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Source Apportionment and Elemental Composition of $PM_{2.5}$ in Chengdu, China

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ABSTRACT

In order to investigate the elemental composition of $PM_{2.5}$ and its sources, $PM_{2.5}$ samples were collected at an urban site in Chengdu, China. The contents of 20 elements including Al, As, Ba, etc. in $PM_{2.5}$ were analysed. Enrichment factor, correlation analysis and principal component analysis were applied to analyse the sources of $PM_{2.5}$. The results showed that the daily mean concentration was 49.2~425.0 $\mu g \cdot m^{-3}$ and annual mean concentration was 165.1±84.7 $\mu g \cdot m^{-3}$. The element concentrations in winter (7,575.0 $ng \cdot m^{-3}$) were higher than those in summer (3,326.8 $ng \cdot m^{-3}$). The results of source apportionment showed that the sources of $PM_{2.5}$ were soil dust and dust storm, traffic emission, combustion of fossil fuels and coal, and metal smelting in Chengdu. The results of multiple linear regression analysis showed that wind speed and temperature had a negative correlation with most trace elements.

INTRODUCTION

In recent years, with the rapid development of economy, population and urbanization, fine particulate matter has become a major air contaminant, which severely affects human health, visibility and climate change (Yang et al. 2013). Due to its large surface area, $PM_{2.5}$ can absorb many kinds of toxic air contaminants such as metal elements and organic compounds (Yang et al. 2015). Although elements contribute less to $PM_{2.5}$ compared with other pollutants, some of the elements, even at extremely low contents, pose a threat to human health (Han et al. 2015). For instance, Zn can lead to arteriosclerosis, heart disease and hypertension, As and Cd have a potential teratogenic effect on the human body, and Pb and Hg have a toxic effect on the fetus (Fang et al. 2010). At present, there are several studies on the source apportionment and elemental composition of $PM_{2.5}$ in domestic and overseas regions. In China, investigations on the source apportionment and elemental composition of $PM_{2.5}$ have been conducted in several main cities and regions, such as Beijing, Shanghai, Guangzhou (Zhang et al. 2009). For example, the ground observation of dust aerosols was conducted in Beijing in spring to investigate the elemental composition and origin of mineral dust. The results showed that most mineral elements of particles (i.e. Mg, Si, Fe, Al and Ti) mainly come from dust events. Gu et al. (2011) studied the chemical composition of $PM_{2.5}$ during winter in Tianjin, and the elevated $PM_{2.5}$ was mainly attributed to combustion sources including vehicle exhaust, heating, cooking and industrial emissions, as well as low wind speeds and high relative humidity. Aldabe et al. (2011) evaluated the $PM_{2.5}$ concentration and its trace element levels during 2009 at three different locations (rural, urban and urban-traffic) in Navarra (North of Spain). The results indicated that 90~96% of the total trace elements present in $PM_{2.5}$ consisted of P, Ti, Cr, Mn, Ni, Cu, Zn, Sr, Sn, Ba and Pb, and the main sources were crustal, secondary sulphate, secondary nitrate, traffic and sea-salt aerosols. However, there is limited data available on the elemental composition of $PM_{2.5}$ in Chengdu city, which is located in the Chengdu-Chongqing Economic Zone and is densely populated with serious air pollution.

In this work, the mass concentrations of $PM_{2.5}$ and its elements from April 19, 2009 to January 31, 2010 were determined to evaluate the levels of $PM_{2.5}$ and its elemental components, as well as its seasonal variation characteristics. Furthermore, enrichment factor, correlation analysis and principal component analysis were applied to analyse the possible sources of $PM_{2.5}$. It is expected that the analysis results will provide a scientific basis to regulate and control $PM_{2.5}$ levels in Chengdu.

