



Contamination, sources and health risks of toxic elements in soils of karstic urban parks based on Monte Carlo simulation combined with a receptor model

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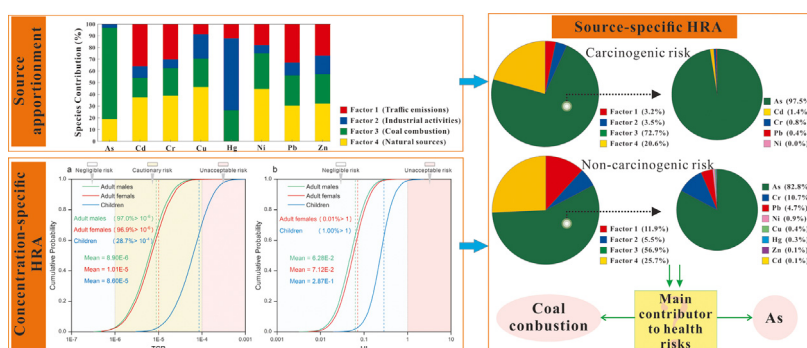
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HIGHLIGHTS

- Hg and Cd posed highest ecological risks in the study area.
- Four sources governing toxic element contamination were identified.
- The health risk to children was higher than adults.
- Arsenic from coal combustion was the main contributor to the human health risks.

GRAPHICAL ABSTRACT



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ABSTRACT

Understanding the health risks of toxic elements (TEs) in urban park soils and determining their priority control factors are crucial for public health and pollution management. Soil samples were collected from 33 urban parks in Guiyang, a typical karstic city. For each park, 15–45 topsoil samples were collected according to the area and then thoroughly mixed to obtain a representative sample. The results showed that the mean concentrations of TEs in park soils (22.5, 0.37, 88.6, 43.7, 0.26, 39.9, 44.7, and 101.0 mg/kg for As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn, respectively) were higher than their background values. Approximately 54.5% and 33.3% of enrichment factor (EF) values reached moderately enriched to significantly enriched levels for Cd and Hg, respectively. Moreover, 54.5% and 42.4% of monomial potential ecological index (EI) values were at considerable to high risk levels for Cd and Hg, respectively. These results illustrate that Cd and Hg pose high ecological risks. According to the potential ecological risk index (RI) values, 21.2% of the parks were exposed to considerable ecological risk and 48.5% were at moderate risk. Based on the positive matrix factorization (PMF) model, four sources governing TE contamination (including coal combustion, natural sources, traffic emissions, and industrial activities) were identified, with contribution rates of 32.3%, 31.0%, 19.6%, and 17.1%, respectively. A probabilistic health risk assessment showed acceptable non-carcinogenic risks and high levels of carcinogenic risk in all populations. Based on the source-specific health risk assessment, arsenic from coal

Abbreviations: TEs, toxic elements; PMF, positive matrix factorization; HRA, health risk assessment; MCS, Monte Carlo simulation; EF, enrichment factor; RI, potential ecological risk index; EI, monomial potential ecological index; NCR, non-carcinogenic risk; CR, carcinogenic risk; TCR, total carcinogenic risk index; HQ, hazard quotient; HI, hazard index; ADD, average daily intake dose; BV, background values; GV, guide value; CV, coefficients of variation.

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combustion was determined to be a major contributor to human health risks. Although several efforts have been made by the local government to eliminate coal-borne arsenicosis, our results revealed that the accumulation of arsenic in the soil due to coal combustion poses a potential threat to human health.

1. Introduction

Toxic elements (TEs) in urban park soils have a profound effect on human health. Owing to their pronounced cytotoxicity, persistence, insidiousness, and bio-accumulation, TE contamination has raised widespread concern (Han et al., 2021b; Tang et al., 2020; Xiong et al., 2017; Zeng et al., 2022a; Zhao et al., 2022). With rapid industrialization and urbanization, large amounts of TE contaminants have been released into the environment, and urban soils are considered the most prominent ultimate sinks for these TEs (Hiller et al., 2017; Wang et al., 2019b). In China, urban parks are vital recreational spaces for urban dwellers to enjoy outdoor activities, because most residents live in high-rise buildings (Qu et al., 2020; Zhao et al., 2020a). Local residents and frequent visitors to parks are susceptible to exposure to TEs in soil through dermal contact, oral ingestion, or inhalation (Hiller et al., 2017; Huang et al., 2021; Luo et al., 2012). Therefore, understanding the health risks of TEs in urban park soils and determining their priority control factors are critical for public health and pollution management.

To control the risk of TEs in soil to human health, it is necessary to first clarify their source apportionment (Long et al., 2021). Because of the inability to quantify the sources of contaminants, several existing approaches (e.g., cluster analysis, factor analysis, and principal component analysis) are not capable of source-specific risk assessment (Han et al., 2021a). However, the positive matrix factorization (PMF) model developed by U.S. EPA (2014) has been proven to solve this shortcoming by obtaining non-negative results and apportioning the source contribution to each element (Sakizadeh and Zhang, 2021; Song et al., 2021). In this study, geostatistical methods, correlation analysis, and the PMF model were combined to enhance the accuracy of the source apportionment.

The probabilistic health risk assessment (HRA) model provides a reliable means for characterizing the potential human health effects of contaminant exposure. Conventional HRA methods typically rely on deterministic exposure parameter values. However, this may lead to less reliable and informative results because of the inability to avoid uncertainty in HRA (Tong et al., 2018). Instead, Monte Carlo simulation (MCS), a valid probabilistic method, considers the uncertainty and sensitivity of different exposure pathways and thus provides a more realistic risk assessment (Ganyaglo et al., 2019; Kumari and Kumar, 2020). Recently, this probabilistic method has been employed by researchers to examine the potential health risks of contaminants in soils and other aspects of the environment (Huang et al., 2022; Orosun, 2021; Zhang et al., 2021b).

Identifying the main contributors to human health risks is a prerequisite and key step in the prevention and control of soil contamination. Owing to the different contents and toxicity coefficients of TEs, each individual source of contamination can lead to widely varying health risks (Huang et al., 2021; Huang et al., 2018a; Zhao et al., 2020a). Therefore, clarification of the relationship among health risks, TEs, and contamination sources is urgently needed to determine the primary health hazard factors (Sun et al., 2022). However, systematic studies of this relationship are limited. Therefore, this study was conducted to identify the factors that have the greatest impact on human health by elucidating the effects of individual TE and contamination sources on harm risks.

