostly from leaded gasoline. Furthermore, it has been verified that airborne Pb can be carried globally through long-range transport, since it tends to concentrate on fine particles that have a long residence time in the atmosphere (Cong et al., 2010).

Mixed source particles: There also some small soot in many samples. They are clearly distinguishable from other aerosol types due to its unique morphology, shown in Fig. 2d. The morphology of this kind of particle varies from short chains to complex clusters, which depend on different types of fuels, burning conditions, and atmospheric processing (Chakrabarty et al., 2006; Yue et al., 2006). In our study, a considerable percentage of soot particles contain S, Cr, Zn, Pb and Ni with varying crustal elements (e.g., Al, K and Ca), which was observed by EDX analysis, indicating a mixed source of these particles. Paoletti et al. (2003) suggested that the S content in soot aggregates was probably caused by a gas-to-particle conversion process during the transport (Pósfai et al., 2003). Other unrecognized particles could contain organic particles with light elements (such as C, N and O) or biological particles.

3.4 Variations in the Daily Percentage of Three Categories

Variations of calculated daily percentage of crustal originated metals (I), mixed source metals (II) and pollution derived metals (III) are shown blow (Fig. 4) based on the EFc analysis and the morphology results. Generally, the daily percentage of these three categories exhibited pronounced difference with the order of (I)>(III)>(II). The crustal originated metals (alkali metals and alkali earth metals) showed the highest average daily percentage at 69.7%, ranged from 51.0% to 76.0% during the sampling period. The mixed source metals including transition metals and lanthanoids was found to be the lowest with an average percentage of 11.2%, while pollution derived heavy metals accounted for 19.1% of the total metal concentrations. The largest daily variability of pollution derived metals (III) was found (ranged from 11.0% to 41.0%) when compared to crustal originated metals (I) and mixed source metals (II) during the sampling period (Fig. 4), indicating significant day to day variations in source strength of these pollutants.

