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Original Research

Bioaccessibility and Health Risk Assessment of Inhalable Heavy Metals in Soil from the Bayan Obo Mining Area, Northern China

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Abstract

Soil contamination caused by mining activities may pose potential risks to human lungs. This study investigated the pollution levels and inhalable bioaccessibility of selected metal elements in the soils of Bayan Obo, a representative mining city, and Baotou, an industrial city in China. The human health risks of metal elements were also evaluated. The results indicated that La, Ce, and Eu exhibited the highest levels of enrichment, with significant spatial variations. The inhalable bioaccessibility of the metals followed the order: As>Mn>Ni>Pb≈Zn>Cu>Cr>La>Ce>Eu. Health risk assessment results indicated that evaluating human health risks based on total metal concentrations rather than bioaccessible concentrations could not only overestimate the risk of soil metals but, more critically, lead to misidentification of high-risk pollutants. The measured inhalable bioaccessibility data revealed that both non-carcinogenic and carcinogenic risks for children and adults were below safety thresholds, with Mn and As being the primary contributors to non-carcinogenic and carcinogenic risks, respectively.

Keywords: soil pollution, bioaccessibility, health risk assessment, rare earth elements, simulated epithelial lung fluid

Introduction

The sources of heavy metals in soil are diverse and can be categorized into natural sources (volcanic eruptions, rock weathering, atmospheric deposition, and biological processes) and anthropogenic sources (mining activities, vehicle emissions, industrial discharges, and fossil fuel combustion) [1]. Mining activities are a major contributor to heavy metal contamination in soils, making soil one of the primary "sinks" for these pollutants [2]. Heavy metal pollution in soils caused by mining has become a global environmental issue. The concentration

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of heavy metals in mining cities is significantly higher than in non-mining cities and far exceeds that in rural areas. The heavy metal concentrations in mining city soils are closely related to the type of mineral resources. For example, the mining of lead-zinc deposits results in Pb, Zn, and Cd contamination [3], while major pollutants in copper mining areas include Cu, Mo, Ag, and Cr [4]. In rare earth mining, substantial amounts of rare earth elements (REEs) migrate into the soil environment during extraction [5]. Metal concentrations in soil can be classified into "total concentration" and "bioaccessible concentration". The "total concentration" refers to the overall amount of heavy metals in all forms, whereas "bioaccessible concentration" represents the fraction of heavy metals that can be absorbed into the biological circulatory system through dermal contact, ingestion, or inhalation [6]. Metals present in the soil may pose potential health risks to humans [7]. Metals can enter the human body through three primary exposure pathways: ingestion, inhalation, and dermal contact. Since the human body cannot fully absorb the total metal concentration present in soil, the concept of "bioaccessibility" has been introduced [6, 8]. Although ingestion is the primary pathway for human exposure to soil metals, most ingested metals can be excreted through the digestive system [9]. In contrast, inhaled soil particles are more likely to deposit in the respiratory tract. Due to the limited clearance capacity of alveolar tissues, this may trigger a range of diseases [10]. Therefore, compared to oral bioaccessibility, the inhalation pathway poses a potentially higher health risk and warrants greater attention [11].

Prolonged exposure to airborne dust or particulate matter may adversely affect human health through the respiratory tract. Epidemiological studies indicate that such exposure is closely associated with elevated heavy metal concentrations in blood and urine, suggesting that heavy metals can enter the bloodstream through the respiratory tract [12, 13]. Further studies have shown that inhalation of heavy metals may pose health risks. For instance, research by Lee et al. (2024) found that inhalation of toxic metals could lead to reduced lung function and even induce lung cancer [14]. In the study by Onan et al. (2024), inhaled heavy metals accumulated in the kidneys, significantly increasing the incidence of glomerulonephritis [15]. During inhalation, soil particles primarily distribute into at least one of two "compartments" [16]. To simulate the neutral extracellular environment of deep lung regions, a commonly used simulated lung fluid is Gamble's solution (GS) with a pH of 7.4 [17, 18]. Another commonly used simulated lung fluid is artificial lysosomal fluid (ALF), which mimics the lysosomal fluid of alveolar macrophages with a pH of 4.5 [19]. These two simulated fluids have been widely used in assessing human exposure to inhaled pollutants. In subsequent studies, Gamble's solution has been continuously refined, leading to variants such as "modified Gamble's solution" [20], "simulated lung fluid" [21], and "pseudo alveolar fluid" [22]. In these simulated fluid models, the added inorganic salts typically include NaCl, $CaCl_2$, NaH_2PO_4, $NaHCO_3$, KCl, $MgCl_2{\cdot}6H_2O$, and Na_2SO_4, with concentrations closely approximating those found in a healthy human body [23-25]. This study employs the simulated epithelial lung fluid (SELF) developed by Boisa et al. (2014), which maintains the same inorganic salt concentrations as the aforementioned models [16]. Several studies have shown that proteins, organic acids, and certain macromolecular lipids play a crucial role in inhalation bioaccessibility [16, 20, 26, 27]. Albumin and mucin are the most abundant proteins in the human body [16, 28], and they readily bind to metal ions in solution, thereby influencing the bioaccessibility of metals [29]. The primary organic acids in the human body include glycine and cysteine [30, 31]. These organic acids help maintain the balance of simulated fluids and exhibit anti-inflammatory properties during endothelial inflammation [16]. In a study by Cross et al. (1994) [32], the concentrations of ascorbic acid, uric acid, and glutathione in healthy human respiratory tract fluid were reported as 17.6 $mg{\cdot}L^{-1}$, 15.1 $mg{\cdot}L^{-1}$, and 30.7 $mg{\cdot}L^{-1}$, respectively. The concentrations of these components in SELF were set accordingly. SELF was developed based on previous studies, incorporating dipalmitoylphosphatidylcholine (DPPC) as a major lipid in human respiratory tract fluid [33, 34], where it functions as a surfactant in simulated epithelial lung fluid [35]. Therefore, this study employs simulated epithelial lung fluid (SELF) containing DPPC to assess the inhalation bioaccessibility of metals in the study area's soil.

The Bayan Obo mine is located in Baotou, Inner Mongolia, China. The Bayan Obo mining area is renowned for its abundant reserves of rare earth elements (REEs), niobium, thorium, and iron, with REE reserves accounting for approximately 97% of China's total, making it the world's largest known REE deposit [36]. Open-pit mining at Bayan Obo began in 1957, with ores rich in strategic materials such as Fe, Nb, and REEs transported by rail to Baotou Steel Plant, providing essential raw materials for steel production. Leveraging the abundant mineral resources of Bayan Obo, Baotou has undergone years of development and has become a major global center for rare earth research, production, processing, and application. [37]. Both Baotou and Bayan Obo have a semi-arid, mid-temperate continental climate, characterized by cold temperatures, aridity, and strong winds (predominantly from the northwest) [38]. Mining activities have led to urban soil contamination, and fine sand beneficiation processes have further exacerbated soil degradation. Therefore, remediation of surface soil has become an urgent necessity. Due to the region's dry climate and frequent strong winds, Baotou and Bayan Obo have long suffered from recurrent sandstorms. Under these conditions, workers and residents exposed for extended periods are more likely to inhale soil particulates than those in other regions. However, existing studies mainly focus on the total metal concentrations in pollution assessments, with insufficient attention to the bioaccessibility of metals in soil particulates and their associated health risks, making it difficult to comprehensively assess the health risks in this region.

