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ORIGINAL PAPER

Areal Health Risk Assessment Using Soil Bioaccessible Heavy Metal(loid)s Around Industrial Area in Nanjing, Southeast China

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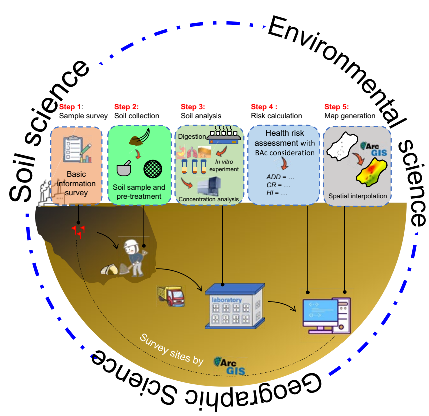
Abstract

A systematic protocol for conducting area-specific health risk assessments associated with soil heavy metal(loid)s contamination, incorporating total and bioaccessible metal(loid)s, is outlined in this study. We evaluated total and bioaccessible heavy metal(loid)s levels, along with the associated community health risks, in urban soils around two industrial areas, LH01–LH10 and QX01–QX11, in Nanjing, Southeast China. Our findings unveiled substantial disparities in metal(loid)s concentrations, pollution sources, and population health risks across these sampled areas. Soils from LH03, LH05, LH09, QX09, and QX10 exhibited unacceptable carcinogenic (*CR*) and non-carcinogenic risks (*HI*) based on total metal(loid)s concentrations. However, most heavy metal(loid)s were proved non-bioaccessible through oral ingestion, inhalation, and dermal contact, challenging the over-conservative use of total concentrations in risk assessment. Considering bioaccessible metal(loid)s, *CR* values decreased from 0.181×10^{-4}–1.371×10^{-4} to 0.023×10^{-4}–0.080×10^{-4}, and *HI* decreased from 0.163–3.400 to 0.069–1.328 for children. Spatial interpolation underscored variability in health risks, shaped by geographical and climatic conditions, soil properties, and toxic metal(loid)s concentrations, prompting our proposal for a more comprehensive area-specific health risk assessment protocol that integrates bioaccessible metal(loid)s considerations. These findings encapsulated a comprehensive and feasible risk assessment procedure, and provided valuable insights for refining approaches, remediating contaminated soils, and optimizing local and regional environmental policies.

Graphical Abstract



Keywords Soil · Heavy metal(loid)s pollution · Health risk assessment · In vitro digestion · Spatial interpolation

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Introduction

Heavy metal(loid)s play crucial roles in diverse industrial processes. For instance, chromium (Cr) and nickel (Ni) find applications in electroplating (Zhao et al. 2019), lead (Pb) in batteries and alloys (Cheng and Hu 2010), and arsenic (As) in pesticide and fertilizer manufacturing (Rahaman et al. 2022). These metal(loid)s contribute to environmental challenges stemming from open-air waste weathering, sewage discharges, and polluted air precipitation, posing significant environmental issues (Huang et al. 2015). Soil heavy metal(loid)s often go unnoticed due to their lack of flavor, odor, and color. Even worse, their persistence and difficulty in removal once introduced into soil ecosystems (Jiang and Xu 2013), present ongoing threats to public health and the environment. Contaminated urban soil poses threat to local residents, which has attracted global attention, sparking increased research interest (Cai et al. 2016; Moller et al. 2005).

Various modelling techniques, including the risk exposure model, Monte Carlo simulation, and positive matrix factorization, have been employed to evaluate public health risks associated with soil heavy metal(loid)s through incidental ingestion or food-chain accumulation (Ding et al. 2022; El Fadili et al. 2024; Liang et al. 2023a). The risk-exposure model, originating from the US Environmental Protection Agency (USEPA), dissects the incidental ingestion exposures of local residents from polluted soil, considering pathways like oral ingestion, inhalation and dermal contact (US Environmental Protection Agency 1989; Liang et al. 2023b). Among these, oral ingestion is recognized as the primary route (Wang et al. 2023; Yang et al. 2022). Notably, industrial and transportation dust, as well as precipitation, may serve as significant potential sources of heavy metal(loid)s exposure via inhalation and dermal contact, posing health risks to vulnerable groups including infants, children, municipal sanitation workers, and associated factory employees without adequate protection (Gyimah et al. 2022; Moller et al. 2005). Although human health risk assessments have predominantly focused on total heavy metal(loid)s concentrations in soil, the bioaccessibility of toxic metal(loid)s (BAc, the ratio of bioaccessible and total metal(loid)s) is greatly influenced by the properties of ingested soil. A more rational protocol for population health risk assessment considering bioaccessible factors has been underutilized (Villegas and Zagury 2023), despite a substantial research on BAc evaluation in environment studies (Cui and Chen 2011; Jardine et al. 2013; Yu et al. 2012).

Spatial interpolation technology has been extensively applied in the fields of surveying, mapping and geo-information sciences, and it has also been integrated into the realm of soil environmental science to model the spatial distribution of pollutants (Cheng et al. 2007). Spatial interpolation serves as a vial tool for creating continuous area cartogram, effectively representing multi-point datasets. One specific interpolation method, known as Inverse Distance Weighting (IDW), utilizes an inversely proportional distance weighting scheme to provide accurate estimates and predictions of limited pollutant concentration and environment outcomes (Shokr et al. 2022). However, few endeavors aimed at assessing areal health risk while considering BAc have been made, which is of significant implications for both environmental preservation and human health.

The BAc of heavy metal(loid)s is primarily influenced by their chemical fractions and soil characteristics. For example, in alkaline soil, BAc of Pb and Cd significantly reduced due to soil particle repulsion, with increased BAc as soil pH decreased (Cui et al. 2023). Conversely, higher bioaccessible fraction of arsenate oxyanion was observed in acidic soil conditions (Das et al. 2013). Literatures on soil properties' impact on metal(loid)s BAc may sometimes yield conflicting results. For instance, deprotonated carboxyl and hydroxyl functional groups, carrying net negative charges, may increase arsenate BAc (Lu et al. 2011). However, soil organic matter (SOM) can decrease oxyanion BAc through cation bridge formation (Peel et al. 2017; Xiao et al. 2023). Thus, when assessing health risks, bioaccessible levels of heavy metal(loid)s prove a superior index compared to total concentrations.

