



Risk Assessment and Source Analysis of Soil Heavy Metal(oid)s Pollution in Beijing, China

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Abstract Investigating heavy metal(oid)s (HMs) in urban soils is vital to safeguarding human life and health. This study investigated HMs in soils in urban areas of Beijing, assessed their ecological and health risks, and combined principal component analysis, geostatistical analysis, and positive definite matrix factor decomposition models to analyze them retrospectively. Although the Cu, Zn, and Pb in the soils of the study area were all below national

quality standards, the average levels were 1.55, 1.17, and 1.21 times higher than the background values for HMs in Beijing soils, respectively, indicating the presence of HMs enrichment in the surface soils. Neither of the nine HMs in this study caused ecological risk. The health risk assessment results show that the nine HMs in this study do not yet pose a non-carcinogenic risk. Ni and Cu have significant carcinogenic effects on humans, and children are more susceptible to carcinogenic risks, with direct ingestion being the main route of HMs exposure. The main sources of HMs pollution in soils are industrial activities, soil-forming matrices and historical sites, which account for 32.89%, 44.16%, and 22.95% of total HMs accumulation respectively. We have found that V, Mn, Co, As, Ni, and Cr are mainly from parent material, Zn and Pb are mainly from industrial emissions, and Cu is from historical remains.

Danyang Meng and Yang Shao contributed equally to this study.

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1 Introduction

Heavy metal(oid)s (HMs) pollution has become one of the most severe soil pollution problems, and has been listed as priority pollution for detection and control. It is difficult to remove HMs pollution by chemical and biological degradation methods because of its long residual period, irreversibility, ease of migration,

toxicity, insidiousness, chemical complexity, and high bioavailability (Bolan et al., 2014; Dai et al., 2018; Yu et al., 2020). HMs can affect human health by entering the body directly through direct ingestion, inhalation and dermal contact, or indirectly through biogeochemical cycles (Mao et al., 2019; Rosell et al., 2009). An excessive amount of Pb can cause neuropathy, affect the endocrine and immune systems, and predispose the body to cancer. The long-term intake of As can cause skin cancer, peripheral vascular disease. An excessive intake of Zn will affect the balance of cholesterol. Cr, As, Pb, Cu, Zn, and Ni have been listed as priority pollutants for control by the United States Environmental Protection Agency (USEPA) and are increasingly concerned in the EU countries and Japan.

Soils act as a temporary sink for HMs, whether they are from artificial or natural sources (Palansooriya et al., 2020). For soil HMs, the spatial distribution is influenced by natural and anthropogenic factors. Natural factors include soil texture, high background values, soil physicochemical properties, rock aging, and surface runoff. Anthropogenic factors mainly include industrial production and emissions, mining and smelting of mineral resources, HMs use of fertilizers and pesticides (Rezapour et al., 2019), traffic emissions, and vehicle abrasion. Since HMs in soils have complex sources, their distribution features can be influenced by a combination of factors and have substantial spatial heterogeneity. Mehr et al. (2017) found that Cr and Ni were mainly derived from natural geological factors by PCA and PMF analyses of HMs in urban soils in Isfahan Province, Iran. Gu et al. (2014) studied that Ni is usually present in alloys and that weathering of urban infrastructure may lead to the enrichment of Ni in soils. Cu, Pb, and Zn in the soil surface layer are mainly derived from vehicle emissions and atmospheric deposition, as indicated by a study on soil HMs in Puning city, Guangdong Province, China (Wang et al., 2019a). Dong et al. (2017) studied that the primary source of As was industrial activities in the soils of Tongliang District, Chongqing. Jahandari et al. (2020) found that Cr can be sourced from mixed anthropogenic sources, in the soils of Shiraz City, Iran.

Studies show that 50% of the global land area is polluted with HMs or metalloids, especially in the developing countries (Quinton & Catt, 2007). According to the National Soil Pollution Survey Bulletin,

the total exceedance rate of HMs in Chinese soil was 16.1%, of which the exceedance rates of Cr, Hg, As, Cu, Pb, Cr, Zn, and Ni were 7.0%, 1.6%, 2.7%, 2.1%, 1.5%, 1.1%, 0.9%, and 4.8% respectively (Ministry of Ecology and Environment of the People's Republic of China and Ministry of Natural Resources of the People's Republic of China, 2014). The surface soils have higher levels of HMs than the deep soils (Dong et al., 2017). The current studies on HMs mainly focus on agricultural soils because of their bio transport effect (Huang et al., 2018a). As living areas, cities have been studied mainly for HMs in atmospheric particulate matter, while the number of studies assessing HMs in urban soils is still low. Rapid economic and population growth has fuelled the expansion of urban built-up land. It is estimated that by 2030, the global urban surface area will increase by 1.2 million square kilometers compared to the beginning of the 21st century (Cambou et al., 2018). By 2050, about two-thirds of the world's 50-year-old population will be living in cities (Edmondson et al., 2014). China has undergone rapid urbanization and industrialization in recent decades due to reform and opening up. The imperfect environmental protection measures in early production and living led to an increase in the release of HMs into the environment and an increase in the concentration of HMs in the surface soil. Rapid urbanization transformed much-contaminated land into commercial, urban parks, and residential land. Zhang et al. (2018) found that the distribution pattern of HMs in the soil of China's provincial capitals is consistent with the famous "Heihe-Tengchong Line" of population, geology, and economic boundaries. Li et al. (2022) showed that health risks due to HMs were distributed similarly to the level of urban development in their study of soils in Wuxi, with high health risk values occurring in the urban center area and decreasing in a circular pattern. HMs pollution in cities has higher human health risks due to their high population density and poses a challenge to the sustainable development and public health of cities (Vega et al., 2022). Compared with the naturally developed land, HMs in urban soils have larger difference of concentration over shorter distances (Li et al., 2013). Urban areas have many functional areas, such as transport, parks, and housing. Furthermore, the sources of HMs are influenced by a combination of factors such as industry, transport, construction, and domestic waste, making the study of HMs in urban

soils more complex than in agricultural soils (Yesilonis et al., 2008).

Beijing has been rated as a first-tier city by the World Urban Research Institute, with a developed tertiary industry, high population density, and congested traffic. Zhang et al. (2018) classify Beijing as one of the nine provincial capitals where HMs should be controlled with priority. A soil contamination survey undertaken by China National Environmental Monitoring Centre (CNEMC) during 2005–2013 revealed that the surface soil in Beijing shows medium pollution with the HMs of As, Pb, and Cu (Chen et al., 2016). A study by Xie et al. (2018) found that Cu, Pb, and Zn were the most accumulated HMs in the residential areas of Beijing. In order to better understand the potential risks and sources of HMs in urban surface soils of Beijing, a sampling study was carried out within the 5th Ring Road area of Beijing. The main objectives are as follows: (1) To find out the current accumulation and distribution conditions of soil HMs in urban areas in Beijing. (2) To evaluate the ecological and human health risks arising by HMs in urban surface soils in Beijing. (3) To identify the sources of HMs in the urban surface soils of Beijing. This study will provide a scientific basis for taking reasonable management measures to control HMs pollution in the surface soils of big cities.

