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Original Research

Risks to Human Health of Exposure to Heavy Metals through Wheat Consumption near a Tailings Dam in North China

Jumei Liu[1, 2], Wantong Si[2]*, Zhen Xiao[2], Runlian Tang[2], Pan Niu[2], Yu Fu[2], Weishou Shen[1]**

[1]Nanjing Institute of Environmental Sciences, Ministry of Ecology and Environment, Nanjing 210042, China
[2]College of Chemistry & Environmental Engineering, Chongqing Key Laboratory of Environmental Material and Restoration Technology, Chongqing University of Arts and Sciences, Chongqing 402160, China

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Abstract

Heavy metals can accumulate in nearby farmland soil through leakage of tailings and suspended dust, thereby posing a threat to local agriculture and human health. This study measured the heavy metal content (mercury, Hg; copper, Cu; zinc, Zn; lead, Pb; cadmium, Cd; selenium, Se; manganese, Mn) of farmland soils and wheat from twelve villages near a tailing dam in Baotou, China to evaluate the potential risks to the local ecosystem and human health. The results of the Contamination Factor (C_f) and Geo-accumulation Index (I_{geo}) indicated serious soil Hg, Se, and Cu contamination. Nemerow Composite Index (*NCI*) values showed that ±75% of the total sampling sites suffered heavy metal pollution. Soil Potential Ecological Risk Index (*IRs*) to the west of the tailings exceeded those to the southwest, demonstrating higher levels of Hg and Cd. The Hazard Quotient (*HQ*) of Mn for adults and children exceeded 3.0, whereas the *HQ* of Cu for children exceeded 1.0; the Hazard Index (*HIs*) for both adults and children exceeded 2.0. The results indicate that long-term consumption of wheat grains contaminated with heavy metals in the study area may result in adverse non-carcinogenic health risks, in particular among children. The accumulation of Cd posed carcinogenic risks to human health in two villages ($>1 \times 10^{-4}$). There is an urgent need to regulate the pollution of farmland soil near the tailings dams to mitigate risks to human health.

Keywords: farmland soils, wheat, heavy metals, health risk assessment

*e-mail: siwt02@163.com
**e-mail: shenweishou@163.com

Introduction

China has diverse and abundant mineral resources, and exploitation of these resources has played an important role in the economic development across China. However, mineral mining has come at a cost of serious environmental damage, including contamination of land and groundwater, particularly by heavy metals (HMs) originating from waste rocks, tailings sands, and mineral dust [1-5].

HMs tend to be toxic and accumulate and persist in the environment and within the food chain, thereby reducing the quality of air, water bodies, soils, and crops and posing a substantial risk to the ecosystem and human health [2, 4, 6-8]. Humans can readily assimilate HMs by inhalation of contaminated dust or by ingestion of food crops grown in contaminated soil. Several previous studies have identified the risks of accumulation of HMs such as Hg, Se, Pb, Cd, Zn, and Cu, to the environment and human health [9-11]. In particular, excessive intake of Hg, Pb, and Cd can have significant impacts on human health [9, 10]. Human assimilation of Hg can result in gastrointestinal irritation, kidney problems, and brain damage, with symptoms including ataxia, attention deficit, blindness, deafness, dementia, dizziness, and loss of memory [9, 12]. Moreover, ingested Pb (or Pb^{2+} in a compound form) is dissolved by stomach acids, following which it enters the bloodstream, soft tissue, and bone tissue, thereby causing damage to multiple organs [13]. Pb^{2+} in the bloodstream leads to an elevated blood-lead level (BLL), which can interfere with the synthesis of hemoglobin and the biochemical processes associated with hematopoiesis. Long-term exposure to elevated levels of Pb, even if BLL does not reach hazardous levels ($\geq$ 5.00 μg/dL for young children), can still result in significant damage to the human body, in particular to the nervous system, leading to a loss in intelligence and dementia [14, 15]. Previous studies have associated long-term human exposure to Cd with various diseases, including lung cancer and pulmonary diseases, prostatic proliferative lesions, osteoporosis, itai-itai disease, renal dysfunction, hypertension, and diabetes [16, 17]. Hence, soil HM pollution and the associated risks to human health through the consumption of contaminated vegetables and grains have been a matter of widespread global concern [9, 10, 18].

Baotou City (Inner Mongolia, China) is a semiarid and continental climate zone with annual average precipitation and potential evaporation equal to 240-400 mm and 2,100-2,700 mm, respectively. Due to its arid climate and extremely limited water resources, agricultural irrigation using groundwater is common in Baotou. However, contamination of groundwater by leakage of tailings' impoundment has continued to be a serious environmental problem in the area [1, 3]. Field research conducted in the present study has revealed that irrigation using local groundwater resulted in the death of crop seedlings, and many areas were found to be unsuitable for human habitation. Moreover, large areas of local wasteland around the tailings dam were occupied by smelting and chemical plants. These industrial plants as well as the tailings dam released contaminants into surface soil through wastewater irrigation and atmospheric sedimentation. Some villages, inhabited primarily by elderly people and children, are situated nearby the tailings dam, regardless of contamination issues. Despite this, to date, there haven't been any studies on the potentially harmful effects of HMs on human health through the consumption of food crops grown in this area.

The present study aimed to evaluate the risks of HM contamination in the study area to the local ecosystem and human health. The main objectives of the present study were to: (1) assess HM contamination of farmland soil and the associated potential environmental risks; (2) analyse the HM (Hg, Cu, Zn, Pb, Cd, Se, and Mn) composition in farmland soils and wheat grains in the vicinity of the tailings dam; (3) evaluate the risks of HMs to human health through consumption of wheat grains. The present study can act as a reference for future studies on land use and the remediation of soil pollution.

Materials and Methods

Site Description and Sampling

The tailings dam located in the study area is in Dala Hai Xia village, situated 12 km west of Baotou City, at the junction of the Kundulun River, the alluvial-diluvial fan of the Hadamen area, and Jiuyuan district (Fig. 1). The tailings dam is a surface reservoir that first began operation in 1965. The dam has an area of 11 km^2 and extends at a distance of 3.2 km from east to west and 3.5 km from south to north. It has an effective capacity of ±70 million m^3 [3]. No effective anti-seepage measures have been implemented on this dam and there is no vegetation cover on its surface. In this area, ore collected from the Bayan Obo mine was dressed and smelted by Baotou Iron and Steel, and the leftover slag was stored in the tailings dam. Thus this tailings dam has become a typical pollution source and is responsible for the increasing risks of contamination of the surrounding habitats and population through water leakage, atmospheric dust, rainwater erosion, and runoff [1, 19]. The major food crops grown in the saline-alkaline soils of the study area are wheat (*Triticum aestivum*), maize (*Zea mays*), and some vegetables [7]. However, the present study focused on wheat as a study crop.