Guizhou Province is located in the center of the world's largest karst region, and includes a unique distribution pattern of soil and water resources and ecological vulnerability (Wang et al., 2020b; Xia et al., 2021). Guizhou contains concentrated energy and mineral resources. Intensified mining activities, rapid urbanization, and industrialization have led to increasingly serious TE contamination in the region (Ma et al., 2020; Ning et al., 2021; Xia et al., 2020). As the capital city of Guizhou Province, Guiyang is the first national forest city and is famous for its good ecological environment and resort tourism. It is also a central industrial city in Southwest China (Tian et al.,

2021). Numerous small-scale non-ferrous metal smelters, metal-processing industries, and cement plants exist in the city (Xu et al., 2016). Owing to the rapid growth of vehicles and the limited urban area, traffic in Guiyang is very congested. In addition, Guiyang is rich in coal resources, and coal was extensively used before 2000. However, little attention has been paid to the HRA of soil TE contamination in urban parks in typical karst regions.

Therefore, using Guiyang, a provincial capital city in a karst region as a case study, the present study aimed to: (1) investigate the contamination characteristics of multiple TEs in urban park soils, (2) quantitatively derive their sources, (3) assess the health risks due to exposure to TEs, and (4) determine the major environmental contributors to human health risks in this region.

2. Materials and methods

2.1. Study area and sample collection

Guiyang, the capital city of Guizhou Province, is located in a typical karst area in southwest China. It is a prominent tourism city and has been given the title of 'Summer Capital of China.' At an elevation of 1050 m, the city is controlled by a westerly wind belt year-round. The mean annual temperature of Guiyang is 15.3 °C and the annual precipitation ranges from 1100 to 1400 mm (Tang et al., 2007). Owing to the extensive use of coal, the city has suffered from severe acid rain for many years (Tian et al., 2021). Industry is the backbone of Guiyang's economy, and includes coal-fired power plants, cement, non-ferrous metal smelting and deep processing, tobacco products, pharmaceuticals, and rubber products (Xu et al., 2016). By 2020, the permanent population of Guiyang was 5.99 million, with an urbanization rate of 76%. The urban area of Guiyang covers the districts of Nanming, Yunyan, and several parts of Guanshanhu, Baiyun, Huaxi, and Wudang (Fig. S1).

Samples were collected from 33 parks in the urban area of Guiyang (Fig. S1). Depending on the size of the park and the points at which tourists often gather or pass by, 15–45 sub-samples of topsoil (0–5 cm) were collected from each park (Gu et al., 2016; Huang et al., 2021). The sub-samples were then thoroughly mixed with equal weights from each sub sample to obtain a representative sample (Beroigui et al., 2020; Hiller et al., 2017; Liu et al., 2020). The total weight of each composite soil sample was not less than 2 kg. All soils were stored in polyethylene Ziplock bags and transported to the laboratory for further processing.

2.2. Sample preparation and analysis

All soil samples were air-dried at room temperature, ground with an agate mortar, and homogenized through a 100-mesh nylon sieve (0.15 mm) after unnecessary materials, such as plant debris, stones, and gravel, were removed. Ground soil samples were digested with aqua regia. The concentrations of TEs were measured using ICP-MS (NexION 1000G) and AFS-9750, and the Al content was detected using ICP-OES (Avio 200). Method blanks, duplicates, and the national soil standards (GBW07390, GBW07981, and GBW07404a, respectively) were used to verify the accuracy of the digestion. The recoveries of these elements were 90–110%, and the relative differences among the soil replicates was <5%.

2.3. Soil TE contamination indices

The enrichment factor (EF) was calculated to evaluate the enrichment level and anthropogenic influence on soils as follows (Gujre et al., 2021; Sun et al., 2018):

$$EF = \frac{(C_i/C_x)_{Sample}}{(C_i/C_x)_{Background}} \quad (1)$$

where (C_i/C_x) is the concentration ratio of a particular element (C_i) to the normalized element (C_x) in the soil sample and the reference background. Owing to its large component in soil and immobility, Al is usually chosen as the normalized reference element (Huang et al., 2021; Ma et al., 2020; Tang et al., 2010).

The potential ecological risk index was applied to evaluate the ecological risks of TEs in soil and was calculated as follows:

$$EI = T_r^i \times \frac{C_i}{S_i} \quad (2)$$

$$RI = \sum_{i=1}^n E_r^i \quad (3)$$

where EI represents the monomial potential ecological index, T_r^i is the toxic response coefficient obtained from Buch et al. (2021). C_i and S_i are the concentrations of each TE in the soil samples and reference background material, respectively. Additionally, n denotes the number of TEs. The evaluation criteria for the EF, EI, and RI are listed in Table S1 (Wu et al., 2021).

2.4. Positive matrix factorization (PMF) model

Receptor models are mathematical techniques that have been extensively adopted in recent years to analyze the contributions of various sources based on sample compositions or fingerprints (Cheng et al., 2020; Wang et al., 2019c). The PMF is a particularly efficient receptor model and a multivariate factor analysis tool recommended by the U.S. EPA for the quantitative apportionment of sources (Huang et al., 2018b; Ran et al., 2021). It offers significant advantages in data preprocessing, including its ability to handle datasets with missing data, analyze data rationality, and estimate uncertainty (Wang et al., 2020a). In this study, EPA PMF 5.0 was applied to obtain potential sources and quantify the contribution from each source to urban park soils. The detailed calculations and explanations of this model are described in the corresponding user guide (U.S. EPA, 2014).

2.5. Health risk assessment (HRA) model

In this study, the HRA model was utilized to calculate the potential health risks (including non-carcinogenic (NCR) and carcinogenic risks (CR)) posed by TEs via different pathways (Adimalla et al., 2020; Men et al., 2021). Considering their behavioral and physiological differences, local residents were classified into three groups: children, adult males, and adult females (Men et al., 2018; Sun et al., 2022). Potential NCR was evaluated based on the hazard quotient (HQ) and hazard index (HI) (Eq. (4)). The potential CR and total carcinogenic risk index (TCR) were quantified using Eq. (5) (Han et al., 2021a; Penteado et al., 2021), as follows:

$$HI = \sum HQ_i = \sum \frac{ADD_i}{RfD_i} \quad (4)$$

$$TCR = \sum CR = \sum ADD_{ij} \times SF_{ij} \quad (5)$$

where ADD is the average daily intake dose of TEs via direct ingestion, dermal contact, and inhalation. Detailed calculations of ADD are provided in the Supplementary Materials, and the corresponding indices are provided in Table S2. RfD_i and SF_{ij} are the reference dose and slope factor values, respectively, for each TE (Table S3). Serious non-carcinogenic effects to the public may occur if the HI (or HQ) value is greater than 1; otherwise, the non-carcinogenic effect posed is insignificant (Gu et al., 2016; Gujre et al., 2021). If the TCR (or CR) value exceeds 1×10^{-4} , the carcinogenic risk is unacceptable; if the TCR (or CR) value is less than 1×10^{-6} , there is no apparent risk to human health (Adimalla et al., 2020).