In vitro assays are a valuable tool for evaluating the impact of human digestion on contaminants, overcoming limitations of in vivo approaches such as ethical concerns, extended time requirements, and poor repeatability (Li et al. 2015). This method evaluates the fraction of contaminants in the soil matrix that can be absorbed by the human body, representing bioaccessible metal(loid)s. The Unified BARGE Method (UBM), proposed by the Unified Bioaccessibility Research Group of Europe, was proved a practical and efficient for assessing dissolution and adsorption risks of heavy metal(loid)s, saving time and labor, showing strong correlations with in vivo results (Denys et al. 2012; Zhu et al. 2019). Although efforts have improved BAc understanding to reduce assessment errors (Ruby et al. 1996; Tian et al. 2020), health risk assessment with BAc considerations for soil heavy metal(loid)s contamination are rarely included in practice.

Hence, the primary objective of this study was to formulate a comprehensive protocol for assessing areal health risk, incorporating BAc, in regions affected by heavy metal(loid)s pollution. The specific objectives were to: (1) determine total concentrations of heavy metal(loid)s, identify their sources, and evaluate the associated health risk in two designated areas; (2) establish correlations between soil properties and bioaccessible fraction of heavy metal(loid)s; (3) conduct assessments and create health risk maps, considering total and bioaccessible metal(loid)s, in the vicinity of chemical industry parks in Nanjing, Southeast China.

Materials and Methods

Areal Environment, Sample Sites, and Soil Characterization

Sample areas 1 and 2 are adjacent to chemical plant parks, operational for a dozen and over 80 years, respectively. Positioned along the Yangtze River, one lies in the north and the other in the south. According to information from the chemical company's official websites, they employ various raw materials like crude oil, minerals, light hydrocarbons, and natural gas for production petrochemicals, smelting products, asphalt, fertilizer, and more. Transportation involves pipelines, internal trains, and land and water transport. Due to the presence of assisted living facilities and communities nearby, heterogeneity in the pollution landscape is inevitable.

Hence, sampling sites were strategically selected in the proximity of these two chemical plants situated in Nanjing, Southeast China. Soil samples were collected from diverse locations, encompassing areas outside the factory, road greenbelts, abandoned vegetable plots, and residential neighborhoods. The collected surface soil samples (0–20 cm), 10 from area1 and 11 from area 2, were air-dried, ground and sieved through a 60-mesh sieve. These soil samples were subsequently labeled as LH01–LH10 and QX01–QX11, respectively.

Soil pH was determined in a 1:2.5 (w/v) aqueous solution using Orion A211 pH meter (Thermo Scientific, MA, USA). Soil cation exchangeable capacity (CEC) and SOM were measured using ammonium chloride-ammonium acetate, and potassium dichromate oxidation methods, respectively (Pansu and Gautheyrou 2007). Soil total carbon (TC) and total nitrogen (TN) were analyzed following the Dumas combustion protocol (NC 600, Shanghai Baoying Scientific, China) (Cui et al. 2023). Dithionite-citrate-bicarbonate solution was used to extract soil free Fe, Al and Mn oxides (Pansu and Gautheyrou 2007). HF-$HClO_4$-HNO_3, aqua regia and sodium carbonate-sodium hydroxide solutions were used for digesting total concentrations of Pb, Cu, Cr, Cd, Ni, As, Cr(VI), Fe, Al, and Mn, respectively (Pansu and Gautheyrou 2007; Shi et al. 2003; US Environmental Protection Agency 1996). Atomic absorption spectroscopy (AAS, novAA350, Analytik Jena AG, Germany), inductively coupled plasma-optical emission spectrometry (ICP-OES, Optima 8000, PerkinElmer, Inc., USA), and hydride generation atomic fluorescence spectroscopy (HG-AFS, AFS-230E, Beijing Haiguang Instrumental Company, China) were employed for the concentrations of Pb/Cu/Cr/Cd/Ni, Fe/Al/Mn, and As determination, respectively. The concentration of Cr(VI) was determined using atomic absorption spectroscopy after Cr(III) precipitation (Shi et al. 2023a).

Nemerow Pollution Index

Individual pollution index (P_i) and Nemerow composite pollution index (P_N) were employed to evaluate the pollution level of the 21 sample soils, as described by Eqs. 1 and 2 below.

$$P_i = \frac{C_i}{S_i} \tag{1}$$

where C_i is the concentration of the i-th heavy metal(loid)s in soil (mg kg^{-1}); and S_i denotes the evaluation criterion for the i-th heavy metal(loid)s (mg kg^{-1}). The specific background values of soil heavy metal(loid)s in Nanjing, as reported by Wu et al. (2007), were utilized in this study. Pollution levels were classified as insignificant, slight, mild, moderate, and extreme, corresponding to Pi values within the ranges $P_i \le 1$, $1 < P_i \le 2$, $2 < P_i \le 3$, $3 < P_i \le 5$, and $P_i > 5$, respectively (Nemerow 1974).

$$P_N = \sqrt{\frac{(P_i)_{ave}^2 + (P_i)_{max}^2}{2}} \tag{2}$$

where $(P_i)_{ave}$ and $(P_i)_{max}$ signifies the mean and maximum values of P_i, within the dataset, respectively. The insignificant, slight, mild, moderate, and extreme pollution is referred at $P_N \le 0.7$, $0.7 < P_N \le 1$, $1 < P_N \le 2$, $2 < P_N \le 3$, $P_N > 3$, respectively (Nemerow 1974).

Health Risk Assessment

The health risks associated with heavy metal(loid)s are influenced by oral ingestion, inhalation and dermal contact. The daily exposure to contaminated soil particles through each of these pathways can be estimated as follows (US Environmental Protection Agency 1989):

$$ADD = ADD_{oral} + ADD_{inh} + ADD_{dermal} \tag{3}$$

where ADD_{oral}, ADD_{inh} and ADD_{dermal} are the daily intake of heavy metal(loid)s Pb, Cu, Cr, Cd, Ni, and As (mg kg^{-1} d^{-1}) through oral ingestion, inhalation and dermal contact pathways, respectively. Subsequently, assessing the carcinogenic risk (CR) and non-carcinogenic risk (HI) associated with these heavy metal(loid)s from soil particles involves consideration of human exposure parameters and pollutant toxicity values (US Environmental Protection Agency 2007).

$$CR_i = CR_{oral} + CR_{inh} + CR_{dermal} \quad (4)$$

$$CR = \sum CRi \quad (5)$$

$$HQ_i = HQ_{oral} + HQ_{inh} + HQ_{dermal} \quad (6)$$

$$HI = \sum HQi \quad (7)$$

CR_{oral}, CR_{inh}, and CR_{dermal} signify the total carcinogenic risks associated with heavy metal(loid)s through oral ingestion, inhalation and dermal contact, respectively. HQ_{oral}, HQ_{inh}, and HQ_{dermal} represent the total non-carcinogenic risks for heavy metal(loid)s via various exposure pathways. CR_i and HQ_i indicate the specific carcinogenic and non-carcinogenic risks associated with the i-th heavy metal(loid)s in the soil sample, respectively. Unacceptable, acceptable, and non-significant carcinogenic risks are referred to as $CR > 1 \times 10^{-4}$, $1 \times 10^{-6} < CR < 1 \times 10^{-4}$, and $CR < 1 \times 10^{-6}$, respectively. $HI \leq 1$ and $HI > 1$ indicate potential non-carcinogenic risks do not and possibly occur, respectively (US Environmental Protection Agency 2020). Detailed human exposure parameters and pollutant toxicity values are provided in supplementary materials and methods, Tables S2 and S3, respectively.