The present study selected a total of twelve villages (S1-S12) to the west and southwest of this tailings dam for sampling (Fig. 1), with an overall area of sampling equivalent to ±60 km^2. Farmland soils (0-20 cm) and wheat grains were collected from the twelve villages at the maturity stage. The wheat grains were brought

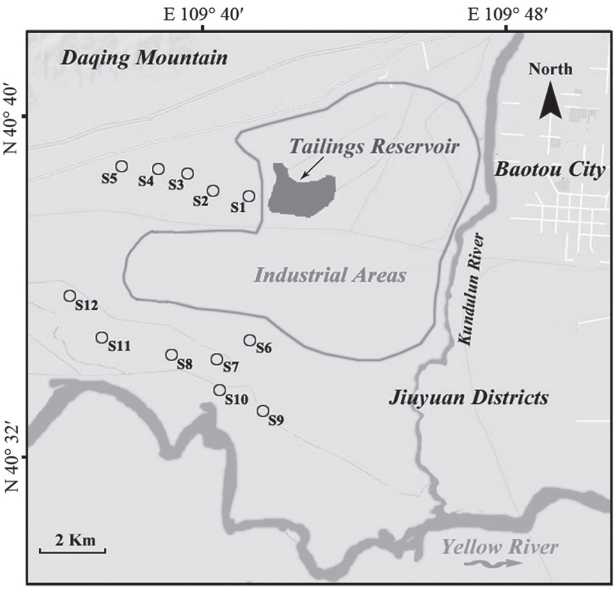


Fig. 1. The spatial locations of sampling sites in Baotou city, Inner Mongolia, China.

to the laboratory, cleaned thrice with deionized water, dried in a constant temperature box at 70ºC for 72 h, crushed and ground, and sieved using a 0.149-mm mesh. Soil samples were air-dried at room temperature and sieved using a 0.149-mm soil mesh for the measurement of metal contents.

Analyses of the Contents of Soil and Crop Heavy Metals

Soil and crop samples were digested in an open digestion oven using HNO_3-HCl-HF-$HClO_4$ (9:3:10:3, v/v) and HNO_3-H_2O_2 (4:1, v/v), respectively. All reagents used were of analytical grade and all solutions and dilutions were prepared in ultrapure water (Milli-Q, Millipore1, USA). Sample concentrations of Hg, Cu, Zn, Pb, Cd, Mn, and Se were determined using an inductively coupled plasma optical emission spectrometer (ICP-OES, Icap6000, Thermo Fisher Scientific, USA). A standard solution (China Iron & Steel Research Institute Group) was used to construct a standard curve for quality control (R^2 = 0.999). During measurement, each standard solution was used to calibrate the instrument for each of the ten samples, and the recovery was within 100±10%.

Evaluation Methods

Heavy Metal Pollution Index

The present study used the Contamination Factor (C_f) [20], Index of Geoaccumulation (I_{geo}) [21], Nemerow Composite Index (*NCI* or *F* values) [22], and the Potential Ecological Risk Index (*Er*, *IR*) [20] to evaluate soil HM contamination.

The C_f can be used to assess the main pollutants in the environment and is an indicator of the degree of contamination of a particular HM. C_f was calculated as the ratio of HM in the soil (C_i) to the soil background values reported for China or Inner Mongolia [7]:

$$C_f = C_i / C_{background} \quad (1)$$

Igeo, commonly known as the Muller index, not only considers the impact of natural geological processes upon HM background values but also considers the impact of human activities on HM [2]:

$$Igeo = \log_2 (C_i / 1.5C_{back\ ground}) \quad (2)$$

NCI or *F* values were calculated as:

$$F = \sqrt[2]{({C_{f\ max}}^2 + {\bar{C}_{f\ average}}^2)/2} \qquad \bar{C}_{f\ average} = \frac{1}{n}\sum_{i=1}^{n} C_f \quad (3)$$

where $\bar{C}_{f\ average}$ and $C_{f\ max}$ are the average and the maximum values of individual pollutant indices, respectively.

IR was computed as:

$$IR = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} (T_r^i \times C_f) \quad (4)$$

where T_r^i is toxic response factor for a particular HM (40, 5, 1, 5, 30, 1, and 1 for Hg, Cu, Zn, Pb, Cd, Se, and Mn, respectively) [23]; E_r^i is the potential ecological risk for a single element.

Table 1 shows the classifications of the above indices.

Health Risk Assessment

Bioconcentration Factor

The rates of transfer of HMs from soils to crops were measured using the bioconcentration factor (*BCF*) [8, 9]. *BCF* is calculated as the ratio of element contents in grains of wheat (C_{wheat}) to that in soils (C_{soil}):

$$BCF = C_{wheat} / C_{soil} \quad (5)$$

Non-Carcinogenic Risk Assessment

The Hazard Quotient (*HQ*) and Hazard Index (*HI*) are commonly used to measure non-carcinogenic risks posed to human health by pollutants, with values of less than 1.0 have no effects, whereas those greater than or equal to 1.0 indicating adverse non-carcinogenic effects on the health of the exposed population [10].

$$HI = \sum_{i=1}^{n} HQ_i = \sum_{i=1}^{n} (ADI_i / R_f D_i) \quad (6)$$

where ADI (mg kg^{-1} day^{-1}) is the average daily intake for humans.

$$ADI = C_{\text{metal}} \times D_{\text{wheat intake}} / B_{\text{average weight}} \quad (7)$$

where R_fD is the chronic toxicity reference dose (mg kg^{-1} day^{-1}) of the pollutant (R_fD_i values of 0.0003, 0.04, 0.3, 0.0014, 0.001, 0.005, and 0.14 mg kg^{-1} day^{-1} for Hg, Cu, Zn, Pb, Cd, Se, and Mn, respectively) [24]; C_{metal}, D_{wheat} intake, and $B_{average}$ weight represent metal concentration in wheat grains, daily wheat intake, and an average weight of residents, respectively. The present study investigated the annual total wheat intake of more than 200 residents. The results of the present study showed that the average daily intake of adults aged 20-59 years old was 0.47 kg person^{-1} day^{-1}, whereas that of children aged 3-6 years was 0.16 kg person^{-1} day^{-1}, and the average weights of adults and children were 64.06 and 19.14 kg, respectively [25].

Carcinogenic Risk Assessment

Carcinogenic risk is estimated by calculating the increased probability of developing cancer over a lifetime, for an individual who is exposed to a potential carcinogen [8]. According to the classification of the International Agency for Research on Cancer (IARC)

Table 1. Classification of indexes for the soil pollution assessment.