Considering the spatial heterogeneity of TEs in park soil and the inter-individual variability of exposure parameters, HRA results may be subject

Table 1

Concentrations of toxic elements (mg/kg) in soils from Guiyang urban parks.

	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Mean	22.5	0.37	88.6	43.7	0.26	39.9	44.7	101.0
Median	18.1	0.38	87.2	41.8	0.19	37.5	43.6	100.2
Max.	79.8	0.71	169.0	77.3	0.79	72.3	81.9	206.8
Min.	4.0	0.12	34.1	13.6	0.08	12.8	18.6	41.2
SD	17.8	0.15	28.1	19.5	0.19	17.0	17.8	34.1
CV	0.79	0.41	0.32	0.45	0.73	0.43	0.40	0.34
BV	13.3	0.13	86.6	25.7	0.10	33.7	29.3	82.4
GV	20.0	20.0	n/a	2000.0	8.0	150.0	400.0	n/a
Over BV/%	72.7	97.0	51.5	81.8	93.9	57.6	72.7	72.7

Abbreviations: SD, standard deviation; CV, coefficient of variation; BV, background values for toxic element concentrations in soils in Guizhou Province; GV, guide values of soil environmental quality for development land (GB36600–2018); Over BV, over background value rate.

to high uncertainty or bias (Tarafdar and Sinha, 2018; Tong et al., 2018). To address these shortcomings, a Monte Carlo simulation (MCS, a probabilistic risk approach) was implemented in the HRA model (Gope et al., 2020; Zhang et al., 2021b). The exposure parameters listed in Table S2 were probabilistically simulated. The TE concentrations were also fitted to a lognormal distribution (Table S4). In this study, both concentration- and source-specific health risk assessments were conducted using a MCS in the HRA model.

2.6. Statistical analysis

Descriptive statistics were implemented using SPSS 26. Plots were constructed using GraphPad Prism 8 and Origin 2018. The spatial distributions of the TE content, EF, EI, and RI values were determined using ArcGIS 10.7 with inverse distance weighted interpolation (Zeng et al., 2022b). The MCS was conducted using the Oracle Crystal Ball software, and the simulation was run for 10,000 iterations (Sun et al., 2022; Zhang et al., 2021a).

3. Results and discussion

3.1. Concentrations of toxic elements in soils

The content of most TEs in the soils of the Guiyang urban parks was relatively high. The mean concentrations of TEs ($\text{mg}\cdot\text{kg}^{-1}$) occurred in the following order: Zn (101.1) > Cr (88.6) > Pb (44.7) > Cu (43.7) > Ni (39.9) > As (22.5) > Cd (0.37) > Hg (0.26) (Table 1). Except for Cr and Ni, the concentrations of the elements were notably higher than their background values (BV) in Guizhou ($p < 0.01$, one-sample *t*-test) (Wang and Wei, 1995). In particular, the concentrations of Cd, Hg, Cu, As, and Pb were 2.7-, 2.5-, 1.7-, 1.7, and 1.5 times higher than their BVs, respectively. The

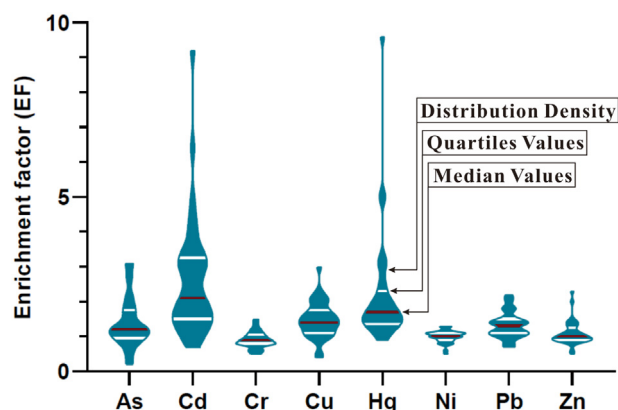


Fig. 1. Violin plots of toxic elements (TEs) with enrichment factor (EF) values in urban park soils of Guiyang.

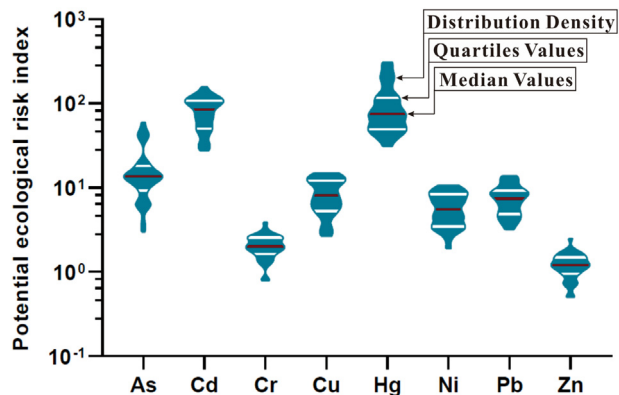


Fig. 2. Violin plots of TEs with monomial potential ecological index (EI) values in urban park soils of Guiyang.

soil concentrations of Cd, Hg, and Cu exceeded their BVs in 97.0%, 93.9%, and 81.8% of the parks, respectively, while the ratios were 72.7% for As, Pb, and Zn. These results indicate that there was a relatively obvious enrichment of elements in addition to Cr and Ni in soils from Guiyang urban parks. Furthermore, the soil As content exceeded the guide value (GV) in 30.3% of the studied parks, suggesting that As may generate potential hazards to the ecological environment and human health.

High heterogeneity was observed in the spatial distributions of elements. All elements had relatively high coefficients of variation ($CV > 0.3$), especially As and Hg, which had CV values of 0.79 and 0.73, respectively. This suggests that these elements exhibit high spatial variation, and that the distribution of As and Hg might be influenced by external forcing and point sources of contamination (Liu et al., 2021). This result was confirmed by the spatial distribution of the soil element concentrations in the study area (Fig. S2). The spatial distributions of elements can be used to gain insight into the degree of soil contamination and to support the development of remediation methods for managing contaminated soils in the study area (Ran et al., 2021; Zeng et al., 2022b). The spatial patterns and hotspots of Cr, Cu, Ni, Pb, and Zn were highly similar. Moreover, the Pearson correlation coefficients revealed significant positive correlations among most of the TEs in soils, suggesting similar sources or controlling factors (Han et al., 2020; Zeng et al., 2020). Furthermore, all TEs except Cd showed significant positive correlations with Al ($0.52 < r^2 < 0.91, p < 0.01$), indicating a possible influence of soil grain size (Zhao et al., 2021). Overall, the spatial distribution of TEs in park soils differed from west to east, with the element content being higher in the northwest. In addition to the influence of grain size, this pattern may also be related to intensive industrial activity in the northeast. This result is discussed further in Sections 3.2 and 3.3 of this paper.