2 Materials and Methods

2.1 Study Area

Beijing (115.7°–117.4°E, 39.4°–41.6°N), located in northern China, is situated in a warm temperate semi-humid region with a continental monsoon climate influenced by the Mongolian high pressure. The summers are hot and rainy, and the winters are cold and dry. The average annual temperature is 12–13°C, and the annual precipitation is 644 mm. The resident population of the central city is about 10.99 million, and the tertiary sector dominates the industrial structure. The main roads in Beijing's urban areas show a circular distribution. Within the 2nd Ring Road is the core area of Beijing, which belongs to the old city of Beijing. The 4th Ring Road is a crucial partition between Beijing's central and non-central areas. According to Beijing's 14th Five-Year Plan, the 5th Ring Road will become an "urban park ring" with many urban green

areas and parks along the 5th Ring Road. The study of HMs content in soils within Beijing's 5th Ring Road is vital for understanding the status of HMs pollution in densely populated areas of Beijing, as well as accumulating relevant data for changes in soil HMs pollution in urban areas of Beijing in response to urban construction and development.

2.2 Sample Collection and Preparation

The samples were collected in October 2021 and April 2022. The locality of the sample sites is shown in Fig. 1. Soil samples were collected in 0–20 cm depths. In order to ensure the homogeneity of the samples, a composite sample was made from 3–5 subsamples within 100 m of the sampling point. The original weight of each sample was more than 1 kg, and all samples were finally placed in cloth bags and brought back to the laboratory. After removing plant roots, leaves, and stones, the sample was placed in a calm, ventilated place and air-dried to constant weight. Grind the sample using an agate mortar and pass it through a 200 mesh sieve.

2.3 Chemical Analysis and Quality Control

(1) Weigh 100 mg of ground and sieved soil samples accurately in a PTFE digestion tube, add 6 ml HNO_3 , 3 ml HCl and 3 ml HF, respectively, and pre-digest for 2 h. (2) Place the anechoic tube with the sample in the microwave anechoic apparatus for anechoic digestion. After the anechoic digestion process, transfer the anechoic solution to a PTFE beaker and wash the anechoic tube three times with 5 ml of 2% mass fraction HNO_3 . Finally, transfer the washing solution together to the PTFE beaker. (3) Transfer the PTFE beaker to a hotplate and heat it at 180°C. Add 1–2 drops of H_2O_2 during the acid removal process in 2–3 min until approximately 1 ml of digestion solution remains. Dilute the concentrated digestion solution with 2% HNO_3 to a concentration suitable for the assay. Filter the diluted digestion solution with filter heads of 0.45 μm pore size to remove incompletely digested particulate matter.

The concentration of HMs (V, Mn, Co, Ni, Cr, As, Cu, Zn, Pb) was quantified using inductively coupled plasma mass spectrometry (ICP-MS, Thermo-X7, Thermo Fisher, USA). For the quality guarantee of analysis, standard reference soil materials (GBW

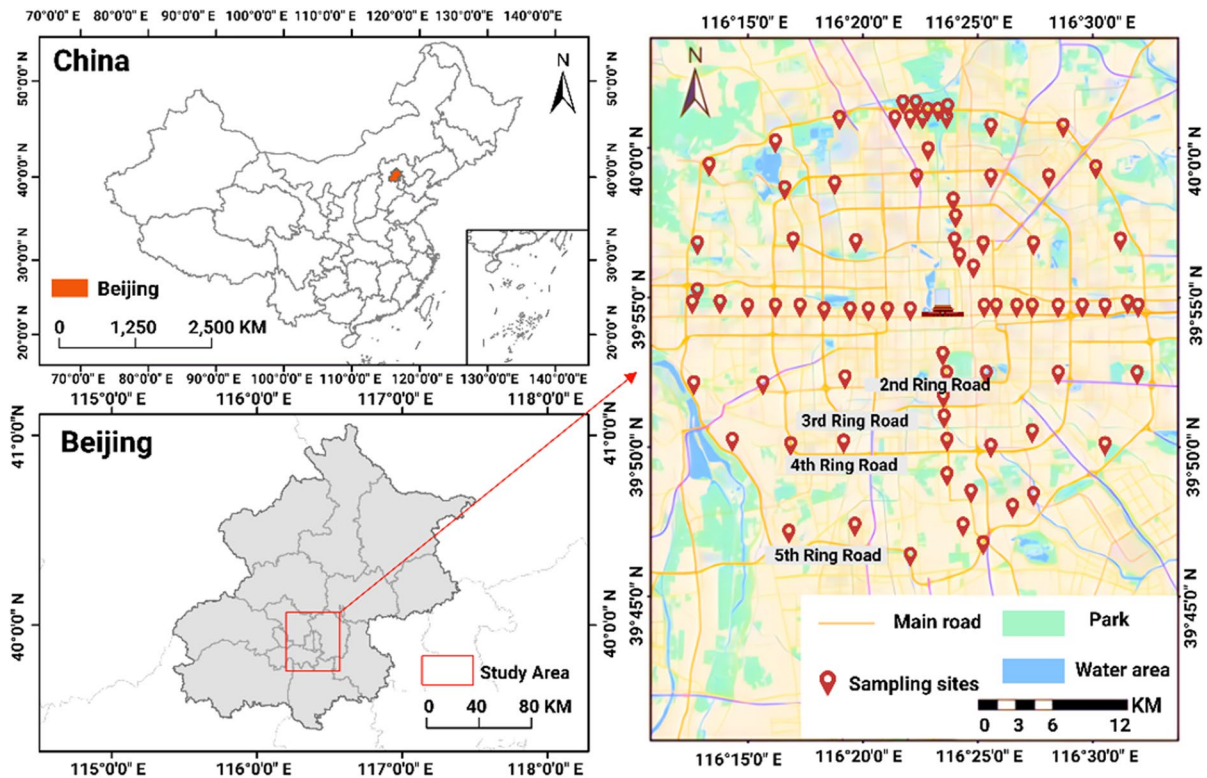


Fig. 1 Study area and sampling sites

08302) from the National Institute of Metrology of China were used to verify the accuracy of elemental analysis. Apparent recovery of V, Cr, Mn, Co, Ni, Cu, Zn, As, and Pb were 110–120%, 95%–107%, 101–111%, 96–105%, 87–97%, 91–124%, 90–108%, 94–119%, and 82–103%, respectively. To ensure the accuracy of the analytical results, each batch of samples included three reagent blanks and randomly selected 10% of the total sample size to prepare parallel samples with a relative standard deviation (RSD) of less than 10%.

2.4 Methods of Heavy Metal(oid)s Pollution Assessment

2.4.1 Single-Factor Pollution Index

Using the Single-Factor Pollution Index (P_i) method for pollution assessment of a single HMs, the equation is as follows (Zhao et al., 2020):

$$P_i = C_i/S_i$$

P_i is the environmental quality index of pollution in soil, C_i is the measured mass fraction of pollution ($\text{mg}\cdot\text{kg}^{-1}$), and S_i is the evaluation standard value of pollution. This study uses the background values of HMs in Beijing (CNEMC, 1990) as the standard for evaluating HMs accumulation and pollution.

