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Ecological and human health risk assessment of tungsten and other heavy metal(loid)s in farmland around a typical tungsten mining area in southern Jiangxi, China

*Qi Li[1,2,3], Li Tian[1,2,3], Xiaojun Zheng[1,2,3], Weijiang Chen[1,2,3], Buchan Zhou[1,2,3], Ming Chen[1,2,3]**

[1]*Collaborative Innovation Center for Development and Utilization of Rare Metal Resources Co-sponsored by Ministry of Education and Jiangxi Province, Ganzhou, Jiangxi, P.R. China*
[2]*Jiangxi Key Laboratory of Mining and Metallurgy Environmental Pollution Control, Ganzhou, Jiangxi, P.R. China*
[3]*Jiangxi University of Science and Technology, College of Resources and Environmental Engineering, Ganzhou, Jiangxi, P.R. China*
**Corresponding author: jxlgcm@163.com*

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Abstract: The ecological and human health risks of heavy metal(loid)s (HMs) in soils around tungsten (W) mining sites have often disregarded the presence of W. In this study, we aimed to investigate the concentrations of 10 HMs (including W and other accompanying elements) in 18 agricultural soil samples obtained around a W mining site in southern Jiangxi, China. Furthermore, we determined the contamination status, source identification, and ecological and health risks of HMs in soils. Our findings revealed that HMs were extensively accumulated in soils within the study area, with the highest mean concentrations of W found. W concentrations were above background values at all sites. Multivariate analysis revealed that W mining activities, including extracting and transporting W ore, were the primary source of HMs in the soil (61.40%). The ecological risk assessment revealed that the potential ecological risk across the survey area exhibited a high risk, and the cadmium (Cd) and W should be prioritised as control pollutants for soils around the W mine site. The human health risk assessment displayed that 73.43% of children with an unacceptable non-carcinogenic risk, and W contributed the most to the overall non-carcinogenic risk (42.32%), followed by Cd and arsenic (As). In addition, 22.03% of children and 13.4% of adults were under a significant carcinogenic risk. Overall, our findings emphasise the importance of considering element W in future studies investigating the contamination of HMs around W mining areas. As such, we calculated a safe limit value for element W in soil (141.01 mg/kg) to facilitate the conservation and development of soils in W mining areas in China. Our study provides valuable information for pollution prevention and soil contamination risk mitigation in W mining areas.

Keywords: toxic element; toxicity; soil remediation; hazardous waste; safety limit

Tungsten (W), a transition metal known for its toughness, elevated fusion point, and chemical inertness, is primarily found in the scheelite ($CaWO_4$) and wolframite ($[FeMn]WO_4$) forms in the terrestrial system. Its unique properties make it a valuable resource in various fields, such as metallurgy, electronics, and military equipment (Petruzzelli and Pedron 2021). China has the largest reserves of

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W in the world, with over 252 mine sites located in 21 provinces or regions (Liu et al. 2022). The southern Jiangxi region, known as the "world's tungsten capital", has over a century of W exploitation history (Shao et al. 2013). Due to the disordered mining activities, improperly treated W ores waste and smelting slag continuously release various elements into the environment under natural forces, accumulating in soils through pathways such as groundwater transport and atmospheric particle deposition. Therefore, the soils around the W mining area have been polluted with heavy metal(loid)s (HMs), including arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), molybdenum (Mo), nickel (Ni), lead (Pb), zinc (Zn), and W (Zhao et al. 2015, Arslan et al. 2018, Zhang et al. 2018, Guo et al. 2021). These HMs, once in the soil, cannot be biodegraded and may cause prolonged negative effects on the ecological systems and human health. Therefore, ascertaining the degree of pollution and assessing the risks to human health caused by the HMs in the soil around the W mining area is paramount.

Previous studies have investigated the pollution status of HMs in soils around a W mining area. Still, they did not include W in the calculation when assessing HMs' pollution status and human health risks (Lin et al. 2014, Arslan et al. 2018, Zheng et al. 2020, Guo et al. 2021). This omission may result in misestimating ecological and human health risks. Despite being considered a green metal that is environmentally stable and non-toxic to human health for a prolonged period, more and more literature has shown that W has poisonous effects on living organisms (Sheppard et al. 2007, Bednar et al. 2008, Datta et al. 2017, Lindsay et al. 2017, Koutsospyros et al. 2019). Soils with excessive W may directly or indirectly affect plants, animals, and microbes, adversely affecting their growth, survival, and reproduction (Strigul et al. 2005). Moreover, the microbial community may change when the W content in the soil exceeds 1% (Ringelberg et al. 2009). Epidemiological investigations have demonstrated a significant correlation between increased concentrations of W in human urine and the occurrence of health issues (Koutsospyros et al. 2019, Li et al. 2023).

Thus, the main objective of this study is to investigate the concentration of HMs in soils of a typical W mining area, including W and other accompanying elements. The specific objectives are (1) to characterise the HMs' concentration in soils; (2) to evaluate the potential ecological risks of HMs in soils, and (3) to determine the human health risk of the individuals residing in the W mining area from soil HMs exposure. The study's outcomes will be valuable in informing soil management and health risk control.

MATERIAL AND METHODS

The study area and sample analysis

Study area. The study site is located in a township situated downstream of a W mine pit (Figure 1) in Ganzhou City, southern Jiangxi province, China

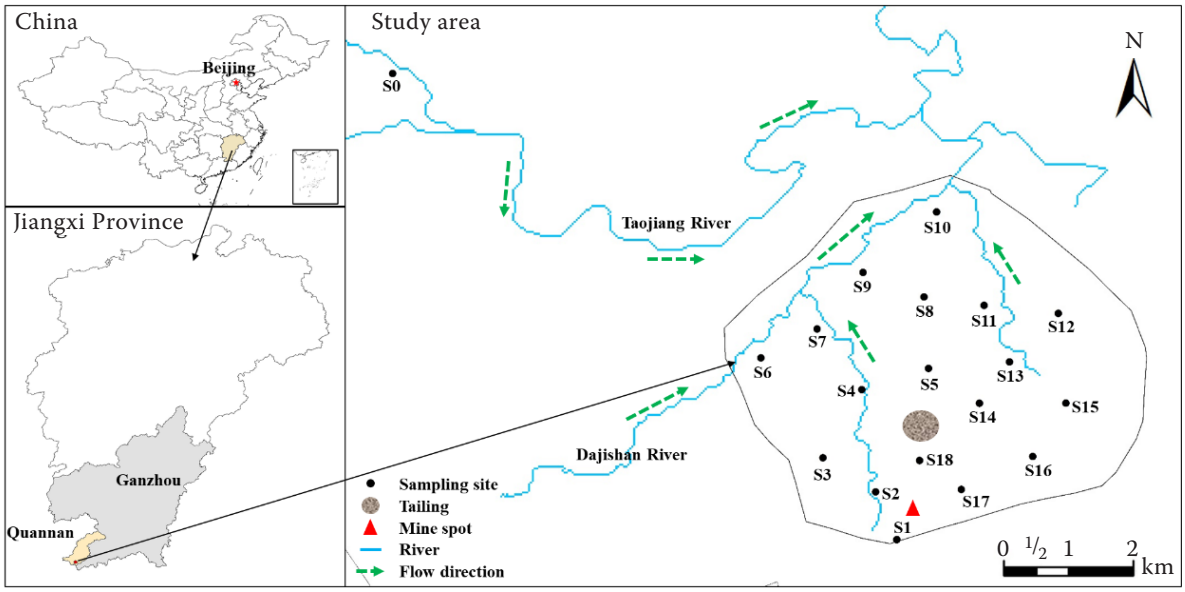

Figure 1. Location of the study area and spatial distribution of sampling sites

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(114°22'32"N, 24°37'26"E). The mine pit is the world's largest known wolframite deposit and has been exploited for over 70 years (Wu 2017). The climate in the study area is characterised as a subtropical monsoon with a prevailing south-southwest (SSW) wind direction. The average yearly temperature and rainfall amount are 18.6 °C and 1 665 mm, respectively. The study area includes a waste rock heap and a mining tailings pond (Figure 1), which can release HMs to surrounding agricultural soils through runoff and aeolian erosion.