3.2. Environmental risks of toxic element contamination in soils

Cd and Hg had higher contamination levels than other TEs in the soils. The average EF values of the studied elements decreased in the following order: $Cd > Hg > Cu > As > Pb > Zn > Ni > Cr$ (Fig. 1). Approximately 54.5% and 33.3% of the samples had moderate to significant enrichment levels of Cd and Hg, respectively, which indicated that Cd and Hg occur at relatively high contamination levels. The EF values for Cr and Ni were always below 2, and their mean values were less than 1, indicating that the enrichment of Cr and Ni in park soils was relatively low, which might be influenced by the relatively low level of anthropogenic activity related to these elements compared to that of other elements (Ma et al., 2020). The average EI values of the TEs decreased in the following order: $Hg > Cd > As > Cu > Pb > Ni > Cr > Zn$ (Fig. 2). The average EI values for Hg and Cd were 101.6 and 82.3, respectively, signifying considerable risk. Moreover, approximately 54.5% and 42.4% of the EI values were at considerable to high-risk levels for Cd and Hg, respectively. Therefore, the results of both the EF and EI values illustrated that Cd and Hg pose high ecological risks to the park soils of Guiyang.

Generally, the RI value is related to human activity. The mean RI value for TEs in the study soils was 226.1, indicating a moderate risk. Overall, 21.2% of the parks were exposed to considerable ecological risk and 48.5% were at moderate risk. The spatial variability of the RI value indicated that more seriously contaminated areas occurred in the northwest and central areas of the city (Fig. S3). Dominated by residential and commercial real estate, the central city is the most densely populated, highly visited, and heavily trafficked area (Huang et al., 2021). The northwestern area has the highest concentration of industrial enterprises. The TE contents of park soils in these areas strongly disturbed by anthropogenic activities and were higher in these than in other regions (Liu et al., 2020). Therefore, it is necessary to reinforce the conservation of the soil environments in these regions.

3.3. Source apportionment

Correlation and geographical statistical analyses are effective tools for source apportionment of soil contaminants. As previously noted (Section 3.1), the distribution of TEs in the study area was potentially influenced by soil grain size. Therefore, to eliminate the effect of grain size on TE enrichment, a partial correlation analysis was performed with Al as the control variable (Zhao et al., 2020b; Zhao et al., 2021), and the results are displayed in Fig. 3c. In addition, the EF values for TEs can be used to indicate different sources, including natural or anthropogenic sources (Ma et al., 2020). Geographical statistical analysis, which can be used to visualize the local conditions, is a useful method for identifying contamination hotspots and sources (Huang et al., 2021; Wang et al., 2020c). Hence, spatial variations in the EF values of TEs were determined using GIS methods (Fig. 4).

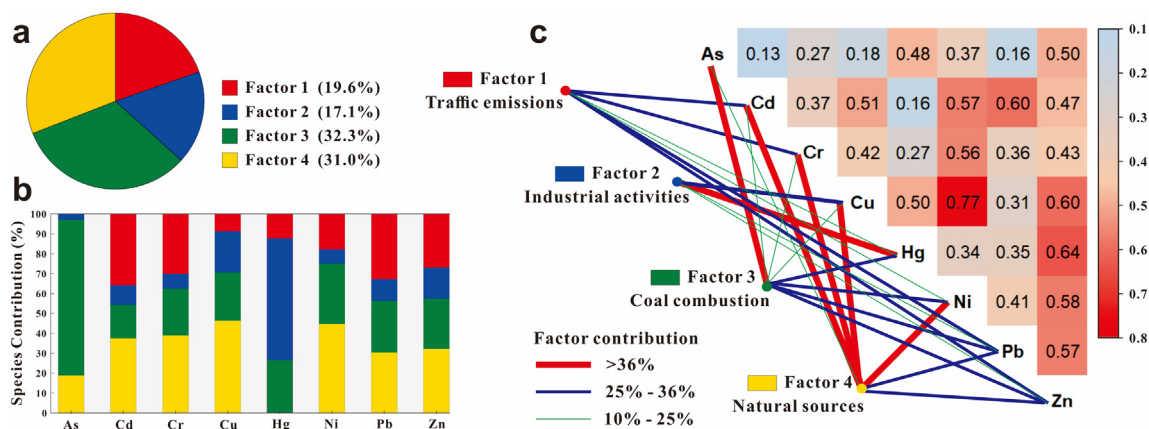


Fig. 3. Source apportionment of TEs in urban park soils of Guiyang. (a) The percentage of different pollution sources. (b) Species contribution of 4 pollution sources for TEs. (c) Combined illustration of 4 sources and partial correlation analysis (Al as the control variable) among TEs.

