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Annual Characteristics, Source Analysis of PM_1-Bound Potentially Harmful Elements in the Eastern District of Chengdu, China

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Abstract

Chengdu, a megacity in southwestern China, experiences severe air pollution; however, knowledge of the seasonal variation in mass concentration, extent of potentially harmful elements (PHEs) contamination, and sources caused by heavy metals remains lacking. This study adopted a weighting method to calculate the daily mass concentration of PM_1 and used ICP-MS to determine PHE concentrations. Results indicated that PM_1 mass concentration was in the range 5.44–105.91 μg/m^3. Seasonal PM_1 mass concentration could be arranged in the following order: winter > fall > spring > summer. The concentrations of PHEs in the PM_1 sample mostly showed the same seasonal variation characteristics as mass concentration. The average concentration of each PHE decreased as follows: Cu (107.44) > Zn (81.52) > Pb (22.04) > As (8.17) > Sb (1.91) > Ni (1.87) > Cr(VI) (0.84) > Cd (0.40) > Tl (0.33) (ng/m^3). Enrichment factor values varied markedly from mild to anomalous enrichment. Principal component analysis revealed mainly derived from the fossil fuel combustion (55.215%).

Air pollutants, especially fine particulate matter (PM), are increasingly identified as detrimental to human health. Recent research has shown that PM_1 (aerodynamic diameter < 1 μm) accounts for more than four-fifths of the $PM_{2.5}$ mass in China (Yang et al. 2019). Chen et al. (2018) found that China is experiencing severe PM_1 and $PM_{2.5}$ air pollution, with a high $PM_1/PM_{2.5}$ ratio, and in Northeastern China, North China Plain, Eastern Coastal areas, and Sichuan basin, $PM_1/PM_{2.5}$ ratio is high (i.e., > 0.9), whereas in remote areas in Northwestern and North China, the ratio of $PM_1/PM_{2.5}$ is low (i.e., < 0.7). Due to its smaller size, higher surface area, and volume ratio, PM_1 is more harmful to humans than $PM_{2.5}$; it can carry more toxins from anthropogenic emissions deeper into lung alveoli (Lin et al. 2016; Agudelo-Castañeda et al. 2017; Qin et al. 2018; Wang et al. 2019). Evidence has shown that exposure to PM_1 in a long run may increase asthma and asthma-related symptoms (Yang et al. 2018), increase the risk of hypertension (Yang et al. 2019), lead to excessive mortality (Hu et al. 2018), and the risk of preterm birth (Du et al. 2020). Given the more serious adverse effects on the human body of PM_1 in comparison with $PM_{2.5}$, investigation of environmental PM_1 should receive greater attention.

Very fine particles (PM_1) can cause more damage to human health than fine particles, partly because they contain relatively higher levels of toxic chemicals, such as potentially harmful elements (PHEs) (Wang et al. 2018). Potentially harmful elements (PHEs), such as cadmium, chromium, arsenic, lead, and nickel, which are nondegradable and genotoxic, can induce carcinogenic outcomes in humans (Galindo et al. 2018). Usually, PHEs are recognized as tracers in particle source studies because of their unique source characteristics (Diapouli et al. 2017; Salcedo et al. 2016). Coal combustion, steel production, vehicular exhaust/non-exhaust emissions, power plants, and metal smelters are the principal sources of PHEs, such as cadmium, copper, chromium, nickel, and lead (Dall'Osto et al. 2008; Dai et al. 2015).

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Chengdu, an important central city in western China, accounts for approximately 19% of the total population of Sichuan Province. Located in the west of the Sichuan Basin, Chengdu is encircled by plateaus and mountains (Tian et al. 2019) that affect atmospheric dispersion (Chen et al. 2017). The factor may lead to the serious air pollution experienced in the city. Previous research on PM in Chengdu focused primarily on PM_{10} and $PM_{2.5}$ (Cheng et al. 2018), whereas few studies have investigated PM_1. Therefore, we collected PM_1 samples at a site in Chengdu over a 12-month period in 2018–2019. The study had three primary objectives: (1) to characterize the variation of PM_1 mass concentration and determine the concentration of nine PHEs using ICP-MS; (2) to evaluate the level of pollution of PM_1-bound PHEs based on enrichment factors (EF), and (3) to investigate the likely sources of such PHEs.