In Vitro BAc Assay

The UBM method was employed to assess the bioaccessible metal(loid)s through oral ingestion. Briefly, 0.600 g soil sample (< 250 μm) was weighed into a 100 ml polypropylene centrifugation tube, 9 ml stimulated saliva solution was added with 30 s vigorous hand shaking. Subsequently, 13.5 ml stimulated gastric solution was introduced into the mixture, and the pH was adjusted to a range of 1.1–1.3 using concentrated hydrochloric acid. The mixture underwent homogenization through agitation on a vertically rotating shaker (QB128 rolling incubator, Kylin-Bell Ltd., China) at 37 °C for 1 h. Afterward, 27 ml stimulated duodenal juice and 9 ml bile were added to the mixture, maintaining the suspension pH within 5.8–6.3, and left for 4 h. The supernatant was filtered through 0.45 μm filters after centrifugation, stored at 4 °C for subsequent analysis (Shi et al. 2023b). Gamble solution (simulating the extracellular environment of alveol) and biomimetic sweat (simulating apocrine sweat) were utilized to investigate the bioaccessible metal(loid)s through inhalation and dermal contact, respectively. Each portion of 0.400 g (< 100 μm) and 1.000 g (< 425 μm) soil samples were mixed with 40 ml gamble solution and 10 ml biomimetic sweat, shaken at 37 °C and 150 rpm, for 24 and 2 h, respectively (Yu et al. 2022). The supernatant was filtered through 30–50 μm fiber filters after centrifugation, stored at 4 °C for further analysis.

Data Quality Control, IDW Spatial Interpolation, and Statistical Analysis

All experiments were performed in duplicate, and the resulting averages were recorded. The accuracy of heavy metal(loid)s concentrations was guaranteed using certified reference materials for chemical composition of soil (GBW07405) and a blank sample for correction, consistently yielding values within the 90–110% range of the certified values. The standard solutions from Guobiao Testing & Certification Co., Ltd., Beijing, China, used for metal(loid)s determination via AAS, ICP-OES and HG-AFS methods were tested after every tenth sample to ensure the accuracy of the analysis. Spatial maps depicting health risks were generated through IDW interpolation, implemented in ArcGIS 10.2 (ESRI, Redlands, CA, USA). Principal component analysis (PCA) incorporated heavy metal(loid)s concentrations from soil samples as input variables. Kaiser–Meyer–Olkin (KMO) statistics (> 0.5) and Bartlett's test (< 0.05) were applied to confirm the adequacy of PCA (Field 2000). Statistical analysis, including Spearman and Pearson correlation analysis after Shapiro–Wilk test, one-way analysis of variance with Duncan's multiple range test, PCA, KMO and Bartlett' test, were performed using IBM SPSS 26.0 software (IBM Corp., Armonk, NY, USA). All figures, except for the spatial map, were generated using Origin 2018 software (Origin Lab, Northampton, MA, USA).

Results and Discussion

Soil Background Investigation

The basic physicochemical properties of the studied soil samples (classified as Alfisol) are presented in Table 1. The data indicated a wide range for soil CEC and SOM, spanning from 8.40 to 20.70 and 6.25 to 16.27 cmol kg^{-1}, and from 5.73 to 53.38 and 13.92 to 194.57 g kg^{-1}, in the studied area 1 and area 2, respectively. Notably, QX08 and QX10 exhibited particularly high SOM contents, reaching 194.57 and 100.34 g kg^{-1}, respectively, attributed to historical vegetable garden cultivation.

Furthermore, TC and TN were determined within the ranges of 1.94–58.76 and 0.27–1.54 g kg^{-1} in area 1, and 7.84–81.98 and 0.54–3.37 g kg^{-1} in area 2, respectively. The

Table 1 Physicochemical properties of the studied soils collected from two areas

Area	Sample	pH	SOM (g kg^{-1})[a]	CEC (cmol kg^{-1})[b]	Fe (g kg^{-1})	Al (g kg^{-1})	Mn (g kg^{-1})	Free Fe (g kg^{-1})[c]	Free Al (g kg^{-1})[d]	Free Mn (g kg^{-1})[e]	TC (g kg^{-1})[f]	TN (g kg^{-1})[g]
Area 1	LH01	8.15	25.72	17.43	10.60	4.35	0.26	14.36	1.96	0.52	15.31	1.38
	LH02	8.57	5.73	14.15	16.82	4.92	0.35	14.80	2.08	0.51	1.94	0.27
	LH03	7.59	23.57	15.54	60.62	5.60	0.90	39.68	2.22	0.42	13.10	1.28
	LH04	7.94	34.30	11.90	17.64	2.77	0.92	15.46	1.77	0.81	25.89	1.45
	LH05	8.01	39.21	15.76	22.75	2.71	0.34	31.70	2.01	0.44	22.10	1.54
	LH06	7.64	21.23	20.70	15.64	4.12	0.46	16.64	1.78	0.49	12.39	1.50
	LH07	8.15	16.52	14.98	16.60	4.83	0.43	17.45	2.02	0.54	8.62	1.00
	LH08	8.04	37.88	8.40	19.42	5.64	0.36	15.07	1.52	0.30	58.76	1.49
	LH09	7.57	27.19	12.84	10.75	5.60	0.51	10.91	1.36	0.40	11.80	1.02
	LH10	8.09	53.38	11.05	33.05	5.31	0.78	29.75	1.62	0.48	32.51	1.30
Area 2	QX01	8.15	24.54	13.09	17.64	5.04	0.51	17.41	2.01	0.52	7.84	0.54
	QX02	8.37	28.98	13.45	18.90	3.48	0.57	13.13	1.90	0.36	23.87	2.01
	QX03	8.60	16.35	12.20	21.64	6.34	0.60	14.26	1.94	0.48	13.24	0.68
	QX04	7.65	21.86	13.97	18.30	6.23	0.51	9.80	1.06	0.44	11.49	1.42
	QX05	7.99	35.02	15.23	20.60	5.07	0.75	12.50	1.54	0.56	20.69	1.98
	QX06	8.41	13.92	10.61	24.23	5.00	0.73	15.19	1.69	0.57	10.09	0.59
	QX07	8.59	20.03	10.46	23.86	4.12	0.85	14.76	1.49	0.57	12.96	0.66
	QX08	7.55	194.57	8.42	20.08	5.01	0.96	12.04	1.67	0.28	81.98	3.37
	QX09	8.27	79.01	6.25	18.16	3.73	0.64	11.98	1.60	0.32	54.50	1.70
	QX10	7.95	100.34	16.27	20.16	4.32	0.85	14.28	1.59	0.49	56.10	3.08
	QX11	8.16	29.80	12.15	10.60	5.17	0.30	16.65	1.83	0.64	14.03	1.10

