

Chemical characterization and sources of PM_{2.5} at 12-h resolution in Guiyang, China

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Received: 17 July 2017 / Revised: 12 October 2017 / Accepted: 30 October 2017 / Published online: 7 November 2017
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Abstract The increasing emission of primary and gaseous precursors of secondarily formed atmospheric particulate matter due to continuing industrial development and urbanization are leading to an increased public awareness of environmental issues and human health risks in China. As part of a pilot study, 12-h integrated fine fraction particulate matter (PM_{2.5}) filter samples were collected to chemically characterize and investigate the sources of ambient particulate matter in Guiyang City, Guizhou Province, southwestern China. Results showed that the 12-h integrated PM_{2.5} concentrations exhibited a daytime average of $51 \pm 22 \mu\text{g m}^{-3}$ (mean $\pm$ standard deviation) with a range of 17–128 $\mu\text{g m}^{-3}$ and a nighttime average of $55 \pm 32 \mu\text{g m}^{-3}$ with a range of 4–186 $\mu\text{g m}^{-3}$. The 24-h integrated PM_{2.5} concentrations varied from 15 to 157 $\mu\text{g m}^{-3}$, with a mean value of $53 \pm 25 \mu\text{g m}^{-3}$, which exceeded the 24-h PM_{2.5} standard of 35 $\mu\text{g m}^{-3}$ set by USEPA, but was below the standard of 75 $\mu\text{g m}^{-3}$, set by China Ministry of Environmental Protection. Energy-dispersive X-ray fluorescence spectrometry (XRF) was applied to determine PM_{2.5} chemical element concentrations. The order of concentrations of heavy metals in PM_{2.5} were iron (Fe) > zinc (Zn) > manganese (Mn) > lead

(Pb) > arsenic (As) > chromium (Cr). The total concentration of 18 chemical elements was $13 \pm 2 \mu\text{g m}^{-3}$, accounting for 25% in PM_{2.5}, which is comparable to other major cities in China, but much higher than cities outside of China.

Keywords Trace elements · PM_{2.5} · Source apportionment

1 Introduction

Associations between atmospheric fine fraction (aerodynamic diameter $\leq 2.5 \mu\text{g}$) particulate matter (PM_{2.5}) and adverse human health outcomes have been extensively reported (Pope et al. 2004; U.S EPA 2009; Stanek et al. 2011; Solomon et al. 2012; HEI 2013). Due to its ability to penetrate deep into the lungs and its solubility, components of PM_{2.5} are considered to be respirable and bio-available (Raes et al. 2000; Kelly and Fussell 2012). Moreover, PM_{2.5} can contain toxic heavy metals such as chromium (Cr), cadmium (Cd), titanium (Ti), manganese (Mn), nickel (Ni), lead (Pb), arsenic (As), and zinc (Zn) (Kim et al. 2004; Figueroa et al. 2006). Those PM_{2.5} associated toxic elements have been demonstrated to detrimentally affect cardiovascular and respiratory health risks, as well as pose a lung cancer risk (Gao et al. 2015a, b; Costa and Dreher, 1997; Kodavanti et al. 1998; Adamson et al. 2000; Lewtas, 2007; Wallenborn et al. 2009). PM_{2.5} can also remain in the atmosphere for a considerable period time and transport over regional and global scales, hence it plays an important role in the dispersion of its combined toxic elements in the environment.

Since PM_{2.5} in ambient air is a broadly recognized environmental and public health issue, it has received considerable attention worldwide and is currently regulated

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in most industrialized countries (WHO 2005). Numerous studies on $\text{PM}_{2.5}$ and its toxic element constituents were conducted in many countries (Toledo et al. 2008; Crilley et al. 2014; Morishita et al. 2011; Cristaldi et al. 2013). China is the world's largest emitter of atmospheric $\text{PM}_{2.5}$, and the government has been increasingly taking actions to reduce emissions (Li et al. 2016). Investigations into ambient $\text{PM}_{2.5}$ air pollution have focused primarily on developed capital cities, such as Beijing, Xi'an, Shanghai, and Guangzhou. Those studies mainly focused on $\text{PM}_{2.5}$ mass and major components, temporal and spatial distribution characteristics, and identification of its sources (Dan et al. 2004; Huang et al. 2015; Chan et al. 2016; Lai et al. 2016). For example, Huang et al. (2014) revealed that severe haze events were largely driven by secondary aerosol formation. Gao et al. (2015a, b) reported the risk levels for the carcinogenic heavy metals such as $\text{Cr} > \text{Cd} > \text{Ni}$, suggesting that Cr may pose a carcinogenic risk in Beijing daily $\text{PM}_{2.5}$. Ye et al. (2003) reported weekly $\text{PM}_{2.5}$ mass concentrations and components with a $\text{PM}_{2.5}$ mass ranging from 21 to $147 \mu\text{g m}^{-3}$. Results indicated that coal combustion was the largest contributor to $\text{PM}_{2.5}$ in ambient air in Shanghai. Wang et al. (2006) reported an average concentration of $98 \mu\text{g m}^{-3}$ of $\text{PM}_{2.5}$ in Guangzhou, observing that the major components were organic matter and sulfate. Shen et al. (2014) investigated day-night diurnal variations of water soluble ions in PM_{10} in Xi'an and found low $\text{NO}_3^-/\text{SO}_4^{2-}$ ratios, suggesting that coal combustion was the major contribution.