Materials and Methods

Sampling Collection

Continuous collection of samples was undertaken at Chengdu University of Technology (30° 40′ 26.55″ N, 104° 08′ 22.04″ E), in the eastern district of Chengdu between the second and third ring roads of Chengdu city, Sichuan Province (Fig. 1). The area is a complex mixture of roads and both residential and business buildings, including traffic congestion (diesel and gasoline emissions), demolition of old buildings and construction emissions, and industrial pollution. The observation site is on top of a five-story building, and there are no other high-rise buildings within 150 m. Therefore, the site represents a regular urban area in the eastern district of Chengdu. The PM_1 samples were obtained using moderate-volume (100 L/min) air samplers (Wuhan Tianhong Co., TH-150C). The annual average temperature is 21.7 °C. The samplers, which captured PM_1 on 90-mm quartz filters (Whatman QM-A), ran for 24 h (from 08:00 to 08:00 local time). Our sampling activity was divided into four periods representing the four seasons: fall (October 1–November 30, 2018, and September 1–30, 2019), winter (November 16, 2018 to February 28, 2019), spring (March 1 to May 31, 2019), and summer (June 1 to August 31, 2019). Overall, 114 valid samples were gathered.

PHE Analysis and QA/QC

Before sampling, the filters were combusted in a muffle furnace at 500 °C for 4 h to remove contaminants. They were placed in a chamber for 48 h under conditions of 50% ± 5% relative humidity and 25 ± 5 °C both before and after sampling (Cheng et al. 2018). For determining the PM_1 mass, an electronic microbalance (Sartorius CPA225D, Germany) was used to weigh the filters before and after sampling. The variation between duplicate weighing was < 0.00004 g. Thereafter, the filters were transferred in a plastic sampling filter box and kept refrigerated at − 20 °C until subsequent chemical analysis. Using $HF\text{-}HNO_3$ system, 25% of each sample was digested. Then with Milli-Q® water, the digestion solutions were diluted to a final volume of 10 ml, to be analyzed for PHEs using ICP-MS (PE 6000, Perkin Elmer Inc., CT, USA), as described previously (Wang et al. 2020). To assess the quality of the procedure, the GSS-4 standard also was analyzed. To ensure that the experimental process conformed to the standard, a blank experiment was performed. We defined a group of ten field samples as a sequence, and one standard sample was analyzed for each sequence to ensure the repeatability of the analytical instrument. For every sequence, each field sample was analyzed twice to ensure the relative error was not > 20% and the relative standard deviations was not > 10% (Table S1).

Data Processing Methods

Enrichment Factors

Enrichment factors (EF) are used widely to distinguish between anthropogenic and natural sources of trace metals in atmospheric PM (Othman et al. 2016; Wang et al. 2019). Generally, EF values are defined as the ratio of PHE content in a sample to the PHE content of the background. In this study, EF values for the PHEs in the PM_1 samples were calculated using Eq. (1):

$$\mathrm{EF} = \left(C_s/C_{\mathrm{Mn}}\right)_{\mathrm{PM_1\ sample}} / \left(C_s/C_{\mathrm{Mn}}\right)_{\mathrm{background}}, \quad (1)$$

where C_s = concentration of the element considered in the PM_1 sample or the background. C_{Mn} = concentration of reference elements (Mn) in the PM_1 sample or the background (Zajusz-Zubek et al. 2017). The contents of elements in soil in Chengdu were taken as the background values (Zhu et al. 2004). According to Yang et al. (2016), enrichment of elements can be categorized into five levels ranging from one to five. The classification values are given in Table S2.

Statistical Treatment

We applied principal component analysis (PCA) and correlation to determine the source apportionment of the PHEs in the PM_1 samples, while testing the initial data using the Kaiser–Meyer–Olkin test of sampling adequacy and the Bartlett test of sphericity. Statistical software package SPSS statistics 22.0 is used for statistical processing.