2.4.2 Nemeru Composite Pollution Index

The Nemeru composite pollution index (P_N) reflects the overall contamination status of HMs in the soil and can highlight the role of seriously contaminating HMs pollutants. The expressions are as follows (Zhao et al., 2020):

$$P_N = \sqrt{\frac{1}{2}(P_{iMax}^2 + P_{iAve}^2)}$$

$$P_{iAve} = \frac{1}{n} \sum_{i=1}^n P_i$$

P_N is the combined pollution index of the sampling sites; P_{iMax} is the maximum value of the single pollution index of the sampling sites; P_{iAve} is the average of the single pollution index of the sampling sites. This study uses the background values of HMs in Beijing (CNEMC, 1990) as the standard for evaluating HMs accumulation and pollution. Pollution levels were classified into five levels based on the Nemerov combined pollution index (Zhao et al., 2019):

- $P_N \leq 0.7$, safety;
- $0.7 < P_N \leq 1.0$, alarming;
- $1.0 < P_N \leq 2.0$, lightly pollution;
- $2.0 < P_N \leq 3.0$, moderate pollution;
- $P_N > 3.0$, heavy pollution.

2.4.3 Index of Geoaccumulation

The geoaccumulation index (Igeo) considers natural geological processes and human activities contributing to HMs pollution. This parameter is vital in identifying the impact of human activities (Muller, 1969). It is calculated using the following expression (Liu et al., 2019):

$$I_{geo} = \log_2 \left(\frac{C_n}{k B_n} \right)$$

C_n is the concentration of HMs n in the sample; B_n is the geochemical background value of HMs n in the study area; k is the correction index ($k = 1.5$), which is often used to characterize sedimentary features, rock geology, and other effects to minimize the possible impact of changes in background values (Doabi et al., 2018). This study uses the background values of HMs in Beijing (CNEMC, 1990) as the standard for evaluating HMs accumulation and pollution.

The Geoaccumulation Index can be divided into seven classes representing different pollution levels (Keshavarzi et al., 2021; Wang et al., 2019a, b). $I_{geo} < 0$, the pollution level is 0, indicating no pollution; $0 \leq I_{geo} < 1$, the pollution level is 1, indicating no pollution to moderate pollution; $1 \leq I_{geo} < 2$, the pollution level is 2, indicating moderate pollution; $2 \leq I_{geo} < 3$, the pollution level is 3, indicating moderate pollution to intense pollution; $3 \leq I_{geo} < 4$, the pollution level is 4, indicating intense pollution; $4 \leq I_{geo} < 5$, the pollution level is 5, indicating strong

pollution to extreme pollution; $I_{geo} \geq 5$, the pollution level is 6, indicating extreme pollution.

2.4.4 Potential Ecological Risk Assessment

The potential ecological risk index (RI) (Hakanson, 1980) is proposed based on HMs' nature and environmental behavior characteristics, considering the toxicity of different HMs, environmental sensitivity to HMs pollution, HMs content, etc. It is widely used in environmental risk assessment and is calculated as follows (Chen et al., 2021; Wang et al., 2020):

$$E_i = T_r^i (C_i / C_0)$$

$$RI = \sum E_i$$

C_i is the measured concentration of the HMs element; C_0 is the relevant background concentration of the HMs element; T_r^i is the toxicity factor of a particular HMs element ($As = 10$, $Co = Pb = Cu = Ni = 5$, $V = Cr = 2$, $Zn = Mn = 1$) (Hakanson, 1980); E_i is the potential ecological hazard factor of HMs; RI is the combined potential ecological hazard of multiple HMs at a point in the sediment index. This study uses the background values of HMs in Beijing (CNEMC, 1990) as the standard for evaluating HMs accumulation and pollution. Potential ecological risks can be classified into five classes:

- $E_i \leq 40$, $RI \leq 90$ is a low potential ecological risk.
- $40 < E_i \leq 80$, $90 < RI \leq 180$ is moderate potential ecological risk;
- $80 < E_i \leq 160$, $180 < RI \leq 360$ is considered strong potential ecological risk;
- $160 < E_i \leq 320$, $360 < RI \leq 720$ is very high potential ecological risk;
- $E_i > 320$, $RI > 720$ is an extremely high potential ecological risk.

2.4.5 Health Risk Assessment

Human health risk values can assess and predict the potential adverse health risks to humans when exposed to specific contaminants (Agyeman et al., 2021). The health risk assessment model recommended by the US EPA is used to assess the health risks caused by HMs in the study area (USEPA, 1997). Carcinogenic and non-carcinogenic risks are quantified based on HMs' average daily dose (ADI)

via different exposure routes (direct ingestion, inhalation, dermal contact). The average daily intakes were calculated as shown below (Han et al., 2016):

$$ADI_{ing} = \frac{C \times IR \times EF \times ED \times CF}{BW \times AT}$$

$$ADI_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT}$$

$$ADI_{der} = \frac{C \times SA \times ABS \times AF \times EF \times ED \times CF}{BW \times AT}$$

ADI_{ing} , ADI_{inh} , and ADI_{der} are the average daily intake of soil HMs by the direct ingestion, inhalation, and dermal contact routes, respectively; C is the soil HMs concentration ($\text{mg}\cdot\text{kg}^{-1}$); IR is the daily soil intake (children: $200 \text{ mg}\cdot\text{day}^{-1}$, adults: $100 \text{ mg}\cdot\text{day}^{-1}$); EF is the exposure frequency ($365 \text{ day}\cdot\text{year}^{-1}$); ED is exposure duration (children: 6 years, adults: 24 years); CF is concentration conversion factor (10^{-6}); BW is body weight (children: 23.97 kg, adults: 57.9 kg); AT is the average life expectancy, $AT=70 \times 365$ when applied to carcinogenic risk calculations, $AT=24 \times 365$ when applied to non-carcinogenic risk calculations for adults, and $AT=6 \times 365$ when applied to non-carcinogenic risk calculations for children; $InhR$ is daily soil inhalation rate (children: $7.5 \text{ m}^3\cdot\text{day}^{-1}$, adults: $14.5 \text{ m}^3\cdot\text{day}^{-1}$); PEF : particle emission factor ($1.36 \times 10^9 \text{ m}^3\cdot\text{kg}^{-1}$); SA is skin exposure area (children: 2448 cm^2 , adults: 5075 cm^2); ABS is skin absorption factor (0.001); AF is skin adhesion factor (children: $0.2 \text{ mg}\cdot\text{cm}^{-1}\cdot\text{day}^{-1}$, adult: $0.07 \text{ mg}\cdot\text{cm}^{-1}\cdot\text{day}^{-1}$). Non-carcinogenic risks are assessed using the following equation (Wang et al., 2022):

$$HI = \sum HQ_i = \sum \frac{ADI_i}{R_f D}$$

Hazard index (HI) represents the non-carcinogenic risk due to multiple routes; hazard entropy (HQ) is the non-carcinogenic risk from a single route; $R_f D$ is the reference dose, which is the maximum dose per unit of time per unit of weight required to avoid adverse effects when absorbed; and i represents the route of intake. The values of $R_f D$ are shown in Table 1. If $HI > 1$, it indicates that the metal may pose a non-carcinogenic risk to public health (Kharazi et al., 2021). Carcinogenic risk can be assessed using the following equation (Kharazi et al., 2021):

$$TCR = \sum CR_i$$

$$CR_i = ADI_i \times SF_0$$

CR is the value of carcinogenic risk, and SF_0 is the carcinogenic slope factor. The values of SF_0 are shown in Table 1. If $CR < 1 \times 10^{-6}$, it means no significant carcinogenic risk; $1 \times 10^{-6} < CR < 1 \times 10^{-4}$, it means the carcinogenic risk is tolerable; if $CR > 1 \times 10^{-4}$, it indicates high carcinogenic risk.