With ambient levels of $\text{PM}_{2.5}$ continuing to increase in China, the identification of responsible sources has become an area of active and necessary research. In Xi'an, source apportionment of $\text{PM}_{2.5}$ using the positive matrix factorization (PMF) receptor model showed that motor vehicular emissions, coal combustion, secondary inorganic aerosol, and fugitive dust were the major sources, accounting for 80% of total $\text{PM}_{2.5}$ mass (Xu HM et al. 2016). The authors also indicated that the contribution of coal combustion decreased from $31\% \pm 5\%$ in 2006 to $24\% \pm 3\%$ in 2010, owing to the coal-fired power plants' flue gas controlling implement, while the mobile source contribution remained essentially the same from 2006 ($19\% \pm 5\%$) to 2010 ($21\% \pm 5\%$). In Beijing, eight sources of $\text{PM}_{2.5}$ were identified, which consisted of biomass burning (11%), secondary sulfates (17%), secondary nitrates (14%), coal combustion (19%), general industrial (6%), motor vehicles (6%), road dust (9%), and yellow dust (Song et al., 2006). Yang et al. (2015) investigated the chemical composition of $\text{PM}_{2.5}$ in Beijing and reported that the total mass concentrations of metals in $\text{PM}_{2.5}$ ranged from 0.4 to $13.2 \mu\text{g m}^{-3}$. Gu et al. (2011) investigated chemical composition of $\text{PM}_{2.5}$ during the winter in Tianjin and found that SO_4^{2-} , NO_3^- , Cl^- , and NH_4^+ were four major

ions, accounting for 17, 12, 5, and 6% of total $\text{PM}_{2.5}$ mass, respectively.

Currently, few studies of ambient $\text{PM}_{2.5}$ have been conducted in Guiyang, a quickly developing inland capital city of Guizhou Province, in southwestern China. The primary goals of this pilot study were to (1) characterize the temporal distribution pattern of potentially toxic PM elemental components, (2) investigate elemental composition and their enrichment factors, and (3) identify potential emission sources and affecting meteorological factors of ambient $\text{PM}_{2.5}$ in Guiyang.

2 Experimental methods

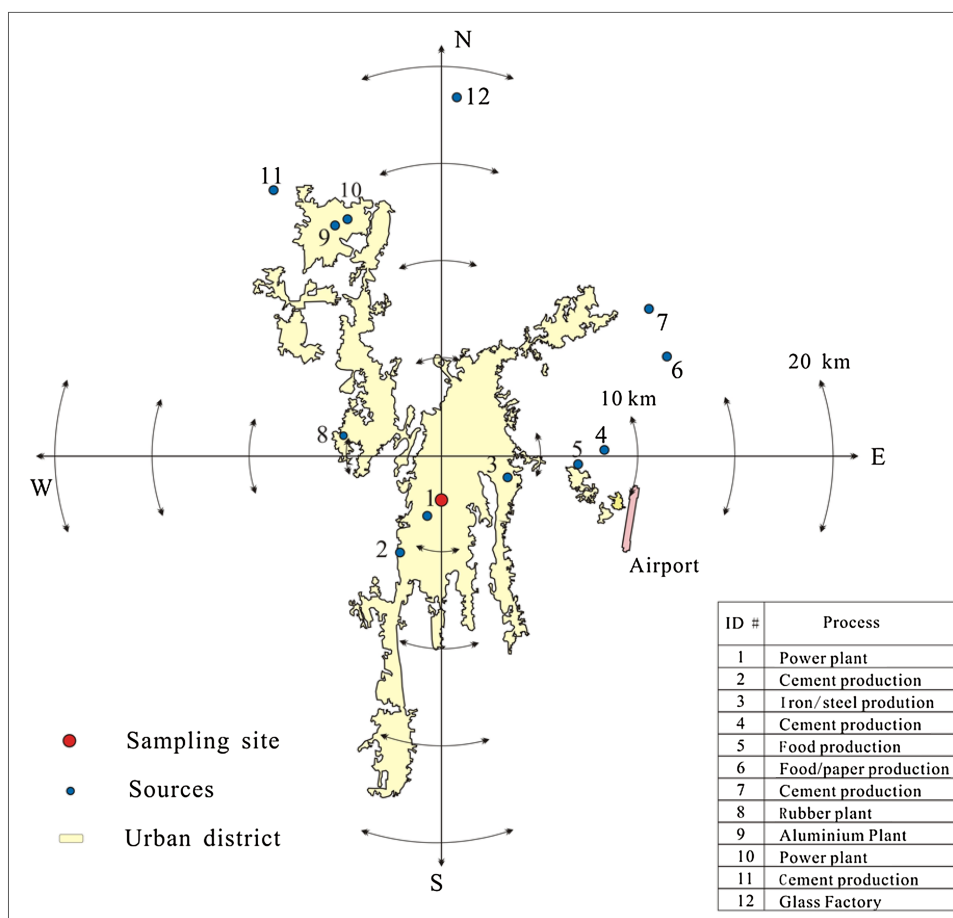
2.1 Study domain

Guiyang, the capital city of Guizhou Province, is a large and rapidly growing inland city in southwestern China (Fig. 1) with an estimated population in 2014 of approximately 4.3 million within an area of 8030 km^2 . The climate in Guiyang is characterized as subtropical humid monsoon. Annual predominant winds are from the northeast and south in summer/spring and are more northeasterly in autumn and winter (Xu XH et al. 2016). Major industries in Guiyang include petrochemical, metallurgy, energy extraction, electricity generation, and medicine. The sampling site is located at the Guiyang Environmental Monitoring Station (EMS), which is a residential area and approximately 100 m away from a roadway ($106^\circ 43' 03'' \text{E}$; $26^\circ 33' 56'' \text{N}$). It is also located approximately 7.0 km from a coal-fired power plant. In addition, a cement production plant is located to the southwest of the sampling site, and an iron/steel plant and another cement production plant are located in the east.

2.2 Methods and materials

A total of 180 twelve hour (12-hr) integrated $\text{PM}_{2.5}$ filter samples were collected daily during the fall season of September, October and November using a URG Corporation (Chapel Hill, NC, USA) Model 2000-01 J Weekly Air Particulate Sampler (WAPS). The WAPS sampler was located on the roof of an approximately 30 m high building. The WAPS sampled at a 16.7 L min^{-1} flow rate onto 47 mm Teflon filters (Millipore, PGTRISLIDETM). During the sampling campaign, the 12-h integrated samples were collected from 08:00–20:00 (daytime) and 20:00–08:00 (nighttime). After collection, samples were stored in a freezer (-20°C) waiting for shipment. When the sampling campaign was finished, all filters were shipped at ambient temperature to U.S. EPA Office of Research and

Fig. 1 Sampling location in Guiyang located in southwestern China and surrounding major emission sources



Development, National Exposure Research Laboratory for weighing and chemical analysis.