Sample collection and preparation. Eighteen agricultural topsoil samples (0–20 cm) were collected in December 2021. The sampling sites were randomly selected to represent the entire study area. Besides, a control site (S0) was chosen upstream of the Taojiang River and far away from the W mining area (Figure 1). The soil samples were collected using a plastic scoop and a stainless-steel shovel. A composited soil sample (about 1 kg) was procured at every sampling point by aggregating five sub-samples from a 20 m × 20 m region. Each collected soil was placed in sampling bags and transferred to the laboratory. All samples were dried indoors at room temperature after removing the plant or wood wastes and stones. Then, grind them into fine particles with an agate mortar, followed by homogenisation and sieving through a 0.15 mm mesh. All dried samples were stored in a desiccator before metal analysis.

Chemical analysis. This study focuses on the HMs of W and other accompanying elements, including As, Cd, Co, Cr, Cu, Mo, Ni, Pb, and Zn, in soil samples. The W content in soils was analysed using the USEPA standard procedure 3050B (USEPA 1996). Briefly, a 1.000 g dried soil sample was precisely weighed into a Teflon digestion vessel and digested on a graphitic panel heater at 95 ± 5 °C for 30 min without boiling after adding 10 mL 1:1 (*v*/*v*) HNO_3. The step of adding 5 mL concentrated HNO_3 and heating the mixture for 30 min was repeated four times. Then, 3 mL of 30% H_2O_2 was added, and the samples were heated for 15 min. After that, 2 mL 30% H_2O_2 was added, and the slurry was mixed and heated for 2 h. After the sample was thoroughly cooled, solid particles in the digested solution were separated by filtration, and the resulting filtrate was diluted to 50 mL with 1% HNO_3 before subsequent analysis. Finally, the W concentration was measured by the ICP-MS (Agilent 7800, Santa Clara, USA).

The concentrations of the other HMs in soils were measured according to a previously reported method (Xiang et al. 2020). A 0.500 g soil sample was precisely dissolved with a blend of HNO_3/HF/$HClO_4$ (10/5/2, *v*/*v*/*v*) in a 200 mL polytetrafluoroethylene tube. The digestion process was conducted at 110 °C for 30 min, 140 °C for 30 min, and 180 °C until the soil was completely digested. The clarified digestion fluid was diluted with deionised water to 25 mL, and the concentration of HMs was measured using ICP-MS (Agilent 7800).

All HNO_3, HCl, $HClO_4$, HF, and H_2O_2 reagents were purchased from the Xilong Scientific Co., Ltd. (Guangdong, China). The standard solutions for each HM were purchased from the General Research Institute for Nonferrous Metals (Beijing, China). Deionised water was utilised throughout the experiment, and all plastic containers and glassware utilised for soil digestion were soaked overnight in 25% HNO_3 and washed three times with deionised water before use. Reagent blanks and standard reference material (GBW07451) were applied for quality assurance.

Potential ecological risk evaluation

In this study, we employed a novel method for evaluating potential ecological risks, referred to as the Nemerow integrated risk index (NIRI) (Men et al. 2020). This approach was derived from the combination of the Nemerow integrated pollution index (NIPI) and the potential ecological risk index (RI) methods, which are commonly utilised for quantifying soil HMs contamination and ecological risk (Yang et al. 2011, Zhao et al. 2015, Wang et al. 2017, Li et al. 2019). However, each of these methods has significant limitations. For example, the NIPI fails to account for differences in the toxicity of individual HMs, while the RI does not consider the influence of the number of HMs. The NIRI method was developed to overcome these limitations, which comprehensively considers both the toxicity factor and the number of HMs. The calculation of NIRI follows the Eqs. 1–3:

$$ERI_i = T_r^i \times \frac{C_i}{S_i} \quad (1)$$

$$RI = \sum ERI_i \quad (2)$$

$$NIRI = \sqrt{\frac{(ERI_{max})^2 + (ERI_{average})^2}{2}} \quad (3)$$

Where: NIRI – Nemerow integrated risk index of HMs in soil; ERI_i – ecological risk index of HMs *i* in soil; T_r^i – toxicity coefficient of HMs *i*; C_i – detected concentration of HMs *i* in soil, mg/kg; S_i – reference values (the background values were used in this study) of HMs *i* in soil, mg/kg. The T_r^i and S_i values were listed in Table 1.

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Table 1. The toxicity coefficient (T_r^i) and reference values (S_i) values for each heavy metal(loid)s (HMs) in this study

HMs	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	W	Zn
T_r^i	10[a]	30[a]	5[a]	2[a]	5[a]	1[b]	5[a]	5[a]	2[c]	1[a]
S_i (mg/kg)	8.85	0.09	9.14	34.56	15.17	0.6	12.35	34.19	5.1	58.05

[a]Xu et al. 2008; [b]Song et al. 2021; [c]Li et al. 2023

Following the definition of the ecological risk index (ERI), the upper limit for the first level threshold was set at the highest T_r^i value among all the determined HMs in this study (30 for Cd). Subsequently, the higher-level limit values for ERI were adjusted accordingly. The classification scheme for the NIRI index used in this study was based on previous research (Men et al. 2020). The classification schemes for ERI and NIRI are presented in Table 2.

Deterministic exposure and health risk assessment models

Exposure dose. This study assessed HMs exposure among populations *via* soil by evaluating three pathways: ingestion, inhalation, and dermal contact. To account for differences in physiology, two distinct population groups were assessed, namely children (4–18 years old) and adults (> 18 years old). Eqs. 4–7 were utilised to determine the average daily dose (ADD) of HMs for each exposure pathway.

$$ADD_{ing} = \frac{C_i \times IR_{ing} \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (4)$$

$$ADD_{inh} = \frac{C_i \times IR_{inh} \times EF \times ED}{PEF \times BW \times AT} \quad (5)$$

$$ADD_{der} = \frac{C_i \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (6)$$

$$ADD_{total} = ADD_{ing} + ADD_{inh} + ADD_{der} \quad (7)$$

Where: ADD_{ing}, ADD_{inh}, ADD_{der} and ADD_{total} – average daily dose of soil through ingestion, inhalation, dermal contact, and the comprehensive way, respectively, mg/(kg·d); C_i – concentration of HMs in soil, mg/kg. The definition and the reference value of other parameters of these equations are listed in Table 3.

