CASE STUDY



Contamination and human health risk assessment of heavy metal(loid)s in topsoil and groundwater around mining and dressing factories in Chifeng, North China

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Abstract

Chifeng is a concentrated mining area for non-ferrous metal minerals, as well as a key prevention and control area for heavyduty enterprises. This situation necessitates an effective ecological and human health risk assessment of heavy metal(loid) s driven by the wide distribution of metal ore processing, mining, and smelting factories in Hexigten Banner and Bairin Left Banner. We conducted surveys to assess the levels of heavy metal(loid)s (Cr, As, Pb, Cd, and Hg) in the topsoil and groundwater of the areas. The results indicated that the concentrations of As, Cd, and Pb in partial soil samples exceeded the environmental quality standards of Grade II. Based on contamination assessments, such as geoaccumulation indices and pollution indices, we inferred that Cd, Pb, and As were primary pollutants in topsoil. Potential ecological risks when considered as part of the average risk indices (RI) are up to 1626.40 and 2818.76, respectively, in the two areas. Comparative analysis revealed that Cd posed a very high potential ecological risk, followed by As. Moreover, the evaluation showed that the three exposure pathways of carcinogenic and non-carcinogenic risk followed a descending order: inhalation > ingestion > dermal contact, except for Pb. Arsenic in topsoil posed a potential non-carcinogenic risk to human health, while there were no adverse effects of As in groundwater. In addition, the average total carcinogenic risk for As in the two areas, as well as the risk of Pb in the topsoil of Bairin Left Banner and all the five heavy metal(loid)s in groundwater, exceeded human tolerance. Pb-Zn mines caused higher human health risks. In addition, the tandem contamination of heavy metal(loid)s in soil and groundwater was not obvious. This research study provides a basis for pollution remediation to control heavy industry-induced ecological and health risks of heavy metal(loid)s.

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Highlights

- Quantified the level of heavy metal(loid) pollution around the metal ore processing, mining and smelting factories in Chifeng.
- (2) Heavy metal(loid) risks were assessed based on optimized ecological and health risk model.
- (3) Cd, As and Pb posed a relatively serious threat to the region.
- (4) Targeted emission reduction and soil remediation measures are required to reduce heavy metal(loid) pollution in factories.

Keywords Heavy metal(loid)s · Contamination indices · Ecological risks · Human health risks · Chifeng

1 Introduction

Heavy metal(loid)s are produced in large quantities in industrial production activities, such as mineral smelting. They are suspended in the atmosphere around the factories and then deposited in the topsoil through rainfall or sedimentation. They may also enter the groundwater through the hydrological cycle. Heavy metal(loid)s have received widespread attention due to their toxicity, carcinogenicity, persistence, ubiquity, bioaccumulation, and biomagnification (Jitar et al. 2015; Li et al. 2021; Peng et al. 2020), which cause noteworthy ecological risks and human health risks. Compared to moving and dynamic road dust in field studies (Li et al. 2015; Yadav et al. 2019; Kolakkandi et al. 2020; Han et al. 2020), it is warranted to discuss the relationship among contamination indices, ecological risk factors, and human health risks from metal(loid)s in topsoil and groundwater.

Inner Mongolia is located in northern China with a vast territory and rich mineral resources. It is an important metallurgical industry base. Mineral resource exploitation and environmental pollution in this area have thus attracted scientific attention (Wang et al. 1999; Zhao et al. 2008; Guo et al. 2008; Jadambaa et al. 2014; Li et al. 2017; Jiang et al. 2022). Chifeng is a typical mining area concentrated in nonferrous metal minerals, and thus, it is a key prevention and control area for heavy-duty enterprises. There are a large number of smelters for mining and dressing factories of lead-zinc, tungsten-molybdenum, iron, and tin ores in the area. These factories extensively produce dust particles during production and operation. Human activities or natural conditions, such as climate, influence the movement and spatial distribution of particulate matter. Furthermore, the distribution of heavy metal(loid)s is affected by particulate matter, resulting in a high content of heavy metal(loid)s, such as As, Pb, Hg, Cd, and Cr, around the factories.

A number of methods such as the over-standard rate method, the single factor index method, the Nemerow pollution index method, the geo-accumulation index method (Yang et al. 2018; Wu et al. 2018), the pollution index method (Krek et al. 2018; Kolakkandi et al. 2020), and the enrichment factor method (Li et al. 2015; Jiang et al. 2016), have been extensively used to assess the pollution degree, temporal and spatial distribution characteristics and development trend of heavy metal(loid)s in soil in previous studies. Moreover, water environment quality evaluation, pollution source evaluation, single index pollution index method, comprehensive pollution index method and classification comprehensive index method are selected to evaluate the current status of heavy metal(loid) pollution in groundwater. Hakanson (1980) used potential ecological risk index of a single metal (E_r^{i}) and total potential ecological risk index (RI) as a diagnostic tool for water pollution control for the first time in a lake or freshwater system. After that the method was used in the evaluation of heavy metal pollution (Yi et al. 2011; Monferran et al. 2016; Jaiswal and Pandey 2019). However, bacause of different ecotoxicity and toxic-response for different metals and different biological tolerance and sensitivity to heavy metal(loid)s (Peng et al. 2020), the biological indicators are not completely consistent with the pollution indicators. More convincing results can be acquired from other methods of research, such as multivariate statistical analysis, health risk assessment and quantitative source apportionment, etc.