To determine the mass of $PM_{2.5}$ that was collected, filters were equilibrated for a minimum of 24-h in a temperature-controlled room at 25 °C with 40% relative humidity. Filters were weighed before and after sample collection using a Mettler Toledo (Columbus, OH, USA) Model UMX2 microbalance. Weights were only deemed valid if duplicates were within 3 μg of each other on another day. Sampled filters were individually stored in petri dishes and sealed in polythene bags for preservation. Once $PM_{2.5}$ mass was determined, sample filters were analyzed by energy dispersive X-ray fluorescence spectrometry (XRF) for total elemental concentrations (Oakes et al., 2016). For the filters measured by XRF, the limit of determination (LOD; deposition area = 6.61 cm^2 , flow rate = 10 L min^{-1}) varied by element from 2.0 ng m^{-3} for Zn to 230 ng m^{-3} for Na.

The meteorological parameters were measured from a 10 m meteorological tower operated at the monitoring site and provided by the Guiyang Meteorological Administration, using an ambient air quality automatic measurement system (model Ambient gas MS, USA). The 12-h

integrated wind speed, wind direction, ambient temperature, and relative humidity were calculated for each filter sampling period. Precipitation was also obtained at the sampling site. All study data analyses were performed using SPSS 11.0 (SPSS Inc., USA) and Microsoft Excel 2007 (Microsoft Corp., USA).

2.3 Quality assurance

The data for the particulate matter samples were acquired using a Kevex energy-dispersive XRF spectrometer, as described by Landis et al. (2001). To monitor the accuracy of the spectrometer, NIST certified reference standard, Standard Reference Material SRM1833-1111 and SRM 1832-249, were analyzed at the beginning and end and met the accepted criterion for accuracy. Average sample concentrations were generally higher than or comparable to average lot and field blank concentrations (within the standard deviation) for all elements measured. The limit of detection (LOD) was calculated as three times the standard deviation of a set of field blanks.

3 Results and discussion

3.1 Characteristics of PM_{2.5}

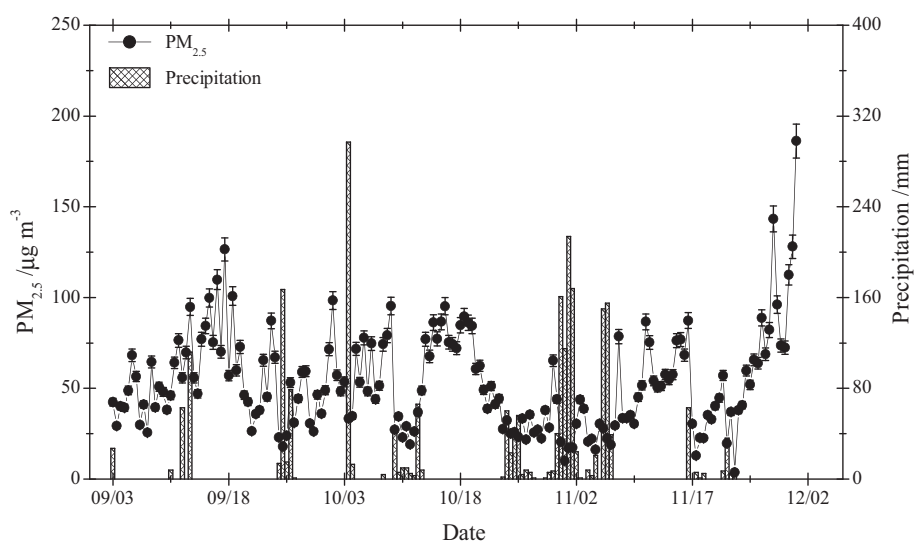
The 12-h integrated PM_{2.5} concentrations showed a daytime average of $51 \pm 22 \mu\text{g m}^{-3}$ (mean $\pm$ standard deviation) ranging from 17 to $128 \mu\text{g m}^{-3}$ ($n = 89$), and a nighttime average of $55 \pm 32 \mu\text{g m}^{-3}$ ranging from 3.7 to $186 \mu\text{g m}^{-3}$ ($n = 89$), respectively. Nighttime PM_{2.5} concentrations exhibited a wider range in concentration compared to daytime PM_{2.5} concentrations. Generally, low PM_{2.5} concentrations were observed during episodes of precipitation, highlighting the importance of rain washout. The lowest mean daily value of PM_{2.5} concentration of $15 \mu\text{g m}^{-3}$ was recorded on October 31 and was the result of washout from a significant wet deposition event (Fig. 2). Large variations of PM_{2.5} observed in Guiyang were a function of differences in meteorological dispersion and the intensity of anthropogenic emissions.

In Guiyang, the daily 24-h (defined as 08:00–08:00) integrated PM_{2.5} levels varied from 15 to $157 \mu\text{g m}^{-3}$, with a mean value of $53 \pm 25 \mu\text{g m}^{-3}$, which was comparable with our recent average value of $68 \pm 30 \mu\text{g m}^{-3}$ observed in the fall of 2014 (unpublished data). The daily average concentrations of PM_{2.5} in Guiyang were lower than results obtained in other domestic developed capital cities, such as Beijing, Shanghai, Guangzhou, and Xi'an, which exhibited as much as 119 ± 14 , 94 ± 52 , 108 ± 94 , and $141 \pm 109 \mu\text{g m}^{-3}$, respectively (Yan et al. 2016; Zhao et al. 2015; Wang JZ et al. 2016; Wang et al. 2015). Compared to cities abroad, however, the PM_{2.5} concentrations in Guiyang were higher than those observed in Steubenville, Ohio ($26 \pm 11 \mu\text{g m}^{-3}$), USA, but lower than concentrations measured in Korogocho ($166 \mu\text{g m}^{-3}$), Kenya, and Agra ($71 \mu\text{g m}^{-3}$), India (Egondi et al. 2016).