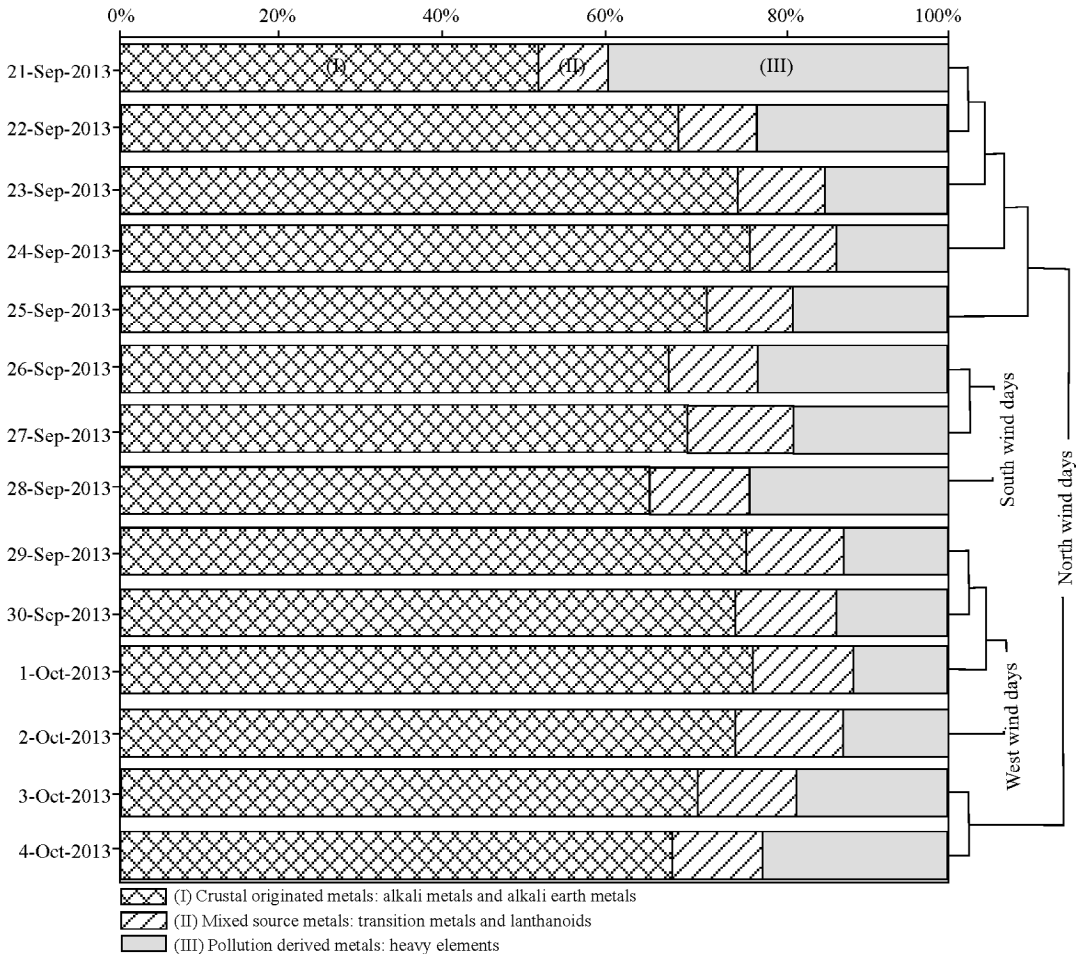

Figure 4. Daily variations in the percentage of crustal originated metals, mixed source metals and pollution derived metals during the sampling period from Sep. 21 to Oct. 4, 2013.

3.5 Possible Source Investigation

In order to investigate potential source regions of pollution derived heavy metals in FK aerosols, 5-day backward trajectories were computed using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT-4) model developed by the National Oceanic and Atmospheric Administration (http://www.arl.noaa.gov/ready/hysplit4. html; Reanalysis Meteorological data). The trajectories were calculated at 500 m above ground level during the period of Sep. 21–Oct. 4, 2013 (Fig. 5). It is apparent that pollution derived heavy metals respond independently to different pollutant sources, day-to-day variations of individual heavy metal concentrations thus have been considered (Fig. 6).

In general, the majority of air mass was from north, west and south. Combining them with the daily percentage variations in different categories (Fig. 4) and the variations of individual heavy metal concentrations (Fig. 6), the air masses arriving at the sampling site can be classified into three types.

(1) Air masses come from the north. Samples of these north wind days exhibited high daily percentage of pollution derived heavy metals, especially on Sep. 21 (41%) (Fig. 4), as well as high concentration of all individual heavy metals (Sb, Ag, Se, As, Hg, Ta, Tl, Sn and Pb) (Fig. 6). For Sb, Ag, Se, their concentrations were less varied during the campaign period except for September 21st and October 4th when peaks were found during these two north wind days (Fig. 6), indicating an anthropogenic sources from the north. The trajectories ending on the Sep. 21–25 and Oct. 3–4 represented the long-range aerosol transport, originating from the northwest regions near Siberia, passing through East Kazakhstan, Karamay Oil Field, Gurbantunggut Desert and south edge of Jungger Basin. Especially, these air masses traveled across the Semipalatinsk test site (Figs. 1 and 5). The Semipalatinsk test site (STS) (50º7′N, 78º43′E) was the primary testing venue for the former Soviet Union's nuclear weapons (Zhang and Edwards, 2011; Olivier et al., 2003). It is located on the Northeast Kazakhstan, south valley of the Irtysh River. The nuclear test site were located around 150 km west of the Semipalatinsk Town, near the border of the East Kazakhstan Province and Pavlodar Province, with most of the nuclear tests taking place at various sites further to the west and south, some as far as into Karagandy Province. While according to the data derived from backward air trajectories from the Tianshan, the typical transit time from possible major source regions (such as Gobi Desert in Mongolia and Badain Jaran Desert in northern China) to the Tianshan (about 1 000 km distant) is 0.5–1 day in summer (Sun et al., 2001). Our sampling site is therefore assumed to be also affected by long range transport of Sb, Ag, Se, Hg, Tl and Sn on Sep. 21 and Oct. 4 emissions from Karamay Oil Field and the nuclear test sites like Semipalatinsk.