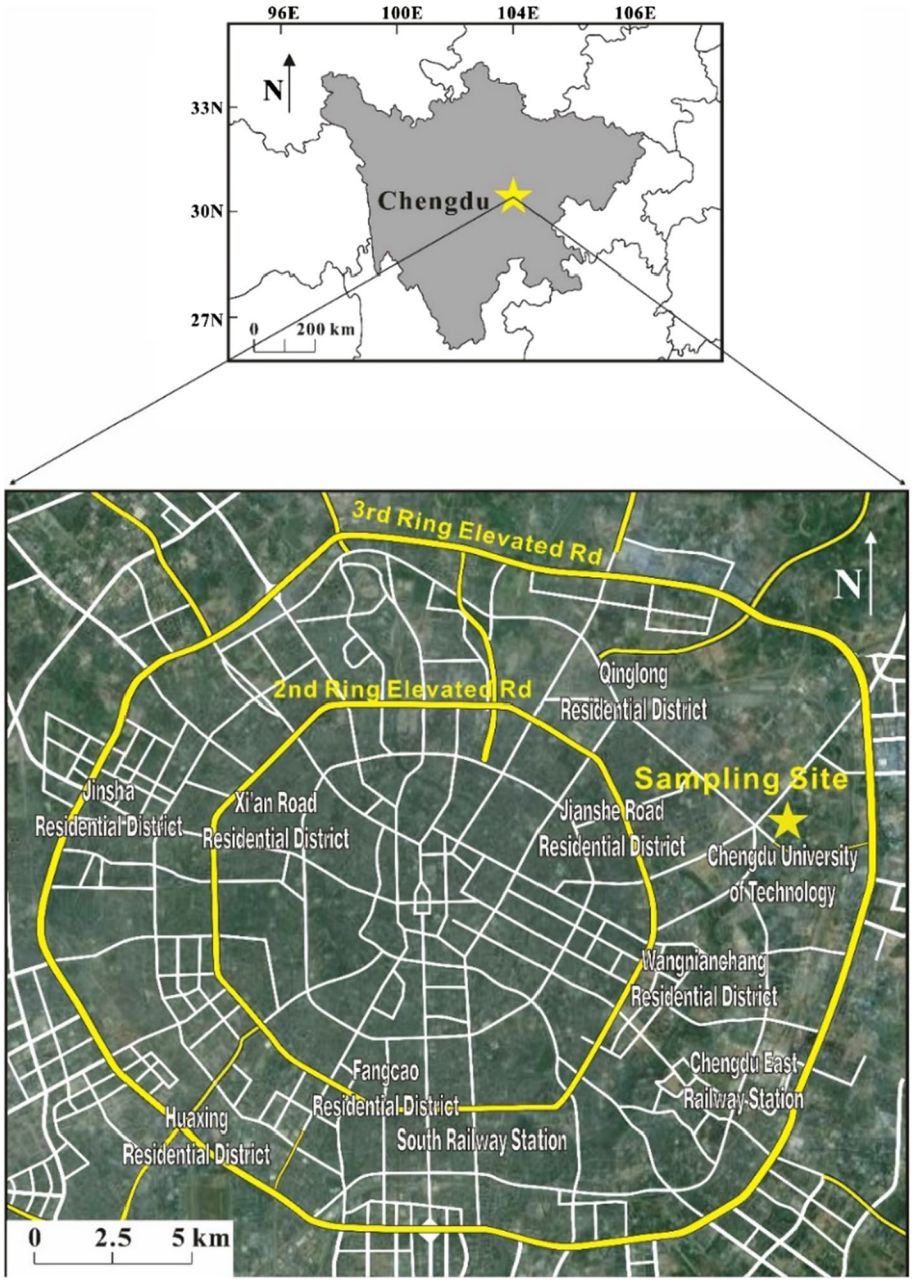

Fig. 1 Sampling site in the eastern district of Chengdu

Results and Discussion

Seasonal Variation of PM_1 Mass Concentrations

During our sampling period, the PM_1 mass concentration in spring, summer, fall, and winter was 15.25–77.60 (mean: 40.54 ± 15.38), 5.44–55.85 (mean: 29.50 ± 11.35), 19.22–96.72 (mean: 46.46 ± 20.62), and 22.54–105.91 (mean: 69.09 ± 20.97) μg/m^3, respectively. The annual average concentration was 46.50 ± 22.68 μg/m^3. Variation of PM_1 concentration with season was evident (Fig. 2), i.e., the mass concentration was lowest in summer, followed by spring, fall; the highest mass concentration was observed in winter. Compared with other seasons, the relatively high mass concentration of PM_1 in winter is mainly due to weak solar radiation, a low boundary layer, high frequency of calm weather, and limited diffusion of pollutants. In addition, the lower amount of rainfall in winter is also favors pollution buildup. Conversely, in summer, solar radiation is relatively strong, the boundary layer is vulnerable to destruction, and atmospheric convection is enhanced (Huang et al. 2020), which finally leads to a decrease in the mass concentration of PM_1.