Negative correlations among PM_{2.5} and wind speed ($r = -0.31$, $p < 0.0001$) and relative humidity ($r = -0.46$, $p < 0.0001$) were observed (Fig. 3) during this study, and this highlights the influence of meteorological dispersion on local anthropogenic emissions. Precipitation accompanied by high wind can remove and spread a significant portion of PM_{2.5} from the air due to below cloud scavenging (Charron and Harrison 2005; White et al. 2013), and the efficiency of removal is a function of aerosol size, morphology, and hydrophobicity (Chate et al. 2003; Jung et al. 2003). Hence, meteorological conditions will result in changes to the composition of PM_{2.5} in the air. Guiyang is a city characterized by high annual average rainfall between 110 and 130 cm, with up to 178 days rainy days per year (Xu et al. 2008). The PM_{2.5} concentrations in Guiyang City were lower than other domestic cities (Shen ZX et al. 2016; Wang YL et al. 2016) and might be attributed to the frequent rainfall events.

WHO (2005) has set three interim target values for PM_{2.5} of 24-hr, including interim target-1 (IT-1), interim target-2 (IT-2), and interim target-3 (IT-3). The highest limit value is $75 \mu\text{g m}^{-3}$ for IT-1. However, air quality guidelines (AQG) for human health protection recommended a 24-h PM_{2.5} limit of $25 \mu\text{g m}^{-3}$. The current U.S. EPA National Ambient Air Quality Standard (NAAQS) for 24-h integrated PM_{2.5} is $35 \mu\text{g m}^{-3}$ (USEPA, 2012). Currently, China MEP has proposed a national target for 24-h average PM_{2.5}, which is $75 \mu\text{g m}^{-3}$. Although the overall study average 24-h PM_{2.5} concentration of $53 \pm 25 \mu\text{g m}^{-3}$ observed in Guiyang during the study period met the WHO's IT-1 standard, the 24-h concentration limit was exceeded 20 times. In addition, the observed concentrations were much higher than the values set by the AQG and U.S. EPA NAAQS.

Fig. 2 The distribution pattern of 24-h integrated PM_{2.5} concentrations incorporated with precipitation depth during the sampling campaign



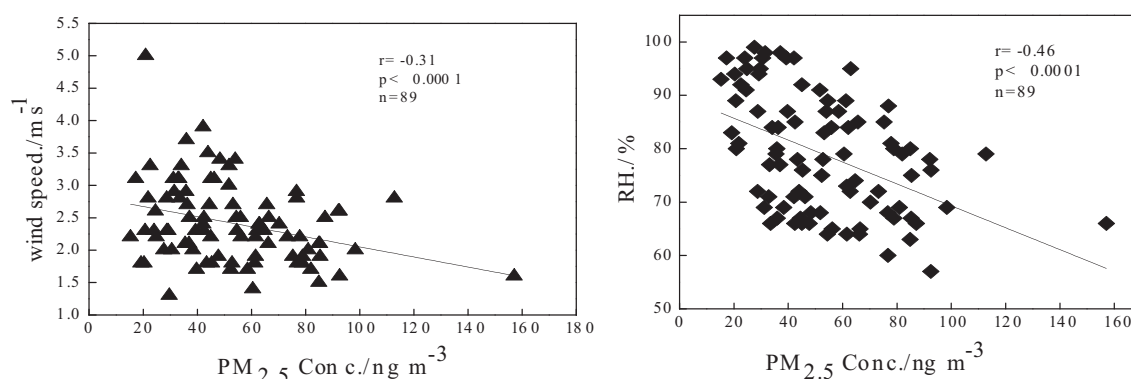


Fig. 3 Ambient $\text{PM}_{2.5}$ concentrations relative to wind speed and relative humidity

3.2 $\text{PM}_{2.5}$ chemical composition

The measured elemental mass (the total mass of specific oxides, Table 1) of 18 elements constituted an average of 25%, varying from 22% to 29% of the total $\text{PM}_{2.5}$. As expected, S, and K showed high average concentration of $6.7 \mu\text{g m}^{-3}$ with a range of $0.61\text{--}16 \mu\text{g m}^{-3}$, and an average of $1.7 \mu\text{g m}^{-3}$ and a range of $0.14\text{--}8.0 \mu\text{g m}^{-3}$, respectively. In general, K in particles mainly originated from biomass combustion (Yamasoe et al. 2000) and other anthropogenic sources like iron/steel slag and sintering operations (Pancras et al. 2013). During the study period, local agricultural burning of crop residues, such as rice and

corn straws, was observed and resulted in relatively high concentration of K in particles. Similarly, major elements of Si, Fe, Ca and Al, which are considered as crustal elements (Zíková et al. 2016), also exhibit high concentrations with averages of 2.2, 0.63 and $0.34 \mu\text{g m}^{-3}$, and range from $0.16\text{--}10$, $0.016\text{--}3.8$ and $0.014\text{--}1.2 \mu\text{g m}^{-3}$, respectively. The high concentrations combined with large ranges of Si and Ca may indicate significant contributions of construction dusts and resuspended mineral particles to $\text{PM}_{2.5}$ in Guiyang City.