Health risk assessment. In accordance with USEPA guidelines, the potential health impacts of toxic chemicals can be divided into two categories: carcinogenic risk (CR) and non-carcinogenic risk (NCR). Generally, the total carcinogenic risk (TCR) is determined by summing the CR values of individual chemicals, and the cumulative NCR associated with a mixture of chemicals is quantified by the hazard index (HI). Eqs. 8 and 9 can be used to calculate the TCR and HI, respectively.

$$TCR = \sum CR_i = \sum \left(ADD_{ing} \times SF_{ing} + ADD_{inh} \times SF_{inh} + ADD_{der} \times SF_{der}\right) \quad (8)$$

$$HI = \sum HQ_i = \sum \left(\frac{ADD_{ing}}{RfD_{ing}} + \frac{ADD_{inh}}{RfD_{inh}} + \frac{ADD_{der}}{RfD_{der}}\right) \quad (9)$$

Where: CR_i – potential carcinogenic risk of HM i; HQ_i – hazard quotient of HM i; SF and RfD – carcinogenic slope factor and reference dose of the ADD through different exposure ways, respectively (Table 4).

Regarding CR, a TCR value greater than 1.00E-04 signifies a significant risk of cancer, while a TCR value lower than 1.00E-06 is deemed to have a negligible cancer risk. Concerning NCR, a maximum acceptable threshold value of 1 is established for the HI. If the HI value exceeds 1, it indicates possible adverse health impacts, and caution should be exercised.

Table 2. The classification of ecological risk index (ERI) and Nemerow integrates risk index (NIRI)

Ecological risk index	Nemerow integrates risk index	Description
ERI ≤ 30	NIRI ≤ 30	low risk
30 < ERI ≤ 60	30 < NIRI ≤ 60	moderate risk
60 < ERI ≤ 120	60 < NIRI ≤ 120	considerable risk
120 < ERI ≤ 240	120 < NIRI ≤ 240	high risk
ERI > 240	NIRI > 240	extreme risk

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Table 3. The definition and the reference value of parameters used in the health risk assessment model

Parameter	Definition	Unit	Children	Adult	Reference
IR_{ing}	ingestion rate	mg/day	TRI (66, 103, 161)	TRI (4, 30, 52)	(Yang et al. 2019)
IR_{inh}	inhalation rate	m^3/day	UN (8.4, 13.3)	UN (11.2, 19.6)	(Duan 2016, Zhao 2015)
EF	exposure frequency	day/year	TRI (180, 345, 365)		(Chen et al. 2019)
ED	exposure duration	year	6	24	(Huang et al. 2018)
PEF	particle emission factor	m^3/kg	1.36×10^9		(Huang et al. 2018)
SA	exposed skin area	cm^2	2 800	5 700	(Gu et al. 2016)
AF	adhesion factor of the skin	mg/(m^2·d)	0.2	0.07	(Huang et al. 2018)
ABS	dermal absorption factor	–	0.001 (non-carcinogenic); 0.01 (carcinogenic)		(Huang et al. 2021)
BW	average body weight	kg	UN (17.5, 51.8)	UN (42.3, 77.8)	(Duan 2016, Zhao 2015)
AT	average exposure time	day	365 × ED (non-carcinogenic); 365 × 70 (carcinogenic)		(Huang et al. 2021)

UN – minimum, maximum, uniform distribution; LN – mean, standard deviation, lognormal distribution; TRI – minimum, likeliest, maximum, triangular distribution

Conversely, if the HI value is less than 1, it generally indicates minimal adverse health impacts.

Monte Carlo simulation. To consider the variability of the factors mentioned above, a Monte Carlo simulation method was employed to calculate the exposure dosage of HMs, CR, and NCR for the populations with 20 000 iterations (Huang et al. 2021). The concentrations of HMs in soil followed a lognormal distribution. The optimal probability distribution of exposure factors such as ingestion rate (IR_{ing}), inhalation rate (IR_{inh}), exposure frequency (EF), skin adherence (SA), adherence factor (AF), and body weight (BW) was determined and presented in Table 4. These factors were incorporated into the Monte Carlo simulation, which used Matlab R2022a (The MathWorks Inc., Natick, USA).

Statistical analysis. The statistical analysis was performed using SPSS Statistics 23 software (Armonk, USA), including multivariate analysis, correlation analysis using Pearson's correlation, principal component analysis (PCA), and hierarchical clustering analysis (HCA). The source identification was conducted by factor extraction with eigenvalues greater than 1 after varimax rotation. The normal distribution of the data was evaluated using the Shapiro-Wilk test.

Table 4. Corresponding slope factor (SF) and reference dose (RfD) values of heavy metal(loid)s (HMs) in soil by different pathways

Element	SF			RfD		
	ingestion (mg/(kg·d))	inhalation (mg/m^3)	dermal (mg/(kg·d))	ingestion (mg/(kg·d))	inhalation (mg/m^3)	dermal (mg/(kg·d))
As	1.50E+00[a]	1.51E+01[c]	3.66E+00[c]	3.00E-04[a]	1.50E-05[a]	1.23E-04[b]
Cd	1.60E+01[d]	6.30E+00[b]	n/a	1.00E-04[a]	1.00E-05[a]	1.00E-05[b]
Co	n/a	9.80E+00[c]	n/a	3.00E-04[a]	6.00E-06[a]	1.60E-02[e]
Cr	5.00E-01[a]	4.20E+01[b]	n/a	3.00E-03[a]	1.00E-04[a]	6.00E-05[b]
Cu	n/a	n/a	n/a	4.00E-02[a]	4.02E-02[b]	1.20E-02[b]
Mo	n/a	n/a	n/a	5.00E-03[a]	2.00E-03[a]	1.90E-03[e]
Ni	n/a	8.40E-01[b]	n/a	2.00E-02[b]	2.06E-02[b]	5.40E-03[b]
Pb	8.50E-03[a]	n/a	n/a	3.50E-03[b]	3.52E-03[b]	5.25E-04[b]
W	n/a	n/a	n/a	8.00E-04[a]	n/a	n/a
Zn	n/a	n/a	n/a	3.00E-01[a]	3.00E-01[b]	6.00E-02[b]

n/a – data not available; [a]USEPA 2022; [b]Gu et al. 2016; [c]Lu et al. 2014; [d]Fan et al. 2017; [e]Ferreira-Baptista and De Miguel 2005