[a]SOM: Soil organic matter;

[b]CEC: Cation exchange capacity;

[c]Free Fe: Free iron oxides, calculated as Fe_2O_3;

[d]Free Al: Free aluminum oxides, calculated as Al_2O_3;

[e]Free Mn: Free manganese oxides, calculated as MnO;

[f]TC: Total carbon;

[g]TN: Total nitrogen

concentrations of free metal oxides displayed variability, ranging 10.91–39.68 and 9.80–17.41 g kg^{-1} Fe, 1.36–2.22 and 1.06–2.01 g kg^{-1} Al, 0.30–0.81 and 0.28–0.64 g kg^{-1} Mn in area 1 and 2, respectively. Pearson correlation analysis among soil properties revealed strong correlations between TC and TN with SOM, with correlation coefficients (r) of 0.88 ($P<0.01$) and 0.83 ($P<0.01$), respectively (Fig. S2).

The average concentrations of heavy metal(loid)s (mg kg^{-1}) in area 1 and area 2 followed the order: Pb (201) > Cu (141) > Cr (105) > As (42) ≈ Ni (35) > Cd (4) ($P<0.05$), and Pb (123) > Cr (82) > Cu (56) ≈ As (37) ≈ Ni (37) > Cd (5) ($P<0.05$), respectively (Fig. 1). The average values (excluding As) were below the risk screening values outlined in the Soil Environmental Quality-Risk Control Standard for Soil Contamination of Development Land of China (GB 36600–2018), which set Pb, Cu, Ni, and Cd levels at 400, 2000, 150, and 20 mg kg^{-1}, respectively, for the first type of development land (sensitive land). However, it's noteworthy that concentrations of Pb in LH05, LH09, as well as As in all soil samples, exceeded the screening values (with As set at 20 mg kg^{-1}) but remained lower than the intervention values (set at 800 and 120 mg kg^{-1} for Pb and As, respectively). Consequently, the total concentration of heavy metal(loid)s in the studied areas raises concerns regarding potential environmental and human health implications.

Pollution Source Analysis

Previous studies have demonstrated that source apportionment of heavy metal(loid)s helps quantify the contribution of pollution sources stemming from both natural and anthropogenic activities. Spearman correlation analysis unveiled significant positive correlations ($P<0.05$) among the concentrations of Cd-As, Cd–Pb, and Cu-As in area 1 (Fig. 2a), while Cd and Ni concentrations displayed a significant negative correlation ($P<0.05$). These findings suggest a strong

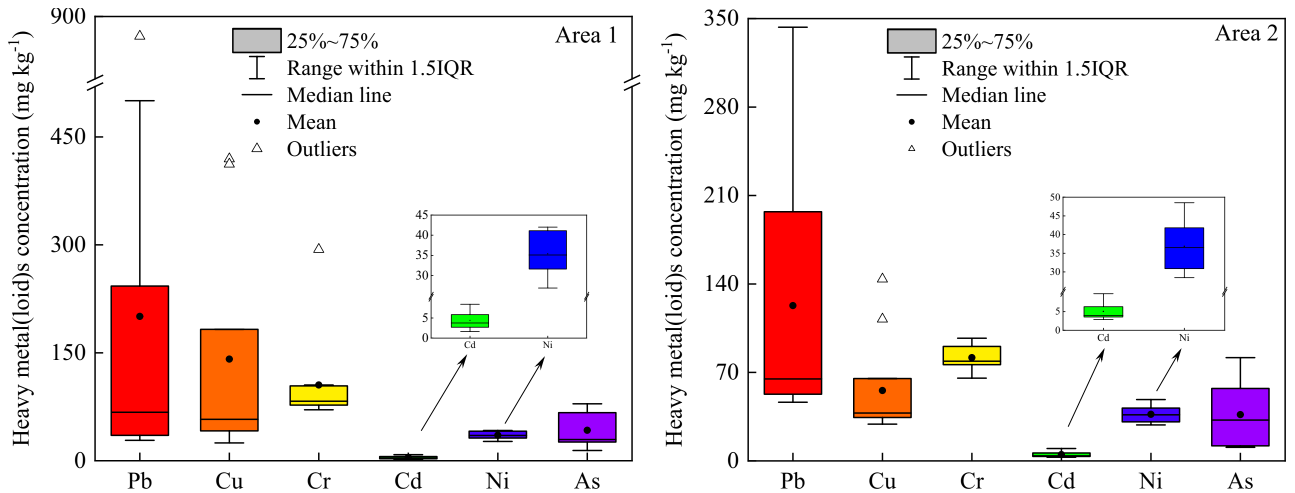

Fig. 1 The concentration of heavy metal(loid)s (Pb, Cu, Cr, Cd, Ni and As) in the two studied areas. The bottom and top of boxes are the 25th and 75th percentiles, respectively. The box represents interquartile range (IQR). Whiskers extending from the bottom and top of the boxes are set at 1.5 times of the IQR. The circle and solid lines inside the box respectively indicate the mean and median metal(loid) s concentrations

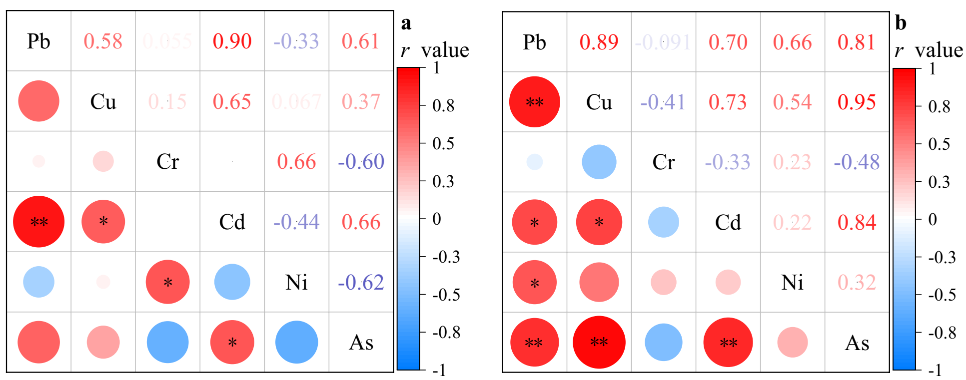

Fig. 2 Heat map of heavy metal(loid) concentrations in area 1 (**a**) and 2 (**b**). The * and ** on the grid indicated significance at 0.05 and 0.01 levels, respectively

connection between Cd, As, Pb, and Cu, likely originating from a common source of these metal(loid)s, contrasting with Cr and Ni (Yan et al. 2022). Similarly, concentrations of Cd, As, Pb, and Cu in area 2 were significantly positively correlated with each other ($P<0.05$, Fig. 2b), further emphasizing the likelihood of a shared source of these metal(loid)s.