Besides, PM_1 measured in this study was also taken into compared with that of other Chinese cities. As shown in Fig. 2, the annual mass concentration of PM_1 on average is

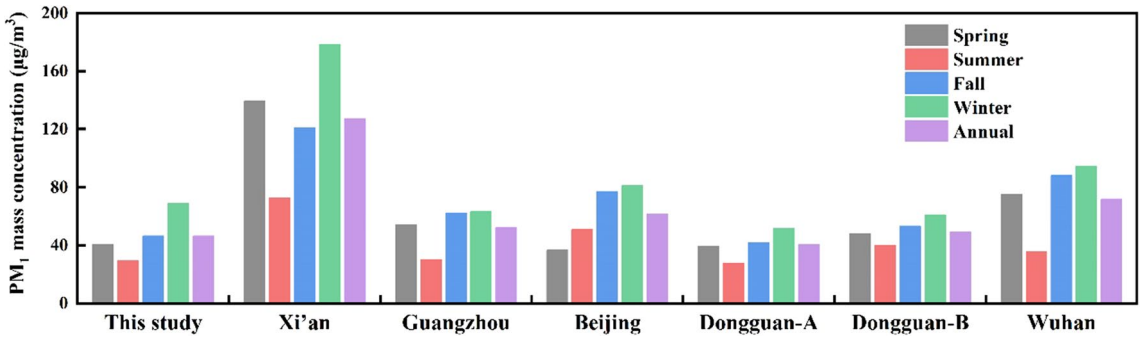

Fig. 2 Mass concentrations (μg/m^3) of PM_1 in selected Chinese cities. PM_1 mass concentration values reflect the period 2007–2008 in Xi'an (Shen et al. 2010), 2009–2010 in Guangzhou (Tao et al. 2012), 2016–2017 in Beijing (Fan et al. 2018), 2011–2012 in Dongguan-A (residential areas) and B (industrial areas) (Liu 2016), and 2013 in Wuhan (Liu 2016)

less than that of the other cities considered, except in residential areas of Dongguan. In winter, the PM_1 mass concentration in all the cities considered was higher than in other seasons. Although the concentration of PM_1 in the eastern district of Chengdu was not remarkably high, PM_1 has a more important role than larger PM in terms of air quality, the deterioration of visibility, climate change, and adverse effects on human health (Zhang et al. 2018); therefore, actions to reduce PM_1 emissions should be considered.

Characteristics of Elements in PM_1

The average concentration of each of nine PHEs measured in our PM_1 samples in different seasons during the entire study period is given in Fig. 3 and Table S3. The seasonal variation of Cd, Sb, Tl, Pb, Zn, and Cu in PM_1 sample was found similar and consistent with the seasonal variation of the mass concentration of PM_1, i.e., largest in winter and smallest in summer. Conversely, the concentration of both Cr(VI) and Ni was highest during summer, attributable primarily to resuspension of soil and road dust during the hot months. It was found that Tl does not exhibit significant seasonal variation. However, Tl is an extremely toxic metal element that requires investigation because of the lack of previous reports in relation to PM_1 particles.

The annual average concentration of PM_1 PHEs in the eastern district of Chengdu is given in Fig. 4. The mean concentration of the PHEs decreased in the following order: Cu (107.44) > Zn (81.52) > Pb (22.04) > As (8.17) > Sb (1.91) > Ni (1.87) > Cr(VI) (0.84) > Cd (0.40) > Tl (0.33) (ng/m^3). The annual average concentration of Cr(VI) and As in PM_1 was higher than the NAAQS limit of 0.025 and 6 ng/m^3, respectively, whereas the average concentration of Ni and Pb was lower than the WHO limits of 25 and 500 ng/m^3, respectively. Similarly, the average concentration of Cd was also lower than the NAAQS and WHO limit of 5 ng/m^3.