In this study, we studied the urban soils of Bayan Obo and Baotou. The objective was to examine the inhalation bioaccessibility of eight heavy metals (Cd, Zn, Cr, Pb, As, Ni, Cu, and Mn) and rare earth elements in the soil, as well as to assess the human health risks associated with these heavy metals. Since most rare earth elements had concentrations below the detection limit in bioaccessibility tests, only La, Ce, and Eu were detected, and thus, this study focuses solely on these three elements.

Material and Methods

Sample Collection and Preparation

This study selected four surface soil samples from Bayan Obo and five from Baotou, designated as Y1#~Y4# and T1#~T5# (as shown in Fig. 1). Y1# was collected from open land near the Bayan Obo railway station, Y2# from surface soil near a residential area, Y3# from open land near a wastewater treatment plant in the urban area, and Y4# from suburban soil south of Bayan Obo. Sampling points T1#~T5# were set starting from the east side of the Baotou tailings pond, with one sampling site placed every 3 km toward the Yellow River. Each sample (10 kg) was collected using a stainless steel shovel and stored in labeled polyethylene bags. After being transported to the laboratory, the samples were air-dried at room temperature for 72 hours, manually cleaned of foreign materials (such as stones, roots, and insect remains), sieved through a 2 mm mesh, and stored for further analysis.

Sample Characterization

The determination of soil pH and electrical conductivity was conducted using the following methods: The soil samples, sieved through a 2 mm mesh, were mixed with water at a soil-to-water ratio of 1:2.5, shaken for 10 minutes, and left to settle. The pH of the supernatant was then measured using a LICHENPH-100B pH meter. Another aliquot of the pre-treated soil was mixed with water at a soil-to-water ratio of 1:5, and the electrical conductivity of the mixture was measured using a LEICIDDB-303A conductivity meter. Soil organic matter was determined using the potassium dichromate oxidation-external heating method: a sieved soil sample was weighed, mixed with a potassium dichromate-sulfuric acid solution, and digested using an oil bath heating method. The remaining oxidant

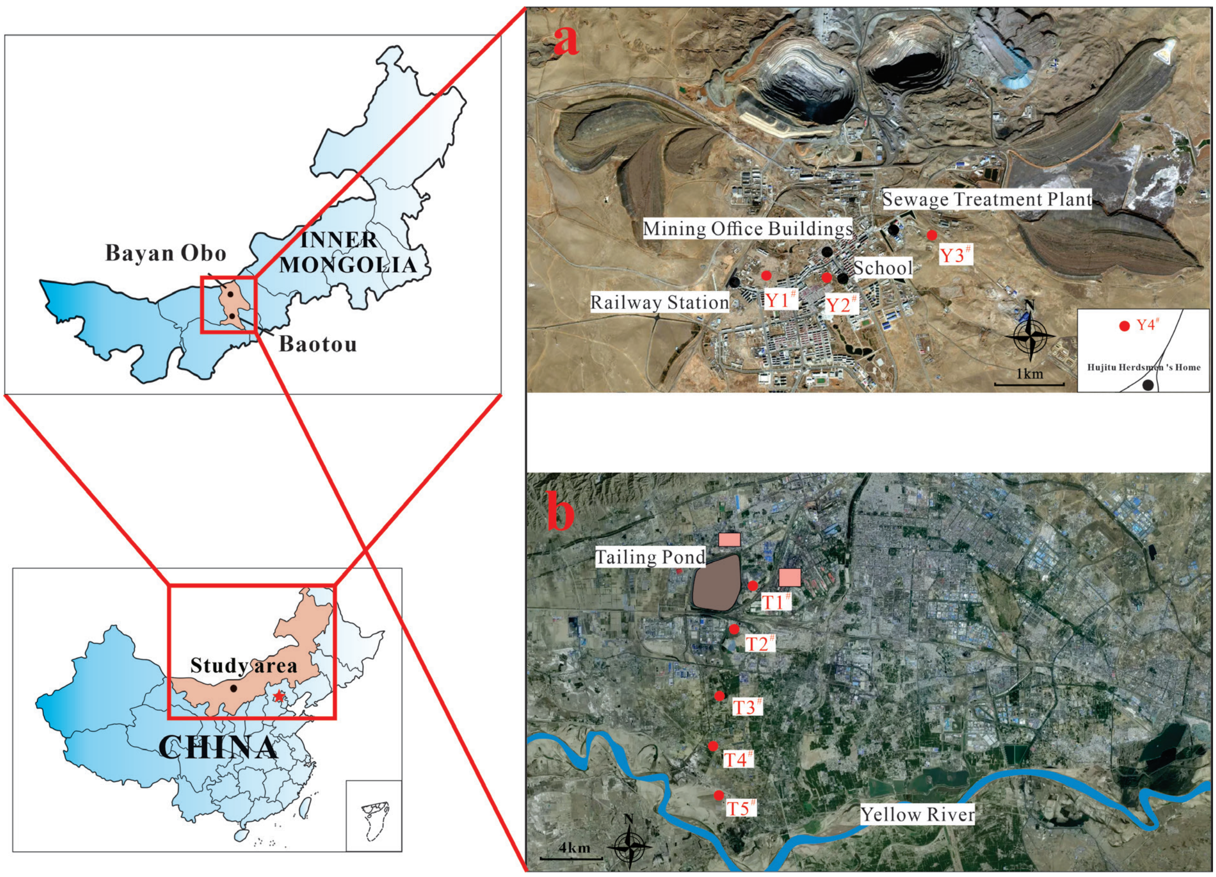


Fig. 1. Sampling sites in the Bayan Obo district (Y1#–Y4#, a) the core urban area of Baotou (T1#–T5#, b).

was titrated with ferrous sulfate, and the organic matter content was calculated based on oxidant consumption. Particle size analysis was performed using sieves of different mesh sizes (2 mm, 1 mm, 0.5 mm, 0.25 mm, 0.074 mm) in sequence. The soil retained on each sieve was weighed to calculate the particle size distribution, and the remaining soil was sealed and stored for future use. All tests were repeated three times, and the final results were averaged.

To determine the metal concentrations in soil, 50 mg of dried soil (sieved through a 2 mm mesh) was ground to a particle size of less than 0.05 mm and placed in an Erlenmeyer flask. The sample was digested with a mixed acid solution of HNO_3, $HClO_4$, HF, and HCl on a hot plate at 130-185°C for 3 hours. After cooling, the digestion solution was diluted to a specified volume with deionized water for analysis. As and Cr were measured using hydride generation atomic fluorescence spectrometry (HG-AFS) and X-ray fluorescence spectrometry (XRF), respectively. Mn and Zn were analyzed by inductively coupled plasma optical emission spectrometry (ICP-OES), while Cd, Cu, Ni, Pb, and rare earth elements were determined using inductively coupled plasma mass spectrometry (ICP-MS).