Index	Value	Soil quality
C_f [21]	<1	Practically uncontaminated
	$1 \leq C_f < 2$	Uncontaminated to moderately contaminated
	$2 \leq C_f < 3$	Moderately contaminated
	$3 \leq C_f < 4$	Moderately to heavily contaminated
	$4 \leq C_f < 5$	Heavily contaminated
	$5 \leq C_f < 6$	Heavily to extremely contaminated
	≥6	Extremely contaminated
I_{geo} [21]	<0	Practically uncontaminated
	$0 \leq I_{geo} < 1$	Uncontaminated to moderately contaminated
	$1 \leq I_{geo} < 2$	Moderately contaminated
	$2 \leq I_{geo} < 3$	Moderately to heavily contaminated
	$3 \leq I_{geo} < 4$	Heavily contaminated
	$4 \leq I_{geo} < 5$	Heavily to extremely contaminated
	≥5	Extremely contaminated
NCI (*F*) [22]	<0.7	Clean
	$0.7 \leq F < 1.0$	Warning value
	$1.0 \leq F < 2.0$	Lightly polluted
	$2.0 \leq F < 3.0$	Middle to highly polluted
	≥3.0	Heavily polluted
E_r [20]	<40	Practically no risk
	$40 \leq E_r < 80$	Moderate risk
	$80 \leq E_r < 160$	Moderate to heavy risk
	$160 \leq E_r < 320$	Heavy risk
	≥320	Extreme risk
IR [20]	<150	Practically risk
	$150 \leq IR < 300$	Moderate risk
	$300 \leq IR < 600$	Heavy risk
	≥600	Extreme risk

Note: C_f, Contamination Factor; I_{geo}, Index of Geoaccumulation; *NCI (F)*, Nemerow Composite Index. E_r, potential ecological risk for single element; *IR*, Potential Ecological Risk Index non-cancer causing HMs investigated in the present study include Pb, Zn, Cu, Mn, Cd, Hg, Se, whereas only Cd was regarded as having potential carcinogenic risk [26]. *SF* is the carcinogenicity slope factor and is a good measure of the average daily intake of a toxin during a lifetime translated to an increased risk of cancer. The *SF* for Cd is 0.38 mg^{-1} kg day.

$$Risk = ADI \times SF \tag{8}$$

Risks values of $<1 \times 10^{-6}$ indicate the non-existence of a health risk; a risk value lying between 1×10^{-6}-1×10^{-4}, specifies an acceptable range of health risk; whereas those $>1 \times 10^{-4}$ are considered an unacceptable risk to health [24].

Statistical Analyses

Results were presented as mean±standard deviation. All statistical analyses were conducted in the SPSS 19.0 software One-way Analysis of Variance (ANOVA) followed by Duncan's multiple Comparisons was used to detect differences. Pearson correlation analysis were performed to analyze correlation factor between the heavy metal concentration in soils and its concentration in the wheat samples. $p<0.05$ was regarded as statistically significant. Principal component analysis (PCA) was performed using the CANOCO version 4.5 software. Radar chart was constructed using the software Microsoft Excel 2010.

Results and Discussion

Distribution of Heavy Metals in the Soils of the Twelve Villages

The present study showed the cumulative influence of HM pollution and the associated health risk in farmland soils near the tailings dam, Baotou, China. This tailings dam has accumulated large quantities of waste rocks and tailings sands over nearly 60 years. This has consequently resulted in an altered accumulation of various HMs in the surrounding soil. Fig. 2 shows the contents and distribution of HMs. And the soil background values of China and Inner Mongolia were used as baseline to evaluate the content of HMs, respectively [27].

There were significant differences in the soil concentrations of HMs among the twelve villages, in comparison to the background value. In particular, soil Hg of all twelve sampling sites exceeded the background values of Inner Mongolia (0.0278 $mg \cdot kg^{-1}$) and China (0.040 $mg \cdot kg^{-1}$), and ranged between 0.084-0.389 $mg \cdot kg^{-1}$ (Fig. 2a). In contrast, soil Cu, Zn, and Se exceeded the soil background value for Inner Mongolia, except for Cu in S11 and S12 and Zn in S12 (Fig. 2(b, c, f)). Soil Pb in S1, S2, S3, and S4 exceeded the soil background value of Inner Mongolia (15.0 $mg \cdot kg^{-1}$), whereas soil Cd in S1, S2, S3, S4, S5, and S10 exceeded the soil background value of Inner Mongolia (0.037 $mg \cdot kg^{-1}$), however, those for other soils were within limits (Fig. 2d,e). For all soil samples, the content of Mn was lower than the background value of China (482.0 $mg \cdot kg^{-1}$) and ranged between 221.00-449.00 $mg \cdot kg^{-1}$, except for S1 (Fig. 2g). These results suggested severe Hg and Se contamination, particularly in the soils acquired from the sites located to the west (S1-S5; Fig. 2a) and southwest (S6-S10; Fig. 2f) of tailings, respectively, indicating different distributions of HMs in the chosen soils.

The present study applied PCA to further identify the sources of soil HMs [8]. The results of PCA clearly showed three clusters among the soil samples of the twelve villages (Fig. 2h). Among them, S1, the site nearest to the tailing reservoir, and most affected by the tailings, was isolated as a group; S5 to the far west of the tailings and S11 and S12 to the far northwest of tailing were classified into another group; S2 to S4 and S6 to S10 formed a tight cluster (Fig. 2h). The combined pollution of soil HMs showed a direct relationship to the distance to tailing pond and was consistent with previous findings [10].

Assessment of Soil Heavy Metal Pollution and Environmental Risk

The pollution characteristics and potential ecological risks of HMs in the soils of the twelve villages were assessed using C_f, I_{geo}, *NCI* (*F*) (Fig. 3), and *IR* (Fig. 4).

In general, a C_f of ±1.0 is an indicator of soil HM enrichment due to processes of crustal materials or natural wear. A C_f exceeding 1.5 is an indicator of anthropogenic input being the main source of HM enrichment [8]. The C_f ranges of Hg, Cu, Zn, Pb, Cd, Se, and Mn were respectively equal to 3.01-13.99, 0.90-2.26, 0.88-2.21, 0.46-1.68, 0.29-2.51, 1.18-4.19, and 0.50-1.50. The HMs content according to mean C_f was: Hg>Se>Cu>Zn>Cd>Pb>Mn (Fig. 3a). These results showed that the soils from twelve villages near the tailing dam were seriously polluted by Hg and Se, with an extreme (C_f>6) and heavy (C_f>4) levels of contamination (refer to Table 1). The high C_f of Se in local farmland soil observed in the present study has not been reported in previous studies (Fig. 3a). Se is a trace element, and the content of Se in the soil environment is closely related to the health of humans and animals. A deficiency of Se or redundancy can lead to various endemic diseases, including Keshan disease and endemic fluorosis, respectively [28], with the latter widespread in humans and animals in the Baotou region of Inner Mongolia [19]. Previous studies have associated the environmental pollution of Se, Cu, and other HMs with the prevalence of fluorosis. This indicates the need for further emphasis on the prevention of Se and Cu pollution. Although the means C_fs of Cu, Zn, Cd, and Pb were relatively low, the levels may still be classified as moderate contamination ($2 \leq C_f <3$) at some sites, such