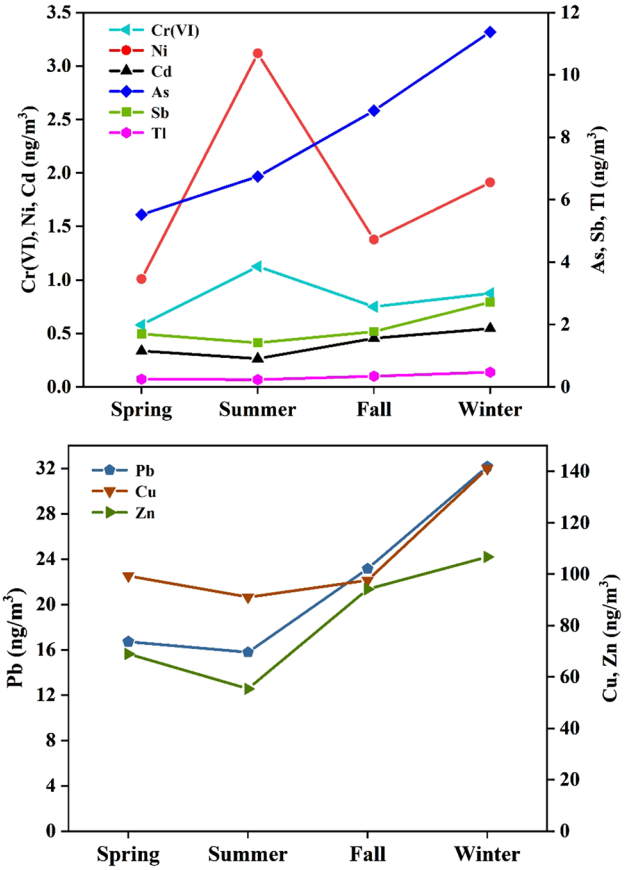

Fig. 3 Seasonal mean of PHE concentrations in PM_1

The EF for the PHEs of Cu, Sb, Cd, Zn, As, Pb, Tl, Cr(VI), and Ni in the PM_1 samples were calculated as 150.73, 115.58, 98.32, 41.63, 33.92, 33.16, 18.35, 0.44, and 2.35, respectively (Fig. 5). The EF value of both Cu and Sb exceeded 100, indicating anomalous enrichment of these PHEs, attributable primarily to anthropogenic activity. The EF values of Zn, As, Pb, Cd, and Tl reflect moderate

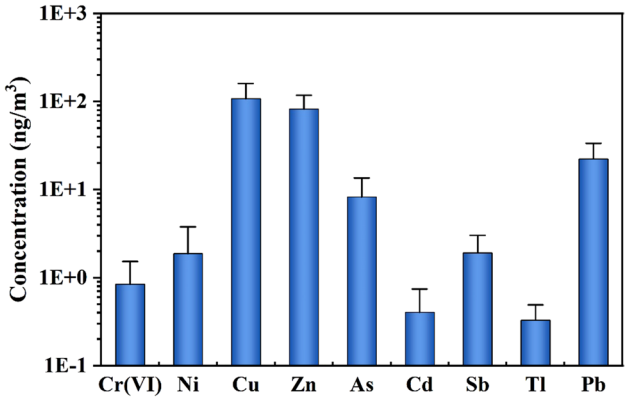

Fig. 4 Annual averages of PHE concentrations in PM_1 sampled from the eastern district Chengdu

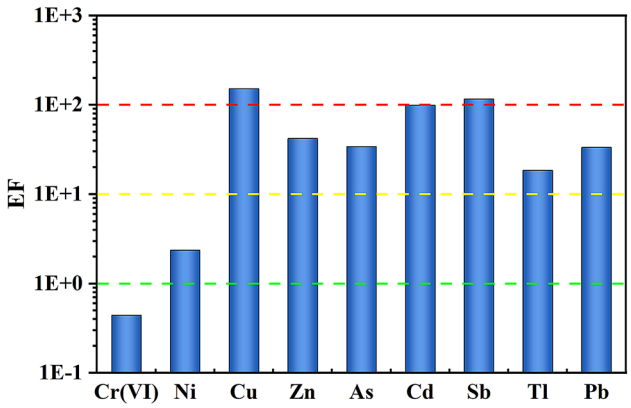

Fig. 5 Enrichment factors (EFs) of PHEs in PM_1

enrichment ($10 < EF \leq 100$), indicating that anthropogenic activity was an important source of these PHEs. The EF values for Ni, and Cr(VI) were < 10, indicating that these metals have only mild enrichment, attributable partly to the natural background (crust or soil) and partly to anthropogenic activity.

Source Identification

Correlation and Principal Component Analysis

A correlation matrix for the PHEs in PM_1 in the eastern district of Chengdu is illustrated in Fig. S1. In Chengdu, we found significant strong positive correlation between Cr(VI)–Ni (0.774), Cu–Zn (0.415), Cu–As (0.324), Cu–Cd (0.346), Cu–Sb (0.391), Cu–Tl (0.459), Cu–Pb (0.464), Zn–As (0.779), Zn–Cd (0.667), Zn–Sb (0.724), Zn–Tl (0.890), Zn–Pb (0.888), As–Cd (0.617), As–Sb (0.599), As–Tl (0.750), As–Pb (0.745), Cd–Sb (0.588), Cd–Tl (0.729), Cd–Pb (0.735), Sb–Tl (0.792), Sb–Pb (0.793), and Tl–Pb (0.998) at the $p < 0.001$ level. The data suggest that the nature of the sources, enrichment, and transportation of these PHEs could be similar.