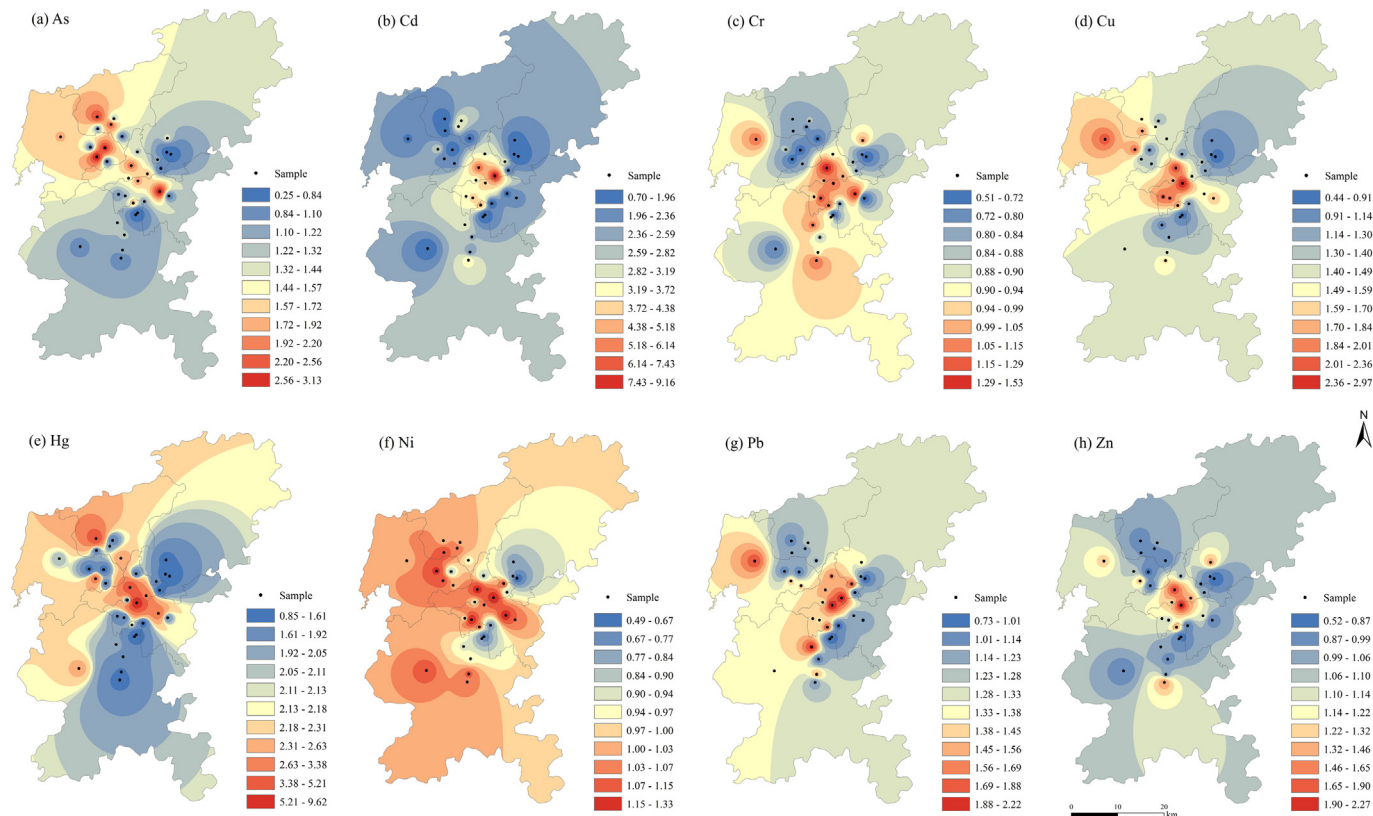


Fig. 4. Spatial variations in enrichment factors of the TEs in urban park soils of Guiyang.

PMF analysis was employed to determine the sources and quantify the contributions of TEs in the Guiyang urban park soils. The number of factors was set from three to five and run 20 times until a minimum and stable Q value was obtained (Ran et al., 2021). Almost all of the residuals were between -3 and 3 , and the R^2 values were 0.99, 0.45, 0.73, 0.98, 0.99, 0.95, 0.91, and 0.88 for As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn, suggesting an accurate and rational fit for PMF modeling (Liu et al., 2021; Yang et al., 2020). Consequently, four factors were extracted via the PMF model, with contribution rates of 19.6%, 17.1%, 32.3%, and 31.0%, respectively (Figs. 3 and S4).

Factor 1 may have been associated with traffic emissions. Accounting for 19.6% of the total variance, this factor was characterized by Cd (35.8%), Pb (32.7%), Cr (30.1), and Zn (26.8%) (Figs. 3 and S4). Relatively high correlation coefficients were observed between these elements (Fig. 3c). Moreover, parks with higher EF values were more concentrated in the central city, which has a high traffic volume, according to the GIS map (Fig. 4). Pb and Cd are major markers of a traffic source (Huang et al., 2021). Vehicle emissions are estimated to account for approximately two-thirds of global lead emissions (Fei et al., 2020). Although the use of leaded petrol was prohibited in 2000 in China, considerable levels of Pb still exist in soil due to the centuries-long half-life of Pb (Guan et al., 2018; Wang et al., 2020a). Tire dust, oil burning in automobiles, and braking-related emissions from trains could induce significant non-point Cd contamination in urban areas (Javed et al., 2017; Liang et al., 2017). Zinc is common in automobile tires, lubricant oils, and galvanized parts (Hu et al., 2018; Nazarpour et al., 2019; Thapalia et al., 2015). Hence, the contamination hotspots of Cd, Pb, Cr, and Zn in the central city may be the consequence of traffic emissions.

Factor 2 was allocated to industrial activities such as non-ferrous metal smelting. Accounting for 17.1% of the total variance, this factor had high loading values for Hg (61.2%), followed by Cu (20.7%), and low loadings for Cr (7.3%), Ni (7.0%), and As (2.9%) (Figs. 3 and S4). Mercury, which originates primarily from smelting, coal combustion, and cement production in China, is commonly used to indicate industrial emissions (Fei et al., 2020; Sun et al., 2020). Feng and Qiu (2008) noted that coal combustion and metal smelting were the two main anthropogenic sources of Hg emissions in

Guizhou and were responsible for the high Hg content in local ecosystems. Guizhou is a province with abundant coal production and consumption and its coal often contains high levels of arsenic and mercury (Cao et al., 2021). During coal burning, large amounts of contaminants such as As, Cr, and Ni are discharged into the environment, accompanied by the release of mercury (Tian et al., 2014). However, the rather low weight of Cr, Ni, and especially, As, in this factor indicates an industrial source with little impact from coal combustion. In the past few decades, industries such as non-ferrous metal smelting and deep processing, chemical plants, rubber, cement, medicine, and tobacco production have been the pillars of the Guiyang economy. Therefore, this factor was considered to reflect industrial activity.

Factor 3 can be interpreted as coal combustion. Accounting for 32.3% of the total variance, this factor was dominated by As (78.3%), Ni (30.5%), Hg (26.6%), Pb (25.9%), Zn (25.2%), Cu (24.3%), Cr (23.8%), and Cd (16.8%) (Figs. 3 and S4). As has been extensively used as a tracer for coal burning (Olise et al., 2019). Coal in China, especially in Guizhou, has high levels of arsenic compared with other regions of the world (Liang et al., 2017). During coal combustion, large amounts of As are released into the air, water, and soil, resulting in serious environmental contamination. Industries, coal-fired power plants, and residential areas in China are estimated to emit 522, 252, and 21 tons of arsenic into the atmosphere each year, respectively (Kang et al., 2011). Tian et al. (2014), Tian et al. (2011) reported that approximately 550 and 335 t of As were emitted from coal-fired power plants in China in 2007 and 2010, respectively. In addition, numerous studies have demonstrated that many hazardous elements, such as Ni, Hg, Pb, Cr, and Cu, are released during coal combustion (Fei et al., 2020; Wang et al., 2020a). As a large coal-fired city, there are three main combustion systems for coal in Guiyang: large-scale coal-fired power plants (34% of the annual coal supply), medium-to-small-sized coal-fired steam boilers (44% of the annual coal supply), and domestic users (22% of the annual coal supply) (Tang et al., 2007). Emissions of As, Ni, Hg, Pb, Cr, and Cd from coal-fired power plants were estimated to be 8.52, 12.39, 5.2, 13.54, 9.51, and 0.43 t, respectively, in 2010 in Guizhou Province (Tian et al., 2014). The GIS map of EF values confirmed that the hotspots of As were clustered in the central and northwest