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RESULT AND DISCUSSION

The concentration of HMs in soil. The statistical data of HMs concentration, background values (BGV) in Jiangxi province, and the risk screening values (RSV) and risk interventional value (RIV) for soil pollution in the farmland are shown in Table 5. On average, the HMs concentrations follow the sequence: W > Zn > Pb > Cr > Cu > As > Ni > Co > Cd > Mo. W has the highest concentration, accounting for 29.1% of the total HMs content. In comparison to BGV values, Cr, Cu, and W were higher than BGV at all sites, while for As, Cd, Co, Ni, and Pb, the concentrations exceeded BGV in 72.22, 88.89, 77.78, 72.22, and 94.44% of the sampling sites, respectively, indicating that these HMs were highly enriched in the soil. Except for Cd, none of the other HMs in the soil exceeded the RSV, indicating the negative impacts of these HMs on agricultural product quality and crop growth. However, the mean values of Cd concentration in soil were 15.47 and 3.09 times higher than the RSV and RIV, respectively, indicating that the soils in this study area were seriously contaminated. Furthermore, Cd concentrations exceeded the RIV at 50.00% of the sampling sites. According to the definition of RSV in the Chinese national standard (GB 15618-2018) (CMEE 2018), these contaminated soils are unsuitable for agricultural production due to the high risk of agricultural products not meeting quality and safety standards, requiring strict control measures. Generally, the coefficient of variation (*CV*) is associated with the spatial variance of HMs in soils and reflects the natural or extrinsic factors (Zhao et al. 2010). In this study, the *CV* of Cd, As, Mo, W, and Cu is larger than 70.00%, suggesting that anthropogenic activities significantly impacted the distribution of soil HMs.

A comparison of the soil HMs concentration between this study and previous studies that examined the degree of HMs pollution in the soil in the vicinity of a W mining area was presented in Table 6. It revealed that HMs were found to contaminate soils at all sites, and the distribution of HMs varied between mining sites. The HM distribution in this study was similar to that in the W mines in Jiangxi Province but differed significantly from those in other provinces or countries. This may be attributed to wolframite being the primary W ore in Jiangxi, while scheelite is the primary W ore in other areas. Additionally, the concentrations of As and Pb in the soil around the Xianghualing W mine were nearly ten times higher than in other mines (Table 6). Similar conclusions were drawn regarding the W concentrations in the Etibank Wolfram and the Carrock Fell mines (Table 6). Alongside the W mining activities, W smelting and military applications were also important sources of W pollution. Du et al. (2021) revealed that the W concentration in topsoil in an abandoned smelting plant reached a maximum value of 3 191.00 mg/kg.

Table 5. Concentrations of heavy metal(loid)s (HMs) (mg/kg) in soils from the study area

HMs	Minimum	Median	Maximum	Mean	SD	*CV* (%)	BGV	RSV	RIV
As	5.34	17.94	82.18	25.29	22.06	87.24	8.85	30.00	200.00
Cd	0.07	2.01	27.20	4.64	7.55	162.56	0.09	0.30	1.50
Co	7.02	11.84	38.29	17.70	10.96	61.93	9.14	n/a	n/a
Cr	44.70	67.32	95.72	68.42	14.04	20.52	34.56	250.00	800.00
Cu	19.21	37.61	141.77	49.84	36.05	72.34	15.17	150.00	n/a
Mo	0.02	0.41	1.75	0.55	0.48	86.83	0.60	n/a	n/a
Ni	7.54	16.95	34.31	18.00	6.80	37.77	12.35	60.00	n/a
Pb	28.20	60.78	130.23	71.42	28.74	40.24	34.19	80.00	400.00
W	6.24	125.85	517.20	144.17	124.93	86.65	5.10	n/a	n/a
Zn	27.60	55.50	223.27	71.49	47.06	65.82	58.05	200.00	n/a

n/a – not available; SD – standard deviation; *CV* – coefficient of variation; BGV – background value of heavy metal(loid)s in soil at Jiangxi province (He et al. 2006); RSV and RIV – risk screening values and risk interventional values for soil contamination of agriculture land from GB 15618-2018 (CMEE 2018), respectively, and the value were selected in pH ≤ 5.5

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Multivariate analysis of soil heavy metal(loid)s

A Pearson correlation matrix was calculated and presented in Table 7 to investigate the relationships among HMs in soils. In general, if the correlation coefficients between metals are found to be significant ($P < 0.05$), they may originate from a shared source and demonstrate comparable transportation characteristics (Tholkappian et al. 2018, Baltas et al. 2020). Furthermore, PCA was used to determine the sources of HMs in soil. Both the Kaiser-Meyer Olkin (KMO) and Bartlett's sphericity (BS) tests were conducted to ensure the data was suitable for PCA. The KMO value was 0.55, and the significant level of the BS test was 0.00, indicating that the data were appropriate for PCA. The analysis revealed three major components, with eigenvalues larger than 1, which explained 79.88% of the total system variance (Table 8).

The first principal component (PC1) explained 51.08% of the total variance, with strong positive loadings (> 0.70) for As, Cd, Cu, Mo, W, and Zn. Based on the results of Pearson's correlation analysis, significant ($P < 0.01$) positive correlations were found between W and Cd ($r = 0.74$), Cu ($r = 0.85$), Mo ($r = 0.53$), and Zn ($r = 0.78$), shown in Table 7, suggesting that they may have a common source. Anthropogenic sources, such as untreated waste W ore and smelting slag, continuously release W and its accompanying elements (including Cd and Mo) under the influence of natural forces, leading to their accumulation in the soil through groundwater transportation and atmospheric particle deposition (Clausen and Korte 2009, Datta et al. 2017, Du et al. 2021). Additionally, As was significantly correlated with Cd and Cu (Table 7), suggesting their common source. In addition, previous studies have reported that W mining activities may lead to As pollution (Shao et al. 2013, Wang et al. 2015, Zheng et al. 2020, Guo et al. 2021). Thus, PC1 (including As, Cd, Cu, Mo, W, and Zn) can be attributed to W mining activities.

The second principal component (PC2), which accounted for 23.36% of the total variance, exhibited high positive loadings for Co, Cr and Ni. The significant ($P < 0.01$) positive correlation among these three HMs (Table 8) suggested a possible common natural source. According to Rodríguez et al. (2008), the distribution of Cr and Ni in soils is mainly influenced by the geochemical weathering of parent rock material. In addition, the Co was usually identified

Table 6. The comparison of heavy metal(loid)s (HMs) average concentration (mg/kg) in the soil around the tungsten (W) mining areas

W mine name	Location	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	W	Zn	Reference
Dajishan	Jiangxi Province, China	25.29	4.64	17.70	68.42	49.84	0.55	18.00	71.42	144.17	71.49	this study
Dangping	Jiangxi Province, China	59.00	1.60	/	74.80	51.70	/	/	77.30	80.30	327.50	(Zheng et al. 2020)
Tieshanlong	Jiangxi Province, China	72.05	7.47	/	53.61	152.32	/	21.72	117.77	/	237.41	(Guo et al. 2021)
Xihuashan	Jiangxi Province, China	/	0.72	8.07	70.50	50.70	/	21.80	75.30	/	112.10	(Shao et al. 2013)
Xianghualing	Hunan province, China	2 617.98	3.76	/	68.88	132.53	/	/	790.17	/	784.37	(Zhang et al. 2018)
Lianhuashan	Guangdong province, China	72.20	0.18	11.70	36.30	40.70	1.51	13.60	90.70	22.3	125.6	(Guo et al. 2017)
Dai Tu district	Thai Nguyen province, Vietnam	52.50	1.18	/	20.80	179.00	/	6.10	50.70	/	84.30	(Chu Ngoc et al. 2009)
Etibank Wolfram	Bursa, Turkey	0.95	2.77	9.28	0.50	224.6	0.80	/	/	1 378.60	376.90	(Erdemir et al. 2017)
Carrock Fell	Cumbria, UK	/	/	/	/	/	/	/	/	1 240.00	/	(Wilson and Pyatt 2006)