MATERIALS AND METHODS

Sample collection: $PM_{2.5}$ samples were collected on the roof of a building 15 m above the ground within the Institute of Plateau Meteorology of China, located in the western part of downtown Chengdu in a residential commercial neighbourhood. The $PM_{2.5}$ samples for elemental analysis were collected using a low-flow air sampler (MiniVol TAC,

AirMetrics Corp., Eugene, OR, USA) with a flow rate of 5 $L \cdot min^{-1}$ on 47 mm Teflon filters (Whatman PTFE). The sampling procedure was carried out based on the US EPA National Ambient Air Quality Standard for particulate matter. A total of 121 $PM_{2.5}$ samples and 10 blank samples were collected in spring, summer, autumn in 2009 and winter in 2010. The collection period for each sample was 24 h. The filters were analyzed gravimetrically for particle mass concentrations using a Sartorius MC5 electronic microbalance with a sensitivity of ±1 μg (Sartorius, Göttingen, Germany) after 24 h equilibration at a temperature of 22±1°C and a relative humidity of 40±5%. Each filter was weighed at least three times before and after sampling, and the average weight was calculated to determine the mass concentration of $PM_{2.5}$. All the filters were individually placed into plastic bags and stored in a freezer (-18°C) until the subsequent analysis to limit any contamination.

Elemental analysis: In this study, Energy Dispersive X-Ray Fluorescence Spectrometry (Epsilon 5 ED-XRF, PANalytical B. V., Netherlands) was used to determine concentrations of the elements collected on the $PM_{2.5}$ Teflon filters. In total, 20 elements (i.e. Al, As, Ba, Br, Ca, Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Rb, Sr, Sb, Si, Sn, Ti and Zn) were determined. In addition, quality assurance (QA) and quality control (QC) procedures were carried out, including regular instrument calibration with standard reference materials (SRM), and following the standard QA/QC requirements of the Chinese government. These standard procedures were performed routinely during the entire study. Also, an intensive QC program was implemented to maintain accuracy and precision throughout this project and the ED-XRF spectrometer was calibrated with thin-film standards obtained from MicroMatter Co. (Arlington, WA, USA).

Statistical analysis: A thorough literature survey showed that statistical methodology has been widely used to study possible pollution sources for the atmospheric environment (Lee et al. 2013). In this paper, correlation analysis (CA), principal component analysis (PCA) and enrichment factors (EF) were applied to analyse the possible sources of $PM_{2.5}$ in Chengdu.

RESULTS AND DISCUSSION

Elemental composition of $PM_{2.5}$: During the period of observation, the annual average mass concentration of $PM_{2.5}$ in Chengdu was 165.1 $\mu g \cdot m^{-3}$ (Table 1), which is 4.7 times higher than the National Ambient Air Quality Standard for annual $PM_{2.5}$ (35 $\mu g \cdot m^{-3}$). Daily mean concentrations were 49.2~425.0 $\mu g \cdot m^{-3}$. The number of days which exceeded the second-level ambient air quality standard (75 $\mu g \cdot m^{-3}$, GB3095-2012) issued by the Ministry of Environmental Protection of China accounted for 90.9%. In addition, the total mass concentration of the elements in this study was 6,543.3 $ng \cdot m^{-3}$, which accounted for 4.0% of $PM_{2.5}$ mass concentration. The concentrations of the 20 elements are given in Table 1. It can be seen that the concentrations of the elements followed the order: Fe>Si>Al>Ca> Zn>Pb>Mn>Ti>Br >Cu>Sr>As>Ba>Sn> Cr>Mo>Sb> Cd> Rb>Ni. The concentrations of Fe, Si and Al exceeded 1,000.0 $ng \cdot m^{-3}$ (1,679.0, 1,093.9 and 1,053.0 $ng \cdot m^{-3}$, respectively), which are considered high pollution levels. Then, the concentrations of Ca, Zn, Pb, Mn and Ti were 995.2, 822.9, 320.5, 137.6 and 126.3 $ng \cdot m^{-3}$, respectively. The concentrations of Br, Cu, Sr, As, Ba, Sn, Cr, Mo, Sb, Cd and Rb were lower than 100.0 $ng \cdot m^{-3}$ (ranging from 55.5~9.5 $ng \cdot m^{-3}$). Finally, Ni concentration was the lowest at 5.1 $ng \cdot m^{-3}$. However, As (40.5 $ng \cdot m^{-3}$) concentration was 5.7 times the annual average concentration standard (6.0 $ng \cdot m^{-3}$), and Cd (9.5 $ng \cdot m^{-3}$) concentration was about 2 times the standard (5.0 $ng \cdot m^{-3}$), and they are both typical carcinogenic heavy metal elements. Moreover, the total concentration of the earth's crust elements (including Fe, Si, Al, Ca and Ti in this study) was 4,947.3 $ng \cdot m^{-3}$, accounting for 75.6% of the 20 elements. The results showed that the main elements in atmospheric $PM_{2.5}$ from Chengdu were earth's crust elements and carcinogenic heavy metals, indicating that the pollution was serious.