3 Results and Discussion

3.1 Concentrations and Spatial Distribution of Heavy Metal(oid)s

The HMs contents in the study area soils are shown in Table 2. In the study area, the mean levels of

Table 1 References dose and slope factor for HMs

HMs	$R_f D$ ($\text{mg}\cdot\text{kg}^{-1}\cdot\text{day}^{-1}$)			SF_0 ($\text{kg}\cdot\text{day}^{-1}\cdot\text{mg}^{-1}$)		
	Direct ingestion	Inhalation	Dermal	Direct ingestion	Inhalation	Dermal
As	3.00E-04 (Adimalla, 2020a)	4.29E-06 (Yang et al., 2022)	1.23E-04 (Adimalla, 2020a)	1.50 (Adimalla, 2020a)	15.1 (Yang et al., 2022)	3.66 (Adimalla, 2020a)
Cr	3.00E-03 (Adimalla, 2020a)	2.86E-05 (Adimalla, 2020a)	6.00E-05 (Huang et al., 2021)	0.50 (Huang et al., 2022)	4.20E+01 (Adimalla, 2020a)	2.00E+01 (Adimalla, 2020a)
Cu	4.00E-02 (Adimalla, 2020a)	4.02E-02 (Huang et al., 2021)	1.20E-02 (Adimalla, 2020a)	1.70 (Kan et al., 2021)	–	42.5 (Kan et al., 2021)
Pb	3.50E-03 (Huang et al., 2021)	3.52E-03 (Adimalla, 2020a)	5.25E-04 (Xiao et al., 2017)	8.50E-03 (Adimalla, 2020a)	0.042 (Yang et al., 2022)	–
Zn	3.00E-01 (Adimalla, 2020a)	0.30 (Adimalla, 2020a)	6.00E-02 (Adimalla, 2020a)	–	–	–
Ni	2.00E-02 (Adimalla, 2020a)	9.00E-05 (Zhang et al., 2020)	5.40E-03 (Adimalla, 2020a)	1.70 (Adimalla, 2020a)	0.84 (Yang et al., 2022)	4.25E+01 (Adimalla, 2020a)

Table 2 Descriptive statistical analysis of soil HMs

Metals	Min	Max	Mean	SD	CV (%)	Swness	Krtosis	K-Sp	BV	SV
V	53.84	111.63	79.15	14.31	18.08	0.33	-0.71	0.098	79.2	752
Cr	44.26	124.59	64.74	13.18	20.36	1.49	4.79	0.186	68.1	n/a
Mn	309.39	999.46	594.56	104.41	17.56	0.47	2.04	0.2	705	n/a
Co	7.34	16.71	11.12	2.04	18.35	0.54	-0.06	0.2	15.6	70
Ni	19.01	44.06	28.76	5.42	18.85	0.65	0.32	0.03(0.2)	29	900
Cu	20.1	161.59	36.58	19.46	53.20	4.41	24.55	0	23.6	18000
Zn	50.77	282.49	119.99	43.59	36.33	1.07	2.15	0.2	102.6	n/a
As	5.57	14.74	8.55	1.54	18.01	1.07	2.42	0.026(0.2)	9.7	60
Pb	16.74	121.41	30.73	16.9	55.00	3.33	13.46	0	25.4	800

BV: Beijing soil HMs background value; SV: screening value (GB 36600–2018 Soil Environmental Quality Soil Contamination Risk Control Standards for Construction Land, Trial Implementation)

HMs of V, Cr, Mn, Co, Ni, Cu, Zn, As, and Pb were 79.15 ± 14.31 , 64.74 ± 13.18 , 594.56 ± 104.41 , 11.12 ± 2.04 , 28.76 ± 5.42 , 36.58 ± 19.46 , 119.99 ± 43.59 , 8.55 ± 1.54 , and 30.73 ± 16.90 mg·kg⁻¹. The mean contents of Cu, Zn, and Pb exceeded the background values of soil HMs in Beijing, which were 1.55, 1.17, and 1.21 times higher than the background values, respectively. The screening value recommended by the Soil Environmental Quality Standards for Development Sites in China (GB36600-2018) is the standard value of soil pollutants that exceed the value that may cause harm to human health (no standard for zinc, manganese, and total chromium). The HMs concentrations at all sampling sites in the study area

were lower than the screening values. We compared the concentrations of HMs in this study with similar studies in China and other countries, and the results are shown in Table 3. The average concentrations of Cu and Zn in the study area were higher than those of HMs in the urban soils of Wuxi City (Cu = 25.50 mg·kg⁻¹ and Zn = 90.2 mg·kg⁻¹) (Li et al., 2022). The average concentrations of Cr, Ni, Cu, Zn, and As were higher than the corresponding HMs concentrations in the soils of Xiamen City and Tangshan City (Huang et al., 2018b; Sun et al., 2019). The average concentrations of Pb and As in the study area were similar to those in the urban soils of Hangzhou (Fei et al., 2018). The average concentration of Cr, Ni, Cu, Zn,

Table 3 Comparison HMs concentrations in this study with similar studies in China and other countries (mg·kg⁻¹)

		Cr	Mn	Co	Ni	Cu	Zn	As	Pb
Wuxi, China (Li et al., 2022)	Mean	72.9	851.8	14.2	38.2	25.5	90.2	-	37.6
	Max	99.00	1210	17.9	67.5	37.2	146	-	66.4
Hangzhou, China (Fei et al., 2018)	Mean	52.90	-	-	-	-	-	8.99	31.66
	Max	104.00	-	-	-	-	-	43.20	62.60
Xiamen, China (Huang et al., 2018a, b)	Mean	41.77	-	-	-	23.26	-	5.82	37.35
	Max	972	-	-	-	563	-	18	173
Tangshan, China (Sun et al., 2019)	Mean	36.98	-	-	16.81	22.42	70.31	5.89	22.93
	Max	64.64	-	-	33.38	64.54	136.50	11.30	47.26
India (32 cities) (Adimalla, 2020b)	Mean	412.28	-	-	370.69	159.78	437.44	38.61	77.25
	Max	3194.80	-	-	7163.2	1684.35	3591.35	400.35	665.75
Ancona, Italy (Serrani et al., 2022)	Mean	45.60	-	18.1	50.9	63.9	199.1	-	97.4
Tehran, Iran (Rezayani et al., 2022)	Mean	30	-	-	31	42	138	10.4	46
	Max	65	-	-	73	62	160	15.5	81
Ulaanbaatar, Mongolia (Chonokhuu et al., 2019)	Mean	16.56	-	-	21.26	-	106.11	28.04	43.11

and Pb in the study area was lower than the average concentration of 32 cities in India (Adimalla, 2020b). The level of Cr is 2.16 times higher than that of Tehran, the capital of Iran, and the levels of Ni, Cu, Zn, As, and Pb are lower than those of Tehran (Rezayani et al., 2022). The concentrations of Co, Cu, Ni, Pb, and Zn are all lower than the average concentrations in the central Italian city of Ancona (Co=18.1 mg·kg⁻¹, Cu=63.9 mg·kg⁻¹, Ni=50.9 mg·kg⁻¹, Pb=97.4 mg·kg⁻¹, and Zn=199.1 mg·kg⁻¹) (Serrani et al., 2022). Overall, Beijing has relatively low levels of HMs in its soils compared with other cities.