Note that the concentration data were presented at an average value

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Table 7. The Pearson's correlation analysis between heavy metal(loid)s of soils

	As	Cd	Co	Cr	Cu	Mo	Ni	Pb	W	Zn
As	1.00									
Cd	0.60**	1.00								
Co	–0.02	0.12	1.00							
Cr	0.24	0.19	0.72**	1.00						
Cu	0.67**	0.66**	0.55*	0.53*	1.00					
Mo	0.51*	0.68**	0.12	0.23	0.64**	1.00				
Ni	0.18	0.00	0.82**	0.86**	0.56*	0.13	1.00			
Pb	0.02	0.21	0.49*	0.22	0.51*	0.49*	0.34	1.00		
W	0.55*	0.76**	0.17	0.07	0.60**	0.73**	0.07	0.37	1.00	
Zn	0.51*	0.74**	0.57*	0.43	0.85**	0.53*	0.42	0.43	0.78**	1.00

*$P < 0.05$; **$P < 0.01$

as lithogenic sources (Guo et al. 2017). Thus, PC2 can be interpreted as a natural source.

The third principal component (PC3) accounted for 10.32% of the total variance, with contributions from Pb. As shown in Table 7, Pb was significantly correlated ($P < 0.01$) with Cu and Mo, indicating a common source. Additionally, hierarchical cluster analysis (HCA) was conducted, and the dendrogram was obtained (Figure 2), showing that the HMs in the soil were classified into three clusters. Each cluster contained three (Cu, Zn, and Cd), four (Mo, W, As, and Pb), and three (Co, Ni, and Cr) elements, respectively. Furthermore, the first and second clusters were merged, suggesting they (Cu, Zn, Cd, Mo, W, As, and Pb) may have originated from one source (W mining activities). However, PCA revealed a different Pb pattern than other heavy metals. Therefore, we speculated that Pb may originate from vehicle exhaust emissions during the minerals transportation after W mining, as Pb is a component of gasoline, and previous researches also supports this conclusion (Cai et al. 2015, Jiang et al. 2019, Baltas et al. 2020). Thus, PC3 can be attributed to the source of W mining activities (vehicle exhaust during the transportation of minerals).

Table 8. Varimax rotated the component matrix of heavy metal(loid)s in the study area

Parameter	PC1	PC2	PC3
As	**0.80**	0.16	–0.40
Cd	**0.91**	0.01	0.03
Co	0.06	**0.86**	0.39
Cr	0.15	**0.91**	–0.05
Cu	**0.75**	0.54	0.17
Mo	**0.79**	0.03	0.32
Ni	0.05	**0.96**	0.09
Pb	0.25	0.25	**0.87**
W	**0.88**	–0.03	0.26
Zn	**0.77**	0.43	0.23
Eigenvalues	5.11	2.34	1.03
% of variance	51.08	23.36	10.32
Cumulative %	51.08	74.44	84.77

Ecological risk assessment

As shown in Figure 3, the HMs investigated in this study were ranked in the following order based on their average ERI values: Cd > W > As > Cu > Pb > Co > Ni > Cr > Zn > Mo. Half of the samples displayed high or extreme ecological risks associated with Cd contamination, with the RI contribution from

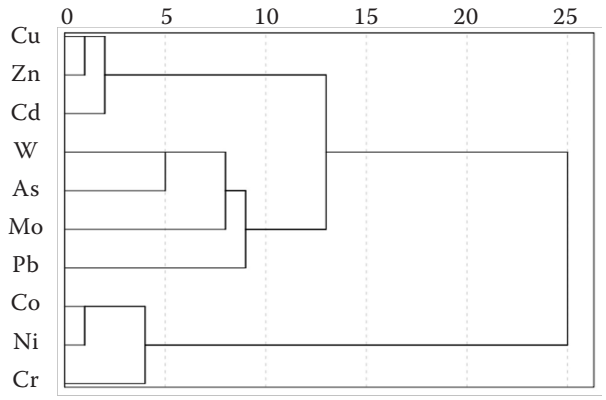

Figure 2. Dendrogram of heavy metal(loid)s concentrations in the topsoils

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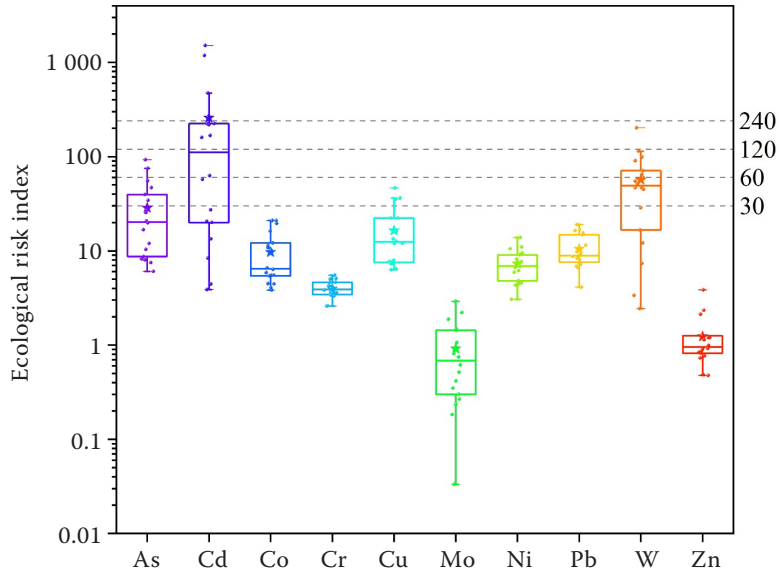

Figure 3. The ecological risk index of heavy metal(loid)s (HMs) in soil (the boxplot exhibits the quartile distribution of ecological risk index (ERI), and the five-pointed star represents the mean value of ERI)

Cd being as high as 89.70% and averaging 42.81%. These findings suggest that Cd pollution poses the greatest threat to the study area, which is consistent with previous studies conducted in several regions of China (Wang et al. 2021, Li et al. 2022, Liu et al. 2022). Moreover, it is noteworthy that the potential ecological risks arising from W contamination cannot be overlooked, given that the ERI values of six samples indicated a considerable or high ecological risk. Conversely, for the remaining HMs, the ERI values of all samples (excluding As at S3 and S13) were below 60, indicating low or moderate ecological risk (Figure 3). Therefore, based on these results, we suggest that Cd and W should be regarded as priority-controlled contaminants in the soil surrounding the W mining area.