The daily average concentration trend of most elements was consistent with the corresponding concentration of $\text{PM}_{2.5}$ mass, suggesting meteorology as one driving factor

Table 1 Monthly average elemental concentrations ($\mu\text{g m}^{-3}$, total mass of specific oxides) in descending order of concentration

Parameter	September	October	November	Whole Sampling Period	
	(n = 28)	(n = 31)	(n = 30)	Mean	Range
S	7.4 ± 3.7	6.7 ± 3.5	5.9 ± 3.5	6.7 ± 3.6	0.61–16
Si	3.1 ± 1.8	2.3 ± 1.8	1.3 ± 0.87	2.2 ± 1.7	0.16–10
K	1.9 ± 1.5	1.6 ± 1.3	1.7 ± 1.3	1.7 ± 1.4	0.14–8
Ca	0.75 ± 0.71	0.47 ± 0.28	0.69 ± 0.67	0.63 ± 0.59	0.016–3.8
Fe	0.36 ± 0.28	0.29 ± 0.17	0.39 ± 0.36	0.34 ± 0.28	0.011–1.9
Al	0.38 ± 0.25	0.33 ± 0.23	0.33 ± 0.28	0.34 ± 0.27	0.0014–1.2
Na	0.33 ± 0.25	0.26 ± 0.15	0.3 ± 0.17	0.3 ± 0.21	0.0058–1.2
Zn	0.4 ± 0.39	0.19 ± 0.19	0.29 ± 0.36	0.29 ± 0.33	0.0067–2.2
Mn	0.32 ± 0.31	0.31 ± 0.59	0.13 ± 0.15	0.25 ± 0.4	0.0023–4
Mg	0.28 ± 0.17	0.23 ± 0.12	0.24 ± 0.17	0.25 ± 0.16	0.012–0.86
P	0.22 ± 0.18	0.22 ± 0.19	0.12 ± 0.11	0.19 ± 0.17	0.000381.1
Pb	0.17 ± 0.17	0.11 ± 0.11	0.12 ± 0.08	0.13 ± 0.12	0.0016–0.92
Ti	0.034 ± 0.026	0.025 ± 0.015	0.031 ± 0.022	0.03 ± 0.02	0.00039–0.16
As	0.021 ± 0.019	0.019 ± 0.016	0.021 ± 0.019	0.02 ± 0.018	0.00087–0.11
Rb	0.014 ± 0.011	0.01 ± 0.008	0.011 ± 0.009	0.012 ± 0.009	0.000031–0.059
Br	0.011 ± 0.009	0.0086 ± 0.004	0.014 ± 0.01	0.011 ± 0.008	0.000072–0.052
Se	0.0065 ± 0.003	0.0053 ± 0.004	0.0079 ± 0.006	0.0066 ± 0.005	0.000054–0.032
Cr	0.0034 ± 0.002	0.0035 ± 0.003	0.0033 ± 0.003	0.0034 ± 0.003	0.000012–0.021

Reported elemental concentrations are those with average values above the limit of detection (LOD) and blank filter concentrations in 12-h integrated samples

influencing overall concentrations of PM and composition. In general, elements measured in the PM_{2.5} samples of Guiyang displayed clear daily variation patterns. It is assumed that local anthropogenic sources and seasonal meteorology led to the temporal variability as shown in Fig. 4. Stable concentrations of Br, which can be associated with emissions from motor vehicle exhaust (Chan et al. 1997), were observed, reflecting a steady contribution of vehicle exhaust emission. All elemental concentrations were relatively high prior to September 20, and the lower concentrations were observed beginning in November. Analysis of meteorological data revealed high rainfall rates in early November with the total precipitation depth reaching 71 cm, which may be responsible for the decreasing concentrations of elements in PM_{2.5}.

The comparisons of PM_{2.5} elemental concentrations during daytime and nighttime sampling periods are presented in Fig. 5a. The daytime average concentration of all elements was $0.75 \pm 0.4 \mu\text{g m}^{-3}$ and the average nighttime concentration was $0.92 \pm 0.6 \mu\text{g m}^{-3}$. Concentrations of elements measured at nighttime typically exhibited higher concentrations than during the daytime. For example, the average concentration of Si was $1.7 \pm 1.0 \mu\text{g m}^{-3}$ in daytime and $2.7 \pm 2 \mu\text{g m}^{-3}$ in nighttime. The higher concentration of elements observed at night during this study was consistent with reported results in Jinshan, Shanghai, China (Zhu et al. 2015) and suggests that nocturnal radiative cooling inversions, which often result in lower nighttime boundary layer heights, decreased atmospheric dispersion and effectively increased concentrations of local PM_{2.5} emissions near the surface (Querol et al. 2001; Marcazzan et al. 2001).

Elements exhibited various monthly temporal variation patterns. Elemental concentration trends of Na, Mg, K, Ca, Ti, Zn, Fe, and Pb exhibited a pattern of September > November > October (Fig. 5b). While other elements (e.g., Al, Si, P, S, Mn) showed a different pattern with the highest observed concentrations in September and lowest concentrations in November. These observations may be indicative of seasonal changes in anthropogenic activities (e.g., local residential coal combustion for heating in winter) and/or meteorological conditions (e.g., precipitation and wind speed (Feng et al. 2004)).

Spearman correlation coefficients among elements and PM_{2.5} mass concentrations in 12-h integrated samples are presented in Table 2. Heavy metals such as Ti, Mn, Fe, Zn, Pb were positively related to PM_{2.5} mass concentrations and to each other, signifying that those elements may have been emitted from the same source type(s). Concentrations of S, likely as sulfate (Pandis and Seinfeld. 1992), also exhibited a strong positive correlation to PM_{2.5}, with a coefficient of determination of 0.905. Sulfate, typically secondary sulfate, is often a relatively large fraction of

PM_{2.5} mass concentrations in areas impacted by major sources of SO₂ (Anderson et al. 2004). High concentrations of PM_{2.5} in the present study suggest the influence of emissions of primary PM_{2.5} and secondary particle formation from precursors gaseous emitted from local and regional sources. Coal combustion is a major source of energy in Guiyang City, and the coal utilized contains significant quantities of toxic elements such as Pb, As, Hg, and S (Zhao et al. 2008; Tian et al. 2015). As a result, the large number of coal-fired power plants, residential coal-burning boilers, and nonferrous smelting activities, such as Pb–Zn producing facilities may play a vital role in the high levels of heavy metals in observed in PM_{2.5} during this study.