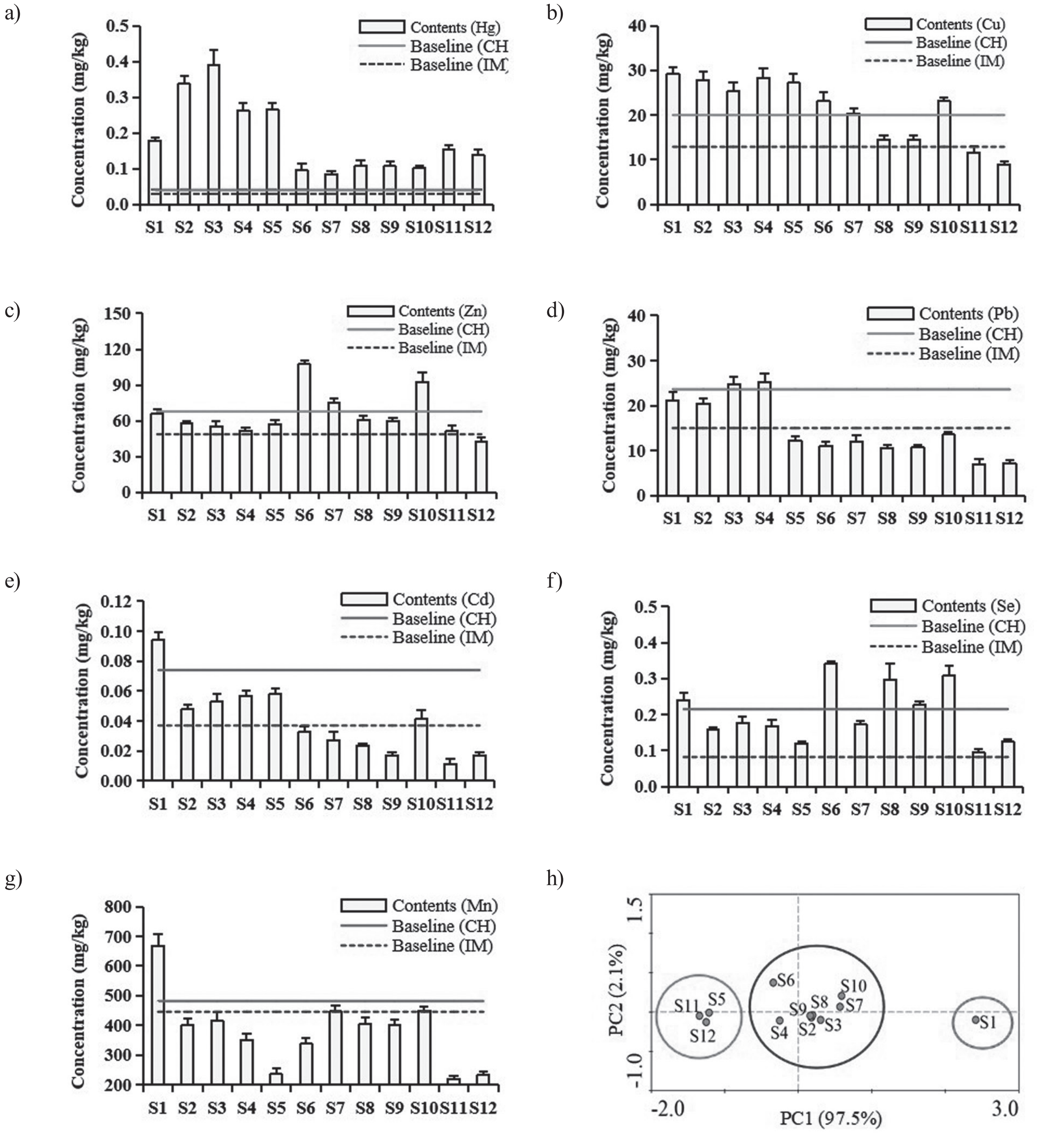


Fig. 2. The content of heavy metals in the soils and results of principal component analysis (PCA) based on the contents of seven heavy metals in each soil sample.

Note: Baseline (CH), soil background values for China for Hg, Cu, Zn, Pb, Cd, Se, and Mn are 0.04, 20.0, 67.7, 23.6, 0.074, 0.215, and 482.0 mg kg^{-1}, respectively [27]; Baseline (IM), the soil background values for Inner Mongolia for Hg, Cu, Zn, Pb, Cd, Se, and Mn are 0.0278, 12.9, 48.6, 15.0, 0.0374, 0.0817, and 446.0 mg kg^{-1}, respectively [27].

as for Cu in S1, S2, S4, and S5, for Zn in S6, and Cd in S1. These results further confirmed that local industrial activities lead to the accumulation of soil HMs.

The HMs content at the twelve sites according to average *Igeo* was Hg>Se>Cu>Zn, Cd, Pb, and Mn. Among them, Hg, Se, and Cu were at moderate levels (0≤*Igeo*<2), whereas Zn, Cd, Pb, and Mn existed below 0 (practically uncontaminated, refer to Table 1) (Fig. 3b). These results suggested that the presence of the tailings dam over the decades had accelerated the accumulation of Hg, Se, and Cu in the surrounding soil, however, it had only minimal effect on Zn, Cd, Pb, and Mn.

Agricultural soil pollution is often the result of multiple factors, whereas C_f and I_{geo} are more suitable for the evaluation of single pollution factors in a region. Therefore, the *NCI* (*F*) is more appropriate in comparison to C_f and I_{geo} for the evaluation of the

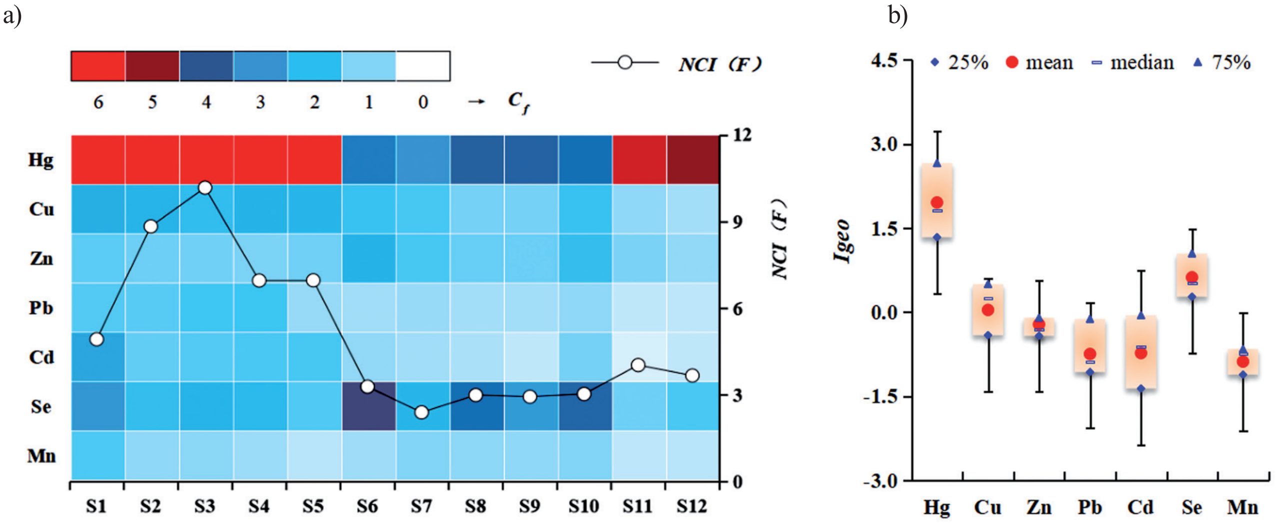


Fig. 3. The C_f, I_{geo} and *NCI* (*F*) values of heavy metals in the study area.
Note: C_f, Contamination Factor; I_{geo}, Index of Geoaccumulation; *NCI* (*F*), Nemerow Composite Index. The C_f of HMs was calculated based on the background values of Inner Mongolia.