Previous studies on Inner Mongolia have mostly focused on the geochemical characteristics of minerals and groundwater (Dai et al. 2002, 2012; Guo et al. 2008), geological structures, metallogenic mechanisms (Liu et al. 2012), ecosystem stability, and vulnerability analysis (Bai et al. 2004; Quan et al. 2013), etc. Therefore, this study aims to evaluate the pollution status and degree of heavy metal(loid)s in the topsoil around the metal ore processing, mining, and smelting factories to assess their potential ecological risk. Moreover, the human health risks in topsoil and groundwater were assessed. Finally, the necessity of implementing pollution remediation and source control measures was considered based on the results.

2 Study area and methods

2.1 Study area

This study was conducted in Hexigten Banner ($42^{\circ} 22'$ 34"-44° 15' 33" N, 116° 34' 26"-118° 12' 59" E) and Bairin Left Banner ($43^{\circ} 51' 04''$ -44° 48' 34" N, 118° 43'



Fig. 1 Location of sampling sites in Hexigten Banner and Bairin Left Banner. The red dots represent topsoil samples and the blue dots represent groundwater samples

00"-119° 46' 51" E), which are mining areas concentrated in non-ferrous metal minerals; they are also key prevention and control zones for heavy metal companies in Chifeng City, Inner Mongolia Autonomous Region of north China (Fig. 1). Hexigten Banner is located in a mid-latitude and mid-temperate semi-arid continental monsoon climate zone with an annual mean temperature of 3.2 °C and an annual precipitation range of 237.0-574.6 mm. The predominant direction of wind throughout the year is from the West, and the annual mean wind speed is 3 m/s. Bairin Left Banner is a semi-arid continental monsoon climate with an annual mean temperature of 5.9 °C and an annual precipitation range of 209.3-677.5 mm. The annual wind direction is generally north-northwest, and the annual average wind speed is 2.6 m/s. As one of China's important bases for the metallurgical industry, the study area possesses rich mineral resources of iron ore, non-ferrous metals, and rare metals. The heavy chemical industry is relatively developed

here, and therefore, the region is a typical representative of heavy metal pollution.

2.2 Soil and groundwater sampling and chemical analysis

Contaminated sites around 35 heavy metal companies in Hexigten Banner and Bairin Left Banner were selected for the sampling survey. At the time of sample collection, 19 enterprises were in production mode, while 16 were in suspension. At least one topsoil sample control was obtained from each of the selected companies. Sampling was performed downstream of the groundwater runoff or downwind of the company. For the metal mining and dressing industries, the focus of the sampling layout was on the tailing pond in the dressing area. Groundwater samples were taken from wells, downstream residential wells, or agricultural irrigation wells that were mainly located downstream of the groundwater, beneficiation areas, tailing ponds, or the surrounding areas of the industries. Sampling points were densely distributed in key areas to cover the main polluted sites. A total of 67 topsoil (0–20 cm) and 38 groundwater samples were collected (Fig. 1).

The groundwater samples were collected in 1-L pickled polyethylene bottles that had been washed thrice. A space of 5-10 mL was left during sampling to avoid opening the bottle stopper with a temperature increase. Two drops of 65% HNO₃ were added to the sampled water for acidification, followed by filtration with 0.45-µm filter paper. Finally, the bottle stopper was tightly sealed with paraffin.

Each topsoil sample was a composite of five random subsamples selected from the upper soil horizon of $1 \text{ km} \times 1 \text{ km}$ square grids. The volume and depth of the sampling points were the same; sand, minerals, and pebbles were removed with a hand shovel. The original weight of each soil sample was > 1000 g. All samples were wrapped in aluminum foil and stored and labeled in self-sealing polyethylene bags in preparation for transportation to the laboratory. The soil samples were sieved by the quarter method using a < 2-mm nylon sieve to remove sand, gravel, plant fragments, and other impurities. After naturally drying in a cool and dark place at 20-23 °C, the soil samples were finely ground with an agate ball mill for homogeneous treatment and stored in the dark at room temperature for chemical analysis (USEPA 1996; ISO 2006). Sample collection, handling, and storage followed the standard procedures recommended by the Chinese Ministry of Water Resources (Wu et al. 2019). These soil samples were digested with concentrated acid, such as HNO₃-HF-HCLO₄, until the silicate mineral shadow disappeared completely. The digested solution was then transferred to a 25-mL polypropylene loop, and 1% acid and deionized water were added to make the samples reach a fixed volume for metal determination. Then the digested samples were filtered by a syringe $(0.45 \,\mu\text{m})$ (Zhang et al. 2020). Afterwards, different methods of determination were applied to measure the concentration of heavy metal(loid)s. Overall, atomic absorption spectrophotometry (AAS-ZCA-1000) was used to analyze Cr, Cd, and Pb (Yang et al. 2020). Cold atomic absorption spectroscopy (LP-CG-1) was selected to analyze Hg. Cyanide-Atomic Fluorescence Spectrometry (AFS-8330) was applied to analyze As (Peng et al. 2020). Detection was performed in triplicates for each sample. These sampling and chemical analysis methods have been widely used previously by a large number of experts and scholars.

3 Contamination assessment

3.1 Geoaccumulation index

Muller (1969) first proposed the concept of geoaccumulation index (I_{geo}) and applied it to the study of heavy

metal pollution in sediments of the Rhine River. Later, this method has been widely used by many scholars in the contamination assessment of heavy metal(loid)s to identify whether the site is polluted and quantify the contamination degree. In this study, the I_{geo} values for the soils at the examined sites were calculated by the following equation:

$$I_{\rm geo} = \log_2(C_{\rm i}/1.5B_{\rm i}) \tag{1}$$

where C_i is the measured concentration of heavy metal(loid)s in soil (mg/kg), and B_i is the geochemical background value of heavy metal(loid)s in local soil (mg/kg). The constant 1.5 is used to explain the potential changes in background values. Based on I_{geo} values, the soil quality is classified into 7 grades listed in Table S1 (Muller 1969; Yang et al. 2018) in Supplementary Material.