Preparation, Extraction, and Calculation of SELF

To prepare 1000 mL of SELF, first, 500 mL of the inorganic phase was prepared, consisting of 6020 mg NaCl, 256 mg $CaCl_2$, 150 mg $NaHPO_4$, 2700 mg $NaHCO_3$, 298 mg KCl, 200 mg $MgCl_2$, and 72 mg Na_2SO_4, all dissolved in deionized water. Then, 500 mg of the organic phase was prepared, containing 18 mg ascorbic acid, 16 mg uric acid, and 30 mg glutathione, also dissolved in deionized water. To better simulate physiological conditions, the inorganic and organic phases were thoroughly mixed, followed by the addition of 376 mg glycine, 500 mg mucin, 260 mg albumin, 122 mg cysteine, and 100 mg dipalmitoylphosphatidylcholine (DPPC). Once all the reagents had completely dissolved, the pH of the solution was adjusted to 7.4.

Next, 0.3 g of soil sample (particle size <0.05 mm) was mixed with 20 mL of SELF in a centrifuge tube and shaken at 37ºC for 96 hours in an orbital shaker to ensure thorough interaction between the solution and the soil. After shaking, the sample was centrifuged at 3000 rpm for 10 minutes, and the supernatant was collected and stored below 4ºC for further analysis.

The percentage inhalation bioaccessibility fraction (%BF) was calculated as follows:

$$BF(\%) = \frac{C \times V}{T \times M} \times 100 \tag{1}$$

where C represents the bioaccessible metal concentration in SELF ($\mu g \cdot L^{-1}$). V represents the volume of the SELF solution (L). T represents the total metal concentration in the soil ($\mu g \cdot g^{-1}$). M represents the mass of the soil (g).

Human Health Risk Assessment

Human health risk assessment establishes a connection between human health and environmental pollution, providing a quantitative analysis of the adverse effects of environmental contaminants on human health. According to the human health risk assessment methodology established by the United States Environmental Protection Agency (USEPA) [39, 40], the daily intake of heavy metals via inhalation from soil (*ADD*inh, mg/kg/day) is calculated using Equation (2).

$$ADD_{inh} = C \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT} \tag{2}$$

where ADD_{inh} represents the daily intake of an element through inhalation and C denotes the bioaccessible concentration of the metal ($mg \cdot kg^{-1}$) or the total metal concentration ($mg \cdot kg^{-1}$). The names and values of other parameters are listed in Table 1.

The hazard quotients (HQ_{inh}, Eq. (3)) and the hazard index (HI_{inh}, Eq. (4)) were used to characterize the non-carcinogenic hazard.

$$HQ_{inh} = \frac{ADD_{inh}}{RfD_{inh}} \tag{3}$$

Table 1. Definitions and reference values of soil heavy metal human health risk assessment parameters.

Parameters	Definition	Values		Reference
		Adult	Child	
InhR	Soil inhalation rate (m^3/day)	20	7.5	[41]
EF	Exposure frequency (day/year)	350	350	[40]
ED	Exposure duration (year)	24	6	[41]
BW	Body weight (kg)	70	15	[40]
AT	Average time (day)	365×ED (non-carcinogen)/ 365×70 (carcinogen)		[40]
PEF	Soil to air particulate emission factor (m^3/kg)	1.36×10^9	1.36×10^9	[40]

Table 2. Summary of reference doses (RfD_{inh}) and slope factors (SF_{inh}) for different pollutants via inhalation.

Metals	RfD_{inh} (mg/kg/day)	SF_{inh} (mg/kg/day)
Cd	1.0×10^{-3}	6.3
Cr	2.86×10^{-5}	42
Pb	3.52×10^{-3}	4.2×10^{-2}[43]
As	3.01×10^{-4}	15.1
Ni	2.06×10^{-2}	0.84
Zn	3.0×10^{-1}	NA
Cu	4.02×10^{-2}	NA
Mn	1.43×10^{-5}	NA
References	[44]	[45]

$$HI_{inh} = \sum HQ_{inh} = \sum \frac{ADD_{inh}}{RfD_{inh}} \quad (4)$$

where RfD_{inh} represents the reference dose of the studied element (mg/kg/day), with values provided in Table 2. When HQ_{inh} or $HI_{inh}<1$, there is no potential non-carcinogenic risk to humans. Conversely, when HQ_{inh} or $HI_{inh}\geq1$, it indicates a non-carcinogenic risk to human health [42].

The carcinogenic risk is represented by CR_{inh}, while TCR_{inh} denotes the sum of the cancer risks induced by all metals, as expressed in Equations (5) and (6).

$$CR_{inh} = ADD_{inh} \times SF_{inh} \quad (5)$$

$$TCR_{inh} = \sum CR_{inh} \quad (6)$$

where SF_{inh} represents the carcinogenic slope factor (mg/kg/day), with values provided in Table 2. When CR_{inh} or $TCR_{inh}\leq10^{-6}$, it is considered to pose no significant carcinogenic risk. When $10^{-6}<CR_{inh}$ or $TCR_{inh}<10^{-4}$, it is considered to indicate a potential carcinogenic risk. When CR_{inh} or $TCR_{inh}\geq10^{-4}$, it is regarded as presenting a significant carcinogenic risk.

Results and Discussion

Soil Chemical Characteristics

Table 3 presents the average physicochemical properties of soils in the Bayan Obo and Baotou regions. The soils in both areas exhibited alkaline characteristics with pH>8, which is consistent with the typical features of saline-alkali soils in northwestern China [46], regarding the highly alkaline nature of Bayan Obo soils. In general, regions with higher electrical conductivity (EC) values tend to exhibit greater salt accumulation, which also corresponds to elevated pH values. This is because alkaline soils are often associated with higher concentrations of carbonates, sulfates, or chlorides, which dissolve in the soil and increase ion concentrations, thereby raising EC [47]. The data in Table 3 corroborate this observation, as both the mean and median values of EC and pH were higher in Baotou compared to Bayan Obo. Alkaline soil environments play a crucial role in regulating the speciation of metals, effectively suppressing the release of metal elements [46, 48]. Previous research has also shown that organic matter can immobilize certain metals through complexation [49]. Therefore, this study analyzed the soil organic matter (SOM) content in the study area. The results revealed that the minimum SOM content in Bayan Obo soils was 19.27 $g\cdot kg^{-1}$, which was significantly higher than the maximum value observed in Baotou soils (17.77 $g\cdot kg^{-1}$). Among all samples, particle size distribution in Bayan Obo soils was relatively uniform, whereas Baotou soils were predominantly composed of fine sand (particle size: 0.25-0.074 mm), with an average mass fraction of 68.66%. The average concentrations of metal elements in soil samples from Bayan Obo and Baotou are shown in Table 4. Except for Cr, the average and median concentrations of other metals in Bayan Obo soils were higher than those in Baotou. In particular, Mn ($p = 0.047$) and Ni ($p = 0.019$) exhibited significant differences, which is consistent with the geological characteristics of Bayan Obo, known for its richness in iron and nickel minerals. However, elements such as As ($p = 0.494$) and Cu ($p = 0.294$), although higher in Bayan Obo, had $p>0.05$, indicating that their spatial variation may be influenced by regional geochemical backgrounds. The lowest enrichment was observed for As, Cu, and Cr, whose concentrations were close to background values; combined with their non-significant $p>0.05$, this suggests that their origins are primarily geological.