cumulative influence of a variety of soil HMs [1, 2, 8]. according to the *NCI* (*F*), the site may be ranked in the following order; S3>S2>S5≈S4>S1>S11>S12>S6>S10 >3.0>S8>S9>S7 (Fig. 3a). Thus approximately 75% of sites had reached a heavy pollution (*F*≥3) status (refer to Table 1). The heavy metal pollution degree of all samples in this study was significantly higher than that of the farmland surrounding the Yellow River wetland, which is far away from the tailings dam (more than 20 kilometers away) in our previous study [7].

The potential risk of a single element to the ecosystem (*Er*) was used to reflect the potential environmental harms of Hg, Cu, Zn, Pb, Cd, Se, and Mn. The *Er* values of Hg in S2-S5 depicted an extreme risk level (≥320) whereas S1, S11-S12 were a heavy risk level ($160 \le E_r < 320$). The E_r values for Hg among the other sites were in between moderate to heavy risk levels ($80 \le E_r < 160$). The *Er* values of Cd in S1, S3-S5 exceeded 40 (40≤Er<80, moderate risk), whereas that for the remaining metals in S1-S12 was lower than the limits of E_r (< 40, nearly no risk) (refer to Table 1) (Fig. 4). The rank of the sites according to the overall potential ecological risk value (*IR*) was: S3>S2>S4>S5>S1>S11>S12>S10>S8>S6>S9>S7. *IR* of S3 was the highest (624.54), approaching an extremely high level of risk (>600). The *IR* values of S1, S2, S4, and S5 indicated high risks (300≤*IR*<600), whereas the *IR* values of S6, S7, S8, S9, S10, S11, and S12 were moderate (150≤*IR*<300) (Table 1; Fig. 4). In general, the potential risks of soil HMs to the ecosystem on the west side of the tailings (S1-S5) exceeded those on the southwest side (S6-S12). The *IR* of S1, the site nearest to the tailings dam, was lower than that of S2-S5. This result could be attributed to the Hg contents of S2-S5 significantly exceeding those in S1. The toxic response factor of Hg (T_r^i = 40) was the highest, and dominated among the seven HMs. The high contamination levels of HMs in the farmland soils have demonstrated the hazard of this tailings dam to the surrounding environment, which is consistent with the previous findings [19].

Heavy Metal Contents in Wheat Grains and the Bioconcentration Factor

Table 2 shows the concentrations of HMs in wheat grains, whereas Fig. 5 shows the average percentage of each HM in wheat grains, the results of the PCA assessment, and the bioconcentration factor (*BCF*) data.

Mn was the most abundant HM in wheat grains (64.37%), followed by Zn (30.31%) and Cu (5.21%), with the remaining HMs collectively accounting for less than 1% of the total content (Fig. 5a). The contents of Hg, Pb, and Mn in wheat grains in S1-S5 exceeded those in S6-S12, whereas Cu contents manifested an opposite trend (Table 2). The results of PCA showed that the twelve sampling sites could be divided into two clusters precisely; (1) S1-S5 to the west of the tailings dam; (2) S6-S12 to the southwest of the tailings dam (Fig. 5b). This result suggested obvious differences in the distributions of HMs in wheat among different geographical locations around the tailings dam. While Xiang et al. [29] reported the contents of the same heavy metal were inconsistent in soil and crops, similarly to those of the present study; and the accumulation of HMs in wheat grains was more complex than in soil and was primarily affected by geographical location, spatial heterogeneity, the ability of crops to accumulate particular HMs, and the interaction of heavy metals. Root system is an important pathway for crops to absorb heavy metals. So, the heavy metal concentration in wheat are ultimately determined by soil. In this study, except for Cu, Pearson correlation analysis showed positively correlation between the heavy elements concentration of Hg, Zn, Pb, Cd, Se and Mn in soils and

Table 2. Concentration of heavy metals in wheat grain (mg·kg^{-1}).

Sampling site	Hg	Cu	Zn	Pb	Cd	Se	Mn
S1	0.0120±0.0002	3.98±0.36	39.30±2.65	0.0540±0.0038	0.0361±0.0024	0.0090±0.0007	100.76±7.61
S2	0.0010±0.0005	3.40±0.28	34.70±2.84	0.0600±0.0049	0.0216±0.0018	0.0160±0.0013	93.28±7.64
S3	0.0130±0.0007	4.26±0.39	32.80±2.14	0.0740±0.0054	0.0177±0.0011	0.0160±0.0012	102.59±7.21
S4	0.0110±0.0009	3.89±0.32	30.30±2.48	0.1440±0.0118	0.0376±0.0031	0.0743±0.0061	120.36±9.85
S5	0.0130±0.0002	5.07±0.45	17.37±1.05	0.0840±0.0062	0.0233±0.0014	0.0640±0.0046	79.04±6.47
S6	0.0035±0.0003	6.54±0.54	32.49±2.66	0.0130±0.0011	0.0218±0.0018	0.0808±0.0066	36.20±2.41
S7	0.0038±0.0003	5.84±0.48	29.26±1.88	0.0190±0.0012	0.0214±0.0013	0.0462±0.0029	34.53±2.28
S8	0.0039±0.0003	5.26±0.43	21.98±1.80	0.0090±0.007	0.0195±0.0016	0.0254±0.0021	32.03±2.62
S9	0.0043±0.0004	5.69±0.45	28.31±1.80	0.0080±0.007	0.0182±0.0011	0.0740±0.0054	38.60±2.48
S10	0.0036±0.0003	4.65±0.38	22.71±1.86	0.0090±0.0009	0.0216±0.0018	0.0177±0.0011	29.68±2.43
S11	0.0035±0.0002	6.21±0.51	32.57±2.13	0.0090±0.0008	0.0144±0.0010	0.0236±0.0019	41.30±2.81
S12	0.0030±0.0002	4.86±0.40	25.46±2.08	0.0090±0.0007	0.0164±0.0013	0.0026±0.0002	29.20±2.39
Hygienic Standard / Limits of Contaminants	0.02[a]	10[b]	50[c]	0.2[a]	0.1[a]	0.3[d]	20[e]