3.3 Enrichment factors

Enrichment factors (EFs) (Soto-Jiménez et al. 2003) of elements relative to the earth's crust were calculated to demonstrate the potential contribution of local anthropogenic sources to the observed elemental levels in the ambient air, which can be applied to identify the potential sources of crustal and anthropogenic components. The EF for each PM_{2.5} element was calculated using Eq. (1):

$$EF_i = \frac{(C_i/C_{Ti})_{PM_{2.5}}}{(C_i/C_{Ti})_{Crust}} \quad (1)$$

where:

EF_i is the enrichment factor of element i, C_i is the concentration of the element investigated in the PM_{2.5} or the crust, C_{Ti} is the concentration of reference element (Ti) in the PM_{2.5} or the crust.

In this study, the concentrations of elements in the local soil were calculated based upon the average composition across the Guizhou Province. In enrichment factor analysis Fe, Ti, and Al are typically used as reference elements (Nováková et al. 2016; Wang J et al. 2016). For this study Ti was chosen as the reference element for calculating enrichment due to its stability and its propensity to be less susceptible to deterioration, alteration, and weathering. As previously suggested (Chester et al. 1999), EF values of less than 10 suggest that the element has a prominent crustal source, while an EF value greater than 10 suggests that a large proportion of that element has anthropogenic origins.

Results indicate that Se was the most enriched element in PM_{2.5}, followed by Pb, Zn, Br, and As (Fig. 6). These elements had high average EFs and ranged from 10² to 10³, suggesting the predominance of non-crustal sources. The EFs of Mg, Cr, Ca, K, Mn, Na, and Rb varied from 10 to 10² on average, which suggest an influence from both soil and non-soil emission sources. The high EFs of Se and As

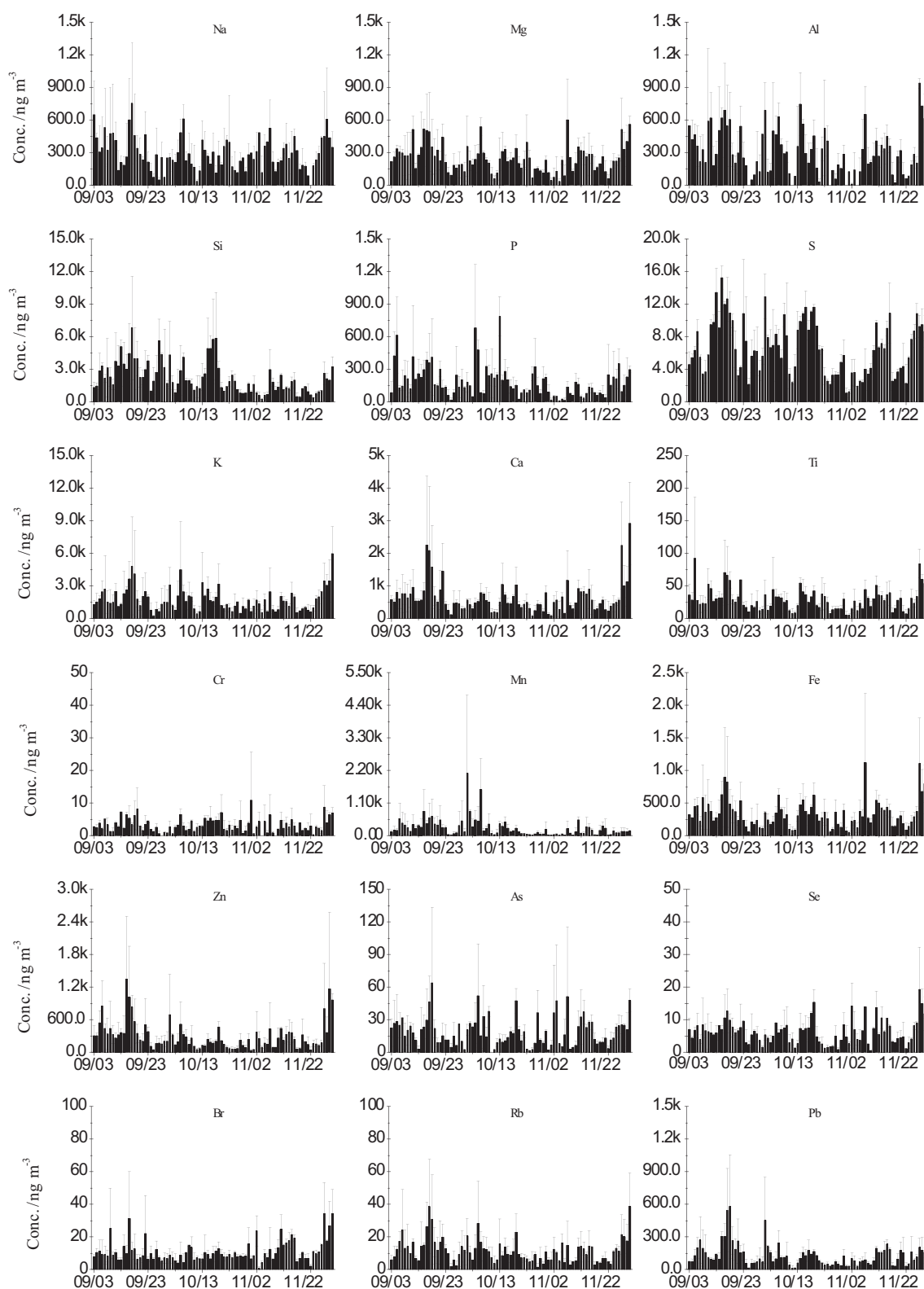
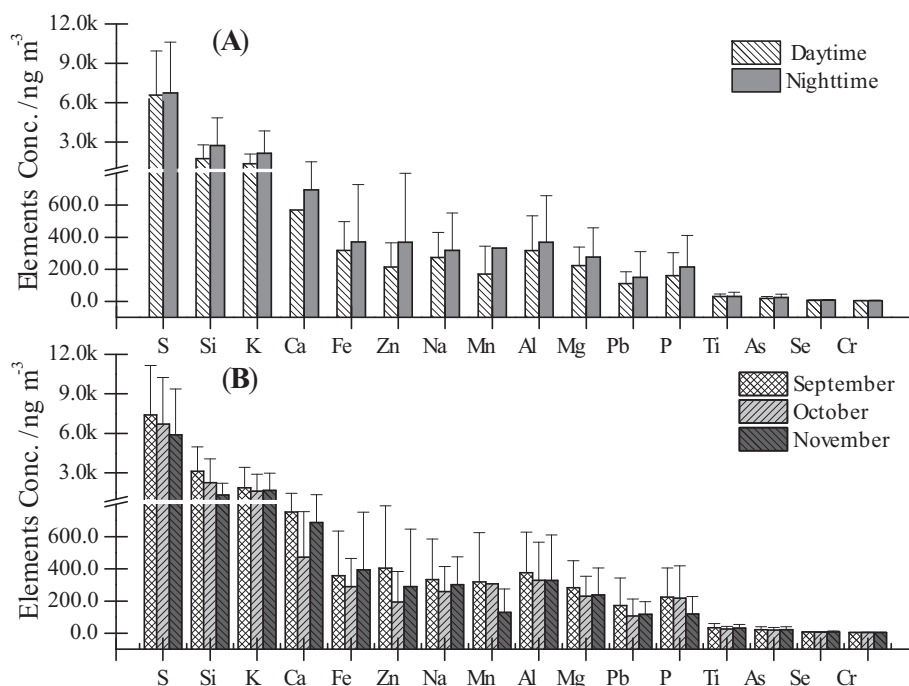