The KMO value was 0.605 (>0.5) and Bartlett's significance yielded a P-value of 1.46×10^{-4} (<0.05), collectively affirmed the adequacy of the sample size, uniformity of variances across samples, and the reliability of metal(loid)s concentrations for PCA analysis (Field 2000). The first three principal components (PC1 + PC2 + PC3) accounted for a substantial 83.31% of the total variance (Fig. S3 and Table S1). PC1, representing 46.00% of the total variance, exhibited robust positive loadings (>0.73) for Pb, Cu, Cd, and As, suggesting a comparable anthropogenic source for these metal(loid)s. This aligns with the earlier observations of a significant positive correlation among these metal(loid)s, indicating a shared source, possibly from activities such as petroleum processing, metal smelting, dust deposition, mining, and transportation (Du and Lu 2022; Guo and He 2013; Zhang et al. 2021). In contrast, PC2, explaining 22.76% of the total variance, revealed significant positive factor loadings for Cr (0.87) and Ni (0.65), implying a similar geological origin for these elements (Cox et al. 2017; Oze et al. 2004; Sun et al. 2019).

Quick Evaluation of Pollution Level

The analysis of heavy metal(loid) concentrations and sources suggests that the studied soils may be contaminated. However, the Nemerow index method, with compares the concentrations in the soil samples to background levels, is a valuable and efficient tool for assessing the environmental contamination levels due to individual and combined heavy metal(loid)s (Nemerow 1974). This method provides a convenient and feasible approach for assessing the degree of

Table 2 Heavy metal(loid) concentrations and pollution levels assessed by single/Nemerow-composite pollution index

Area	Statistics	P_i[c]						P_N[d]
		Pb	Cu	Cr	Cd	Ni	As	
Area 1	Max[a]	32.41	16.14	4.05	6.72	1.39	8.59	23.52
	Min[b]	1.06	0.95	0.98	1.64	0.89	1.55	1.29
	Mean	7.49	5.43	1.45	3.95	1.17	4.59	8.72
		Extreme[h]	Extreme	Slight[e]	Moderate[g]	Slight	Moderate	Extreme
Area 2	Max	12.81	5.53	1.34	9.83	1.61	8.88	9.67
	Min	1.73	1.12	0.90	2.33	0.94	1.17	1.23
	Mean	4.80	2.22	1.11	5.03	1.21	4.25	5.22
		Moderate	Mild[f]	Slight	Extreme	Slight	Moderate	Extreme

Max[a]: Maximum value; Min[b]: Minimum value; P_i[c]: Single pollution index; P_N[d]: Nemerow composite pollution index; Slight[e], Mild[f], Moderate[g], Extreme[h]: The slight, mild, moderate, and extreme pollution was referred at $1<Pi\leq2$, $2<Pi\leq3$, $3<Pi\leq5$, and $Pi>5$, respectively. The corresponding P_N values were $0.7<P_N\leq1$, $1<P_N\leq2$, $2<P_N\leq3$, $P_N>3$, respectively

pollution in countries and districts lacking national pollutant control standards, as well as in regions with elevated background pollution levels (Fei et al. 2023). The results are presented in Table 2. The Nemerow index for the six metal(loid)s followed the order of Pb > Cu > As > Cd > Cr > Ni in area 1 and Cd > Pb > As > Cu > Ni > Cr in area 2, respectively. The higher pollution levels of the first four elements (Pb, Cu, As, and Cd) further support their comparable anthropogenic source, with concentrations exceeding local background values by 2.22–7.49 times, as reported in previous studies (Liu et al. 2019; Shil and Singh 2019). In contrast, the concentrations of Cr and Ni were only 1.11–1.45 times the background levels, combining the observations of PCA analysis, further implying their likely natural origin. The average values of P_N for area 1 and 2 were 8.72 and 5.08, respectively, denoting an extreme pollution level in these areas (Wu et al. 2021). Therefore, assessing the health risks associated with heavy metal(loid)s from accidental exposure is of great practical significance.

Health Risk Assessment Using Total/Bioaccessible Metal(loid)s

The results in Table 3 showed the *CR* and *HI* values for adults and children in their respective areas. The concentration of Cr(VI) accounted for only 0–1.18% of the total Cr (Fig. S1). Consequently, Cr(III) toxicity parameters were applied to assess the health risk of Cr, aligning with the approach proposed by Villegas and Zagury (2023). For adults, the assessment indicated an acceptable carcinogenic risk ($CR = 0.102\times10^{-4}$–0.565×10^{-4}, 0.084×10^{-4}–0.589×10^{-4}) and a non-significant non-carcinogenic risk ($HI = 0.020$–0.365, 0.023–0.147) in the two areas. However, the health risk for children and adolescents was notably higher, with *CR* and *HI* values ranging from 0.238×10^{-4}–1.315×10^{-4}, 0.181×10^{-4}–1.371×10^{-4}, and 0.163–3.400, 0.213–1.368 in area 1 and 2, respectively. Therefore, both the Nemerow index and USEPA health risk assessment consistently indicate that individuals exposed to these soils face a significant health risk.

Clearly, the contaminated soils pose significantly greater health hazards and risks to children and adolescents, who are more susceptible to the effects of heavy metal(loid)s (Wang et al. 2021). This heightened vulnerability is reflected in the higher values of *CR* and *HI*. This susceptibility can be attributed to the higher *ADD* value for children, primarily due to their increased consumption of soil containing heavy metal(loid)s per unit of body weight (US Environmental Protection Agency 2002). For instance, although the daily intake of soil and the ingestion of polluted air through oral ingestion and inhalation were 2 times greater for children (100 and 200 mg d^{-1}) and 2.61 times (20 and 7.65 m^3 d^{-1}), respectively, compared to adults, the weight ratio between the two groups was significantly higher, up to 4.67 times (70 kg for adults compared to 15 kg for children). This weight disparity plays a crucial role in the increased vulnerability of children and adolescents to the health risks associated with heavy metal(loid)s exposure.