[a] National Standards for Food Safety - Limits of Contaminants in Food, National Standard Bureau of PR China (GB 2762-2017); [b] Hygienic Standard for Limit of Copper in Food, National Standard Bureau of PR China (GB 15199-1994); [c] Hygienic Standard for Limit of Zinc in Food, National Standard Bureau of PR Chin (GB 13106-1991); [d] National Standard for Se-Enriched Rice, National Standard Bureau of PR China (DB/T 22499-2008); [e] There is no limit of manganese content in wheat in China, and the content of manganese in the normal human body is 10-20 mg·kg^{-1}, so 20 mg·kg^{-1} has been referred to.

its concentration in the wheat samples. In particular, Hg, Pb and Cd showed significantly positive correlation (the outcomes of correlation were not shown). These results suggest that other heavy metals in soil may inhibited on Cu uptake in wheat. Xiang et al. [29] found Pb and Cr in soil had synergistic effects on Zn uptake in crops, whereas Cu, As, and Hg showed an antagonistic effect on the crop absorption of Zn; the crop absorption of Cu would be inhibited by Ni in soil. It can be seen that complex interactive relationships exist among heavy metals, including synergistic effect and antagonistic effect. Furthermore, it will be crucial to focused on the relationship between heavy metal pollution in soil and crop in the future.

Similar to other reports, the HMs could be ranked in terms of their *BCF* value as follows: Cd>Zn>Cu>Se>Mn>Hg>Pb (Fig. 5c). Si et al. [7] ranked HMs in wheat grains in the Baotou region according to *BCFs* as: Cd>Zn, Cu>Se>Hg>Mn>Pb; Liu et al. [30] ranked HMs in rice as: Cd>Zn>Cu>As; Li et al. [18] ranked HMs in 25 crop types as: Cd>Zn>Cu>Ni>Pb>Cr. The above studies obtained the highest average *BCF* for Cd, which was calculated as 0.698 in the present study.

Although Hg pollution presents an acute ecological risk to local farmland soils, the Hg concentrations of wheat grains (0.001-0.013 mg·kg^{-1}) at all sites were lower than the National Standards for Food Safety of China (GB 2762-2017) (0.02 mg·kg^{-1}) (Table 2). This result could be directly related to the lower *BCF* of Hg. The Hg contents (0.003-0.004 mg·kg^{-1}) of wheat grain from S6-S12 were roughly equivalent to those in wheat grains acquired from areas located along the Yellow River (0.003-0.005 mg·kg^{-1}) [7].

Assessment of the Risk Posed by the Consumption of HMs in Wheat Grain to Human Health

Assessment of the Non-Carcinogenic Risk of the Seven Heavy Metals

The present study randomly surveyed 200 residents from the twelve villages included in the study, to gain basic information on diet, age, body weight, dietary habits, and health problems. The data was used to quantify the average chronic daily intake index (*ADI*), hazard quotient (*HQ*), and hazard index (*HI*). Toxic elements are absorbed and accumulated in food crops through the food chain, thus posing a potential threat to human health [6, 10]. Fig. 6 provides a summary of *HQ* and *HI* of residents of the twelve villages included in the study, resulting from the consumption of wheat grains, and the contribution of *HQ* of each metal to *HI*. The *HQs* of children attributable to wheat consumption exceeded those of adults, an observation consistent with previous research [7].

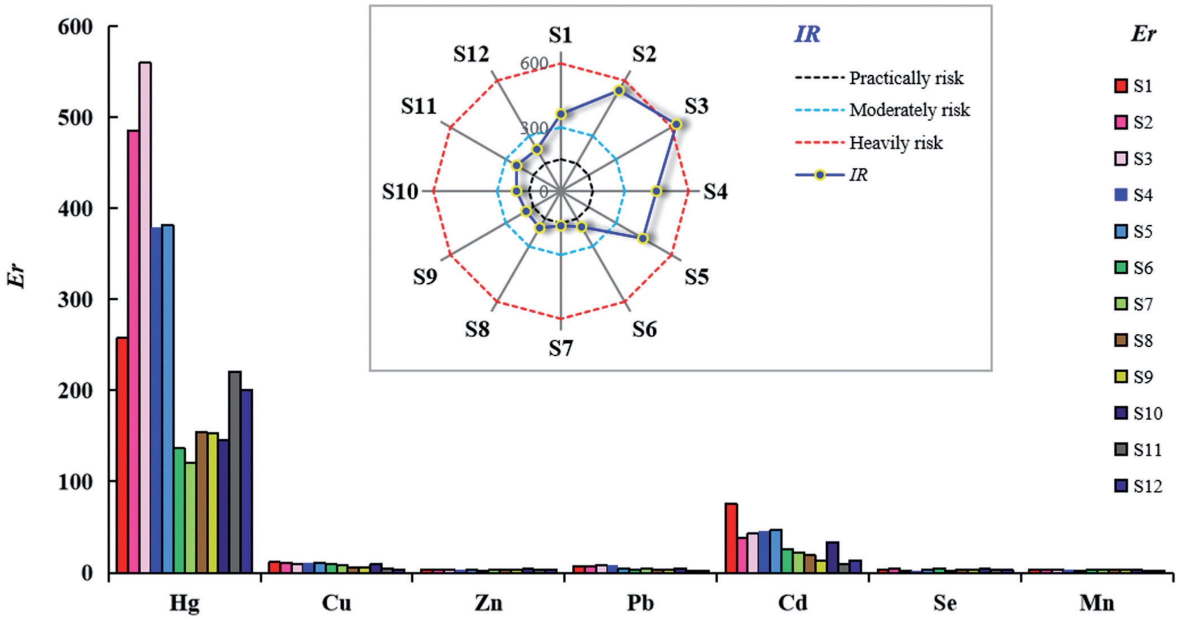
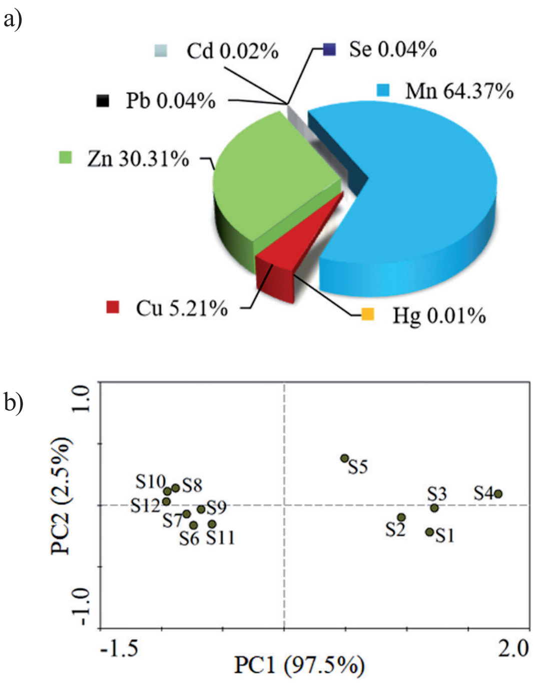