The results showed that the value of the KMO test was 0.841 and that the value of p was less than 0.001. The principal component analysis (PCA) loadings and percentages are presented in Table 1. In this study, we considered only those variables with factor loadings > 0.7 to characterize the pollution sources. Three components were extracted from the PCA to explain 84.347% of the total variance for the eastern district of Chengdu. We found PC1 explained 55.215% of the total variance, comprising Zn, As, Cd, Sb, Tl, and Pb with respective loadings of 0.921, 0.830, 0.798, 0.835, 0.963, and 0.964, which could be related to fossil fuel combustion, vehicular exhaust emissions, and wear and tear of motor vehicle tires (Chakraborty and Gupta 2010; Singh and Gupta 2016). Coal combustion is the primary source of As, Sb, and Tl (Yang et al. 2016; Belzile and Chen 2017). The presence of Cd could reflect industrial activity and might be associated with proximity to electric appliance plants and auto repair plants. Gasoline containing lead has not been used in Chengdu since 2000 (Li et al. 2017). However, of all the cities in China, Chengdu is second only to Beijing in terms of the number of cars registered and thus vehicular exhaust remains the principal source of Pb pollution. Additionally, Pb pollution can be generated by various other sources such as tires and brake dust (Guttikunda et al. 2014; Gope et al. 2018). The PC2, which was dominated by Cr(VI) (0.913) and Ni (0.939), explained 19.635% of the total variance. Although the EF value of both Cr(VI) and Ni was < 10, considering the location of the eastern district of Chengdu, we believe that the primary sources of these PHEs are soil dust and stainless-steel plants (Querol et al. 2004; Chen et al. 2016; Li et al. 2017). The PC3 explained 9.497% of the total variance. Especially during conditions

Table 1 Results of the principal component analysis on PHEs in PM_1

PHEs	Component		
	1	2	3
Cr(VI)	0.248	**0.913**	−0.001
Ni	0.096	**0.939**	0.026
Cu	0.531	−0.049	**0.840**
Zn	**0.921**	−0.078	−0.095
As	**0.830**	0.014	−0.193
Cd	**0.798**	−0.029	−0.112
Sb	**0.835**	−0.051	−0.026
Tl	**0.963**	−0.088	−0.050
Pb	**0.964**	−0.082	−0.044
% Variance	55.215	19.635	9.497
% Cumulative	55.215	74.850	84.347

PCA loading > 0.7 are shown in bold

of traffic congestion, Cu could be emitted from the brake lining of motor vehicles (Sternbeck et al. 2002; Chakraborty and Gupta 2010; Yıldırım and Tokalıoğlu 2016; Singh et al. 2016). Furthermore, it is possible that exhaust gases emitted by the metallurgical industry could be a source of Cu (Yang et al. 2016).

Conclusions

The daily PM_1 samples were collected from October 2018 to September 2019 at eastern district Chengdu in Southwest of China. The mean mass concentration of PM_1 was 40.54 ± 15.38 μg/m^3 for spring, 29.50 ± 11.35 μg/m^3 for summer, 46.46 ± 20.62 μg/m^3 for fall, 69.09 ± 20.97 μg/m^3 for winter, thus showing a seasonal trend. The average PM_1 mass concentration in eastern district Chengdu was lower than that of other cities, except Dongguan A. The cadmium, stibium, thallium, lead, zinc, and copper in PM_1 samples had the same seasonal variation characteristics as mass concentration. The average concentrations for Ni, Cd, and Pb were lower than the WHO limit, whereas Cr(VI) and As were higher than NAAQS limit. The EFs of individual PHEs varied dramatically. Both Copper and Sb were enriched anomalously, Cd, Zn, As, Pb, and Tl were enriched moderately, Cr(VI) and Ni were mild enriched. According to the PCA results, the major sources of PHEs in PM_1 at the studied were combustion of fossil fuel, soil dust, stainless-steel plants, vehicular emissions, and metallurgical industry. Our findings provide indispensable information for the study of PM_1-bound potentially harmful elements in eastern district Chengdu, China.

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