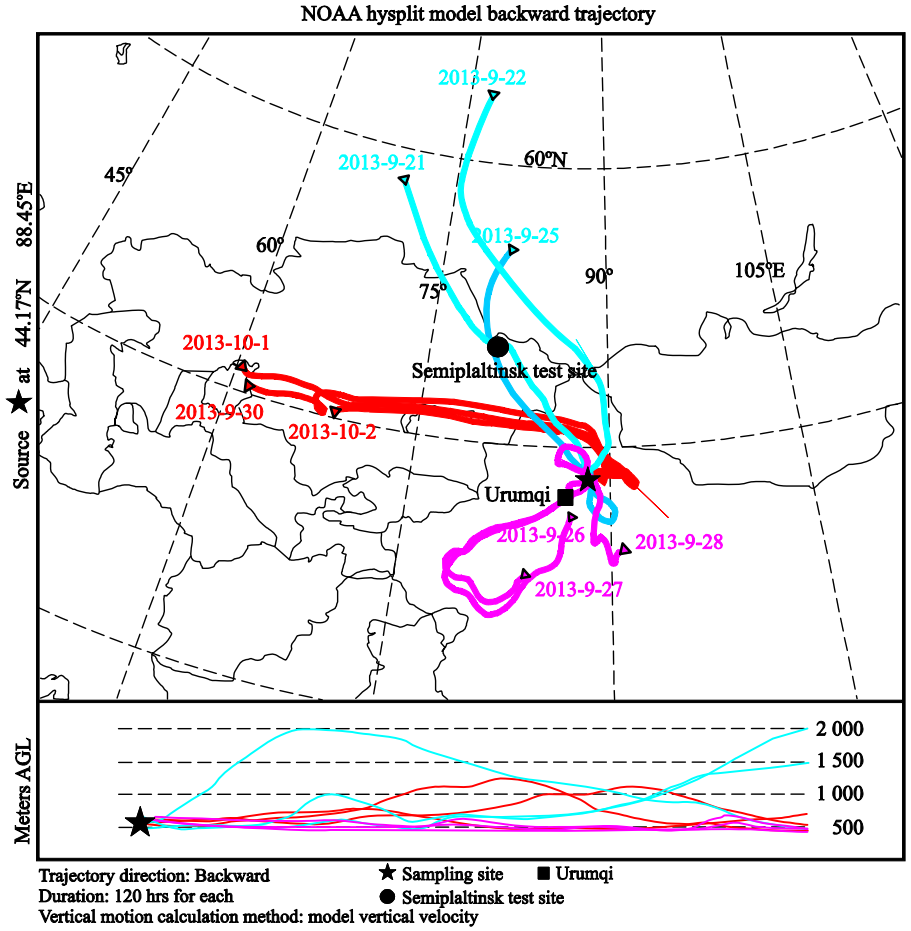

Figure 5. Daily backward trajectories terminating at the sampling site.

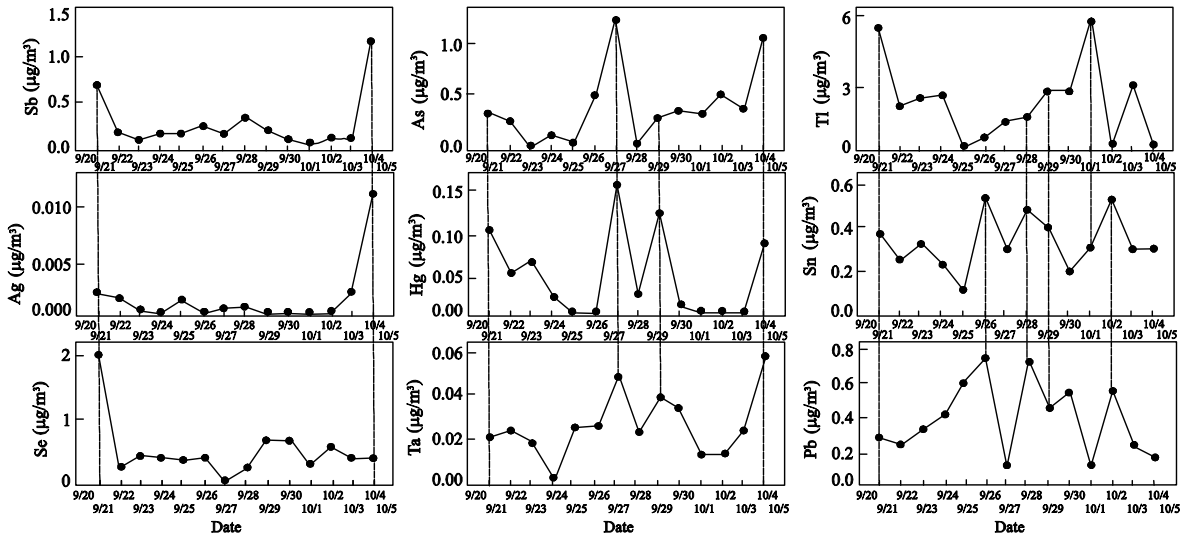

Figure 6. Day-to-day variations in the concentrations of heavy metals during the sampling campaign from Sep. 21 to Oct. 4, 2013.