Fig. 4. The potential ecological risks (*Er*, *IR*) of soil heavy metals in the study area.
Note: *Er*, potential ecological risk for single element; *IR*, Potential Ecological Risk Index.



c)

BCF	Hg	Cu	Zn	Pb	Cd	Se	Mn
S1	0.0670	0.1363	0.5982	0.0025	0.3840	0.0375	0.1511
S2	0.0297	0.1252	0.6003	0.0029	0.4500	0.1019	0.2325
S3	0.0334	0.1684	0.5931	0.0030	0.3340	0.0904	0.2464
S4	0.0418	0.1370	0.5895	0.0057	0.6596	0.4423	0.3439
S5	0.0491	0.1864	0.3058	0.0068	0.4017	0.5333	0.3353
S6	0.0368	0.2833	0.3027	0.0012	0.6673	0.2360	0.1072
S7	0.0454	0.2875	0.3882	0.0016	0.7829	0.2681	0.0772
S8	0.0363	0.3627	0.3624	0.0009	0.8239	0.0854	0.0794
S9	0.0403	0.3906	0.4731	0.0007	1.0706	0.3265	0.0966
S10	0.0355	0.2012	0.2462	0.0007	0.5226	0.0573	0.0661
S11	0.0228	0.5356	0.6324	0.0013	1.3091	0.2450	0.1869
S12	0.0216	0.5420	0.5972	0.0013	0.9647	0.0209	0.1257

Legend: 0.5, 1, 1.5

Fig. 5. The average percentage a), principle component analysis (PCA) based on the contents of seven heavy metals in wheat grains b), bioconcentration factor (*BCF*) c) of seven heavy metals in the wheat grains.

The rank of the HMs according to *HQ* for children was (Fig. 6a): Mn>Cu >1.0>Zn>Pb>Cd>Hg>Se; that for adults was: Mn>1.0> Cu>Zn>Pb>Cd>Hg>Se. In particular, the *HQ* of Mn for both adults and children exceeded 3.0 (Fig. 6a). These values exceeded the values reported by Li et al. [10] in Zhuzhou city, China. This difference can presumably be attributed to the daily intake of wheat grains in the rural study area of the present study exceeding that in urban areas, with the diet of urban residents demonstrating a greater emphasis on rice and vegetables. The rank of the study sites in terms of *HI* for both adults and children was (Fig. 6b, black-white circle figure): S4>S1>S3>S2>S5> S11>S6>S9>S7>S8>S12>S10>2.0 (above the threshold value of 1.0). These results indicated that long-term consumption of wheat grains may cause adverse human

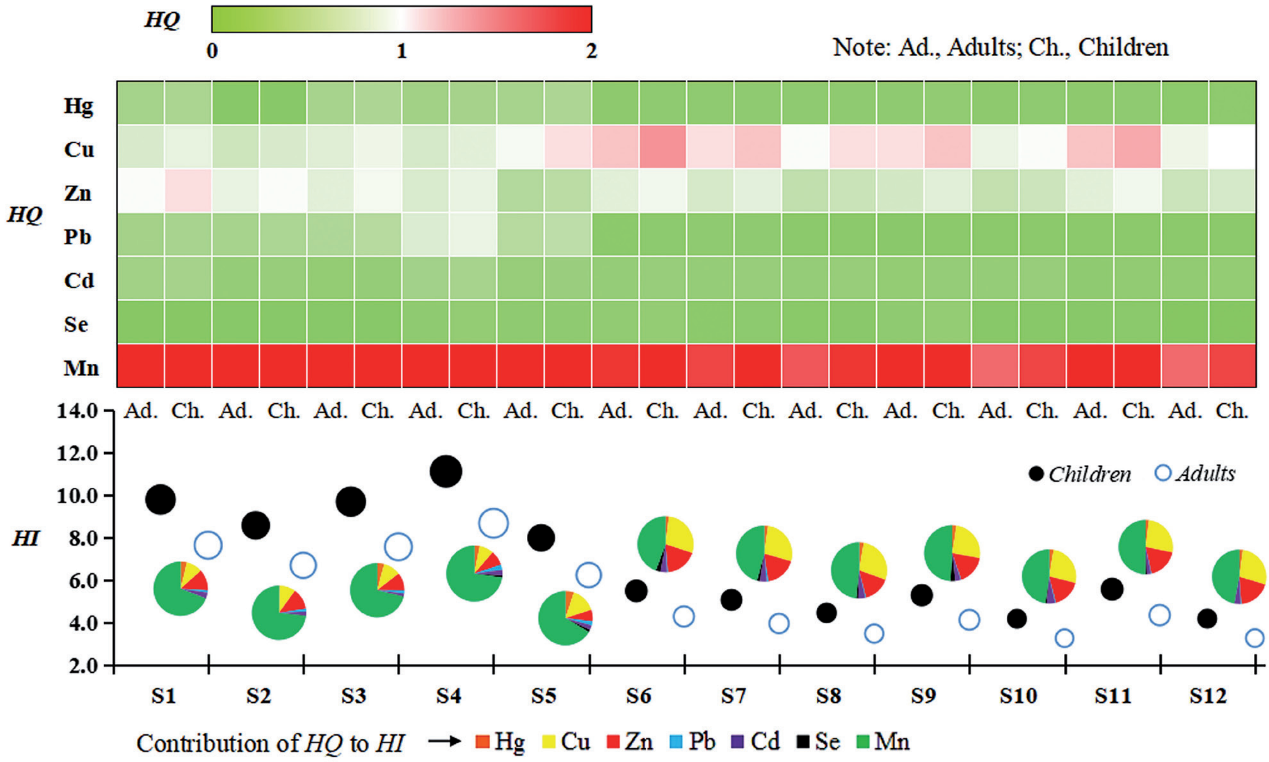


Fig. 6. Non-carcinogenic risk of the seven heavy metals for children and adults in the study area.
Note: Hazard Quotient (*HQ*) values (a), Hazard Index (*HI*) values (b, black-whiter circle figure), and the contribution of *HQ*/*HI* (b, pie chart).

health effects among the residents of all study sites. *HI* showed considerable variation among different villages, with that of children generally exceeding that of adults (Fig. 6b, black-white circle figure). This result suggested that local children encounter an increased risk of non-carcinogenic health hazards. Han et al. [14] similarly found that the consumption of contaminated food is more harmful to children in comparison to adults. This result can be attributed to the greater rate of food intake and more frequent hand-mouth activity in children in comparison to adults and the higher sensitivity of children to environmental pollutants [8]. Therefore,