In the aforementioned USEPA health risk assessment exposure model, the total concentration of heavy metal(loid)s in soil samples was employed, assuming that the loaded heavy metal(loid)s could be ingested and absorbed by the human body at nearly 100%. However, it's crucial to recognize that the absorption of heavy metal(loid)s from the soil matrix can vary significantly due to different levels of heavy metal(loid)s BAc within various soils. This variability is influenced by a combination of factors, including soil characteristics, heavy metal(loid)s types, concentrations and fractions, as well as environmental conditions (Cui et al. 2023; Shi et al. 2023a). Therefore, conducting a health risk

Table 3 Health risk assessment without and with bioaccessibility consideration of the population in the studied two areas

Area	Sample	With BAc consideration				Without BAc consideration			
		CR ($\times 10^{-4}$)[a]		*HI*[b]		*CR* ($\times 10^{-4}$)		*HI*	
		Adult	Child	Adult	Child	Adult	Child	Adult	Child
Area 1	LH01	0.011	0.025	0.011	0.100	0.193	0.450	0.020	0.189
	LH02	0.012	0.028	0.010	0.091	0.186	0.432	0.018	0.163
	LH03	0.018	0.042	0.015	0.138	0.565	1.315	0.074	0.687
	LH04	0.013	0.029	0.014	0.128	0.210	0.488	0.030	0.280
	LH05	0.036	0.084	0.142	1.328	0.508	1.183	0.365	3.400
	LH06	0.016	0.037	0.014	0.134	0.366	0.851	0.028	0.259
	LH07	0.013	0.031	0.011	0.106	0.102	0.238	0.018	0.170
	LH08	0.011	0.025	0.009	0.081	0.213	0.495	0.038	0.353
	LH09	0.034	0.080	0.114	1.061	0.544	1.266	0.208	1.934
	LH10	0.020	0.046	0.034	0.315	0.188	0.437	0.108	1.004
Area 2	QX01	0.012	0.029	0.008	0.072	0.084	0.196	0.031	0.291
	QX02	0.013	0.029	0.008	0.077	0.231	0.538	0.028	0.258
	QX03	0.011	0.026	0.009	0.082	0.086	0.201	0.025	0.231
	QX04	0.012	0.027	0.007	0.069	0.078	0.181	0.023	0.213
	QX05	0.011	0.025	0.011	0.105	0.211	0.491	0.029	0.268
	QX06	0.012	0.028	0.017	0.159	0.116	0.270	0.045	0.419
	QX07	0.012	0.029	0.020	0.187	0.365	0.849	0.061	0.561
	QX08	0.010	0.023	0.006	0.060	0.414	0.964	0.091	0.841
	QX09	0.013	0.030	0.008	0.077	0.461	1.072	0.147	1.368
	QX10	0.011	0.025	0.011	0.106	0.589	1.371	0.114	1.056
	QX11	0.011	0.025	0.012	0.113	0.270	0.628	0.033	0.303

[a] *CR* ($\times 10^{-4}$): Sum of carcinogenic risk;

[b] *HI*: Sum of non-carcinogenic risk

assessment solely based on the total pollutant load, while neglecting the intricate processes of human digestion and absorption, inevitably leads to an overestimation of the potential health deterioration (Martinez-Sanchez et al. 2013; Villegas and Zagury 2023).

The bioaccessible concentrations of the studied heavy metal(loid)s through three exposure pathways are presented in Fig. 3. The result emphasized the prevalence of a stable non-bioaccessible fraction of heavy metal(loid)s as the primary form. For instance, non-bioaccessible Pb fractions, considering oral ingestion, inhalation, and dermal contact, accounted for 11.91–94.63%, 57.34–98.81%, and 95.89–100% of the total concentration of Pb in soils, respectively (Fig. 3). This is consistent with findings from previous studies reporting that non-bioaccessible Pb constitutes a significant proportion, ranging from 58 to 70% via oral ingestion (Cui et al. 2023), 35 to 88% via inhalation (Leal et al. 2018), and 87 to 98% via dermal contact (Pelfrene and Douay 2018), respectively.

A notable difference in bioaccessible concentrations existed among the various heavy metal(loid)s (Fig. 3), where inhalation and dermal contact resulted in a lower bioaccessible fraction for each heavy metal(loid)s compared to the oral ingestion pathway. Specifically, the bioaccessible concentrations of metal(loid)s were in the order of Pb (56.35 ± 19.41 mg kg^{-1}) > Cu (29.67 ± 7.47 mg kg^{-1}) > Cr (1.53 ± 0.75 mg kg^{-1}) ≈ Cd (0.31 ± 0.06 mg kg^{-1}) ≈ Ni (2.48 ± 0.23 mg/ kg^{-1}) ≈ As (1.77 ± 0.12 mg kg^{-1}) ($P < 0.05$). Hence, the BAc of these metal(loid)s were 42.59 ± 5.11%, 32.96 ± 1.50%, 1.15 ± 0.24%, 6.01 ± 0.89%, 6.90 ± 0.57%, 6.29 ± 0.87% for Pb, Cu, Cr, Cd, Ni, and As, respectively. Notably, the heavy metal(loid)s Cr and Ni, originating naturally, had lower average BAc levels in most soil samples. This can be attributed to their inert properties, resulting in a slow release rate.

Pearson correlation analysis conducted between BAc and the measured soil properties in this study revealed strong correlations between BAc and various soil characteristics, including soil pH, CEC, and the concentrations of SOM, TN, TC, and free Fe, Al and Mn oxides. For instance, the BAc of Pb through oral ingestion exhibited an extremely significant negative correlation with SOM concentration, with an *r* value of − 0.622 ($P < 0.01$), which may be attributed to the high affinity between Pb and surface functional groups (e.g.,