Previous studies have reported that most rare earth element (REE) concentrations follow a normal distribution and exhibit significant correlations among themselves [50]. In this study, the concentration range of ∑REEs in Bayan Obo soils was 273.08-2770.01 $\mu g\cdot kg^{-1}$, with an average of 1556.51 $\mu g\cdot kg^{-1}$, which is 683% higher than the average REE concentration in Chinese soils. In Baotou, the ∑REE concentration ranged from 226.69 to 1257.83 $\mu g\cdot kg^{-1}$, with an average of 540.48 $\mu g\cdot kg^{-1}$, 172% higher than the national average. Correlation analysis further indicated that La, Ce, and Eu concentrations in Bayan Obo were significantly higher than those in Baotou ($p<0.01$), suggesting that the impact of rare earth element contamination caused by mining in Bayan Obo is more pronounced than the influence of steel production and tailings in Baotou. Specifically, La, Ce, and Eu in Bayan Obo were 9.15, 10.34, and 4.97 times the Chinese soil background values, and 9.98, 13.53, and 6.02 times the Inner Mongolia background values, respectively. In contrast, although REE concentrations in Baotou soils were lower than those in Bayan Obo, they still exceeded both the Chinese and Inner Mongolia background levels, indicating that steel smelting activities and tailings reservoirs also exerted a considerable influence on surrounding soils.

Differences in enrichment levels are closely related to spatial distribution heterogeneity. As shown in Table 4, the coefficients of variation (CV) of REEs in Bayan Obo averaged 48.36%, whereas those in Baotou averaged 96.93%. Both values fall within the moderate variability range (10%<CV<100%), suggesting substantial spatial heterogeneity likely induced by anthropogenic disturbances such as mining, smelting, and transportation. Further analysis revealed that the highest REE concentrations in Bayan Obo were

Table 3. Descriptive statistics of the Bayan Obo and Baotou soil properties.

	pH	EC	SOM	Particle size distribution (mass %)					
		μs·cm⁻¹	g·kg⁻¹	>2 mm	2-1 mm	1-0.5 mm	0.5-0.25 mm	0.25-0.074 mm	<0.074 mm
Bayan Obo									
Mean	8.44	165.59	30.20	19.91	5.57	12.83	19.02	30.67	11.98
Median	8.46	167.28	21.23	20.06	5.31	13.87	18.28	26.83	10.82
Max	8.52	218.90	59.06	26.89	8.76	14.28	23.95	47.17	16.96
Min	8.34	108.90	19.27	12.64	2.89	9.28	15.58	21.86	9.32
Stdev	0.09	45.22	19.29	6.12	2.43	2.38	3.54	11.25	3.41
Baotou									
Mean	8.69	375.46	10.70	1.69	2.46	5.60	11.05	68.66	10.54
Median	8.68	332.40	8.76	0.41	1.24	4.10	10.58	76.97	10.22
Max	9.01	814.00	17.77	7.17	8.78	18.82	16.75	82.69	15.21
Min	8.40	151.20	5.94	0.00	0.34	0.17	2.45	38.35	6.43
Stdev	0.21	269.47	4.97	3.07	3.56	7.64	5.99	18.57	3.47

Table 4. Soil element concentration in Bayan Obo and Baotou areas (mg/kg).

	As	Mn	Ni	Pb	Zn	Cu	Cr	Cd	La	Ce	Eu
Bayan Obo											
Mean	9.65	1406.60	19.68	65.10	114.05	18.17	38.64	0.30	363.36	707.58	5.12
Median	9.56	1103.39	19.85	50.95	97.85	17.91	38.14	0.27	388.74	730.50	5.07
Max	10.66	2647.73	21.97	128.60	205.32	20.72	41.83	0.54	619.42	1258.80	9.20
Min	8.83	771.89	17.05	29.891	55.18	16.13	36.45	0.13	56.56	110.53	1.14
CV (%)	8.19	60.9	10.3	68.3	57.4	10.5	6.07	57.4	78.0	78.9	73.3
Baotou											
Mean	4.52	513.43	17.05	27.11	62.92	15.77	65.84	0.10	159.27	308.78	2.31
Median	4.33	435.86	14.54	21.21	56.92	14.07	56.18	0.09	81.42	169.81	1.57
Max	5.18	723.92	22.11	48.61	102.86	23.27	105.17	0.17	310.39	588.53	3.81
Min	3.83	387.75	13.61	17.19	40.58	12.05	47.98	0.08	45.42	94.41	1.11
CV (%)	12.4	28.6	24.6	47.2	38.0	27.9	35.6	38.1	82.6	78.8	58.8
Significance (p)	0.494	0.047*	0.019*	0.095	0.163	0.294	0.064	0.090	0.003**	0.004**	0.006**
Inner Mongolia background value [50]	11.2	583	26.9	26.0	74.2	22.6	61.0	0.097	39.7	68.4	1.03
Inner Mongolia background value [51]	7.5	520	19.5	17.2	59.1	14.4	41.4	0.053	36.4	52.3	0.85

*: p<0.05; **: p<0.01.

observed in samples Y1# and Y3#. Y1# was collected near the Bayan Obo railway station, where long-term ore transportation significantly increased metal enrichment in surrounding soils. Y3#, collected near the wastewater treatment plant, likely reflects the influence of REE-rich effluents discharged from mining activities. In contrast, the spatial distribution of REEs in Baotou soils exhibited a decreasing trend with increasing distance from the tailings reservoir, suggesting that the tailings reservoir and steel plant are likely the main sources of REE enrichment in Baotou soils.

Bioaccessibility of Soil Particulates in SELF

Mining activities have contributed to the enrichment of metals in soils, and metals associated with fine soil particles may be inhaled into the human lungs and subsequently translocated into the bloodstream via permeation, thereby posing potential health risks. The results from the SELF simulation reflect the potential concentrations of metals in the human lungs (Fig. 2 and Table 5). Elements not reported were below the detection limit in SELF and are therefore excluded from further discussion.