Fig. 4 Daily concentrations of chemical elements of $PM_{2.5}$ collected in Guiyang

suggest a coal combustion source, while high EFs of Pb, Zn, and bromine (Br) may reflect a large contribution from vehicle exhaust emission (Chao and Wong 2002). With low

EFs, Al and Fe are thought to originate mostly from resuspended local soil. In 2014, our 1 year data verified the high levels of As in $PM_{2.5}$ in Guiyang, with the highest

Fig. 5 Comparison of trace element concentrations measured in PM_{2.5} during the daytime and nighttime, study average (a) and the monthly average (b)



value recorded 10 times higher than the limit of 6.0 ng m^{-3} for ambient air quality standards in China. Hence, the high EFs of Se, particularly As in PM_{2.5} observed in the present study require further investigation.

3.4 Principal component analysis

Principal component analysis (PCA) is commonly employed to reduce the dimensionality of complicated PM_{2.5} datasets, grouping related chemical constituents, and identifying potential sources (Huang et al. 2010). In this research, Varimax rotated PCA was conducted using SPSS 18.0 software, and results are presented in Table 3. Three major components were identified with Eigen values greater than 1 and accounted for a sum of 72% of all variances in the dataset.

The first component had high loadings of Fe, Ca, Se, Br, Al, Ti, Mg, Cr, and Zn, most of which were from the fugitive dust (Kong et al. 2014). This component constituted 32% of the total variance. These elements were closely linked with the PM_{2.5} mass concentrations. In general, this fugitive dust factor likely includes windblown dust, agricultural tilling, construction dust, coal flyash, and resuspended road dust. The element Se is typically considered an element associated with coal-combustion and has a high EFs; however, Al, Fe with low EFs, mainly originated from soil dust (Shen LJ et al. 2016). In general, Cr is an important pollutant from solid waste treatment (Kulshrestha et al. 2009). Zn typically comes from various industrial sources and the abrasion of rubber tires along the

roads (Rogge et al. 1993), and Br mainly originates from vehicle exhaust (Chan et al. 1997). Component 1 is generally considered as typical of urban anthropogenic elements. Interestingly, Cr, Zn, and Br had high loadings (0.618, 0.583, 0.772), demonstrating that the solid waste treatment, industrial emissions, vehicle exhaust, and other pollution sources may somewhat contribute to fugitive dust due to long-term deposition of PM_{2.5} on the surface in this polluted area (Farinha et al. 2009).

Component 2 features loadings of Mn, Si, Pb, and S, comprising 19.7% of the total variance. This group of elements may represent multiple sources of industrial emissions. Usually, Mn, Pb, and S are regarded as indicators of the discharge from fuel burning and vehicle emissions (Pacyna and Pacyna 2002). The element Si is an indicator of emissions from soil dust, road dust, and construction dust.

Component 3 comprises 20% of the total variance and was mainly associated with Na, As, K, and Rb, which could be ascribed to solid waste incineration (Pacyna and Pacyna 2002). Combustion of pressure-treated lumber can emit As and K/Rb, both of which are associated with biomass combustion (Tao et al. 2013). In addition, K, Rb, and As have been associated with iron/steel slag/sintering operations (Pancras et al. 2013).

Table 2 Spearman correlations coefficients among PM_{2.5} mass and trace elements