The NIRI values ranged from 10.89 to 1 075.14, with a median of 85.00, indicating that more than half of the sampling sites exhibited at least considerable ecological risks. Furthermore, the average value of NIRI was 189.80, suggesting a high ecological risk across the entire study area. As presented in Figure 4, though NIRI values exceeded 240 at three sampling points (S10, S1, and S2), the potential ecological risk across the entire study area did not reach a level of high risk after kriging interpolation, the NIRI values of the sampling sites were ranked in the following order: S10 > S1 > S2 > S4 > S11 > S5 > S6 > S3 > S9 > S13 > S7 > S12 > S8 > S14 > S15 > S18 > S17 > S16. S10, although far from mining areas and tailings ponds, has the highest NIRI because of the very high Cd content at this sampling site. Overall, the potential ecological risks gradually increase from southeast to northwest (Figure 4), and this trend aligns with the flow direction of the two tributaries. Therefore, the relatively low risks observed in the southeast region could be attributed to the barrier effect of the river on pollution transportation (Liu et al. 2019).

Health risk assessment

Non-carcinogenic risk assessment. The non-carcinogenic risk assessment results, which include the values of ADD, HQ, and HI, are presented in Table 9. For both children and adults, the distributions of ADD for all exposure pathways in soil were ranked in the following order: W > Zn > Pb > Cr > Cu > As > Cd > Co > Ni > Mo. The ADD_{total} values for children were approximately seven times higher than those for adults. Oral ingestion was the predominant exposure pathway (accounting for 99.5% of the ADD_{total}), consistent with previous studies (Wei et al. 2015, Eziz et al. 2018, Baltas et al. 2020).

The HQ values for all exposure pathways were found to be higher for children than for adults, possibly due to their lower body weight and increased soil intake from hand-to-mouth contact (Zhao et al. 2014, Kusin et al. 2018, Kabir et al. 2021). These results are consistent with previous studies which have reported higher non-carcinogenic risks for children in mining areas compared to adults (Li et al. 2014, Liu et al. 2022). The HQ_{total} values for children and adults ranging from 5.60E-04 to 1.06E + 00 and 8.20E-05 to 1.51E-01, respectively. Regardless of age group, the HQ_{total} value followed the order: W > Cd > As > Co > Cr > Pb > Cu > Ni > Zn > Mo. W contributed

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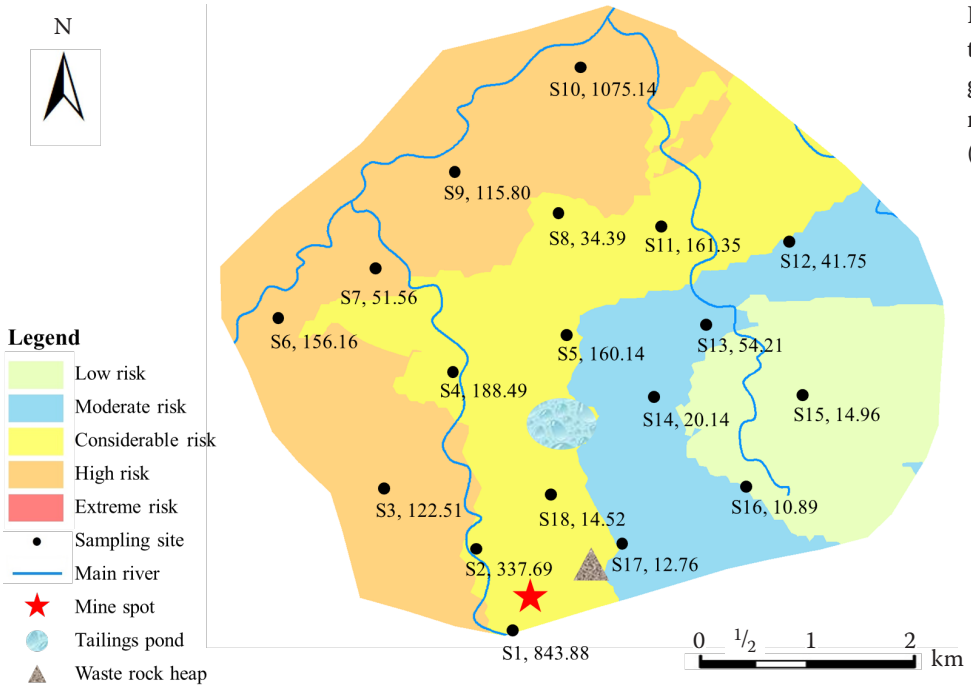

Figure 4. The spatial distribution of the pollution grades based on the Nemerow integrates risk index (NIRI) values

Table 9. Non-carcinogenic risks to humans from soil heavy metal(loid)s (HMs) exposure

Element	Adding		ADD_{inh}		ADD_{der}		ADD_{total}	
	children	adults	children	adults	children	adults	children	adults
As	1.03E-04	1.46E-05	7.44E-09	5.75E-09	5.23E-07	2.02E-07	1.03E-04	1.48E-05
Cd	6.54E-05	9.48E-06	4.74E-09	3.72E-09	3.34E-07	1.31E-07	6.57E-05	9.61E-06
Co	5.90E-05	8.37E-06	4.28E-09	3.31E-09	3.01E-07	1.16E-07	5.93E-05	8.49E-06
Cr	1.98E-04	2.81E-05	1.44E-08	1.11E-08	1.01E-06	3.91E-07	2.00E-04	2.85E-05
Cu	1.72E-04	2.44E-05	1.25E-08	9.63E-09	8.77E-07	3.39E-07	1.73E-04	2.48E-05
Mo	2.76E-06	3.95E-07	2.01E-10	1.56E-10	1.41E-08	5.49E-09	2.77E-06	4.01E-07
Ni	5.48E-05	7.76E-06	3.98E-09	3.07E-09	2.80E-07	1.08E-07	5.51E-05	7.87E-06
Pb	2.18E-04	3.10E-05	1.58E-08	1.22E-08	1.11E-06	4.31E-07	2.19E-04	3.14E-05
W	8.50E-04	1.21E-04	6.16E-08	4.74E-08	4.33E-06	1.67E-06	8.54E-04	1.22E-04
Zn	2.31E-04	3.28E-05	1.67E-08	1.30E-08	1.18E-06	4.57E-07	2.32E-04	3.33E-05
	HQ_{ing}		HQ_{inh}		HQ_{der}		HQ_{total}	
	children	adults	children	adults	children	adults	children	adults
As	3.43E-01	4.85E-02	4.96E-04	3.84E-04	4.26E-03	1.65E-03	3.47E-01	5.05E-02
Cd	6.54E-01	9.48E-02	4.74E-04	3.72E-04	3.34E-02	1.31E-02	6.87E-01	1.08E-01
Co	1.97E-01	2.79E-02	7.13E-04	5.51E-04	1.88E-05	7.27E-06	1.97E-01	2.85E-02
Cr	6.62E-02	9.37E-03	1.44E-04	1.11E-04	1.69E-02	6.52E-03	8.32E-02	1.60E-02
Cu	4.30E-03	6.10E-04	3.10E-07	2.40E-07	7.31E-05	2.82E-05	4.37E-03	6.39E-04
Mo	5.52E-04	7.90E-05	1.00E-07	7.81E-08	7.42E-06	2.89E-06	5.60E-04	8.20E-05
Ni	2.74E-03	3.88E-04	1.93E-07	1.49E-07	5.18E-05	2.00E-05	2.79E-03	4.08E-04
Pb	6.24E-02	8.86E-03	4.50E-06	3.48E-06	2.12E-03	8.21E-04	6.45E-02	9.68E-03
W	1.06E+00	1.51E-01	6.16E-15	4.74E-15	4.33E-13	1.67E-13	1.06E+00	1.51E-01
Zn	7.69E-04	1.09E-04	5.58E-08	4.32E-08	1.96E-05	7.61E-06	7.89E-04	1.17E-04