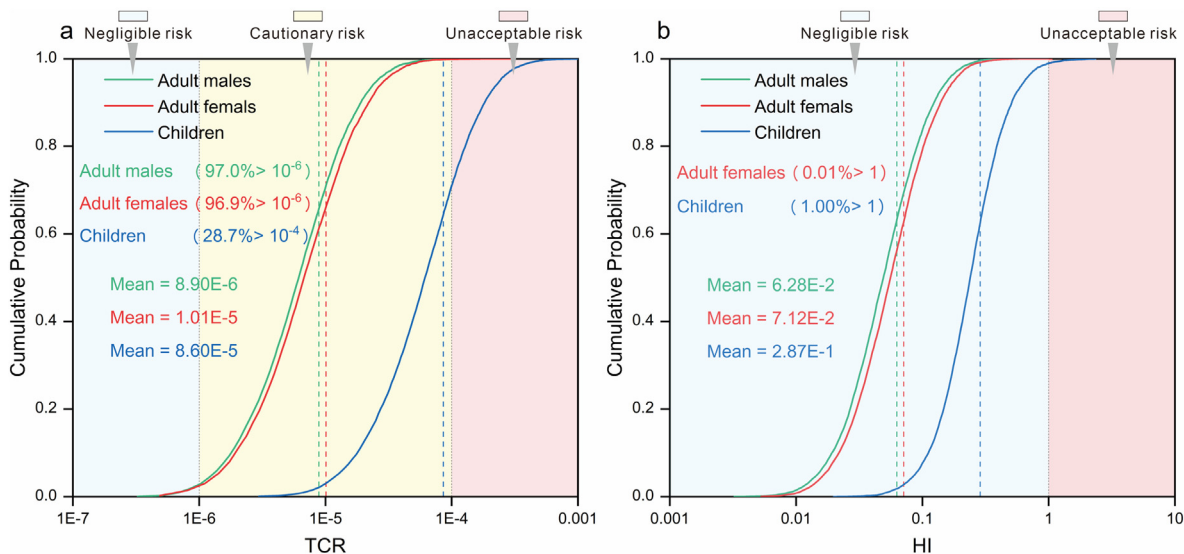


Fig. 5. Cumulative probabilistic estimate of total carcinogenic risk (a) and hazard index (b). The vertical dashed lines in different colors denote the mean values of the different populations.

parts of the city, which have a high density of residents or industrial facilities (Fig. 4). Therefore, this factor was identified as coal combustion.

Factor 4 may be related to natural sources. Accounting for 31.0% of the total variance, this factor was defined as Cu (46.4%), Ni (44.7%), Cr (38.9%), and Cd (37.5%) (Figs. 3 and S4). Relatively high correlation

coefficients were found between these elements (Fig. 3c). The mean Cr and Ni concentrations were similar to their local background values (Table 1). The EF values of Cr and Ni in all parks studied were lower than 2, indicating minimal enrichment (Fig. 1). Moreover, the CV values of Cr were lower than those of the other TEs (Table 1) and the EF values of Ni showed a relatively

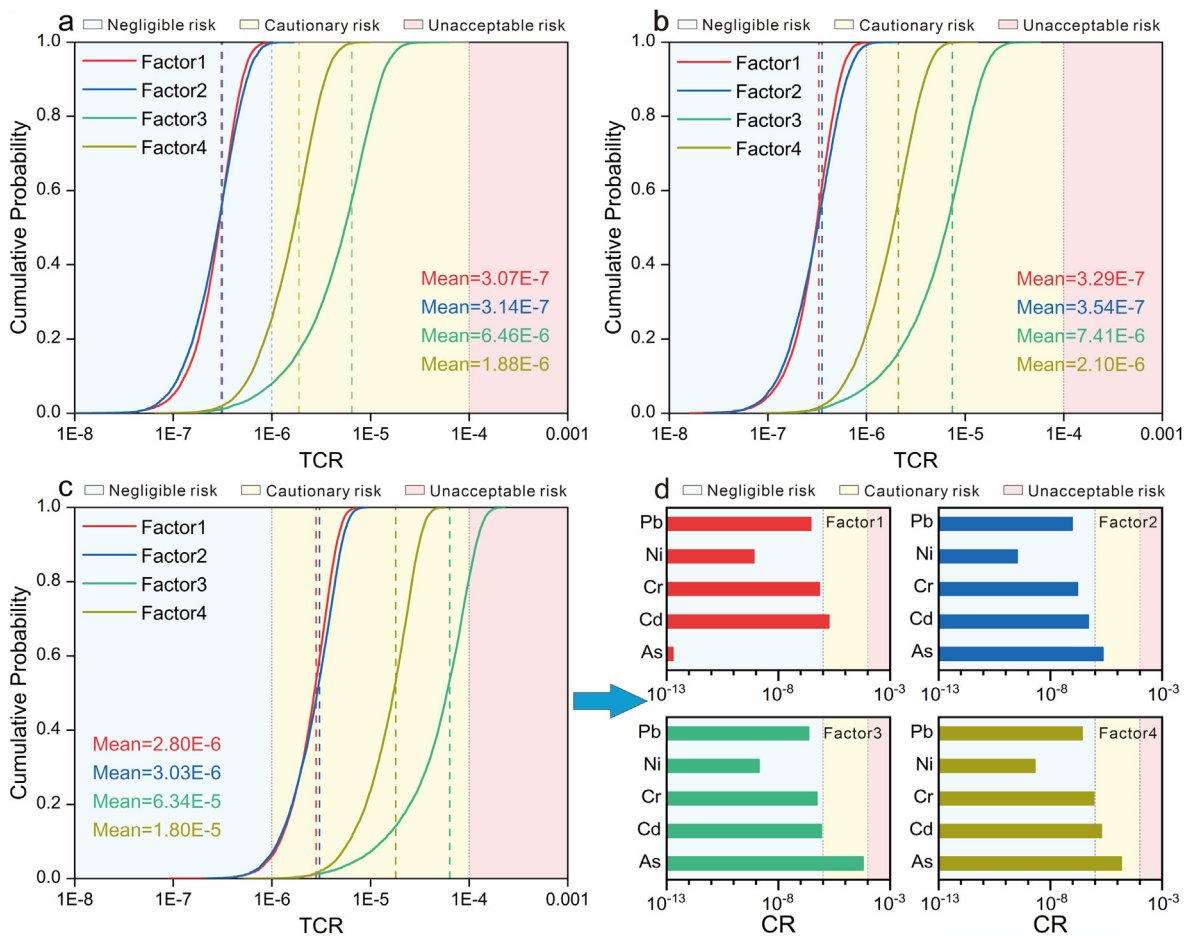


Fig. 6. Cumulative probabilistic estimate of total carcinogenic risk for adult males (a), adult females (b), and children (c) based on source apportionment. (d) The carcinogenic risk of each TE for children according to different sources.