3.2 Pollution index

The pollution index (PI) of heavy metal(loid)s in soils is widely used to quantify the pollution level and degree for protecting the environment and human health, which was computed following Eq. (2):

$$PI = C_i / S_i \tag{2}$$

where S_i is the reference content or reference standard in original soil (mg/kg). The reference values for Inner Mongolia Autonomous Region were: Cr = 41.4, As = 7.5, Pb = 17.2, Cd = 0.053 and Hg = 0.04 (CNEMC 1990). The pollution level was divided into four grade: low pollution (PI < 1), moderate pollution (1 ≤ PI < 3), considerable pollution (3 ≤ PI < 6), and very high pollution (PI ≥ 6) (Islam et al. 2015). On this basis, there introduced the pollution load index (PLI) of all heavy metal(loid)s for a sample, and it was count by Eq. (3):

$$PLI = (PI_1 \times PI_2 \times \dots \times PI_n)^{1/n}$$
(3)

where, PI_1 , PI_2 , ..., PI_n are PI of elements 1, 2, ..., *n*; the PLI values were classified into 6 levels (Table S2 in Supplementary Material) (Wu et al. 2018).

3.3 Potential ecological risk index

The potential ecological risk reflects the toxicity and risk degree of heavy metal(loid)s in the soil and their impact on the environment. In this study, the potential ecological risk indicators were computed by using Eqs. (4)–(6) (Hakanson 1980):

$$C_{\rm f}^{\rm i} = C^{\rm i}/C_{\rm n}^{\rm i} \tag{4}$$

$$E_{\rm r}^{\rm i} = T_{\rm r}^{\rm i} \times C_{\rm f}^{\rm i} \tag{5}$$

$$RI = \sum E_r^i$$
(6)

where, $C_{\rm f}^{\rm i}$ is the contamination factor; $C^{\rm i}$ is the measured concentration of heavy metal(loid)s in soil (mg/kg); $C_{\rm n}^{\rm i}$ is the environmental quality standard or soil background value (mg/kg); $E_{\rm r}^{\rm i}$ is the potential ecological risk index of a single metal (Table S3); $T_{\rm r}^{\rm i}$ is the toxic-response factor for a given heavy metal; $T_{\rm r}^{\rm i}$ of Cd, Hg, As, Pb and Cr were 30, 40, 10, 5 and 2 respectively (Hakanson 1980); $C_{\rm f}^{\rm i}$ is the contamination factor; RI is the total potential ecological risk index of multiple metals (Table S4 in Supplementary Material).

4 Health risk assessment

4.1 Health risk in surface soil

The health risk assessment model used here was originally developed by the United States Environmental Protection Agency. The soil screening guidance was adopted in this study to assess the carcinogenic and non-carcinogenic risks of heavy metal(loid)s. In this study, health risk associated with heavy metal(loid)s exposure in topsoil was noted to occur via three paths, i.e., surface soil ingestion, dermal contact with surface soils, outdoor inhalation of vapors and particulates from surface soils. The average daily exposure dose was calculated using the following models (USEPA 1996, 2002, 2019):

Ingestion:

$$ADD_{ing} = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6}$$
(7)

where, ADD_{ing} is the average daily intake doses by ingestion (mg/kg day); *C* is the concentration of heavy metal(loid) s in soil (mg/kg); IngR is the ingestion rate of soil (mg/day); EF is the exposure frequency (days/year) (250 days/year for adults) (Table S5 in Supplementary Material); ED is the exposure duration (year) (25 year for adults); BW is the average body weight (kg) (61.8 kg for adults), AT is the average time for carcinogenic or non-carcinogenic effect (day) (Table S5 in Supplementary Material).

Dermal absorption:

$$ADD_{dermal} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6}$$
 (8)

where, ADD_{dermal} is the average daily intake doses by dermal contact (mg/kg day); SA is the skin surface area for soil dermal contact (cm²); AF is the soil-to-skin adherence

factor (mg/cm² day); ABS is the dermal absorption factor (unitless).

Inhalation:

Å

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT}$$
(9)

where, ADD_{inh} is the average daily intake doses by inhalation (mg/kg day); InhR is the inhalation rate of soil (m³/day); PEF is the particulate emission factor (m³/kg).

Based on the above three different ways of exposure, Hazard quotient (HQ) is defined as the indicator that characterize the non-carcinogenic health risk of a single heavy metal and a single exposure route.

$$HQ = \frac{ADD}{RfD}$$
(10)

where, RfD is the chronic dermal or oral reference dose (mg/ kg day) (Table S6).

To evaluate the total non-carcinogenic health risk for all exposure of each heavy metal, hazard index (HI) is introduced with Eq. (11) (USEPA 1996, 2019):

$$HI = \sum HQ$$
(11)

where, HI is the sum of HQ of the corresponding heavy metal(loid)s. When HQ < 1 or HI < 1, no adverse health effects are observed, i.e., the potential non-carcinogenic health risk is within an acceptable range. On the contrary, when HQ \geq 1 or HI \geq 1, a non-carcinogenic risk exists and poses a health threat to human health in the area. As the HI values increase, the possibility of such impacts increases.