(2) Air masses come from the west. Elements Tl, Sn and Pb were more pronounced on Sep. 29–30 and Oct. 1–2 in addition to Sep. 21 and 26 (Fig. 6). The trajectories ending on these days were originated from the arid regions of west Central Asia (Fig. 5). It is clear that samples of these west wind days exhibited lowest daily percentage of three categories when compared with those in north and south days (Fig. 4), indicating less pollutant was carried by westerlies although these west air masses resulted in high concentration of Tl, Sn and Pb. In addition, FK sampling site is adjacent to Urumqi Ganquanbao Industrial Park (15 km), which based on petrochemical programs. Air masses come from the west through the industrial park thus probably contributed to high concentration of Tl, Sn and Pb in FK.

(3) Air masses come from the south. For example, compared to Sep. 21 and Oct. 4, As, Hg, Ta, Tl, Sn and Pb were more pronounced on Sep. 26, 27 and 28 (Fig. 6). At the same time, samples of these days exhibited higher daily percentage of pollution derived metals when compared with those from west wind days (Fig. 4). The trajectories arriving on the Sep. 26, 27 and 28 represented the short-range aerosol transport, originating from the south regions near north edge of Qinghai-Tibetan Plateau, passing through the Tarim Basin and Urumqi City. Since there is no anthropogenic activities in the huge Taklimakan Desert, and the long-range pollution influence from South Asia (such as India) was weakly due to the short route of the trajectories in our sampling site, Urumqi City was considered to be the probably sources to As, Hg, Ta, Tl, Sn and Pb at last. Urumqi (86º37′–88º58′E, 42º45′–44º08′N), the capital of Xinjiang Uygur Autonomous Region of China, is in the middle area of Xinjiang, on the north foot of Tianshan and the south edge of the Jungger Basin (Fig. 1). For the past two decades, Urumqi has been suffering from heavy air-pollution and was evaluated as one of the ten heaviest air polluted cities in the world in 1998 (Li et al., 2008; Mamtimin and Meixner, 2007). Enhanced industrial activities in Urumqi might lead to high concentrations of several heavy metals such as As, Hg, Ta, Tl, Sn and Pb.

Besides, the local factories in FK would be absolutely the contributors to the heavy metals of our samples. The advantageous industries of FK City fall to coal production, coal chemical industry, no-ferrous metal production, subsequent deep processing of oil and gas (Wu et al., 2008). Fukang has roughly 28 coal related factories, especially electrolytic nickel production. In addition, FK produced 3 256 t electrolytic nickels in 2005 with 3.87 times of the nation's growth rate (Wu et al., 2008). All of these industrial factories could not shirk responsibility to the heavy metal-polluted aerosols in FK.

4 CONCLUSION

Aerosol samples collected in FK showed that FK has been seriously polluted by heavy metals. Three dominant types of selected 49 elements was determined based on enrichment factors and morphology analysis: (I) crustal originated elements (including alkali metals, alkali earth metals, Si/Al-rich particles and Si/Fe-rich particles); (II) mixed source elements (including transition metals and lanthanoids and mixed source particles); (III) pollution derived elements (including heavy metals and Pb-rich particles). The backward trajectories results indicated the air mass from north was identified as the most polluted source when compared to south and west.

Samples of north wind days exhibited high daily percentage of pollution derived heavy metals, as well as high concentration of all individual heavy metals, such as Sb, Ag, Se, As, Hg, Ta, Tl, Sn and Pb.

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Electronic Supplementary Materials: Supplementary materials (Tables S1 and S2) are available in the online version of this article at http://dx.doi.org/10.1007/s12583-017-0728-6.

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