Comparing the study data with other domestic and global cities (as presented in Table 1), it was found that the $PM_{2.5}$ concentration in Chengdu was higher than Beijing (118.50 $\mu g \cdot m^{-3}$), Guangzhou (81.70 $\mu g \cdot m^{-3}$) and Shanghai (103.10 $\mu g \cdot m^{-3}$). In addition, Si (1,790.0, 2,200.0 $ng \cdot m^{-3}$), Cu (70.0, 60.0 $ng \cdot m^{-3}$), Cr (50.0, 190.0 $ng \cdot m^{-3}$), Cd (50.0, 70.0 $ng \cdot m^{-3}$) and Ni (20.0, 30.0 $ng \cdot m^{-3}$) concentrations in Beijing were higher than Chengdu, respectively. The concentrations of all elements except for Al (2,905.0 $ng \cdot m^{-3}$), Ca (3,008.0 $ng \cdot m^{-3}$) and Sr (206.0 $ng \cdot m^{-3}$) in Shanghai were lower than Chengdu. Also, Ti (110.0 $ng \cdot m^{-3}$), Cu (190.0 $ng \cdot m^{-3}$), As (40.0 $ng \cdot m^{-3}$) and Ba (70.0 $ng \cdot m^{-3}$) concentrations in Chengdu were higher than Guangzhou. Moreover, concentrations of elements in Navi Mumbai were lower than Chengdu, except for Ba (270 $ng \cdot m^{-3}$), Cr (44 $g \cdot ng \cdot m^{-3}$), Mo (23 $ng \cdot m^{-3}$), Rb (11 $ng \cdot m^{-3}$) and Ni (6.3 $ng \cdot m^{-3}$). In Jeddah and Navarra, all the elemental concentrations were lower than Chengdu, except for Si (2,100 $ng \cdot m^{-3}$) and Ni (6 $ng \cdot m^{-3}$) in Jeddah, which were higher than those in Chengdu. The analysis results showed that most of the elemental concentrations in $PM_{2.5}$ in Chengdu were higher than the other domestic and foreign cities, such as Beijing, Shanghai, Guangzhou, Jeddah and Navarra. Thus, Chengdu is suffering from a considerably severe $PM_{2.5}$ pollution problem.

Table 1: Comparison of $PM_{2.5}$ ($\mu g \cdot m^{-3}$) and elements ($ng \cdot m^{-3}$) mass concentrations at urban locations in other domestic and foreign cities.

City Year	Chengdu 2009-2010	Beijing 2005-2006	Shanghai 2009-2010	Guangzhou 2008-2009	Jeddah 2011	Navi Mumbai 2008	Navarra 2009
$PM_{2.5}$	165.1	118.5	103.10	81.7	-	42	15.38
Fe	1679	1130	1328	1850	590	310	-
Si	1093.9	1790	-	-	2100	840	-
Al	1053	790	2905	-	800	450	-
Ca	995.2	900	3008	-	540	310	-
Zn	822.9	530	236	1360	41	64	17.98
Pb	320.5	240	59	450	160	36	2.29
Mn	137.6	90	66	150	19	9.2	2.57
Ti	126.3	80	-	110	55	23	6.31
Br	55.5	30	-	-	9	12	-
Cu	54.1	70	15	190	6	6.2	11.98
Sr	44	-	206	-	4	14	1.16
As	40.5	20	-	40	9	7.1	0.16
Ba	38	210	-	70	34	270	12.08
Sn	18.8	-	-	-	-	-	0.94
Cr	17.9	50	9	70	2	44	2.39
Mo	10.7	-	-	-	-	23	-
Sb	11	-	-	-	-	-	0.7
Cd	9.5	50	1	20	8	-	0.05
Rb	9.9	-	-	-	1	11	0.25
Ni	5.1	20	9	-	6	6.3	1.31