The coefficient of variation (CV) is widely used to describe the degree of global variation in the variable under study, and its value is related to the activity of the human being. CV > 35% indicates a high degree of variation, CV < 15% < 35% indicates a moderate degree of variation, and CV ≤ 15% indicates a low degree of variation (Wang et al., 2019b). The coefficients of variation (CV) of HMs in the study area are Mn < As < V < Co < Ni < Cr < Zn < Pb in order, among which Cu, Zn, and Pb exhibit higher coefficients of variation (CV > 35%). Liu et al. (2016) in 2016 also found high spatial heterogeneity of Cu, Zn, and Pb in urban soils in Beijing, indicating that Cu, Zn, and Pb are mainly influenced by external anthropogenic influences such as traffic and industry.

By using Arcgis to do ordinary kriging interpolation of the soil HMs content data in the study area, an interpolation map was obtained that could reflect the spatial distribution of soil HMs in the study area (Guan et al., 2018). The Kriging interpolation method requires the data to follow a normal distribution to ensure the accuracy of the estimation because high skewness and outliers can jeopardize the spatial continuity of the variational function and affect the prediction accuracy. The K-S test is used to check the normal distribution of the dataset, and the K-Sp > 0.05 indicates that the data conforms to the normal distribution, and the logarithmic transformation is used for the data that does not conform to the normal distribution. In this study, we have used kriging interpolation to create a contamination map to reflect the spatial distribution of nine metals in the urban surface soil of Beijing (Fig. 2). The HMs of V, Mn, and Co have similar spatial distribution patterns, which indicates that these three HMs may have the same source. Hotspots for the HMs of V, Co, and Mn exist in the northwest, southwest, and southeast of the 3rd to 5th Ring

Road. The southwest and northwest areas are close to the Yanshan Mountain Range and Taihang Mountain Range, and the coefficients of variation for V, Co, and Mn are relatively small, so it is tentatively determined that V, Mn, and Co are influenced by the natural environment such as the soil-forming parent material. A similar spatial distribution pattern was observed with Cr and Ni. In the study area, there are high concentrations of Cr and Ni in the western and southeastern directions within the fourth to fifth ring areas, similar to the results of Liu et al. (Liu et al., 2020) for HMs in Beijing's parks. The spatial distribution patterns of Cr and Ni are identical to those of V, Co, and Mn, indicating that natural environmental factors may also influence the sources of Cr and Ni. There are pollution hotspots for Ni, As, Cr, and Zn in the West 5th Ring Road area (Shijingshan District), where giant steel smelters existed before 2004. Coal combustion can produce As and Cr easily (Wang et al., 2016), Ni can enhance the toughness and plasticity of steel (Shin et al., 2000), and ZnO is used for the corrosion protection of carbon steel (Arukalam et al., 2016). Moreover, the wind direction in Beijing is predominantly from west to east, so that steel refining may be the main reason for HMs Ni, As, Cr, and Zn pollution hotspots in the western region. Pollution hotspots of Ni, As, Cr, Zn, Pb, and Cu also exist in the southeastern part of Chaoyang District, which was once an area for chemical industries. In the 20th century, several factories were established in the southeast of Chaoyang District, including chemical plants, coking plants, and glass factories. Such chemical activities may have led to the contamination of the study area with HMs Zn, Pb, and Cu (Yuan et al., 2014). In preparation for the 2008 Beijing Olympic Games, most factories or plants were closed or relocated and have now become an urban renewal area surrounded by residential and commercial areas. Tangshan City, located southeast of Beijing, is a typical site of steel smelting. Thus, industry may be responsible for HMs pollution hotspots in the southeastern part of the study area. High pollution hotspots of As, Cu, Zn, and Pb in the urban center within the 3rd Ring Road may be due to historical sites (Wang et al., 2012a), early urban development in the city center, and high population and traffic flows (Wang et al., 2012b). Pollution hotspots for Pb are also present in Chaoyang Park to the northeast of the study area, with high population flow and dense traffic.