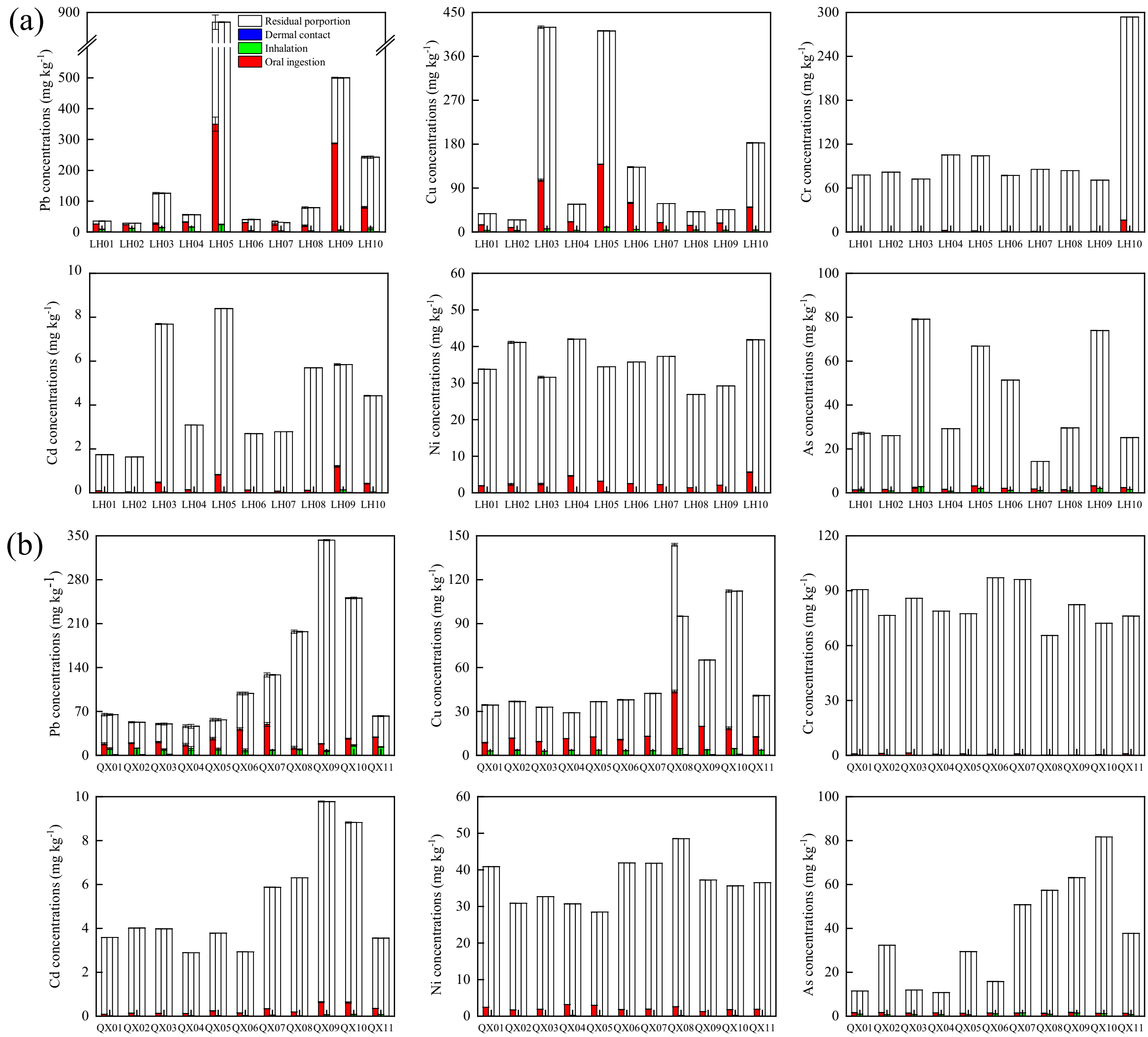

Fig. 3 Bioaccessible concentration of heavy metal(loid)s obtained through three different pathways in areas 1 (**a**) and 2 (**b**), respectively

carboxyl and hydroxyl groups) present on SOM (Cai et al. 2016; Cui et al. 2023; Cui and Chen 2011). However, another intriguing observation was the significant positive correlation between the BAc of Cr, through both inhalation and dermal contact, and the concentration of SOM, with r values of 0.637 and 0.959 ($P < 0.01$), respectively (Fig. S4).

The total Fe, Al and Mn, as well as their oxides, have been suggested to control the BAc of metal(loid)s in the gastric phase (Sun et al. 2023; Zhu et al. 2019). However, no significant correlation was observed between the BAc of heavy metal(loid)s and soil Fe, Al and Mn concentrations (Fig. S4). This may be due to their low and similar concentrations, as shown in Table 1. The coefficient of variation, a measure of variability, classifies it as weak (< 10%), moderate (10–100%), or strong (> 100%) (Rosemary et al. 2017). In this study, the variability of total Fe, Al and Mn, as well as their oxides in the 21 soil samples, was weak or moderate, with coefficients of variation of 44, 16, 25%, and 50, 21, 37% for their concentrations, respectively. These findings indicated nuanced interactions between the BAc of heavy metal(loid)s and soil properties, necessitating further investigation to better comprehend these inconsistencies. Insights gained from additional works could prove valuable in predicting the BAc properties of heavy metal(loid)s in contaminated soil, with practical implications for environmental management and risk assessment.

The findings from the in vitro BAc assay emphasized the crucial role of considering bioaccessible metal(loid)s when assessing health risks associated with heavy metal(loid)s exposure. These results suggested that only a relatively small portion of heavy metal(loid)s can be absorbed by the human body after accidental exposures. Therefore, incorporating bioaccessible metal(loid)s considerations into health risk assessments provide a more rational and accurate estimation of potential health risks, when compared to relying solely on the total amount of heavy metal(loid)s present in the environment (Yu et al. 2012). That said, the *ADD* in Eqs. 3 should be multiplied by the BAc value stands for the bioaccessible concentration of the specific heavy metal(loid)s. These adjustments in health risk assessments indicated a more optimistic evaluation of health risks associated with heavy metal(loid)s exposure. Specifically, the corrected *CR* for adults and children decreased from 0.078–0.589 and 0.181–1.371 to 0.010–0.036 and 0.023–0.084, respectively. Similarly, *HI* decreased from 0.018–0.365 and 0.163–3.400 to 0.006–0.142 and 0.060–1.328, respectively (Table 3). Comparable reductions in health risk have been observed in various environmental contaminants, such as chromite ore processing residue (Yu et al. 2012), lead–zinc ore tailing polluted soil (Liu et al. 2018), and chromated copper arsenate (Villegas and Zagury 2023).

Areal Health Risk Maps Generation

Areal health risk maps were generated for the two industrial areas in Nanjing, Southeast China, using IDW interpolation with and without bioaccessible metal(loid)s consideration (Fig. 4). Notably, the incorporation of bioaccessible metal(loid)s led to a significant reduction in areal health risk, which was of substantial significance to pollution assessment, management, and treatment endeavors. For instance, LH03, LH05, LH09, QX09 and QX10 initially considered to pose an unacceptable carcinogenic risk to children (with $CR > 1 \times 10^{-4}$), were re-assessed to have an acceptable risk level after bioaccessible metal(loid)s consideration (falling within the range of $1 \times 10^{-6} < CR < 1 \times 10^{-4}$). Most prominently, *HI* values for children dropped from 3.400, 1.934, 1.004, 1.368 and 1.056 to 1.328, 1.061, 0.315, 0.077 and 0.106 in LH05, LH09, LH10, QX09 and QX10, respectively. This indicates that significant non-carcinogenic risks were either substantially reduced or eliminated entirely in actual soil samples when bioaccessible metal(loid)s were considered. However, it's crucial to recognize that the BAc