As shown in Table 5, the average concentrations of heavy metals extracted by SELF in the Bayan Obo region were in the order of Mn (4245.84 $\mu g \cdot kg^{-1}$)>As (37.83 $\mu g \cdot kg^{-1}$)>Ni (17.88 $\mu g \cdot kg^{-1}$)>Zn (11.15 $\mu g \cdot kg^{-1}$) >Pb (8.07 $\mu g \cdot kg^{-1}$)>Cr (1.32 $\mu g \cdot kg^{-1}$)>Cu (1.03 $\mu g \cdot kg^{-1}$), all of which were higher than those observed in the Baotou region: Mn (720.37 $\mu g \cdot kg^{-1}$) >As (23.16 $\mu g \cdot kg^{-1}$) >Zn (21.90 $\mu g \cdot kg^{-1}$)>Ni (8.51 $\mu g \cdot kg^{-1}$)>Pb (6.82 $\mu g \cdot kg^{-1}$) >Cu (1.25 $\mu g \cdot kg^{-1}$)>Cr (1.13 $\mu g \cdot kg^{-1}$). Overall, the concentrations of metals extracted by SELF in Bayan Obo soils were significantly higher than those in Baotou, which may be attributed to the higher total metal concentrations present in Bayan Obo soils (Table 4). To further understand the ecological and biological impacts of soil metals, inhalation bioaccessibility was assessed as the ratio of the mass of metals dissolved in the SELF solution to the total metal mass in the tested soil (Fig. 2). Figs 2a) and 2b) illustrate the inhalation bioaccessibility of metals in soils from Bayan Obo and Baotou, respectively. It is evident that in both regions, the bioaccessibility of As and Mn was substantially higher than that of other elements, with As exhibiting an average bioaccessibility of approximately 30%, suggesting a high potential risk. As shown in Fig. 3, As and Mn are predominantly present in the exchangeable and carbonate-bound fractions in soils, which are weakly bound and less stable, making them more susceptible to release under environmental influences such as pH, SOM, or redox potential, and thus exhibit high bioaccessibility [52]. In addition, although Ni, Pb, Zn, and Cu exhibited relatively lower bioaccessibility compared to As and Mn, they may still pose certain regional risks. Compared with Hamad et al. [53], the bioaccessibility of Ni, Pb, and Zn in this study was similar, but Cu was substantially lower. In their work, a simulated macrophage vacuole solution similar to ALF was used to assess nine toxic elements – including As, Cu, Mn, Ni, Pb, Zn, Co, and V – in agricultural and playground soils, with Cu bioaccessibility reaching as high as 2.5%. Guney et al. [54] evaluated the pulmonary bioaccessibility of As, Cu, Fe, Mn, Ni, Pb, and Zn in tailings dam soils using both ALF and GS solutions, and found that most elements were below the detection limit in GS, while the values measured in ALF were generally higher than those obtained in this study, likely due to differences in the pH of the extraction solutions.

However, comparing different elements within the same region revealed that some metals with relatively high total concentrations in soils exhibited relatively low concentrations in SELF extracts. This indicates that the total concentration of metals in soils does not directly determine their bioaccessibility, which is largely governed by the physicochemical properties of the elements themselves, particularly their chemical speciation.

With regard to rare earth elements, their bioaccessibility was significantly lower than that of heavy metals. In the Bayan Obo region, the average SELF-extracted concentrations of Ce, La, and Eu were 2.59, 0.34, and 0.12 $\mu g \cdot kg^{-1}$, respectively, slightly higher than those in Baotou, where Ce and La were measured at 2.40 and 0.29 $\mu g \cdot kg^{-1}$, respectively, while Eu was below the detection limit. However, the bioaccessibility values of La, Ce, and Eu were low, all averaging below 0.2%. This may be attributed to their stable chemical forms in soils [52, 55, 56]. At the same time, this is also related to the intrinsic geochemical characteristics of rare earth elements. In the cold and arid regions where Baotou and Bayan Obo are located, soil pollution caused by mining activities is mainly attributed to aeolian processes [55, 56]. The primary rare earth silicate minerals (fluorocerium silicon apatite, etc.), rare earth oxide minerals (cerianite, etc.), rare earth phosphate minerals (cerium monazite, etc.), and rare earth carbonate minerals (fluorocarbon cerium ore, etc.) present in the Bayan Obo deposit [57] remain relatively stable under the mildly alkaline conditions and oxygen fugacity of simulated lung fluid, which are close to atmospheric environmental conditions (e.g., within the 96-hour experimental period). In particular, as shown in Fig. 3, the three rare earth elements are mainly present in the relatively stable residual fraction (with residual proportions >80%). Metals in the residual phase are difficult to dissolve under normal environmental conditions and are also not readily absorbed by organisms, resulting in low bioavailability. In addition, rare earth elements generally occur as REE^{3+}, while Ce can exist as Ce^{4+} under oxidizing conditions. Therefore, under near-surface conditions, REEs are not easily released into solution, which further reduces their bioavailability [58]. Furthermore, during soil weathering, REEs are readily fixed by clay minerals as well as Fe and Mn oxides [59]. In the process of migration, REE^{3+} ions may also adsorb onto organic colloids through –OH groups [60]. Metals associated with Fe/Mn oxide-bound or strongly organic-bound fractions are generally stable and not easily bioavailable. Since SELF simulates a neutral lung fluid environment (pH = 7.4), it primarily extracts weakly bound metal fractions, thereby providing insight into the actual bioaccessible forms. Consequently, even when the total concentrations of REEs in soils are relatively high, their bioaccessible concentrations remain low due to the intrinsic geochemical characteristics of REEs, coupled with their predominant occurrence in stable phases within the Bayan Obo deposit.

The bioaccessibility values measured by SELF at pH = 7.4 were compared with those reported by Zupančič et al. (2024) [61], in which bioaccessibility was assessed using ALF at an acidic pH of 4.63-4.77. The two studies revealed distinct patterns of inhalation bioaccessibility across their respective research areas. Notably, metal concentrations determined using the acidic ALF were generally higher, with Zn, Pb, and Sb showing bioaccessibility values of up to approximately 40%. This difference can be attributed to the contrasting environmental conditions simulated by the two extraction solutions. ALF mimics the acidic environment of alveolar fluid (pH = 4.63-4.77), which may affect the speciation of certain metals (e.g., carbonate-bound and humic acid-bound forms), resulting in greater extraction of exchangeable metal fractions. In contrast, the neutral SELF solution (pH = 7.4) may promote the formation of insoluble metal complexes, thereby reducing metal bioaccessibility. In studies assessing inhalation bioaccessibility, the definition of particle size varies across research disciplines and regulatory

Table 5. Bioaccessible soil concentrations ($\mu g \cdot kg^{-1}$) in the Bayan Obo and Baotou areas.

	Bayan Obo bioaccessible concentration			Baotou bioaccessible concentration		
	Range	Med	Ave±SD	Range	Med	Ave±SD
As	16.32-53.27	40.86	37.83±17.18	18.42-30.48	20.00	23.16±5.08
Mn	1574.16-10036.67	2686.26	4245.84±3951.58	382.07-1134.40	688.48	720.37±277.93
Ni	14.39-22.51	17.31	17.88±3.75	6.22-10.76	8.48	8.51±1.64
Pb	5.75-10.76	7.89	8.07±2.34	6.02-7.86	6.87	6.82±0.73
Zn	7.04-18.02	9.77	11.15±4.78	12.32-38.28	20.39	21.90±9.80
Cu	0.81-1.37	0.97	1.03±0.26	0.85-1.82	1.11	1.25±0.38
Cr	1.07-1.95	1.14	1.32±0.42	0.85-1.59	1.09	1.13±0.30
Cd	-	-	-	-	--	-
La	0.31-0.37	0.35	0.34±0.03	0.23-0.34	0.31	0.29±0.05
Ce	2.46-2.74	2.57	2.59±0.15	2.30-2.63	2.37	2.40±0.13
Eu	0.10-0.15	0.12	0.12±0.02	-	-	-