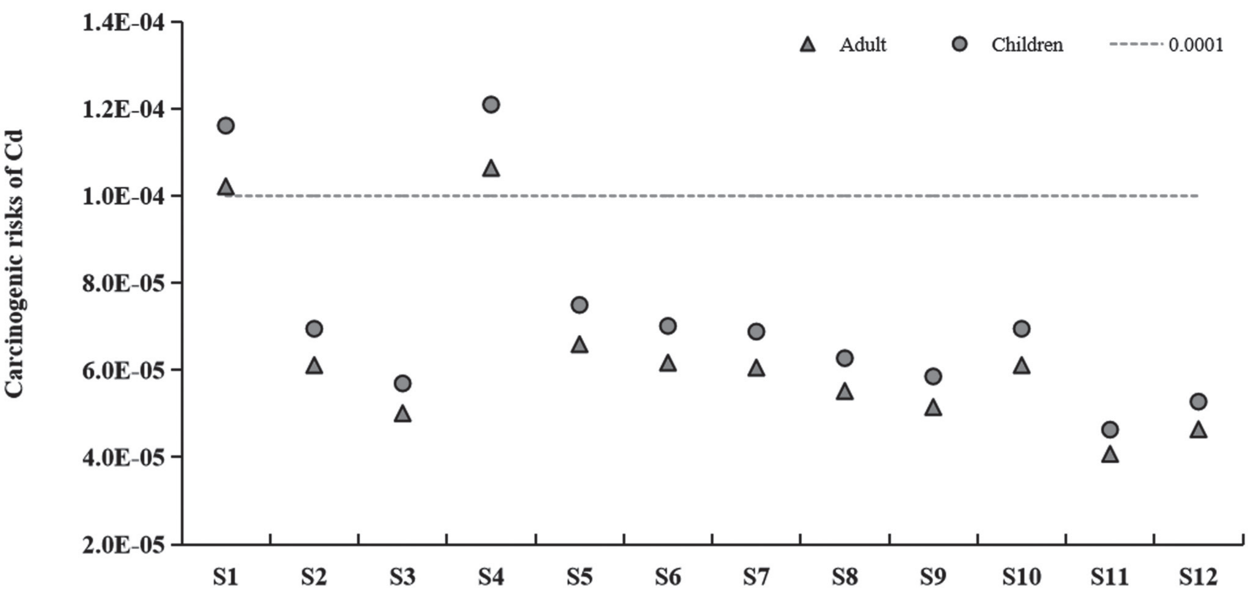


Fig. 7. Carcinogenic risks of Cd for children and adults in the study area.

there is a substantial need for an increased focus on the hazards posed to the health of children by HMs contamination in food.

HMs have been ranked according to their contributions of *HQs* to *HI* in S1-S4 as follows (Fig. 6b, pie chart): Mn>Zn>Cu>Pb>Hg>Cd>Se; in S5-S12 (Fig. 6b, pie chart): Mn>Cu>Zn>Cd>Hg>Pb>Se. This result identifies Mn, Cu, and Zn as the main contributors to non-carcinogenic risk to the health of residents. In particular, Mn and Cu posed high non-carcinogenic risks, with the *HQs* of Mn>3.0 for both adults and children and the *HQs* of Cu>1.0 for children (an *HQ*>1.0 indicates likely non-carcinogenic risks) (Fig. 6a). This result was consistent to those of Si et al. [7], who ranked HMs in spring wheat grain along the Baotou region of the Yellow river according to *HQs* as: Cu>Zn>Mn>Cd>Hg>Pb>Se.

This contribution of the *HQ* of each HM to *HI* was directly related to the different *BCF* and R_fD values of these HMs. Metals migrate from water to soil, and finally into crops via various pathways. In general, the *BCF* and R_fD corresponding to HMs in food crops are positively and negatively correlated to human health risk, respectively. HMs with higher contribution rates are more likely to cause adverse effects on human health. Therefore, there should be an increased focus on the localized hazards posed to human health by Mn, Cu, and Zn in food crops.

Carcinogenic Risk of Cd

As shown in Fig. 7, for both adults and children, the carcinogenic risks of Cd were found to be within an acceptable range (1×10^{-6} to 1×10^{-4}), except in S1 and S4 in which levels of Cd reached the threshold (1×10^{-4}), indicating higher possible carcinogenic risks, particularly to local children. A disproportionate number of cancer deaths have been recorded at S1 (Dalahai Village), with 61 deaths attributable to lung and/or brain cancer between 1999 and 2006 [19]. The high toxicity, accumulation, and carcinogenicity of Cd indicated the need for increased focus on the health risks arising from cadmium exposure [16]. Some studies have reported that the health risks resulting from exposure to Cd in food are age related since younger children consume three to four times more food than the average adult, based on body weight [31]. Thus, exposure to Cd in food, beginning at an early age can result in greater neurotoxic effects. Previous studies have reported that the toxic effect of Cd on biological pathways will be influenced by Ca^{2+} [32]. Cd^{2+} is similar in size and charge to Ca^{2+}, and Cd^{2+} uptake affects Ca^{2+} channels, thereby disrupting Ca^{2+} homeostasis, which is a regulatory messenger for many physiological metabolites. In addition, by disrupting lipid metabolism, Cd exposure also exaggerates diabetes [17].

The results of the present study indicated serious HM soil contamination in the study area (Fig. 3a). Han et al. [14] suggested that soil is an important source of HM exposure in children. There is a need for increased public awareness regarding the dangers of crop cultivation in contaminated soil. Also, children living in heavily polluted areas should be educated to adopt healthy living habits. A study by Zhang et al. [33] on the impact of HM pollution on food safety in agricultural soils in China showed that 10% of arable soil and 14% of grain production were affected by HM pollution. The Chinese government has strengthened the process of prevention of soil pollution in China by issuing the soil environmental quality risk control standard for soil contamination of developmental land (GB36600-2018) and agricultural land (GB15618-2018) in June 2018. These two standards were promulgated on 22 June 2018 and came into effect on the 1st of August, 2018 [34]. The control and mitigation of soil contamination by HMs inevitably require support from the government, industry, academia, and the public. There is a need to facilitate the monitoring of soil environmental quality, the assessment of ecological risk, as well as research into soil remediation materials and technologies, which will therefore help in improving soil quality [4, 8], ensuring food safety and quality [2, 29], and reducing health risks.

Conclusions

This study investigated HMs in soil and wheat grains from twelve farmland areas near a tailings dam. The results were used to characterize the overall levels of HM contamination in soil and wheat and the associated risks to human health. The results indicated that farmland soils around the tailings dam were contaminated by HMs originating from mining activities, with the highest contamination risks associated with the elements Hg, Se, and Cu. Of the twelve sites investigated, 75% manifested serious HM contamination. The potential environmental risks posed to the ecosystem by soil HM contamination in the area situated to the west of the tailings exceeded in comparison to the area located to the southwest. This could be attributed to higher Hg and Cd contamination. Human consumption of wheat grains grown in contaminated soils may pose non-carcinogenic health risks (*HQs* of Mn>3.0 for both adults and children; *HQs* of Cu>1.0 for children; *HI*>2.0 for both adults and children) and higher carcinogenic risks (Cd) to the public, and particularly to children. The results suggest possible carcinogenic risks due to Cd exposure in S1 and S4. The findings of this study highlight the necessity of HM pollution reduction and remediation to preserve inhabitants' health, particularly that of children.

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Conflict of Interest

The authors declare no conflict of interest.

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