uniform distribution in the GIS map (Fig. 4). These results suggest that Cr and Ni mainly originate from natural sources, with little impact from anthropogenic activities. In fact, Cr and Ni generally have been regarded as indicators of lithologic origin because of their wide presence in soil parent materials (Liu et al., 2021). Therefore, Factor 4 was allocated to geogenic sources such as rock weathering and pedogenic processes.

3.4. Health risks assessment

3.4.1. Concentration-specific HRA

All the populations exhibited high levels of CR. The average TCR values for adult males, females, and children were 8.90E-6, 1.01E-5, and 8.60E-5, respectively (Fig. 5a). Meanwhile, TCR values of approximately 97.0% (adult males), 96.9% (adult females), and 100.0% (children) were higher than the acceptable threshold of 1.0E-6. Additionally, nearly 28.7% of the TCR values exceeded the severe risk level of 1.0E-4 for children. These results suggest a high risk of cancer. Among these elements, As exhibited the highest potential carcinogenic risk to all the populations (Fig. S5). Approximately 88.3% (adult males), 87.7% (adult females), and 100.0% (children) of CR values for As were greater than 1.0E-6. In particular, nearly 25.5% of the CR values for As surpassed 1.0E-4 in children.

NCR for all the populations was negligible. The average HI values for adult males, females, and children were 6.28E-2, 7.12E-2, and 2.87E-1, respectively (Fig. 5b). Similar to the carcinogenic risk results, children suffered from a higher NCR than adults. Moreover, only 1.00% (children) and 0.01% (adult females) of the HI values exceeded the threshold of 1. These results suggest that NCR of these TEs barely threatens human health. The mean HQ values of individual TEs for children decreased as follows: As > Cr >

Pb > Ni > Cu > Hg > Cd > Zn, whereas for adults, it decreased as follows: Cr > As > Pb > Cd > Ni > Hg > Cu > Zn (Fig. S6).

3.4.2. Source-specific HRA

The HRA model provided risk levels of contaminants but failed to analyze the contribution of each source to health risk. To compensate for this deficiency, an integrated method was developed by combining the PMF and HRA models (Huang et al., 2021). The results are displayed in Figs. 6-8.

Coal combustion was the dominant source of CR. The change in CR caused by different sources was consistent among different populations, with mean values decreasing as follows: coal combustion source > natural sources > industrial activities > traffic emissions (Fig. 6). For children, the mean CR value of the coal combustion source was 6.34E-5, which was approximately 63 times higher than 1E-6 (the acceptable threshold). Meanwhile, the contribution rates of the four contamination sources to CR decreased in the following order: coal combustion sources (72.7%) > natural sources (20.6%) > industrial activities (3.5%) > traffic emissions (3.2%) (Fig. 8). For specific elements, As showed the highest contribution rates in Factors 2 to 4, with 74.8%, 97.5%, and 83.0% for Factors 2, 3, and 4, respectively. Although the CR value of Factor 1 was mainly contributed by Cd (65.5%), Cr (24.3%), and Pb (10.2%), their contribution to the TCR values was minimal. These results corroborated the view that the health risks were not only related to the content of TEs, but were also closely correlated with the toxicity factor of TEs (Sun et al., 2022).

Similar to the CR results, the most important source of NCR was coal combustion. The change in HI caused by different sources was consistent among the different populations (Fig. 7). In children, the mean HI values of the different sources were 1.64E-1 (coal combustion source), 7.41E-2