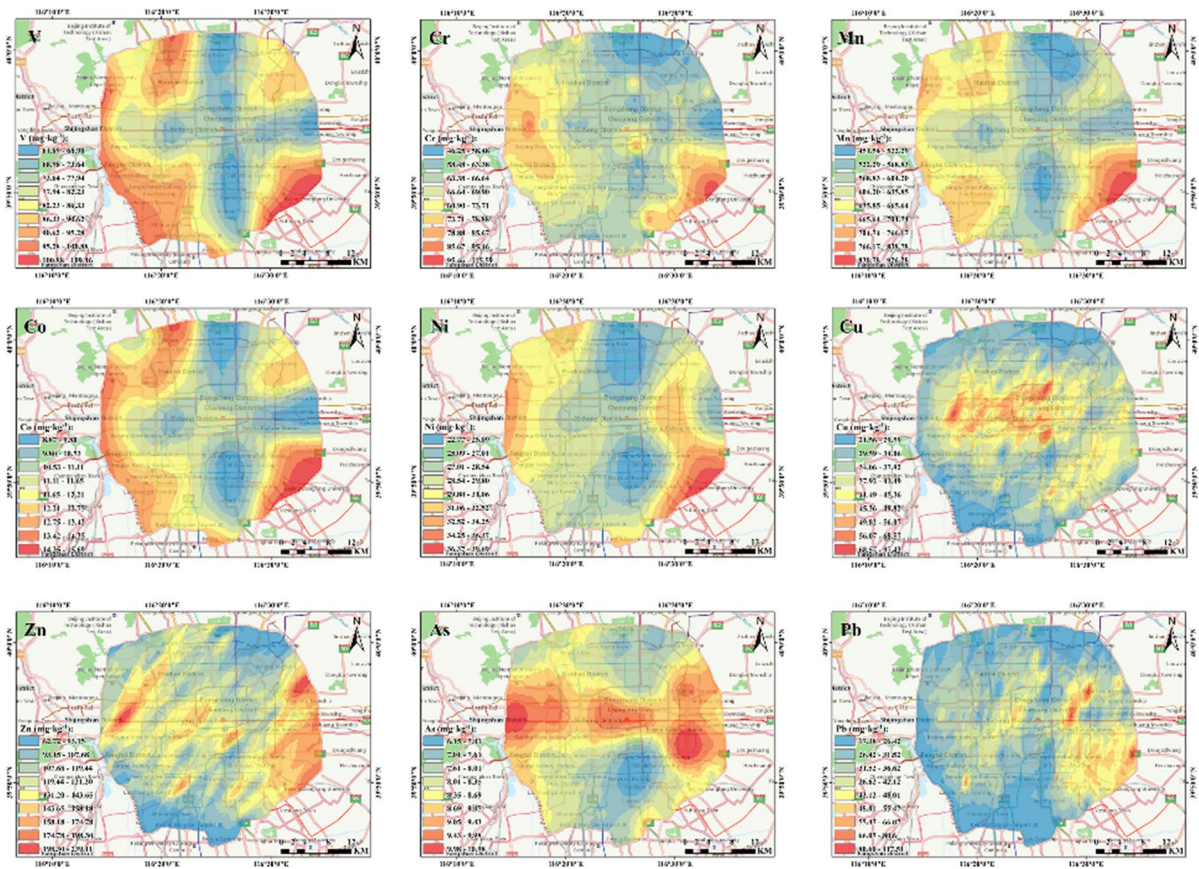


Fig. 2 Spatial distribution of soil HMs concentrations in the study area

3.2 Evaluation of Heavy Metal(oid)s Pollution in Soils

The background values of soil HMs in Beijing were used as guiding indicators for HMs accumulation and pollution assessment. The proportion of different pollution levels of soil HMs in the sampling area was analyzed using the soil HMs single factor pollution index and the Nemerow integrated pollution index (Tables 4 and 5). According to the average of the single-factor pollution index, the degree of contamination of soil HMs in the study area was $Cu > Pb > Zn > V > Ni > Cr > As > Mn > Co$. The average single factor pollution index of Cu, Zn, and Pb in the study area exceeded 1, and Cu was the most polluted, similar to the study by Liu et al. (2016). In the study area, the proportions of HMs exceeded the standard in the following order: $Cu > Zn > Pb > Ni > V > Cr > As > Mn > Co$, indicating that there are differences in the pollution levels of different HMs. The results of the Nemerow Integrated Pollution Index (Table 5) show

that 82.28% of the sampling points in the study area have exceeded the Warning Value, of which 72.15% were lightly polluted, 6.33% were moderately polluted, and 3.80% were heavily polluted, which reveals the extensive accumulation of HMs in the study area.

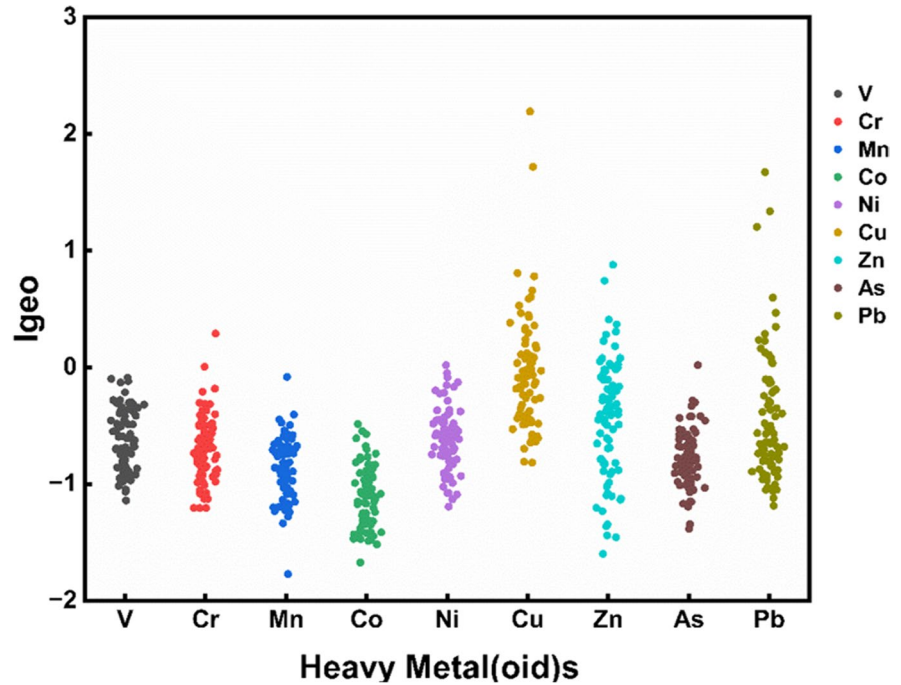
Table 4 Evaluation results of the single factor pollution index for soil HMs in study area

Metals	Min	Max	Mean	Ration (%)
V	0.68	1.41	1.00	44.30
Cr	0.65	1.83	0.95	31.65
Mn	0.44	1.42	0.84	10.13
Co	0.47	1.07	0.71	3.80
Ni	0.66	1.52	0.99	48.10
Cu	0.85	6.85	1.55	89.87
Zn	0.49	2.75	1.17	63.29
As	0.57	1.52	0.88	20.25
Pb	0.66	4.78	1.21	50.63

Table 5 Evaluation results of the Nemer integrated pollution index for soil HMs in study area

Pollution levels	Clean	Precautionary	Slightly polluted	Moderately polluted	Heavily Polluted
	$PN \leq 0.7$	$0.7 < PN \leq 1.0$	$1.0 < PN \leq 2.0$	$2.0 < PN \leq 3.0$	$PN > 3.0$
%	0	17.72	72.15	6.33	3.80

Fig. 3 The result of geoaccumulation index



The results of the geoaccumulation index evaluation show that there is local accumulation of HMs of Cu and Pb in the study area (Fig. 3).

3.3 Ecological and Health Risk Assessments

As shown in Table 6, the results of the ecological risk assessment for HMs show that none of the nine HMs present ecological risks in the study area. Although Beijing has developed at a highly rapid rate over the last 30 years, resulting in the accumulation of HMs in the soil. However, in the last 5 years, the local government has effectively controlled urban soil pollution by implementing a comprehensive strategy to prevent and regulate HMs pollution (Pan et al., 2018).

Non-carcinogenic exposures in adults shows the order: Cr > As > Pb > Ni > Cu > Zn (Fig. 4a); non-carcinogenic exposures in children shows the order: As > Cr > Pb > Ni > Cu > Zn (Fig. 4b). Therefore, As and Cr were the significant contributors to the

non-carcinogenic risk in the study area. The mean value of the total non-carcinogenic risk index in the sampling area was less than 1, indicating that HMs in

Table 6 Results of the ecological risk assessment of HMs in the study area

Metals	The level of potential ecological risk (%)				
	Low	Moderate	Considerable	Very high	Extremely high
V	100	0	0	0	0
Cr	100	0	0	0	0
Mn	100	0	0	0	0
Co	100	0	0	0	0
Ni	100	0	0	0	0
Cu	100	0	0	0	0
Zn	100	0	0	0	0
As	100	0	0	0	0
Pb	100	0	0	0	0

Fig. 4 Contributions to non-carcinogenic risks from different HMs in the study area (a adults; b children)