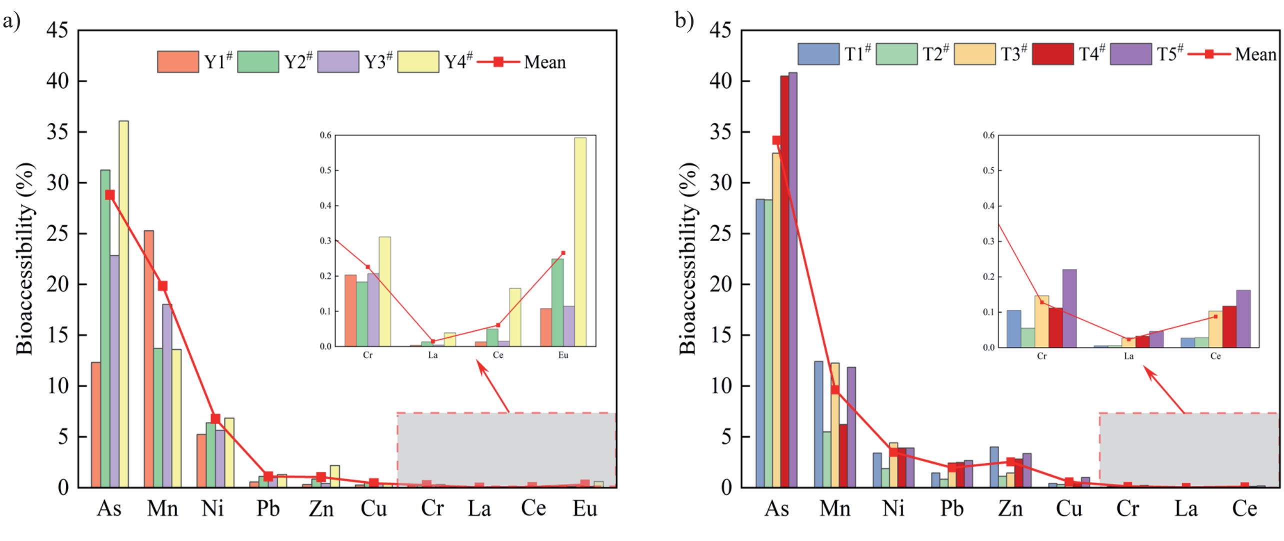


Fig. 2. Lung bioaccessibility of soil in the a) Bayan Obo and b) Baotou areas.

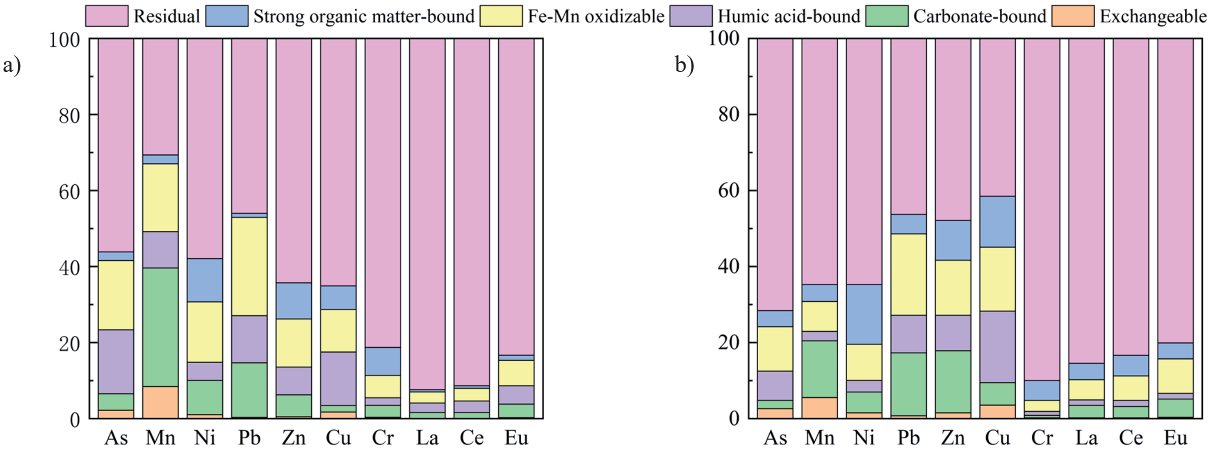


Fig. 3. Metal speciation in soils a) the Bayan Obo district; b) the core urban area of Baotou).

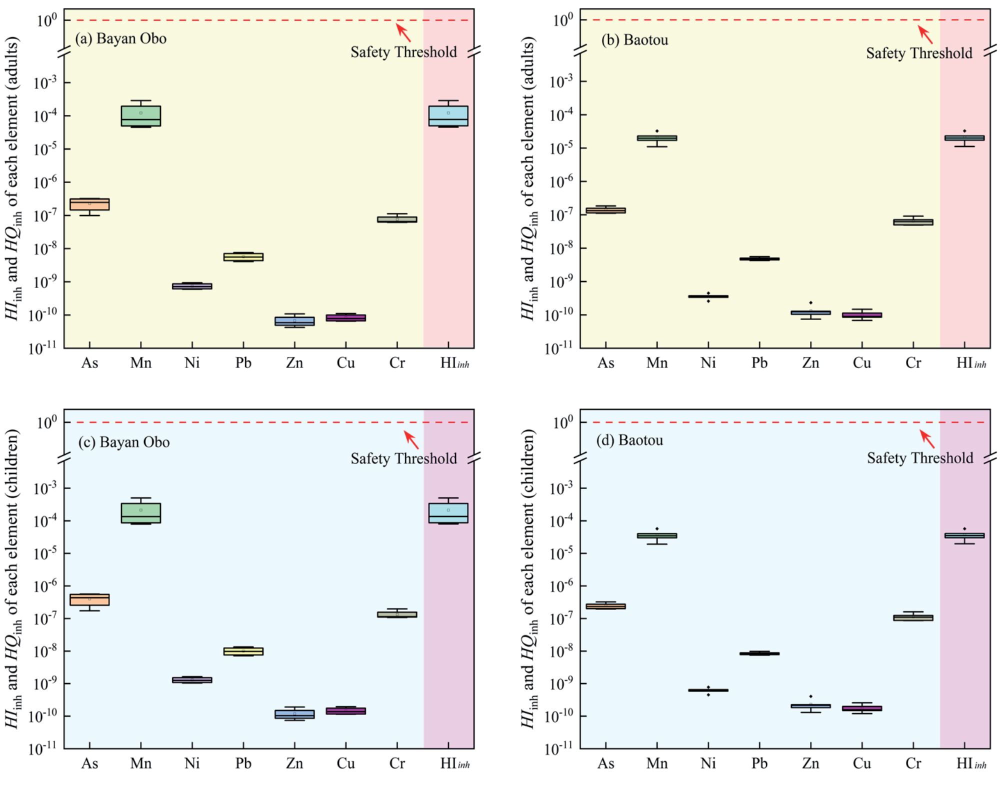
frameworks [62]. Although particles with diameters smaller than 100 µm are generally considered capable of depositing in the respiratory tract [61], larger particles may also settle in the bronchial region under certain site-specific conditions, such as industrial operations or extreme weather. In a European soil study, Menegaki [16] observed that concentrations of nearly all elements increased when soil particle sizes were less than 10 µm, with corresponding increases in the inhalation bioaccessibility of As and Mn. This is consistent with previous findings showing that metal concentrations tend to rise as soil particle size decreases [63]. Such enrichment is likely related to the larger specific surface area and abundance of functional groups on finer particles, which enhance their capacity to adsorb metals. In a study by Kastury et al. (2017) [64], particles with diameters <250 µm were used to evaluate inhalation bioaccessibility. Subsequent work by the same group [65] revealed a negative correlation between particle size and metal concentration, indicating that metal concentrations increased as particle size decreased from <250 µm to 10 µm. Baotou and the Bayan Obo region are located in a cold, arid, sandy area, where the aeolian pollution particles are relatively coarse, which is also one of the reasons why the associated risks are less severe.