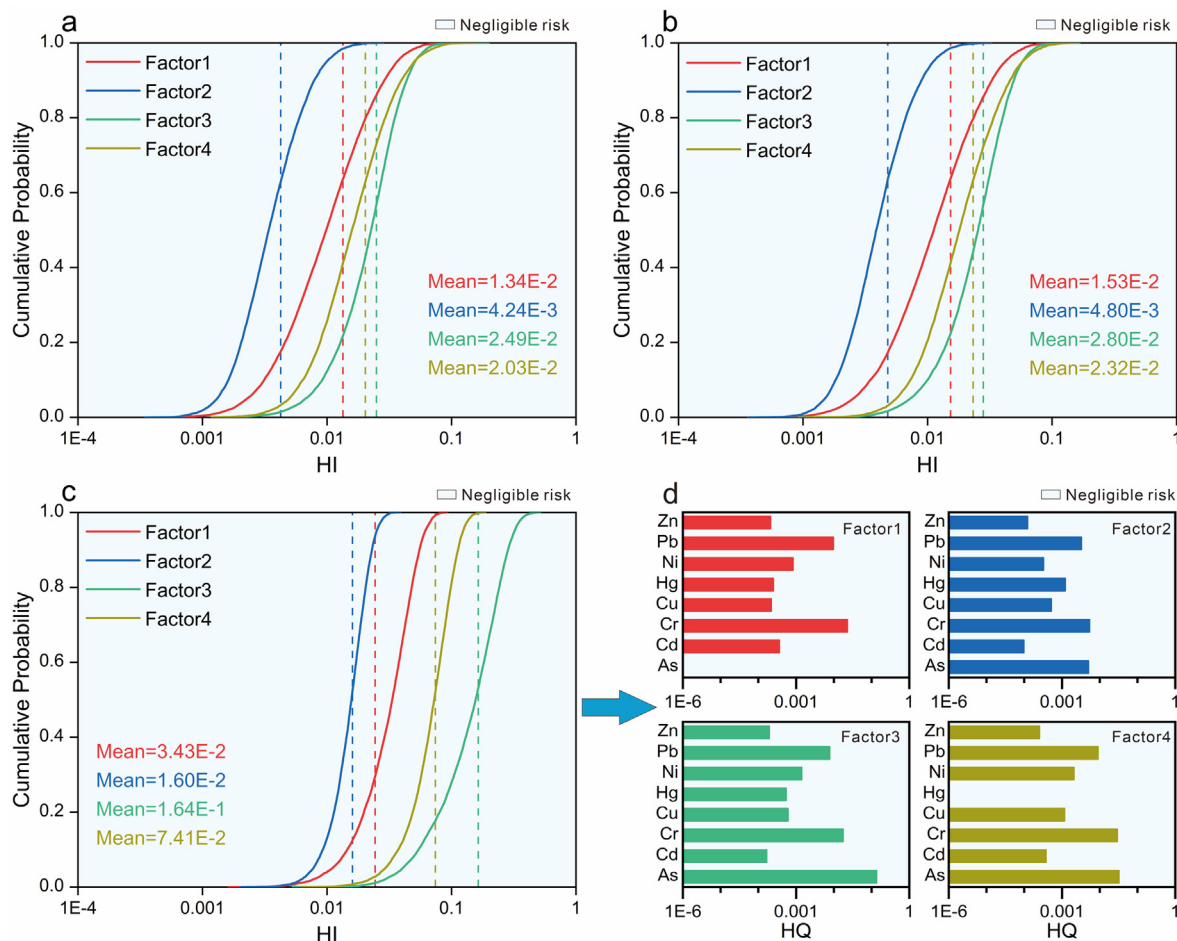


Fig. 7. Cumulative probabilistic estimate of hazard index for adult males (a), adult females (b), and children (c) based on source apportionment. (d) The hazard quotient of each TE for children according to different sources.

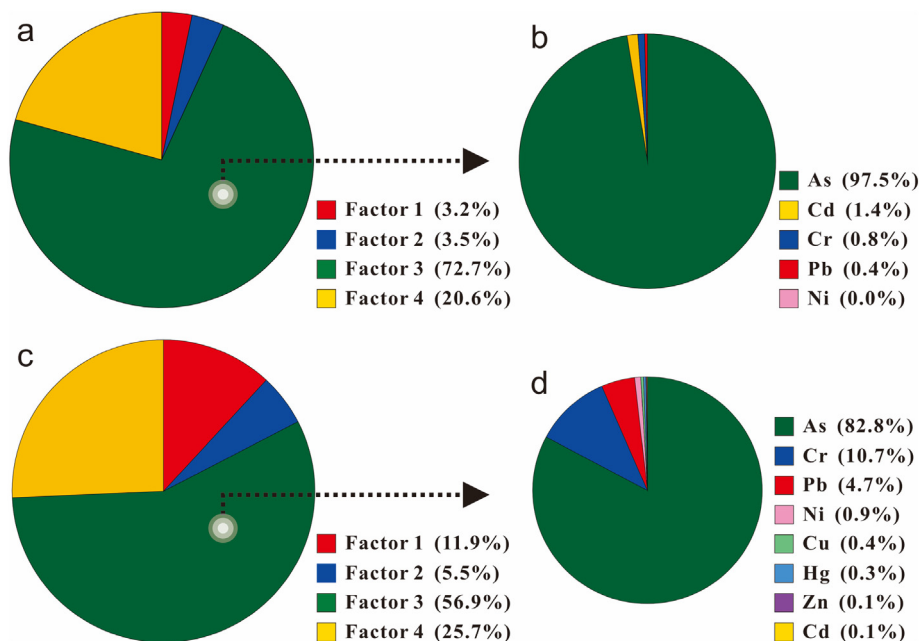


Fig. 8. Contribution percentages of different contamination sources and different TEs to health risk (carcinogenic risk (a and b) and non-carcinogenic risk (c and d)) for children.

(natural sources), $3.43E-2$ (traffic emissions), and $1.60E-2$ (industrial activities). Meanwhile, the contribution rates of the four sources to NCR decreased in the following order: coal combustion source (56.9%) > natural sources (25.7%) > traffic emissions (11.9%) > industrial activities (5.5%) (Fig. 8). The highest contributions to NCR in the coal combustion and natural sources were made by As, and were 82.8% and 43.3%, respectively. Although Cr and Pb mainly contributed to the HQ values in traffic emissions and industrial activities, they contributed the least to NCR.

Coal combustion was identified as the priority contaminant source and As was identified as the priority contaminant. Overall, coal combustion contributed 72.7% to the carcinogenic risk and 56.9% to the non-carcinogenic risk of TEs in park soils. In the coal combustion source, As played the most important role among the different TEs and caused 97.5% of CR and 82.8% of NCR. Chronic As exposure can lead to a variety of adverse consequences, such as cancer of the liver, skin, lungs, prostate gland, and bladder, as well as coronal heart diseases, polyneuropathy, and black foot disease (Li et al., 2012; Matschullat, 2000). In fact, severe endemic arsenism in Guizhou Province has been detected since 1953 owing to the indoor burning of high-arsenic coal (Zhang et al., 2007; Zheng et al., 2005). Approximately 200,000 residents in the three counties of Guizhou Province are at risk of high As exposure from high-As coal combustion (An et al., 2007). In endemic areas, contaminated indoor air and foods such as corn and chili are considered the main sources of As exposure (Li et al., 2012). Although several efforts have been made by the local government to restrict the mining of high-As coal (Wang et al., 2019a), our results highlight that the accumulation of arsenic in soil due to coal combustion still poses a potential threat to human health.

4. Conclusions

The contamination level, source apportionment, and probabilistic health risk assessment of toxic elements in urban park soils were investigated in a typical karst region in China. The results showed that Hg and Cd were at relatively high contamination levels in most parks. Most TEs exhibited strong spatial heterogeneity owing to the influence of different human activities. Four sources governing TE contamination (coal combustion, natural sources, traffic emissions, and industrial activities) were identified, with contribution percentages of 32%, 31%, 20%, and 17%, respectively. A probabilistic health risk assessment showed acceptable non-carcinogenic risks and high levels of carcinogenic risks in all

populations. Based on the source-specific HRA, arsenic from coal combustion was identified as a major contributor to human health risks. Although several efforts have been made by the local government to eliminate coal-borne arsenicosis, our results revealed that the accumulation of arsenic in the soil due to coal combustion poses a great threat to human health.

CRediT authorship contribution statement

Zhenjie Zhao: Conceptualization, Methodology, Resources, Formal analysis, Investigation, Writing - original draft, Writing-review & editing, Visualization, Funding acquisition. **Ming Hao:** Resources. **Yunlong Li:** Investigation. **Shehong Li:** Conceptualization, Supervision, Writing - original draft, Writing-review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.156223>.

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