The seasonal variation of $PM_{2.5}$ concentrations is presented in Fig. 1. Clearly, the concentrations in autumn and winter were higher than those in spring and summer. Seasonal mean concentrations were 133.2±54.5 $\mu g \cdot m^{-3}$ in spring, 113.5±38.7 $\mu g \cdot m^{-3}$ in summer, 188.0±105.2 $\mu g \cdot m^{-3}$ in autumn and 225.5±71.9 $\mu g \cdot m^{-3}$ in winter, and the ranges were 60.0~300.5 $\mu g \cdot m^{-3}$, 49.2~218.9 $\mu g \cdot m^{-3}$, 57.2~425.0 $\mu g \cdot m^{-3}$ and 93.4~376.8 $\mu g \cdot m^{-3}$, respectively. Generally, $PM_{2.5}$ levels are governed by emissions, transportation, chemical transformation and depositional processes, which are all related to meteorological conditions. The effects of meteorological factors (precipitation, wind speed, temperature and humidity) on $PM_{2.5}$ concentrations showed negative correlation. Studies have shown that precipitation can wash away part of atmospheric particulate matter and inhibit the generation of ground dust. Accordingly, ambient air pollutants are dispersed and diluted by frequent rainfall. Wind speed has dual effects on the concentrations of pollutants. Within a certain range, wind speed is favourable to the diffusion and dilution of air pollutants. However, if it is larger than the given range, increased wind speed will lead to a significant increase in $PM_{2.5}$ concentrations in air (Li et al. 2014). In addition, the diffusion of air pollutants in the vertical direction mainly depends on temperature. The atmosphere is unstable when the temperature is higher and the pollutants diffuse upwards under the action of thermal convection, resulting in lower concentrations of pollutants. Conversely, at low temperatures, the atmosphere is stable. Thus, the diffusion of pollutants is restrained, and the concentrations are higher. The precipitation, temperature and humidity values in spring and summer were higher than those in autumn and winter in Chengdu. Therefore, the influence of meteorological factors on $PM_{2.5}$ concentrations was significant. On the other hand, the reason was the local pollution sources.

The total mass concentrations of the elements were 9,911.6 $ng \cdot m^{-3}$ in spring, 3,326.8 $ng \cdot m^{-3}$ in summer, 5,611.3 $ng \cdot m^{-3}$ in autumn and 7,575.0 $ng \cdot m^{-3}$ in winter, accounting for 7.44%, 2.93%, 2.98% and 7.58% in $PM_{2.5}$, respectively. Seasonal variation of elemental concentrations is shown in Fig. 2. It can be clearly seen that the elemental concentrations in spring and winter were higher than those in summer and autumn. The concentrations of Fe (2,385.6, 2233.8 $ng \cdot m^{-3}$), Si (2,281.9, 1011.2 $ng \cdot m^{-3}$) and Ca (1,717.4, 1092.1 $ng \cdot m^{-3}$) in spring and winter, respectively, Fe (1,365.0 $ng \cdot m^{-3}$) in autumn and Al (2,409.1 $ng \cdot m^{-3}$) in spring exceeded 1,000.0 $ng \cdot m^{-3}$, which are high pollution levels. The total concentration of earth's crust elements was 8,987.3 $ng \cdot m^{-3}$ in spring, 2,019.8 $ng \cdot m^{-3}$ in summer, 3,752.5 $ng \cdot m^{-3}$ in autumn and 5,301.7 $ng \cdot m^{-3}$ in winter, accounting for 90.7%, 60.7%, 66.9% and 70.0% of the 20 elements, respectively. Clearly, the effects on the earth's crust elements concentrations in spring were larger compared to the other seasons. The reason for this may be remote transmission. The northern dust storm affected by wind can transfer to Chengdu. The soil dust was transported to Chengdu by northerly air-