Furthermore the carcinogenic risk (CR) and the total carcinogenic risk (TCR) can be expressed by the following mathematical formulas:

$$TCR = \sum_{j=1}^{n} CR_{j} = ADD_{ing} \times SF_{ing} + ADD_{dermal}$$

$$\times SF_{dermal} + ADD_{inh} \times SF_{inh}$$
(12)

where, SF is the carcinogenicity slope factor (mg/kg day). There is no risk of cancer when TCR < 10^{-6} ; The TCR values range from 10^{-6} to 10^{-4} , the total carcinogenic risk is within an acceptable range but risk management measures need to be taken. When TCR > 10^{-4} , the carcinogenic risk has exceeded human tolerance.

4.2 Health risk in groundwater

Health risk associated with heavy metal(loid)s in groundwater occurred via two paths in this study, i.e., (1) groundwater ingestion and (2) groundwater volatilization to outdoor



Fig. 2 Radar diagrams showing concentration spatial trends in major heavy metal(loid)s

ambient air. The carcinogenic risk was associated only with the groundwater exposure pathway in this research. For volatile metals, such as Hg, in addition to considering the intake pathway of groundwater, its volatilization must also be considered to calculate the HQ. TCR and HI are defined as follows:

$$TCR = \frac{C \times SF_{o} \times EF \times ED \times IR_{w}}{BW \times AT_{c} \times 365}$$
(13)

Groundwater ingestion exposure pathway:

$$HQ_{ing} = \frac{C \times EF \times ED \times IR_{w}}{RfD_{o} \times BW \times AT_{n} \times 365}$$
(14)

Outdoor air exposure pathway:

$$HQ_{vol} = \frac{C \times EF \times ED \times VF_{wamb}}{RfC \times AT_n \times 365}$$
(15)

$$HI = \sum HQ_i = HQ_{ing} + HQ_{vol}$$
(16)

where, SF_o is the oral slope factor (mg/kg-day); IR_w is the daily groundwater consumption rate (L/day) ($IR_w = 1$ of adults); RfD_o is the chronic oral reference dose (mg/kg day); RfC is the reference concentration (mg/m³); VF_{wamb} is the groundwater to ambient air volatilization factor (mg/m³ air)/(mg/L water).

5 Spatial distribution of heavy metals

The Kriging method is a geostatistical model based on autocorrelation. It assumes that the distance or direction between the sampling points can reflect the spatial correlation of the surficial changes. The mathematical function is fitted with a specified number of points or all points within a given sampling radius to determine the output value of each position. The main steps of the Kriging interpolation include exploratory statistical analysis, variogram modeling, and surface creation. The Kriging method can be used to weigh the measured values to obtain predictions for the unmeasured positions. The basic principle is as follows:

$$\hat{Z}(s_0) = \sum_{i=1}^{N} \lambda_i Z(s_i) \tag{17}$$

where, $Z(s_i)$ is the measured value at the *i*th position; λ_i is the unknown weight of the measured value at the *i*th position; s_0 is the predicted position; *N* is the number of measurements.

6 Results and discussion

6.1 Contamination indices

6.1.1 *I*_{geo} assessment

The average concentrations of five major heavy metal(loid)s decreased in the order of Pb > As > Cr > Cd > Hg in the topsoil and Pb > As > Cd > Cr > Hg in groundwater in these two study areas (Table S7 in Supplementary Material) (Fig. 2). The average concentrations above the standards for As, Pb, and Cd were 47.79%, 0% and 0% in Hexigten Banner and 151.07%, 14.64% and 284.22% in Bairin Left Banner; the standard considered is the Grade II Environmental Quality Standard for soils (GB 15618–1995). In addition, the concentration of Cd, Pb, or As in 80.00%, 40.00% or 33.33% of the samples was higher than the Grade II values, respectively, in Bairin Left Banner. In Hexigten Banner, 45% of

Location	Industry	Cr	As	Pb	Cd	Hg	Order	Reference
Hexigten Banner	Metal ore mining and dressing plant	- 0.97	1.21	0.45	1.95	- 1.53	Cd > As > Pb > Cr > Hg	This study
Bairin Left Banner	Metal ore mining and dressing plant	- 0.52	1.65	2.68	4.09	- 1.38	Cd > Pb > As > Cr > Hg	This study
Fengcheng	Lead-zinc mine tailings library	- 1.65	2.37	6.79	8.31	1.97	Cd > Pb > As > Hg > Cr	Yang et al. (2018)
Dandong	Wulong gold tailings depot	- 0.23	2.31	1.90	2.70	- 0.58	Cd > As > Pb > Cr > Hg	Yang et al. (2018)
Anshan	Iron ore tailings depot	- 0.36	- 3.02	- 1.23	0.84	- 0.89	Cd > Cr > Hg > Pb > As	Yang et al. (2018)
Huludao	Lead zinc mine tailings library	- 0.26	- 1.24	5.11	6.77	2.23	Cd > Pb > Hg > Cr > As	Yang et al. (2018)
Tianjin	Mining products yard	0.30	5.02	- 0.17	7.16	_	Cd > As > Cr > Pb	Yang et al. (2018)
Jiangxi	Tungsten ore mining area	- 0.02	1.88	0.90	8.69	9.05	Hg>Cd>As>Pb>Cr	Yang et al. (2018)
Shaoguan	Dabaoshan mining area	- 0.64	0.85	1.84	3.95	- 0.23	Cd > Pb > As > Hg > Cr	Yang et al. (2018)
Hunan	Tin mine	- 0.75	0.58	0.47	3.07	6.42	Hg>Cd>As>Pb>Cr	Yang et al. (2018)
Guizhou	Wanshan mercury mine	1.27	- 1.41	- 0.61	- 0.34	6.36	Hg>Cr>Cd>Pb>As	Yang et al. (2018)
Shanxi	Lead-zinc smelter	- 1.11	- 0.69	1.56	0.88	2.64	Hg > Pb > Cd > As > Cr	Yang et al. (2018)
Yunnan	Lead-zinc mine	- 0.03	- 0.13	0.43	2.81	2.20	Cd > Hg > Pb > Cr > As	Yang et al. (2018)
Beijing	Mining areas	1.23	- 2.36	2.61	3.39	2.73	Cd > Hg > Pb > Cr > As	Li et al. (2014)
Henan	Mining areas	- 0.67	- 0.79	0.28	0.16	- 0.50	Pb>Cd>Hg>Cr>As	Li et al. (2014)
Zhejiang	Mining areas	0.26	2.35	5.74	4.71	_	Pb>Cd>As>Cr	Li et al. (2014)
Anhui	Mining areas	- 0.80	3.79	0.34	5.26	1.52	Cd > As > Hg > Pb > Cr	Li et al. (2014)
Sichuan	Mining areas	- 0.42	- 1.88	1.65	3.84	2.33	Cd > Hg > Pb > Cr > As	Li et al. (2014)