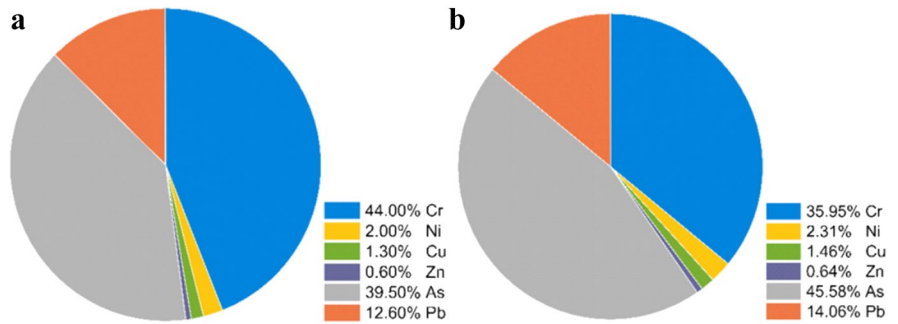


Table 7 Carcinogenic and non-carcinogenic risks of HMs by different routes of intake

	Non-carcinogenic risk (HI)		Carcinogenic risk (CR)	
	Mean for Adults	Mean for Children	Mean for Adults	Mean for Children
Direct Ingestion	1.06E-01	5.14E-01	2.70E-04	1.31E-03
Dermal	2.13E-02	8.70E-03	7.19E-05	2.93E-05
Inhalation	8.44E-04	1.06E-03	5.29E-07	6.61E-07
Total	1.29E-01	5.24E-01	3.43E-04	1.34E-03

the study area do not yet pose a non-carcinogenic risk. The non-cancer risk results for the different routes of intake are shown in Table 7. The order of exposure pathways of both adults and children were ranked as: direct ingestion > dermal > inhalation. The total non-carcinogenic risk values for adults and children were 0.13 and 0.52, respectively, indicating that children are more susceptible to non-carcinogenic risks. This result may be related to specific behavioral patterns of children, such as pica, finger sucking, and high respiratory rates (Kan et al., 2021).

Figure 5 shows the carcinogenic risks of the different HMs. Carcinogenic risk exposures in adults shows the order: Cu > Ni > Cr > As > Pb. For children, the mean values of the individual carcinogenic risk indices of Cr, Ni, Cu, and As were higher than 1×10^{-4} , indicating a high carcinogenic risk for children with these four HMs. For adults, the mean values of the individual carcinogenic risk indices of Ni and Cu were greater than 1×10^{-4} , indicating that the HMs of Ni and Cu pose a high carcinogenic risk to adults. Therefore, Ni and Cu in the study area soils have significant carcinogenic effects on humans. As shown in Table 7, the carcinogenic risk results for the different intake routes were direct ingestion > dermal > inhalation, indicating that the main route of HMs exposure was direct ingestion. The average carcinogenic risk values for all routes of intake and HMs showed that children > adults,

indicating that children are more susceptible to carcinogenic risks, consistent with non-carcinogenic risks.

3.4 Analysis of Heavy Metal(oid)s Sources

3.4.1 Principal Component Analysis

The sources of HMs in the study area were studied using principal component analysis (PCA), as shown in Table 8. The KMO value of 0.809 is above the recommended critical threshold of 0.5, which indicates a

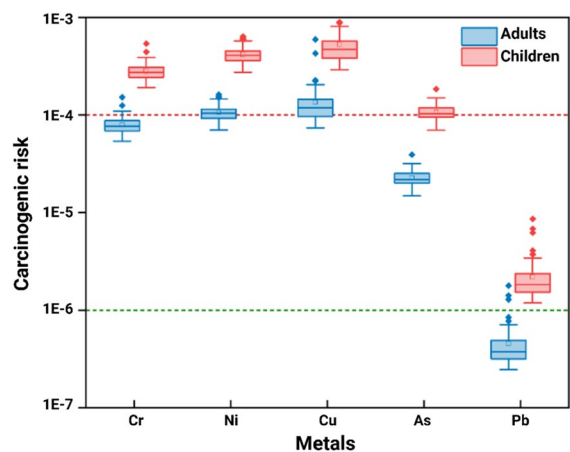


Fig. 5 Carcinogenicity risk index of HMs in the study area

Table 8 The results of the principal component analysis of soil HMs in the study area

Metals	PC 1	PC 2	PC 3
V	0.948	0.009	-0.106
Cr	0.752	0.35	0.163
Mn	0.899	0.233	-0.022
Co	0.963	0.068	-0.033
Ni	0.808	0.194	-0.01
Zn	0.118	0.69	0.448
Cu	-0.057	0.053	0.936
As	0.406	0.411	0.041
Pb	0.121	0.847	-0.085

strong correlation between the variables. The three principal components can reflect 75.08% of the information on the variation of HMs. Factor 1 for soil HMs captures V, Cr, Mn, Co, Ni, and As, with As being influenced by both factor 1 and factor 2. These metals are mainly derived from the soil parent material (Ali et al., 2016;

Jin et al., 2019; Wu et al., 2021), so factor 1 represents the influence of natural factors. A study by Peng et al. (Peng et al., 2013) on HMs in soils in residential areas within the fifth ring area of Beijing found that urbanization did not influence the accumulation of Cr and Ni and was mainly caused by natural background fluctuations. Factor 2 for soil HMs captures As, Pb, and Zn. Factor 3 for soil HMs captures Zn and Cu. Pb, Cu, and Zn mainly came from anthropogenic sources such as traffic pollution, industrial emissions, and building construction (Gu et al., 2016; Yadav et al., 2019). Cai et al. (2013) found that Pb, Cu, and Zn in the surface soils of Guangzhou were strongly influenced by human activities. Therefore, factor 2 and factor 3 may represent two different sources of anthropogenic pollution.

3.4.2 Source apportionment by PMF

PMF model were used in this study to identify the sources and quantify the contribution of HMs in the soils of the study area (Figs. 6 and 7). Input data for