Assessment of Potential Health Risks of Soil Elements

Metals that remain in the lungs over prolonged periods may induce various diseases, such as pneumonia, nephritis, and even cancer [14, 15]. Therefore, in this study, the potential non-carcinogenic and carcinogenic risks associated with inhalation exposure for local residents were assessed based on the bioaccessibility data, using the risk assessment model advocated and validated by the U.S. Environmental Protection Agency (EPA).

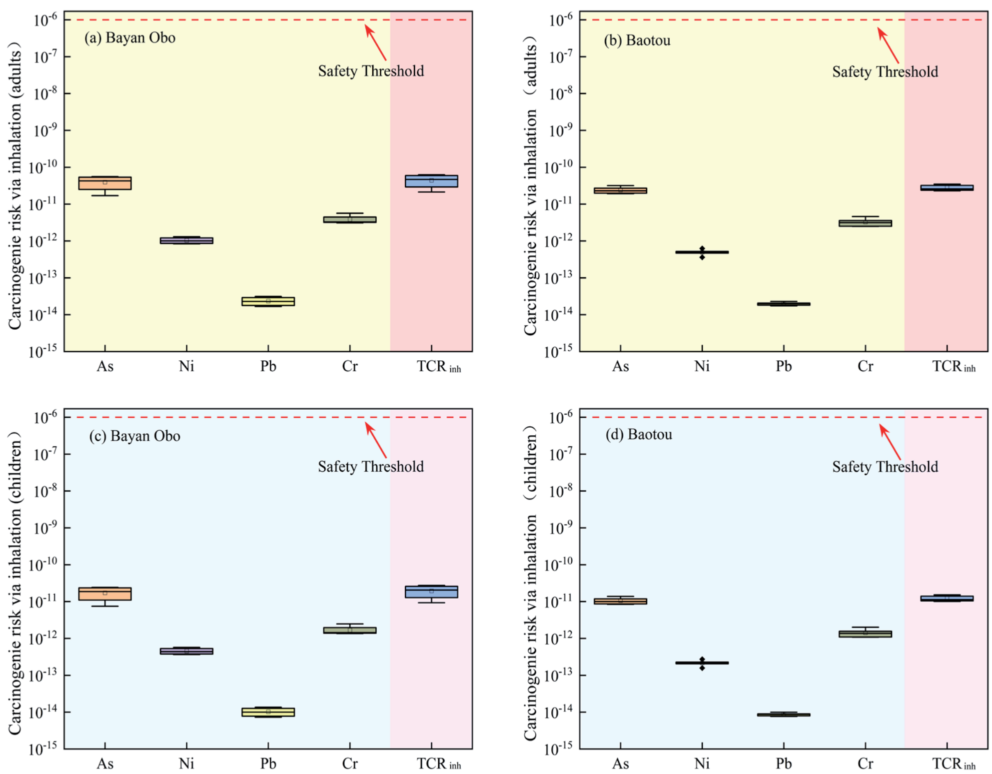
Figs 4 and 5 illustrate the risk assessment results based on the metal bioaccessibility measured using the SELF method, while Figs 6 and 7 show the results calculated using the total metal concentrations in soil. The results indicate that, under the inhalation exposure pathway, both the non-carcinogenic risk (Hazard Index, HI) and the carcinogenic risk (Total Carcinogenic Risk, TCR) of toxic elements were below the safety thresholds recommended by the U.S. EPA (HI<1; TCR<10^{-6}), regardless of whether total or bioaccessible concentrations were used as parameters. This suggests that the health risks posed by individual or combined elements are within acceptable limits. More importantly, substituting total metal concentrations with bioaccessible concentrations in the risk model led to substantial changes in the health risk profile for different elements (Figs 4 vs. 6 and Figs 5 vs. 7). The health risks calculated using total concentrations were generally higher than those based on the bioaccessibility model, as only a small fraction of metals in soil particles are leachable in the lung fluid environment during the exposure period [16, 66]. Notably, the total concentration model may underestimate the hazard posed by As, since the bioaccessibility-based model shows that the risk from As is comparable to or even greater than that of Cr. Therefore, risk assessments based solely on total concentrations may lead to an overestimation of health risks, potentially resulting in unnecessary regulation and remediation efforts.

Compared with adults, children exhibited higher non-carcinogenic risks (Fig. 4), which may be attributed to their higher inhalation rate per unit body weight (*InhR/BW*), a consequence of their physiological characteristics and behavioral patterns. In both regions, the top three contributors to non-carcinogenic risk were Mn > As > Cr, indicating that Mn is the primary element posing potential health risks to humans in these areas. In terms of carcinogenic risk, adults faced higher risks than children, primarily due to their longer exposure duration (ED). Across both study areas, the elements contributing most to carcinogenic risk followed the order As>Cr>Ni>Pb, all of which remained



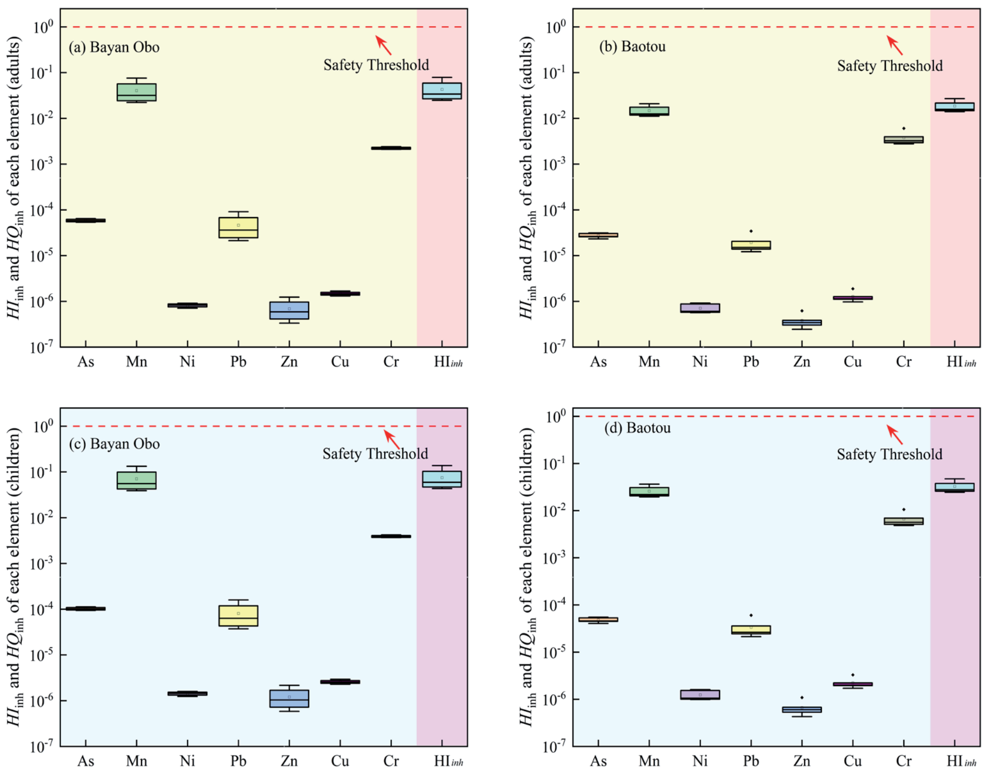
(1)

Fig. 4. Non-carcinogenic hazard quotient (HQ) and hazard index (HI) under respiratory exposure pathway based on bioaccessible concentration were calculated, a) Bayan Obo – Adults, b) Baotou – Adults, c) Bayan Obo – Children, d) Baotou – Children.