Table 1 The average I_{geo} values of heavy metal(loid)s in topsoil in different regions

all the As samples surpassed the Grade II value. The high concentration of heavy metal(loid)s is a result of the superposition of geological and anthropogenic effects (Tepanosyan et al. 2018). The concentrations of Cr and Hg in all the samples were lower than the Grade II standard values. Past studies have shown that the concentration of Cd, As, and Pb in the atmosphere around industrial areas, especially around metal mining and dressing industries; can be significantly higher. They are deposited in the topsoil through rainfall or sedimentation, leading to a high concentration of heavy metal(loid)s in the topsoil (Ivonin and Shumakova 1991; Yang et al. 2018).

It is not reasonable to assess the environmental quality and pollution levels in an area based on the concentration of heavy metal(loid)s only as these levels may also be limited by the geochemical background values of heavy metal(loid) s, land use type, and human activities (Li et al. 2015). Therefore, I_{geo} was introduced for contamination assessment. The average I_{seo} values of Cr, As, Pb, Cd, and Hg in Hexigten Banner and Bairin Left Banner were <2 (Table 1), indicating the soil were practically uncontaminated or moderately contaminated by the five heavy metal(loid)s above. However, in Bairin Left Banner, the average I_{geo} value of Cd was 4.09, suggesting that the degree of contamination ranged from heavily to extremely contaminated. The average I_{geo} value for Pb indicated moderately to heavily contaminated. In addition, the contamination degrees of the five studied heavy metal(loid)s were in the order of Cd > As > Pb > Cr > Hg in Hexigten Banner and Cd>Pb>As>Cr>Hg in Bairin Left Banner, showing that Cd, Pb, and As were key topsoil pollutants. Previous studies have also reported high concentrations of Cd in soils in this area of study (Wei and Yang 2010; Li et al. 2014). In addition, as per the spatial distribution pattern of heavy metal(loid)s presented in Table 1, heavy metal pollution in the soils of southern cities was higher than that in the northern cities (Chen et al. 2015). It is speculated that this phenomenon is related to a higher geochemical background in the southwest and an intensification of human activities in the southeast. These results provide basic data to inform and promote measures for heavy metal pollution control and soil quality improvement.

Figure 3 shows the class distribution of I_{geo} for five heavy metal(loid)s based on Table S7 in Supplementary Material. The results reflected that I_{geo} values of Cr in the two regions and Hg in Bairin Left Banner were in Class 0 for all the sampling sites. For As, all the samples from Hexigten Banner and 86.7% of the samples from Bairin Left Banner were under Class 4. Additionally, 70.0% and 66.7% of the samples fell under Class 2 respectively. Nearly 80% of all the samples were lower than Class 1 in Hexigten Banner for Pb. However, the I_{geo} values ranged from Class 0 to 6 for samples from Bairin Left Banner. Bounded by Class 3, 40% of the sampling sites were under Class 2 and 60% were above Class 4. The class distribution ranges of I_{geo} for Cd in both regions were from Class 1 to Class 6, among which 85% of the samples from Hexigten Banner fell below Class 3, while 79.9% of the samples from Bairin Left Banner were above Class 3. The same number of samples were distributed in Classes 1-3 or 5-6.



Fig. 3 Class distribution of Igeo for heavy metal(loid)s in Hexigten Banner and Bairin Left Banner



Fig. 4 Box-plots of the pollution index (PI) and pollution load index (PLI) of five heavy metal(loid)s in topsoil collected from Hexigten Banner and Bairin Left Banner. The squares represented average

Overall, the I_{geo} values fell below Class 3 for nearly all the samples in Hexigten Banner, which represented that the overall pollution degree in Bairin Left Banner was more serious than in Hexigten Banner. In summary, our results revealed that the factories of metal ore processing, mining, and smelting caused high I_{geo} values, which were mostly above Class 0 for As, Pb, and Cd, suggesting high pollution of the topsoil. This observation is consistent with the average I_{geo} results based on information in Table 1. Due to the limited number of sampling points, these results are not completely representative. Therefore, the pollution index was introduced for further contamination assessment and complementary authentication.

values and black boxes represented outliers. The horizontal line indicated the median line

6.1.2 Pollution assessment

To have a comprehensive understanding of the current status and levels of soil pollution, two indices, the pollution index (PI) and pollution load index (PLI), were introduced. The mean PI values of heavy metal(loid)s in the topsoil were in the order of Cd (9.61) > As (5.91) > Pb (2.62) > Cr (0.84) > Hg (0.62) in Hexigten Banner, proving that the soil was very highly polluted by Cd, considerably polluted by As, moderately polluted by Pb, and only slightly polluted by Cr and Hg. In Bairin Left Banner, the order of PI was Cd (43.50) > Pb (20.00) > As (10.04) > Cr (1.10) > Hg (0.60), implying that the soil was highly polluted by Cd, Pb, and