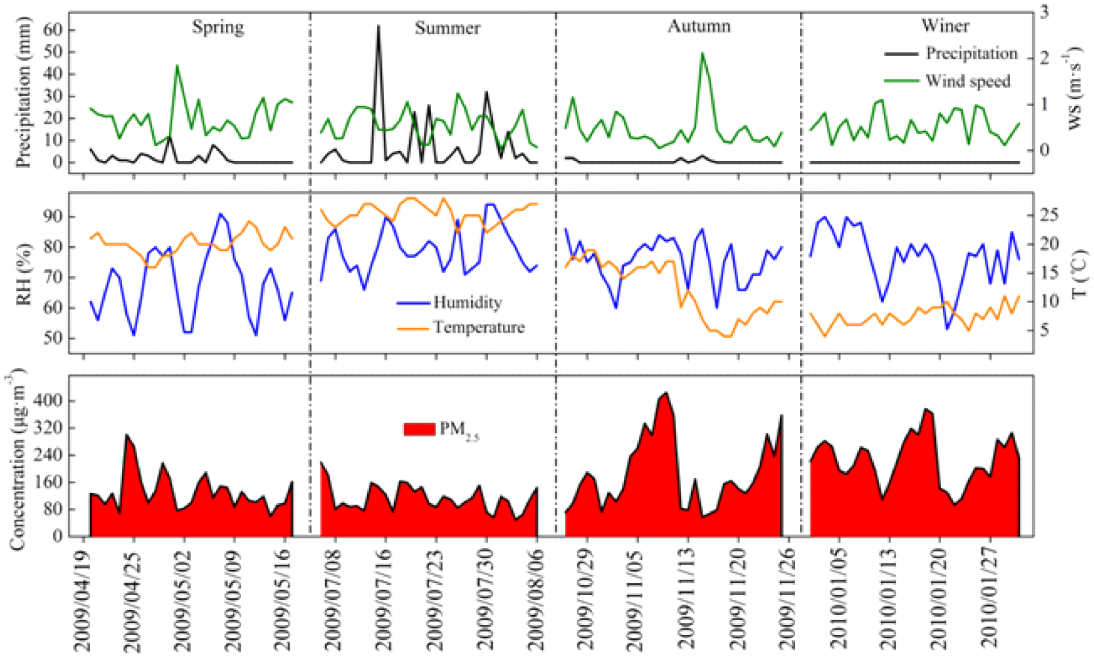

Fig. 1: The seasonal variation of $PM_{2.5}$ concentrations and meteorological factors in Chengdu.

flow, and $PM_{2.5}$ concentrations reached 300.5, 266.0 and 161.1 μg·m^{-3} on April 24, 25 and 26, respectively. Moreover, the concentrations of Al, Si, Ca and Fe on April 24 reached 17,690.1, 18,904.7, 11,474.4 and 14,114.1 ng·m^{-3}, respectively. Therefore, the concentrations of earth's crust elements in spring were higher than those in summer, autumn and winter in this study. In addition, the concentrations of Zn, Pb and Mn (except Zn in winter (1,193.3 ng·m^{-3})) fluctuated between 100.0 to 1,000.0 ng·m^{-3} during the four seasons (Fig. 2). Also, the concentrations of Cu, As, Cr, Sb, Cd and Ni were lower than 100.0 ng·m^{-3}, but As concentrations were 0.30, 4.65, 7.67 and 10.47 times that of the standard in spring, summer, autumn and winter, respectively. Cd concentrations were 0.28, 1.50, 1.58 and 0.18 times the standard, respectively. The reason for this elemental distribution is that waste gas of fired coal, automobile exhaust, rubber abrasion and smelting industry contributed to the atmospheric environment in Chengdu.

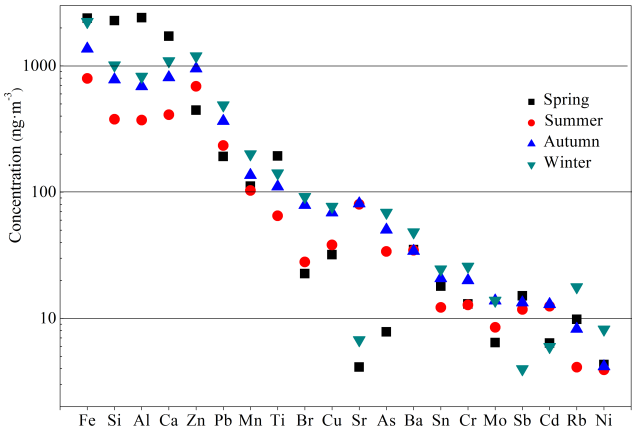

Fig. 2: The seasonal variation of elemental concentrations in Chengdu.