ADD_{ing}, ADD_{inh}, ADD_{der} and ADD_{total} – average daily dose of soil through ingestion, inhalation, dermal contact, and the comprehensive way, respectively; HQ_{ing}, HQ_{inh}, HQ_{der}, and HQ_{total} – hazard quotient through ingestion, inhalation, dermal contact, and the comprehensive way, respectively

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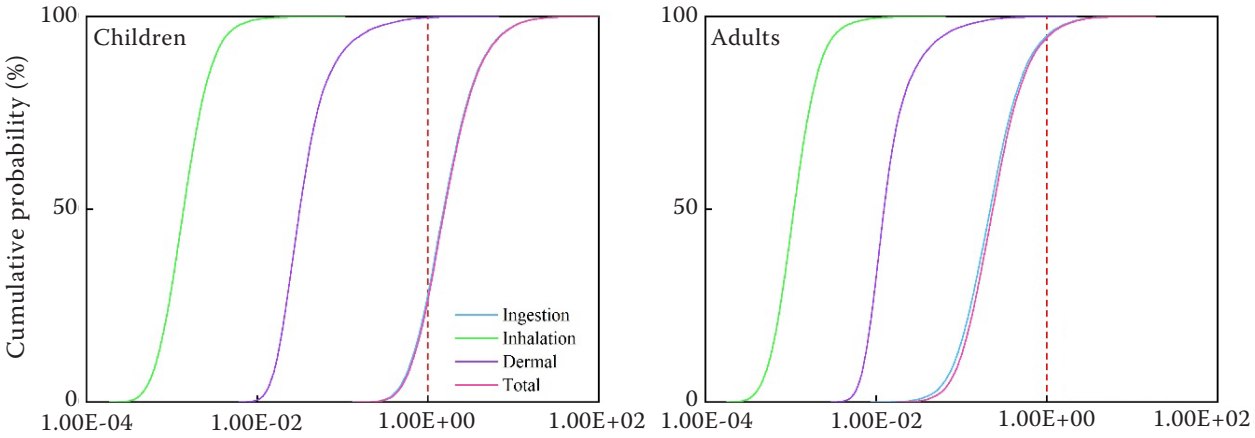

Figure 5. Cumulative probability distribution of hazard index (HI) for the populations

the most (accounting for 42.32%) to the total non-carcinogenic risk, followed by Cd (28.87%) and As (14.01%). Furthermore, no RfD valve of W through inhalation and dermal contact was given in this study (Table 4), but studies have shown that inhalation of W fine particles can cause severe lung diseases (Bolt and Mann 2016). As a result, the non-carcinogenic risk of W may be underestimated. Therefore, the non-carcinogenic risk caused by W exposure should pay closer attention.

According to the cumulative probability distribution of HI for the populations (Figure 5), the total non-carcinogenic risk of 73.43% for children exceeded the maximum acceptable threshold (HI = 1), with an average HI value of 2.49, suggesting a significant non-carcinogenic risk for the children around the W mining area. The non-carcinogenic risk of adults was much lower than children's, and its average HI value was 0.37.

Previous studies have reported the health risk assessment of HMs, including As, Cd, Cr, Hg, Ni, Pb, and Zn, in the soil around the W mining area. Li et al. (2014) reviewed the soil HMs pollution from various mines in China and suggested that W mines should be

Table 10. Carcinogenic risks to humans from soil heavy metal(loid)s (HMs) exposure

Element	Adding		ADD_{inh}		ADD_{der}		ADD_{total}	
	children	adults	children	adults	children	adults	children	adults
As	8.81E-06	4.99E-06	6.37E-10	1.97E-09	4.49E-07	6.94E-07	9.26E-06	5.69E-06
Cd	5.60E-06	3.25E-06	4.06E-10	1.27E-09	/	/	5.89E-06	3.70E-06
Co	/	/	3.67E-10	1.13E-09	/	/	5.31E-06	3.27E-06
Cr	1.70E-05	9.63E-06	1.23E-09	3.81E-09	/	/	1.79E-05	1.10E-05
Ni	/	/	3.41E-10	1.05E-09	/	/	4.94E-06	3.03E-06
Pb	1.87E-05	1.06E-05	/	/	/	/	1.97E-05	1.21E-05
	CR_{ing}		CR_{inh}		CR_{der}		CR_{total}	
	children	adults	children	adults	children	adults	children	adults
As	1.32E-05	7.48E-06	9.62E-09	2.98E-08	1.64E-06	2.54E-06	1.49E-05	1.01E-05
Cd	8.96E-05	5.20E-05	2.56E-09	8.03E-09	/	/	8.96E-05	5.20E-05
Co	/	/	3.59E-09	1.11E-08	/	/	3.59E-09	1.11E-08
Cr	8.51E-06	4.82E-06	5.18E-08	1.60E-07	/	/	8.56E-06	4.98E-06
Ni	/	/	2.86E-10	8.84E-10	/	/	2.86E-10	8.84E-10
Pb	1.59E-07	9.03E-08	/	/	/	/	1.59E-07	9.03E-08

/ – element has no carcinogenic risk from that exposure ways; ADD_{ing}, ADD_{inh}, ADD_{der} and ADD_{total} – average daily dose of soil through ingestion, inhalation, dermal contact, and the comprehensive way, respectively; CR_{ing}, CR_{inh}, CR_{der}, and CR_{total} – carcinogenic risk through ingestion, inhalation, dermal contact, and the comprehensive way, respectively

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prioritised for control due to the high ecological and health risks caused by the accompanying elements of the W mine. However, few studies have investigated the potential health risks associated with W exposure through the soil. Liu et al. (2022) reported that the non-carcinogenic risk from W exposure for workers (adults) in the sand-making area of an abandoned W mine was unacceptable (HI = 1.34), which was higher than that of this study.