Risk index (RI)	Sampling sites	Average RI	Potential ecological risk index (E_r)						
	(Location)		Very high	High	Considerable	Moderate	Low $E_{\rm r}^{\rm i} < 40$		
			$E_{\rm r}^{\rm i} \ge 320$	$160 \le E_{\rm r}^{\rm i} < 320$	$80 \le E_r^i < 160$	$40 \le E_{\rm r}^{\rm i} < 80$			
Extremely high	2 (H)	1626.40	Cd			As>Hg>Pb	Cr		
$RI \ge 1200$	6 (B)	2818.76	Cd		Pb > As		Hg>Cr		
Very high	0 (H)								
$600 \le RI < 1200$	5 (B)	1015.95	Cd		As>Pb		Hg>Cr		
Considerable	5 (H)	391.90		Cd	As		Hg > Pb > Cr		
$300 \le RI < 600$	1 (B)	450.62	Cd				Hg > As > Pb > Cr		
Moderate	9 (H)	232.65			Cd	As	Hg > Pb > Cr		
$150 \le RI < 300$	1 (B)	282.18		Cd			As > Pb > Hg > Cr		
Low	4 (H)	109.12				Cd	Hg > As > Pb > Cr		
RI < 150	2 (B)	123.19			Cd		As > Hg > Pb > Cr		

Table 2 Potential ecological risk index of heavy metal(loid)s in Hexigten Banner (H) and Bairin Left Banner (B)

As, moderately polluted by Cr, and slightly polluted by Hg (Fig. 4). These orders of pollution levels were consistent with median PI values and were similar to I_{geo} values in the two study areas. These findings are consistent with Luo's report on the high level of heavy metal pollution by Cd and Pb in urban soils (Luo et al. 2011). However, the trends noticed here were slightly different from the previously reported trend of Cr > Cd > As > Pb in urban soils around an electronics manufacturing facility in Hubei province (Wu et al. 2018). The trends noticed here were also very different from those in urban soils from Changsha, Linhe, Chongqing, Zhangzhou, Changchun, and Fuzhou (Luo et al. 2011). Figure 4d and c show that Bairin Left Banner had relatively high median PI values for Cd (PI = 28.13) and Pb (PI = 15.03). Figure 4b features similar medians for As with PI=3.27 and PI=3.06 in Hexigten Banner and Bairin Left Banner. In addition, the maximum PI values in Bairin Left Banner were 153.91, 80.12, and 43.57 for Cd, Pb, and As, which were higher than the corresponding maximum values (61.28, 8.99, and 21.81) in Hexigten Banner. Multiple indicators showed that Cd was the most abundant element, followed by Pb and As. A possible reason is that Cd is mainly used in rechargeable Ni-Cd batteries, but Cd-containing products are rarely recycled and are often dumped together with household garbage (Jarup 2003). Owing to the relatively high pollution index, Cd, Pb, and As accumulated in the topsoil were more bioavailable and posed a certain and potential risk to soil quality, ecological environment, food security, and human health. These pollutants might originate from local long-term intensive metal ore mining and smelting activities. These pollution assessment results are the same as the findings of Yang et al. who investigated pollution levels in soils around an e-waste area in Wenling, Zhejiang province (Yang et al. 2020); Their results were comparable with the I_{geo} index recorded here.

PLI stands for multielement enrichment and contamination. In this study, PLI which combined all the PI values of the five elements were 1.95 (average value), 1.86 (median value), 4.65 (maximum value) and 0.62 (minimum value) for Hexigten Banner. However, PLI were 4.48, 4.54, 7.82 and 0.99 respectively for Bairin Left Banner (Fig. 4f). Obviously, Bairin Left Banner was more affected by the pollution of polymetallic elements than Hexigten Banner. It is speculated that the reason may be related to the large-scale shutdown of the factories.

6.2 Potential ecological risks of heavy metal(loid)s

The E_r^i and RI values show the potential ecological risk degree of hazardous heavy metal(loid)s and the sensitivity of organisms to these toxic substances. For a single metal, the mean E_r^i was in the order of Cd>As>Hg>Pb>Cr, corresponding to the value of 288.34, 59.12, 24.90, 13.09 and 1.68 respectively in Hexigten Banner and similar order of Cd > As > Pb > Hg > Cr corresponding to 1304.91, 100.43, 99.98, 23.93 and 2.19 in Bairin Left Banner. The order of $E_{\rm r}^{\rm i}$ was very different from $I_{\rm geo}$ and PI, because of the different ecotoxicities of the different metals. In addition, Hg was found to be significantly related to the organic matter and total organic carbon in the soil (Beckers and Rinklebe 2017). When C/N was low, Hg increased exponentially (Obrist et al. 2011; Zhang et al. 2020). Organic matter is humified and easily decomposed. Therefore, an appropriate amount of Hg is beneficial to decrease ecological risks. As presented in Table 2, the total potential ecological risks posed by the five heavy metal(loid)s in the topsoil in Bairin Left Banner were very high $(RI \ge 600)$ at most sampling sites (73%), which was majorly because of the high level of Cr contamination. In the Hexigten Banner, the ecological risks were considerable and moderate