Possible sources of $PM_{2.5}$: The EF was calculated for each detected element to evaluate the anthropogenic contribution to atmospheric elemental levels. The EFs were calculated using the following equation:

$$EF_i = \frac{(C_i / C_j)_{PM\,2.5}}{(C_i / C_j)_{crust}} \quad ...(1)$$

Where, C_i is the concentration of the element considered in $PM_{2.5}$ or crust and C_j is the concentration of the reference element in $PM_{2.5}$ or crust. Al, Si, Ti and Fe are commonly considered as the reference elements of crustal material. In this study, Al was used as a reference element because it is relatively stable and not affected by contamination. The concentrations of elements in the crust indicate their content in the topsoil or soil in China. EF value lower than 10 is taken as an indication that the element in $PM_{2.5}$ has a significant crustal source, while EF value greater than 10 indicates a significant anthropogenic origin.

The EFs of the elements in $PM_{2.5}$ estimated from this study are shown in Fig. 3. Cd exhibited the highest enrichment factor (>1,000) with EF value of 6,481. Pb, Zn, Br, Sb,

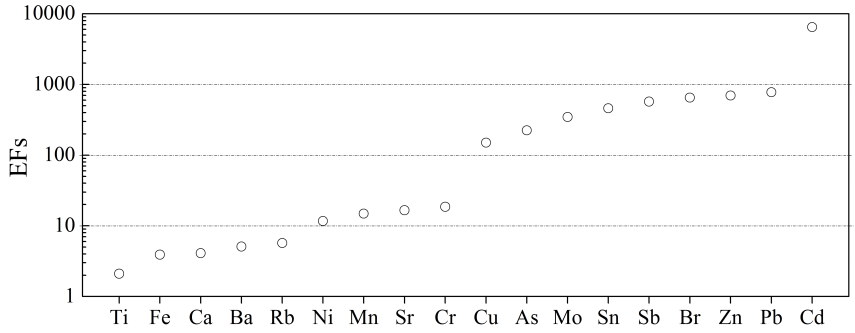

Fig. 3: Enrichment factors of elements in $PM_{2.5}$.

Sn, Mo, As and Cu also showed high enrichment factors (>100). Cr, Sr, Mn and Ni also appeared to be moderately enriched but to a lesser extent (10>EF>100). The high enrichment of these elements suggest that the dominant sources for these elements were non-crustal and a variety of pollution emissions contributed to their loading in the ambient air. Low EF values (in general<10) were found for Ti, Fe, Ca, Ba and Rb, suggesting a negligible contribution from anthropogenic emissions to the ambient levels of these elements. On the other hand, the EFs of carcinogenic heavy metals such as Cd, Pb, As, Cr and Ni were 6,481.2, 776.2, 224.5, 18.6 and 11.7, respectively, indicating that these heavy metals were of anthropogenic origin. For example, Mn is a metal element present in high concentrations in fly ash from coal combustion. In addition, high-temperature processes such as metal smelting and fuel combustion are usually the source of non-crustal volatile metals (e.g. Cd, Ni, Cu) in the atmosphere. Thus, anthropogenic sources were responsible for the main contributions to $PM_{2.5}$ in Chengdu, and natural sources such as soil dust had lesser contribution.

In order to further understand the relationship between elements in $PM_{2.5}$ and its sources, the correlations were analyzed. The results showed that high correlations were observed for Fe, Si, Al, Ca and Ti. For these five elements, the correlation coefficient was greater than 0.9 ($P<0.01$), indicating that these elements in $PM_{2.5}$ were more strongly affected by crustal sources. In addition, Zn was strongly correlated with Pb, Br and Cu, with $R^2=0.891$, 0.854 and 0.824, respectively, and $P<0.01$. Zn is associated with tire wear and tailpipe emissions as a result of its use in motor oil. Pb and Br mainly come from vehicle emissions. Cu is emitted from automobile brake system (Liu et al. 2015). Therefore, motor vehicle exhaust emissions were the main contributors to Zn, Pb, Br and Cu contents in $PM_{2.5}$. As element showed moderate correlations with Zn, Pb, Mn, Cu and Cr, with $R^2=0.860$, 0.890, 0.646, 0.772 and 0.796, respectively, and $P<0.01$. Therefore, these elements in $PM_{2.5}$ had a common source. As is an indicator element of coal (López et al. 2011). Zn can also come from combustion of fossil fuels and waste incineration, indicating that the elements were influenced by combustion of fossil fuels, coal and waste. However, there was a weak correlation for Fe with Mn, $R^2=0.706$, $P<0.01$. Fe not only comes from crustal sources, but also comes from metallurgical processes, suggesting that industrial emissions partly contributed to Fe content in $PM_{2.5}$. Thus, the main sources of $PM_{2.5}$ were soil dust, automobile exhaust, combustion of fossil fuels, coal and waste, and industrial emissions.