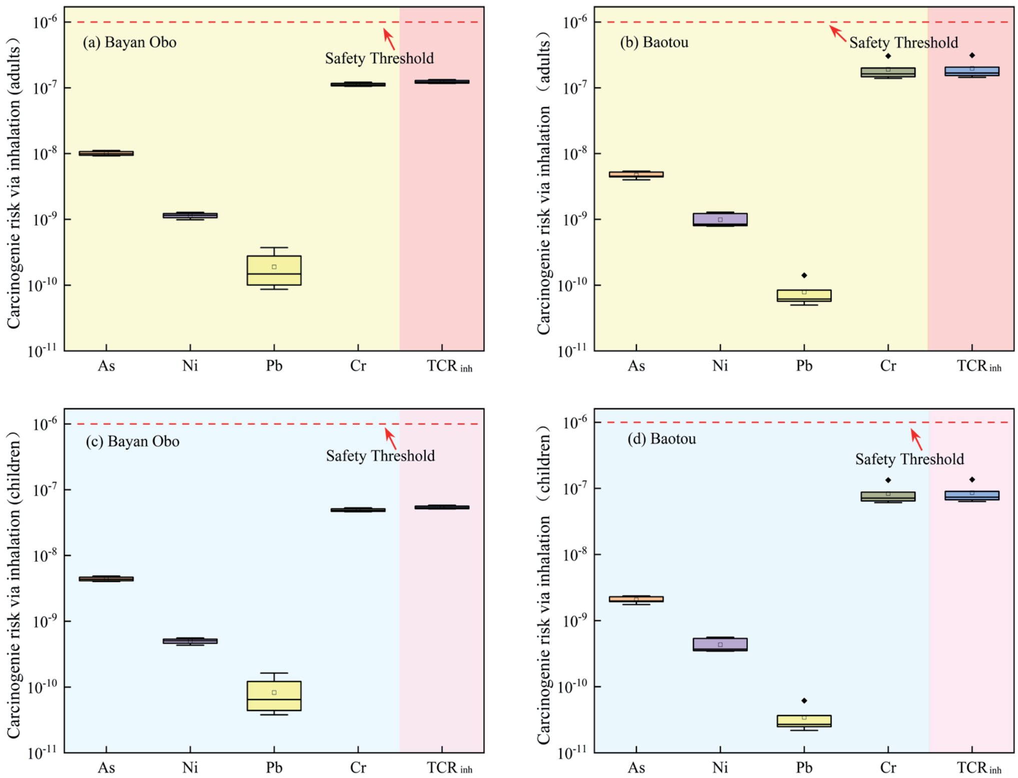
(2)

Fig. 5. Carcinogenic risk under respiratory exposure pathway was calculated based on bioaccessibility concentration a) Bayan Obo – Adults, b) Baotou – Adults, c) Bayan Obo – Children, d) Baotou – Children.



(3)

Fig. 6. Non-carcinogenic hazard quotient (HQ) and hazard index (HI) under respiratory exposure pathway based on metal concentration calculation a) Bayan Obo – Adults, b) Baotou – Adults, c) Bayan Obo – Children, d) Baotou – Children.



(4)

Fig. 7. Carcinogenic risk under respiratory exposure pathway based on metal concentration, a) Bayan Obo - Adults, b) Baotou - Adults, c) Bayan Obo - Children, d) Baotou – Children.

below the acceptable threshold (<10⁻⁶), suggesting no significant inhalation-related carcinogenic risk for either adults or children. A previous study [67] also identified As and Cr as the primary soil-borne carcinogens in certain regions. Combined with the above results, where the inhalation bioaccessibility of As reached 30%±5%, it is worth noting that although the current exposure levels do not exceed the carcinogenic risk threshold, As has a strong potential to accumulate in the lungs over time, which may pose adverse health effects even at low doses. Therefore, precautionary measures should be strengthened.

Conclusions

This study investigated the inhalation bioaccessibility of heavy metals and REEs in soils from the Bayan Obo and Baotou regions, as well as their potential health risks to humans. The inhalation bioaccessibility of major metals in the soils was quantified using the Simulated Epithelial Lung Fluid (SELF) method. According to the extraction results, the concentrations of metals extracted from Bayan Obo soils were higher than those from Baotou, which may be related to the functional zoning differences between the two areas. The calculated bioaccessibility values revealed a decreasing order in lung-simulated conditions: As>Mn>Ni>Pb≈Zn>Cu>Cr>La>Ce>Eu. Among them, As and Mn exhibited relatively high inhalation bioaccessibility and thus posed greater potential risks, whereas REEs – despite their relatively high total concentrations in soil – showed low inhalation bioaccessibility.

The study also found that risk assessments based on total metal concentrations may overestimate actual risks. In contrast, using bioaccessible concentrations offers a more realistic evaluation of inhalation exposure. Health risk assessments based on bioaccessible concentrations indicated that both non-carcinogenic and carcinogenic risks in the two regions were within acceptable limits. Children showed higher non-carcinogenic risks than adults due to their greater physiological sensitivity to inhalation exposure. On the other hand, adults faced higher carcinogenic risks, primarily due to longer exposure durations. Among all elements assessed, both non-carcinogenic and carcinogenic risks were below safety thresholds. Notably, As presented both high inhalation bioaccessibility and relatively elevated carcinogenic risk. Mn was identified as the most concerning non-carcinogenic element, with high total concentrations in soil and bioaccessibility second only to As.

Particular attention should be paid to As and Mn in the soils of both regions, and soil remediation efforts may be necessary if deemed appropriate. Although REEs showed no clear evidence of posing health risks via inhalation in this study, their total concentrations in the soils of Bayan Obo and Baotou were significantly higher than the background values for both China and Inner Mongolia, which also warrants attention.

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Authors' Contribution

De Yao: Conceptualization; Funding acquisition. Rui Ji: Writing – original draft; Visualization; Software; Data curation. Chi Li: Resources; Validation. Jing Luo: Supervision; Visualization. Caifei Liang: Validation; Supervision. Yu Gao: Project administration; Data curation. Shuanhu Li: Supervision. Xiaorong Wang: Supervision. Lei Tian: Supervision.

Conflict of Interests

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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