Heavy metal	HQ _{ing} (mean)		HQ _{der} (mean)		HQ _{inh} (mean))	Hazard index (HI)	
	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner
Cr	2.57×10^{-5}	3.35×10^{-5}	1.19×10^{-5}	1.56×10^{-5}	1.08×10^{-1}	9.13×10 ⁻²	1.08×10^{-1}	9.14×10 ⁻²
As	1.64×10^{-1}	2.78×10^{-1}	3.13×10^{-2}	5.31×10^{-2}	1.28	1.42	1.48	1.75
Pb	3.84×10^{-2}	2.93×10^{-1}	1.55×10^{-2}	1.18×10^{-1}	5.54×10^{-3}	2.75×10^{-2}	5.94×10^{-2}	4.39×10^{-1}
Cd	5.65×10^{-4}	2.56×10^{-3}	1.37×10^{-4}	6.18×10^{-4}	2.21×10^{-2}	6.50×10^{-2}	2.28×10^{-2}	6.82×10^{-2}
Hg	9.20×10^{-5}	8.84×10^{-5}	5.56×10^{-6}	5.35×10^{-6}	2.99×10^{-1}	2.87×10^{-1}	2.99×10^{-1}	2.87×10^{-1}

Table 3 Non-carcinogenic risk of heavy metal(loid)s in the topsoil for the two study areas of three exposure ways

Table 4 Carcinogenic risk of heavy metal(loid)s in the topsoil for the two study areas of three exposure ways

Heavy metal	CR _{ing} (mean)		CR _{der} (mean)		CR _{inh} (mean)		Total carcinogenic risk (TCR)	
	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner
Cr	6.34×10 ⁻⁶	8.26×10^{-6}	2.95×10^{-6}	3.84×10^{-6}	5.83×10^{-5}	4.95×10^{-5}	6.76×10^{-5}	6.16×10^{-5}
As	2.42×10^{-5}	4.12×10^{-5}	4.63×10^{-6}	7.86×10^{-6}	7.44×10^{-5}	8.23×10^{-5}	1.03×10^{-4}	$1.31 \times 10-4$
Pb	4.60×10^{-6}	3.51×10^{-5}	1.85×10^{-6}	1.42×10^{-5}	7.56×10^{-5}	3.76×10^{-4}	8.21×10^{-5}	4.25×10^{-4}
Cd	7.06×10^{-8}	3.19×10^{-7}	1.71×10^{-8}	7.72×10^{-8}	3.58×10^{-7}	1.05×10^{-6}	4.46×10^{-7}	1.45×10^{-6}

(RI < 600) at most sampling points (90%), while no sampling sites were found to have very high levels of contamination.

6.3 Human health risk assessment

6.3.1 Health risk in soil

The basis of health risk assessment is that the consequences of any heavy metal pollution may cause harm to humans. There are many exposure paths for heavy metal(loid)s to enter the human body and cause human health risk, especially surface soil ingestion, dermal contact with surface soils, outdoor inhalation of vapors and particulates from surface soils. As shown in Tables 3, 4 and Fig. 5 for the average health risks, both non-carcinogenic risk and carcinogenic risk caused by the three exposure paths followed the descending order: inhalation > ingestion > dermal contact except Pb. This conclusion was similar with the results of previous researches that inhalation of particulates and vapors presented a higher risk than ingestion (Ferreira-Baptista and Miguel 2005; Cao et al. 2014). While non-carcinogenic risk for Pb decreased in the following order: ingestion > dermal contact > inhalation (Table 3) (Fig. 5a). It was consistent with the previous studies (Mugoša et al. 2015; Wu et al. 2018; Kolo et al. 2018). In this study, the non-carcinogenic risk values for As were 1.48 and 1.75 for Hexigten Banner and Bairin Left Banner respectively. The hazard indices were > 1 and larger than the values calculated by other four metals which revealed that As posed a potential non-carcinogenic risk threat to human health. Other scholars had repeatedly proved that As in topsoil can cause adverse health effects (Carlin et al. 2015; Rinklebe et al. 2019). The HI values for As, Pb and Cd in Hexigten Banner were lower than those in Bairin Left Banner showing smaller health threat. However, the Hazard level for Hg was comparable and for As and Cd were in the same order of magnitude between the two study areas. The orders of HI values were As > Hg > Cr > Pb > Cd in Hexigten Banner, the same as Tang and Rinklebe's views (Tang et al. 2016; Rinklebe et al. 2019) and As > Pb > Hg > Cr > Cd in Bairin Left Banner.

Since Hg is defined as a non-carcinogenic element, only the carcinogenic risk for Cr, As, Pb, and Cd was calculated (Table 4). Inhalation of topsoil appeared to be the most important path for heavy metal(loid) exposure of the public for the two areas, followed by ingestion and dermal contact (Fig. 5b). The average total carcinogenic risk for As in the two areas and for Pb in Bairin Left Banner were higher than 10⁻⁴, implying that the risk induced by the two metals exceeded human tolerance. The exposure dose in these areas was detrimental to people via inhalation, ingestion, and dermal contact. This finding was attributed to the concentrated distribution of Pb-Zn ore mining and dressing factories in the areas. The TCR values for Cr in both areas, Pb in Hexigten Banner, and Cd in Bairin Left Banner were in the range of 10^{-6} to 10^{-4} , which is within an acceptable risk range. There was no risk of cancer from Cd (TCR $< 10^{-6}$) exposure in Hexigten Banner.