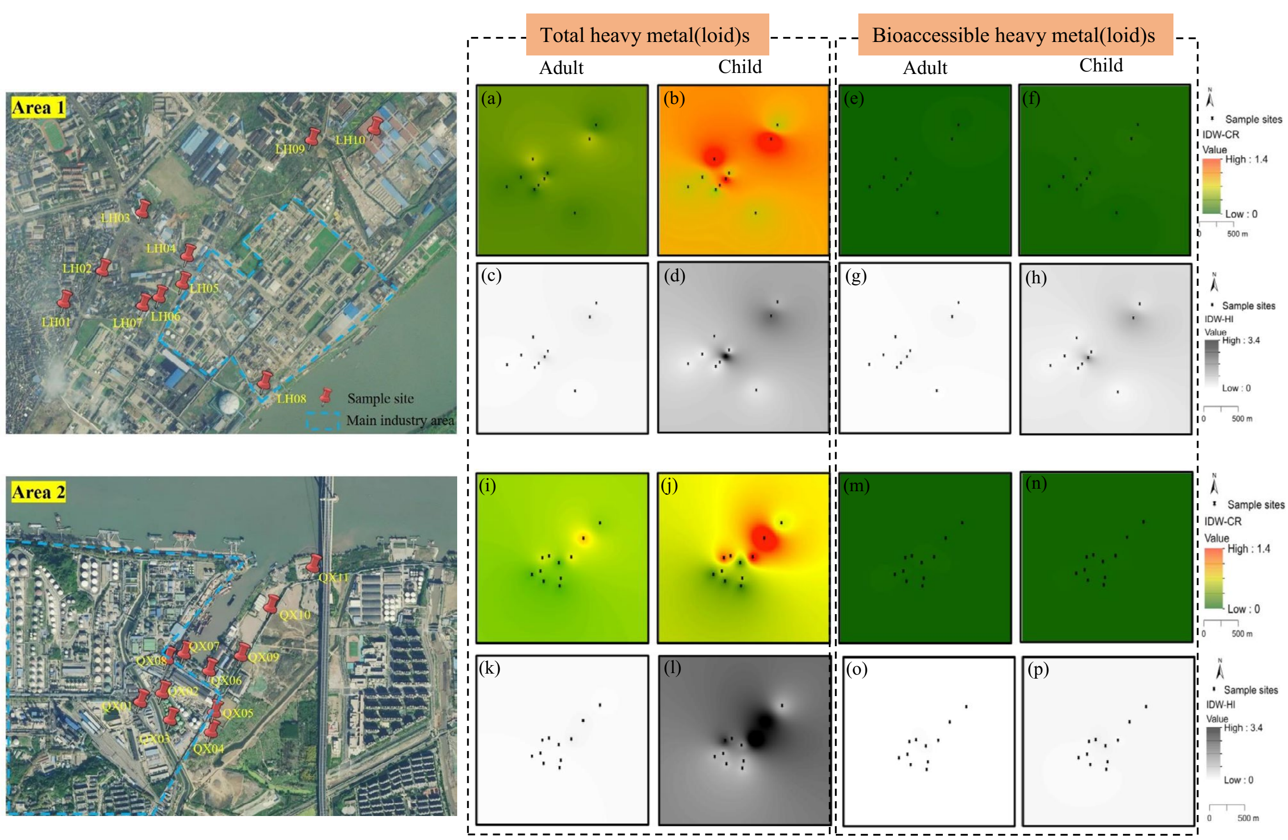

Fig. 4 The geostatistical distribution of health risk associated with heavy metal(loid)s in the two studied areas assessed using IDW. The *CR* ($\times 10^{-4}$) and *HI* values were calculated based on total (a–d, i–l) and bioaccessible (e–h, m–p) concentrations of heavy metal(loid)s

of heavy metal(loid)s can fluctuate due to changes in soil characteristic and prolonged exposure duration. Therefore, the previously dormant non-bioaccessible soil contaminants may become activate, surpassing the toxicological threshold in different soil environments, potentially posing health risks to residents through their daily accidental ingestion of soil particles (Ottosen et al. 2009; Shi et al. 2023a, b).

In Nanjing, the prevailing southeast wind in summer and northwest wind in winter, contributes to the accumulation of polluted soil and dust in the northwest and southeast zones, especially around chemical enterprises (Lu et al. 2013). Notably, LH03, LH05 and LH09 in area 1, as well as QX09 and QX10 in area 2, exhibited higher potential health risks. These areas align with elevated concentrations of priority pollutants such as Pb, Cd, Cu, and As. This emphasized the imperative for effective environmental interventions in these specific zones to mitigate health risks originating from anthropogenic sources for the local population.

An innovative protocol for areal health risk assessment, integrating considerations for bioaccessible heavy metal(loid)s in soil contamination, is proposed. This protocol delineates key distinctions from conventional evaluation methodologies and comprises the following sequential steps: (1) appropriate sample site selection, (2) soil sample collection and treatment, (3) analysis of pollution sources, (4) predominant evaluation environmental contamination levels using Nemerow index, (5) health risk assessment with bioaccessible metal(loid)s consideration, and (6) areal health risk map generation.

Conclusions

The study investigated the distribution of heavy metal(loid) s and the associated population health risks in two industrial areas in Nanjing, considering both total and bioaccessible concentrations of heavy metal(loid)s. The findings indicate: (1) Anthropogenic activities, particularly industrial and transportation processes, are the primary sources of priority pollutants such as Pb, Cu, Cd and As; (2) Parameters including *Pi* and P_N, *CR* and *HI* underscore multiple heavy metal(loid)s contaminants in the studied soils, posing a significant health risk to the exposed population; (3) The consideration of bioaccessible metal(loid)s proves practical and efficient for assessing health risks from these metal(loid)s; (4) The risk map, generated using IDW interpolation, reveals the impact of pollution sources, geography, and climate conditions on the distribution of health risks. This proposed protocol, integrated with disciplines, such as geostatistics, geography, climatology, environment, and specific epidemiology, can provide comprehensive assistance in assessing the environmental quality of urban land, improving the health status of residents, and optimizing the remediation of polluted soil.

The novelty of this study lies in its integration of areal health risk assessment, with bioaccessible metal(loid) s considerations and utilizing IDW interpolation. This innovative methodology enhances the effectiveness and comprehensiveness of pollution and health risk assessment in the studied areas. However, it is crucial to emphasize the need for closer attention to the health status of specific vulnerable groups within the exposed local population. This includes, but is not limited to, children, adolescents, and individuals working in sanitation and industrial sectors in pollution hotspots.

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Data Availability Data are available in this paper and/or from the corresponding author upon reasonable request.

Declarations

Conflict of interest The authors have no relevant financial or nonfinancial interests to disclose.

Ethical approval Not applicable.

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