PM _{2.5}	Na	Mg	Al	Si	P	S	K	Ca	Ti	Cr	Mn	Fe	Zn	As	Se	Br	Rb	Pb
PM _{2.5}	1																	
Na	0.266**	1																
Mg	0.670**	0.535**	1															
Al	0.573**	0.425**	0.736**	1														
Si	0.631**	0.108	0.546**	0.536**	1													
P	0.418**	0.142	0.317**	0.287**	0.411**	1												
S	0.951**	0.198**	0.547**	0.450**	0.564**	0.420**	1											
K	0.749**	0.546**	0.716**	0.651**	0.544**	0.475**	0.662**	1										
Ca	0.703**	0.504**	0.814**	0.694**	0.460**	0.261**	0.579**	0.729**	1									
Ti	0.701**	0.453**	0.754**	0.743**	0.450**	0.242**	0.609**	0.627**	0.842**	1								
Cr	0.538**	0.195*	0.471**	0.413**	0.363**	0.208**	0.462**	0.450**	0.517**	0.429**	1							
Mn	0.677**	0.243**	0.651**	0.470**	0.633**	0.477**	0.643**	0.656**	0.604**	0.517**	0.348**	1						
Fe	0.727**	0.533**	0.827**	0.734**	0.484**	0.199**	0.599**	0.697**	0.881**	0.817**	0.469**	0.580**	1					
Zn	0.694**	0.357**	0.699**	0.495**	0.488**	0.299**	0.632**	0.688**	0.766**	0.612**	0.445**	0.715**	0.734**	1				
As	0.525**	0.463**	0.482**	0.322**	0.218**	0.187*	0.471**	0.576**	0.543**	0.441**	0.314**	0.563**	0.664**	0.563**	1			
Se	0.681**	0.377**	0.538**	0.484**	0.399**	0.198*	0.575**	0.594**	0.708**	0.629**	0.533**	0.724**	0.665**	0.604**	0.425**	1		
Br	0.523**	0.235**	0.476**	0.391**	0.221**	0.127	0.392**	0.524**	0.540**	0.445**	0.285**	0.540**	0.469**	0.425**	0.570**	0.486**	1	
Rb	0.661**	0.509**	0.680**	0.572**	0.497**	0.371**	0.566**	0.906**	0.701**	0.551**	0.409**	0.636**	0.661**	0.583**	0.550**	0.486**	0.721**	1
Pb	0.767**	0.348**	0.702**	0.471**	0.524**	0.234**	0.703**	0.707**	0.743**	0.636**	0.415**	0.793**	0.846**	0.597**	0.656**	0.486**	0.721**	0.721**

Bold typeface indicated correlated > 0.6

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

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

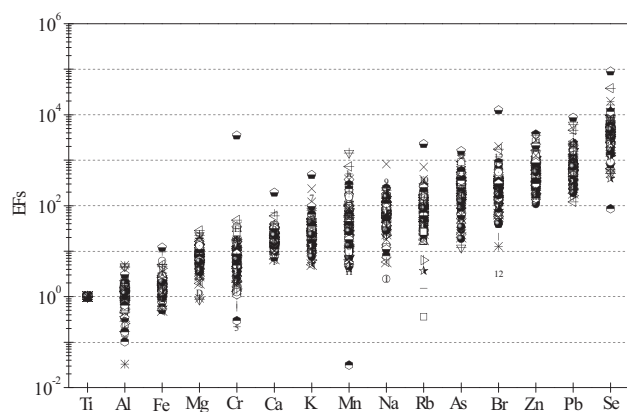


Fig. 6 Enrichment factors (EFs) of elements in PM_{2.5}

Table 3 Varimax rotated principal component factor analysis (PCA) loading matrix for elements in PM_{2.5} from Guizhou

Parameter	Component		
	1	2	3
Na	0.283	− 0.073	0.794
Mg	0.666	0.423	0.377
Al	0.671	0.303	0.324
Si	0.226	0.747	0.131
P	− 0.137	0.435	0.524
S	0.37	0.658	− 0.1
K	0.469	0.508	0.618
Ca	0.828	0.231	0.373
Ti	0.772	0.118	0.182
Cr	0.618	0.318	− 0.079
Mn	− 0.038	0.826	0.133
Fe	0.854	0.176	0.287
Zn	0.583	0.526	0.191
As	0.371	0.139	0.634
Se	0.843	0.106	0.146
Br	0.772	− 0.005	0.212
Rb	0.475	0.496	0.622
Pb	0.316	0.689	0.343
% Variance	32.5	19.7	20.0

Rotation method: Varimax with Kaiser Normalization (Paatero et al. 2005)

4 Conclusions

This pilot study presents characterizations of ambient PM_{2.5} as well as its elemental constituents in samples collected in Guiyang, a developing inland city in southwest China. The 12-h integrated PM_{2.5} concentrations showed a daytime average of $51 \pm 22 \mu\text{g m}^{-3}$ and a nighttime average of $55 \pm 32 \mu\text{g m}^{-3}$, respectively. Nighttime PM_{2.5}

concentrations exhibited a wider range and variance compared to daytime PM_{2.5} concentrations. The 24-h integrated PM_{2.5} levels mean concentrations were $53 \pm 25 \mu\text{g m}^{-3}$. The temporal variation of both PM_{2.5} and its chemical elements suggested the influence of local anthropogenic activities influenced by daily fluctuations in meteorology. Enrichment factor analysis indicated that Se, Pb, Zn, Br, and As likely originated from human activities. The PCA analysis suggested that fugitive dust, combined sources of industrial pollution, motor vehicle emissions, and waste incineration were probably major contributors to atmospheric PM_{2.5} in Guiyang. However, additional PM_{2.5} monitoring covering all seasons and source apportionment modeling will be necessary in the future to further elucidate and refine the atmospheric PM_{2.5} sources in Guiyang. In addition, the development of a local emission inventory from major sources would facilitate deterministic modeling runs that could be compared with receptor-based approaches in Guiyang, thus permitting the further identification of the sources, temporal variability, and chemical composition of ambient PM_{2.5}. Ultimately, studies like these would assist local agencies in developing cost effective and efficient emissions management strategies that would reduce levels of PM to below current standards and better protect public health.

Acknowledgements The U.S. Environmental Protection Agency (EPA), through its Office of Research and Development, partially funded and participated in the research described here through cooperative agreement CR-833232-01 through the U.S. National Science Foundation-National Research Council Research Associateship Award. We acknowledge Dr. Teri L. Conner for X-Ray Fluorescence data analysis with her comments. The views expressed in this paper are those of the authors and do not necessarily reflect the views or policies of EPA. It has been subjected to EPA Agency review and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use. This research was also partially funded by the National Key Basic Research Program of China (2013CB430004) and the National Natural Science Foundation of China (No. 40773067).

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