Heavy metal(loid) s	HQ _{ing} (mean)		HQ _{vol} (mean)		Hazard index	(HI)	Total carcinogenic risk	
	Hexigten Ban- ner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner	Hexigten Banner	Bairin Left Banner
Cr	1.66×10^{-2}	1.78×10^{-2}	_	_	1.66×10^{-2}	1.78×10^{-2}	8.20×10^{-6}	8.77×10^{-6}
As	7.04×10^{-1}	1.94×10^{-1}	_	_	7.04×10^{-1}	2.06×10^{-1}	1.04×10^{-4}	3.06×10^{-5}
Pb	4.40×10^{-1}	4.81×10^{-1}	_	_	4.40×10^{-1}	4.98×10^{-1}	5.27×10^{-5}	5.97×10^{-5}
Cd	2.21×10^{-1}	1.80×10^{-2}	_	_	2.21×10^{-1}	1.94×10^{-2}	1.38×10^{-5}	1.21×10^{-6}
Hg	8.46×10^{-3}	5.50×10^{-3}	3.58×10^{-5}	2.32×10^{-5}	8.50×10^{-3}	5.52×10^{-3}	-	-

 Table 5
 Non-carcinogenic risk and carcinogenic risk of heavy metal(loid)s in groundwater



Fig. 5 Percent Stacked Bar diagram for soil and Stacked Bar diagram for groundwater

Contrarily, pollution levels in Bairin Left Banner presented higher carcinogenic risk than those in Hexigten Banner, except for Cr. Cr and As were in the same order of magnitude between the two places. A decreasing trend was noted for TCR values for carcinogenic risk from As>Pb>Cr>Cd in Hexigten Banner and Pb>As>Cr>Cd in Bairin Left Banner. At the same time, As, Pb, and Cr were the main contributors to carcinogenic risk through inhalation. It can be seen from the results that As is the main element contributing to noncarcinogenic risk, while it also causes a serious carcinogenic risk to the public.



Fig. 6 Spatial zonation of carcinogenic risk and non-carcinogenic risk (**a**–**d** risks in soil; **e**–**h** risks in groundwater; **a**, **c**, **e**, **g** risks in Hexigten Banner; **b**, **d**, **f**, **h** risks in Bairin Left Banner; **a**, **b**, **e**, **f** non-carcinogenic risk (HI); **c**, **d**, **g**, **h** carcinogenic risk (TCR))

6.3.2 Health risk in groundwater

The non-carcinogenic and carcinogenic risks of the five heavy metal(loid)s via groundwater were estimated through the path of intake (i.e. drinking groundwater ingestion). The non-carcinogenic risk of Hg manifested through a unique path, i.e., groundwater volatilization to ambient air. The human health risks in groundwater were calculated separately and are presented in Table 5, where non-carcinogenic risk (<1) can be seen to be within the acceptable range. No adverse health effects were observed. The hazard indices of Cr, As, Pb, and Hg had the same magnitude for the two study areas. The average total carcinogenic risks of the five heavy metal(loid)s exceeded the acceptable levels for humans (1×10^{-6}) , implying that they pose carcinogenic risks to the local population. Arsenic showed a higher carcinogenic risk (1.04×10^{-4}) in Hexigten Banner than other metals. The total carcinogenic risks posed by Cr and Pb were similar at the two sites. In summary, As, Pb, and Cr were the main metals posing a carcinogenic risk to the public. The results of groundwater analysis were consistent with those for topsoil analyses.

6.3.3 Spatial distributions of health risk

The spatial distribution of carcinogenic and non-carcinogenic risks from soil and groundwater are demonstrated in Fig. 6. Soil samples with significant non-carcinogenic and carcinogenic risks were distributed in the midwestern part of Bairin Left Banner (Fig. 6b, d), while the groundwater samples were distributed in the northern part of Hexigten Banner (Fig. 6e, g). The results indicated that Pb–Zn mines caused higher human health risks. On the contrary, Cu and Mo mines were less harmful to human beings. No clear similarity or correlation could be established between the health risks from metal(loid)s via the topsoil or groundwater, proving that tandem contamination is not a strong phenomenon in the study area. Nonetheless, the overall pollution degree in Bairin Left Banner was more serious than in Hexigten Banner. This study suggests that mining has brought non-negligible harm to residents of surrounding areas.

7 Conclusions

Through field research and contamination assessment focusing on five heavy metal(loid)s (As, Pb, Cr, Cd and Hg) in the topsoil and groundwater of Hexigten Banner and Bairin Left Banner, we verified that Cd, Pb, and As are the key pollutants in the topsoil. These metals contribute heavily to the overall pollution degree in Bairin Left Banner, which is more serious than in Hexigten Banner. Based on background values in soil and toxic-response factors obtained from a large number of previous studies, the E_r^i and RI values were used to assess the degree of the potential ecological risk caused by hazardous heavy metal(loid)s, as well as the sensitivity of many organisms to toxic substances. Cd posed a very high potential ecological risk in the two study areas, followed by As. The total ecological risks in Bairin Left Banner were very high at most sampling sites in this region. For health risk assessment, both non-carcinogenic and carcinogenic risks of these metals, except Pb, in the topsoil followed this order: inhalation > ingestion > dermal contact. The five heavy metal(loid)s posed carcinogenic risks to the local population via groundwater as well. However, the tandem contamination of topsoil and groundwater in the study area was not obvious. Pb–Zn mines caused higher human health risks.

Based on the results of this study, it is concluded that heavy metals pose pollution and carcinogenic risks. Considering that the production enterprises are close to sensitive ecosystems, such as farmlands, grasslands, and residential land, necessary measures should be taken for pollution remediation and source control. For example, green isolation belts should be set up downstream of or around the factory to suppress the spread of dust. Multiple seepage cutoff walls and water quality monitoring wells should be set up downstream of these facilities. Production enterprises must build anti-seepage tailings ponds and dust suppression facilities, and tailings wastewater should be subjected to recycling. Tailings sand should be dry discharged. Enterprises that are being suspended or closed should take measures to cover their tailings pond to eliminate any potential environmental safety hazards and reduce pollution hazards.

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Declarations

Conflict of 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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