In this study, the various sources of elements in $PM_{2.5}$ were identified by PCA (Cong et al. 2007), and the results are presented in Table 2. The four principal components with eigen values greater than 1 were extracted and they accounted for 74.7% of the total variance. Factor 1 accounting for 34.3% of total variance was dominated by Zn, Pb, Mn, Br, Cu, As, Ba, Sn, Cr, Mo, Rb and Ni. The elements Br, Cu and Pb mainly came from vehicle emissions and tire wear. The combustion of coal, fossil fuels and waste mainly contributed to Zn, Cr, Ni and As contents in $PM_{2.5}$. The results showed that vehicle emissions, coal combustion, burning of fossil fuels and waste incineration were the primary pollution sources. Factor 2 was characterized by Fe, Si, Al, Ca and Ti, reflecting the effect of crustal sources, and accounted for 27.8%. In addition, Ca is a characteristic element of building constructions. Thus, the secondary pollution sources were identified as soil dust and building dust. Factor 3 was represented by Cd and Sr, accounting for 7.0%. Cd content can be affected by metallurgy and mechanical manufacturing emissions, suggesting that the third pollution source may be mainly industrial emission. Factor 4 was the component of Sb, accounting for 5.6%. Sb is commonly found in brake dust. Therefore, the fourth pollution source is road traffic. The PCA results showed that soil and building dust, automobile exhaust, combustion of coal, fossil fuels and waste, industrial emission, and road traffic were contributors to $PM_{2.5}$ in Chengdu.

Table 2: Rotated component matrix for $PM_{2.5}$ elements.

Elements	Principal components			
	F1	F2	F3	F4
Fe		0.607		
Si		0.809		
Al		0.851		
Ca		0.780		
Zn	0.795			
Pb	0.804			
Mn	0.900			
Ti		0.694		
Br	0.733			
Cu	0.750			
Sr			0.610	
As	0.730			
Ba	0.601			
Sn	0.323			
Cr	0.839			
Mo	0.395			
Sb				0.894
Cd			0.749	
Rb	0.806			
Ni	0.580			
% of variance	34.332	27.793	7.017	5.624
Cumulative %	34.332	62.124	69.141	74.765

CONCLUSIONS

In this paper, we have investigated the elemental composition of $PM_{2.5}$ and its possible sources in Chengdu from April 2009 to January 2010. The total mass concentration of the elements in $PM_{2.5}$ was 6,543.3 ng·m^{-3}, accounting for 4.0% of $PM_{2.5}$ mass concentration. The concentrations of the elements followed the order of Fe>Si> Al>Ca> Zn>Pb> Mn>Ti> Br>Cu>Sr>As>Ba>Sn>Cr>Mo>Sb>Cd>Rb>Ni. The concentrations in autumn and winter were higher than those in spring and summer. The elemental concentrations in spring and winter were higher than those in summer and autumn. The elemental concentrations in the four seasons were 9,911.6, 7,575.0, 3,326.8 and 5,611.3 ng·m^{-3}, accounting for 7.44%, 7.58%, 2.93%, and 2.98% of $PM_{2.5}$, respectively. The results of EF, CA and PCA consistently indicated that the sources of $PM_{2.5}$ in Chengdu were mainly crustal material, construction dust, automotive exhaust emission, coalfire, dust of fossil fuel and refuse burning, and industrial emissions. These sources contributed to the pollution due to the rapid increase in motor vehicles, total energy consumption, smelting industry and dust storm events.

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