Carcinogenic risk assessment. Moreover, this study conducted a carcinogenic risk assessment for As, Cd, Co, Cr, Ni, and Pb, and the mean ADD, CR, and CR_{total} values for these elements were calculated and presented in Table 10. The results indicated that

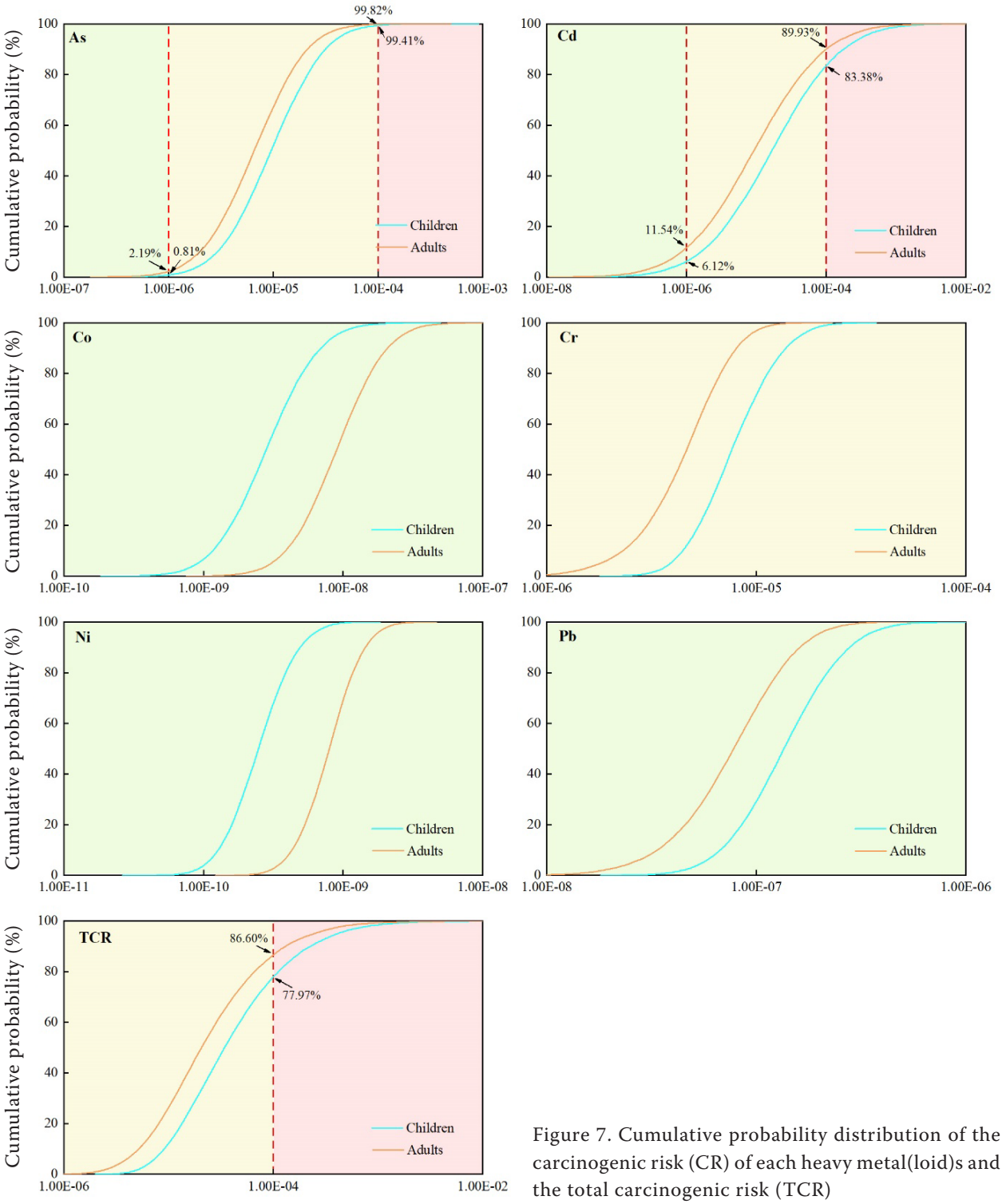

Figure 7. Cumulative probability distribution of the carcinogenic risk (CR) of each heavy metal(loid)s and the total carcinogenic risk (TCR)

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the carcinogenic ADD_{total} value posed by all elements was higher for children than adults, with Pb and Cr having much higher mean carcinogenic ADD_{total} values than the other elements. The ADD_{total} value was ranked as Pb > Cr > As > Cd > Co > Ni for both children and adults. Oral ingestion was the primary exposure route of ADD for both children and adults, accounting for 95.14% and 87.76% of exposure pathways, respectively.

Additionally, as shown in Table 10, the CR_{total} values for children and adults ranged between 2.86E-10 to 8.96E-05 and 8.84E-10 to 5.20E-05, respectively. The CR_{total} value was varied as Cd > As > Cr > Pb > Co > Ni. The probability distribution in Figure 7 showed that the CR_{total} values of Ni, Co, and Pb for children and adults were under the acceptable threshold of 1.00E-06, indicating a negligible cancer risk. In addition, the carcinogenic risk caused by As exposure was within the acceptable range for 97.63% and 98.60% of children and adults, respectively. However, 10.07% and 16.62% of CR_{total} values for Cd exceeded the significant risk threshold (1.00E-04) for children and adults, respectively, suggesting the unacceptable cancer risk. Hence, prioritising Cd and As in the pollutant list is necessary for the study area, consistent with Li et al.'s study (2014). The TCR values for children and adults exceeded the acceptable threshold, indicating that the carcinogenic risk for all populations in the study area cannot be ignored. In particular, 22.03% of children and 13.4% of adults were under a significant carcinogenic risk (Figure 7). Similar conclusions have been found in the soil around a W-Mo mine in Luoyang, China (Wang et al. 2021).

Safety limit value of tungsten in soil

The toxicity and environmental impact of W in soil have been a concern in the United States since 2002 (Seiler et al. 2005), and W was listed as an emerging contaminant in 2008 (USEPA 2008). Although China has listed W smelting slag as a hazardous waste (CMEE 2021), it has not included W in the soil as a contaminant, resulting in a lack of limit standards for W concentration in soil. This situation is not conducive to protecting and exploiting soils in W mining areas in China. Thus, this study aims to calculate a safety limit for W in soil based on the result of a non-carcinogenic risk assessment.

The safety limit value of W in soil was calculated using Eqs. 4 and 9 in inverse deviation. As reported in the Section of "Non-carcinogenic risk assessment", W accounted for 43.34% of the HI_{ing} in the health risk assessment (Table 9). Therefore, the safety limit of W in soil was determined based on the principle that the HQ_{ing} value of W cannot exceed 0.43, resulting in a safety limit of 141.01 mg/kg. This value was higher than the USEPA's regional screening level of 63.00 mg/kg (USEPA 2022) and Li et al.'s study result of 15.26 mg/kg (Li et al. 2021). The differences in the reference values could be attributed to the fact that they were primarily for industrial land use scenarios, where the population was more likely exposed to contaminated soils.

Based on our estimated safety limit, 7 samples (38.89% of total sampling sites) exceeded the limit, indicating that soil remediation in this study area is necessary. As no environmental quality standards for W in soil have been established in China, the limit value obtained in this study may be used as the target value for soil remediation.

Uncertainty analysis

However, there are some limitations to this study. First, we did not consider the bioavailability of HMs in the soil. Second, only direct exposure pathways, such as ingestion, inhalation, and dermal contact with soil, were considered, while the indirect pathway of consumption of contaminated crops was neglected. Third, this study concluded that the Cr is naturally occurring, meaning it mainly existed in the form of $Cr^{(III)}$. At the same time, the cancer risks were calculated assuming the Cr was present as $Cr^{(VI)}$, which has much higher toxicity. All the above factors may lead to the misestimation of the health risk assessment results. Correspondingly, the safety limit value of W calculated based on non-carcinogenic risk may be overestimated or underestimated. We emphasise the importance of ongoing research focusing on human toxicology, the elemental effective state of tungsten, and its bioconcentration factors to better inform soil safety limit values that adequately protect human health and the environment.

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