Fig. 6 Factor profiles and source contributions of HMs from the PMF model

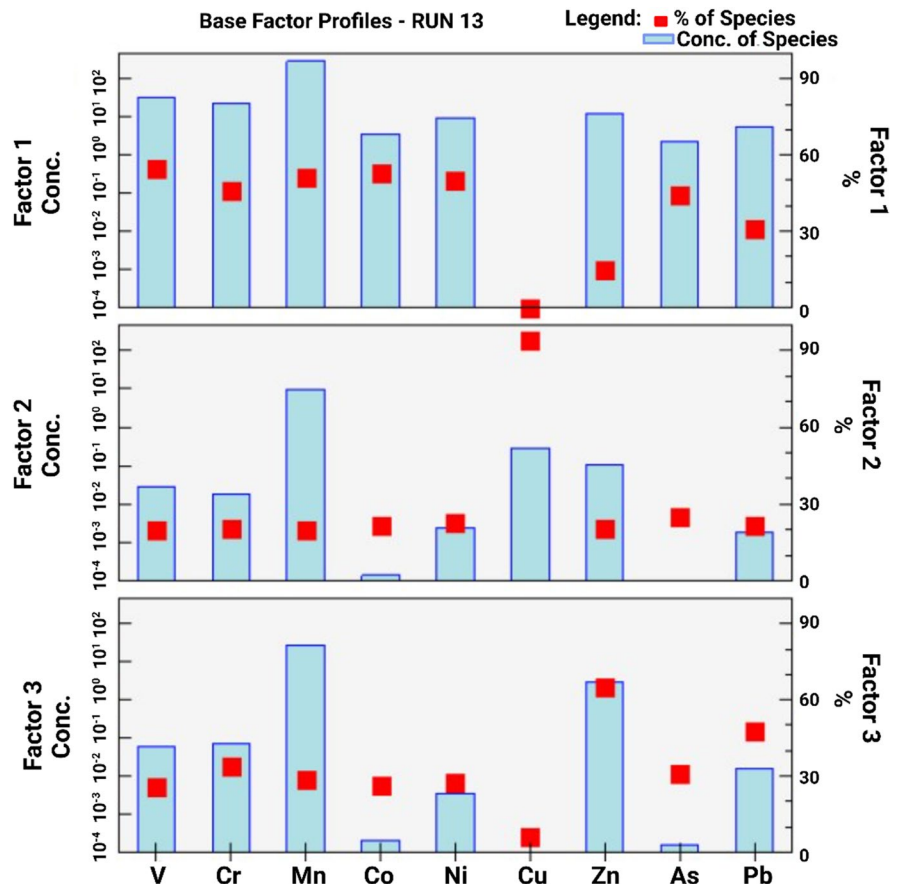
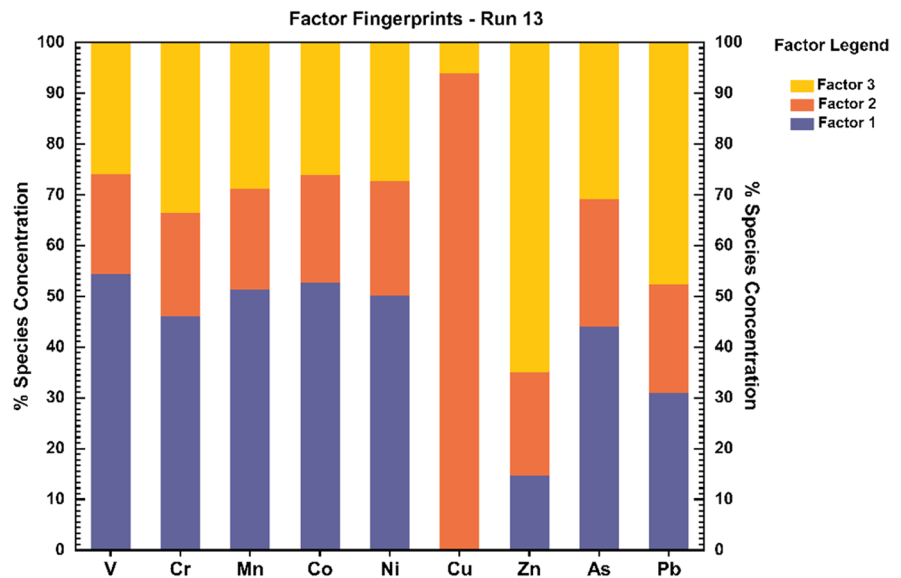


Fig. 7 Attribution of different pollution sources to HMs in the study area



nine HMs from 79 soil samples and the uncertainty data associated with them into the software PMF 5.0. Factor 1 accounts for 44.16% of the total contribution. The contribution rates for HMs V, Cr, Mn, Co, Ni, and As in Factor 1 are 54.47%, 46.11%, 51.37%, 52.82%, 50.24%, and 44.16%, respectively. The mean levels of HMs in V, Cr, Mn, Co, Ni, and As were lower than the background values for soil HMs in Beijing and within the medium range of variability. There were only 10.13% and 3.8% of Mn and Co above background values, respectively, indicating that there were few external pollution sources for Mn and Co. The kriging interpolation results show that Mn, Co, and V distribute mainly in the outer urban areas close to the Yanshan Mountains and the Taihang Mountains and that atmospheric particulate matter from the northwest floods the ground in January (Jin et al., 2019), so Mn, Co, and V may come from parent soil materials blown in from the northern mountains. Peng et al. (2013) studied that the exposure of top soil to air is the main source of HMs in urban soils. The results of the above analysis indicate that Mn and Co are mainly natural sources, which is consistent with the results of Guan et al (2014). Liu et al. (2020) found that soil type and pH were the main influencing factors for HMs of Cr and Ni in Beijing Park soils. Zheng et al. (2005) found that Ni and Cr followed the same accumulation pattern in the soil's chemical or physical composition in a Beijing City study. Beijing lies in an alluvial plain with soils where clay and carbonate can

effectively leach out, predisposing the soil to Cr and Ni accumulation (Alloway, 2013). Thus, factor 1 represents natural factors such as soil parent material and forming processes. factor 2 contributes 22.95% of the total and 93.93% of Cu. Wang et al. (2018) found that Cu accumulation in cities tended to increase linearly with urban age (number of years of establishment). Chen et al. (2005) found that high Cu concentrations were present in parks in the historical central area of Beijing. The casting of bronze ceremonial and ritual objects, weapons, household utensils and coins began more than 4000 years ago. The results of ordinary kriging interpolation show that Cu concentrations are low in the northwest and southwest quadrants of the 4th to 5th ring region, where urbanization is lately. Thus, we attribute factor 2 to historical sites.

Factor 3 was mainly determined by Zn (64.80%), Pb (47.55%), Cr, and As, accounting for 32.89% of the total contribution. The average concentrations of Zn and Pb were higher than the background values of Beijing soils, 1.17 and 1.21 times higher than the background concentrations, respectively. Zn and Pb were in the high variability range, indicating that they were disturbed by anthropogenic activities. Ordinary kriging interpolation results suggest that pollution hotspots are mainly concentrated in the southeast of the study area. The influx of particulate matter in Beijing in July was primarily from the southeast (Chang et al., 2017). Thus, Factor 3 was attributed to industrial sources.

4 Conclusion

In this study, the carcinogenic and non-carcinogenic risks caused by HMs in urban areas of Beijing were assessed using a human health risk assessment model, principal component analysis, PMF model, and geostatistical analysis were used to identify the potential sources of HMs. In the study area, the nine HMs of Mn, Co, V, Ni, Cu, As, Pb, Zn, and Cr did not pose any ecological risk. The results of the human health risk assessment showed that the nine HMs in this study did not yet pose a non-carcinogenic risk; Ni and Cu in Beijing soils have significant carcinogenic effects on human health, and children are more susceptible to carcinogenic risks, with the direct ingestion being the main route of HMs exposure. The PMF model combines geostatistical analysis and principal component analysis to provide a more accurate estimation of the sources of HMs in urban soils in Beijing. Co, Mn, V, As, Ni, and Cr mainly originate from natural factors such as soil parent material, Pb and Zn from industrial emissions, and Cu from historical relics.

Author Contribution Danyang Meng: experiment, data analysis and writing; Yang Shao: methodology and editing; Min Luo: editing; Diandou Xu: supervision, writing-review; Zhiming Liu: visualization, validation; Lingling Ma: funding acquisition, conceptualization, editing.

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Data Availability The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics Approval Not applicable.

Consent to Participate The study does not involve any human participants, human data, or human tissues.

Informed Consent All authors have read and approved the final draft of this manuscript.

Conflict of Interest The authors declare no competing interests.

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