

Review

Status Assessment, Spatial Distribution and Health Risk of Heavy Metals in Agricultural Soils Around Mining-Impacted Communities in China

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Abstract

China holds diversified mineral resources, and long term mining activities can significantly affect the enrichment of heavy metals (HMs) in agricultural soils. A systematic investigation and overall macro-evaluation on HMs contamination in agricultural soils around mining-impacted communities are urgently needed. International and China national databases were retrieved carefully with the search criteria for articles published from 2007 to 2018. Ultimately, 92 articles fit the inclusion criteria and were selected for further analysis. Based on the collected data, this study then discusses the overall status of HMs concentrations in agricultural soils around the mining-impacted communities and evaluates the pollution levels using the geoaccumulation index. To quantify the non-carcinogenic risks of HMs in soils pose to human health via oral ingestion, the recommended model was applied in the process of health risk assessment, along with Monte Carlo simulation and sensitivity analysis were performed. The results revealed that HMs pollution concentrations and associated ecological risks posed by Cd, Pb and Hg were more serious. Meanwhile, the spatial distribution characteristics suggested that HMs (except for Hg) in southern China generally were severer than those in northern China. Based on the result of human health risk assessment, Cd, As and Pb posed health risks to public in some agricultural sampling sites. It should be noted that 34.02%, 32.99% and 30.93% of HI outputs for children, adults females and males respectively was higher than the guideline value of 1, indicating that children were likely to have an exceptionally sensitive of exposure to environmental contaminants because the unique physiological characteristics and behavioral compared to adults. The results from this study may provide insights for policymakers to develop pollution prevention measures and management strategies in China.

Keywords: heavy metals, agricultural soils, mining-impacted communities, spatial distribution, risks assessment

Introduction

Considering the high transfer ability and the unique biological toxicity of heavy HMs in the soil-crop system, the contamination risk of HMs in agriculture soils has become a great concern around the world [1-3], especially in China. According to the First National Soil Pollution Investigation of China between 2005 and 2013, the official agency issued a document in 2014 that 16.1% sample sites exceeded the guideline values in agriculture soils. Among all contaminated samples, cases related to HMs (including copper (Cu), lead (Pb), zinc (Zn), cadmium (Cd), nickel (Ni), chromium (Cr), mercury (Hg), and arsenic (As)) in agricultural soils accounted for 82.4% [4].

Excessive amounts of HMs in agricultural soils are primarily resulted from the anthropogenic activities, such as mineral exploitation, metal smelting, fertilizer application, sewage irrigation and traffic emissions [5, 6]. Due to mining activities on agricultural soils of the pollution intensity more than other pathways, it is thus urgent to investigate systematically the HMs contamination in agricultural soils from the vicinity of mining-impacted communities. Over the past decade, numerous studies have been published on HMs contamination in agricultural soils near mining areas [7-10]. However, most of the above-mentioned studies have been usually paid attention to the HMs contamination investigation in agricultural soils on regional scales. Although review articles have been revealed the status of HMs in agriculture soils on national scale [11-14], the research target uniformly included various categories

of agriculture soils (e.g. normal farmland, greenhouse farmland, facility farmland, sewage irrigation farmland). The accumulation of HMs in agricultural soils around mining-impacted communities (mining & smelting areas) across China was not yet examined in depth. Combine with the above analysis, in this study, scattered literatures were utilized to analyze the contamination status, spatial distribution characteristics and risk assessment of HMs in agricultural soils around the mining-impacted communities in China.

The main aims of this study are (1) to investigate the overall status of HMs concentrations in agricultural soils; (2) to analyze the contamination levels and spatial distribution of HMs based on ArcGIS software; (3) to assess human health risk based on Monte Carlo simulation and conduct cumulative probabilities and sensitivity analyses of risk exposure.

Material and Methods

Literature Searching

An overall retrieval for research papers related to HMs pollution in agricultural soils near the mining-impacted communities in China between 2007 and 2018 was made. Literature searching was performed in English and Chinese languages, and the main literature databases were carried out in the database of Web of Science Core Collection (WoSCC) and China National Knowledge Infrastructure (CNKI). Through pre-analysis and comparison, we retrieved the intended

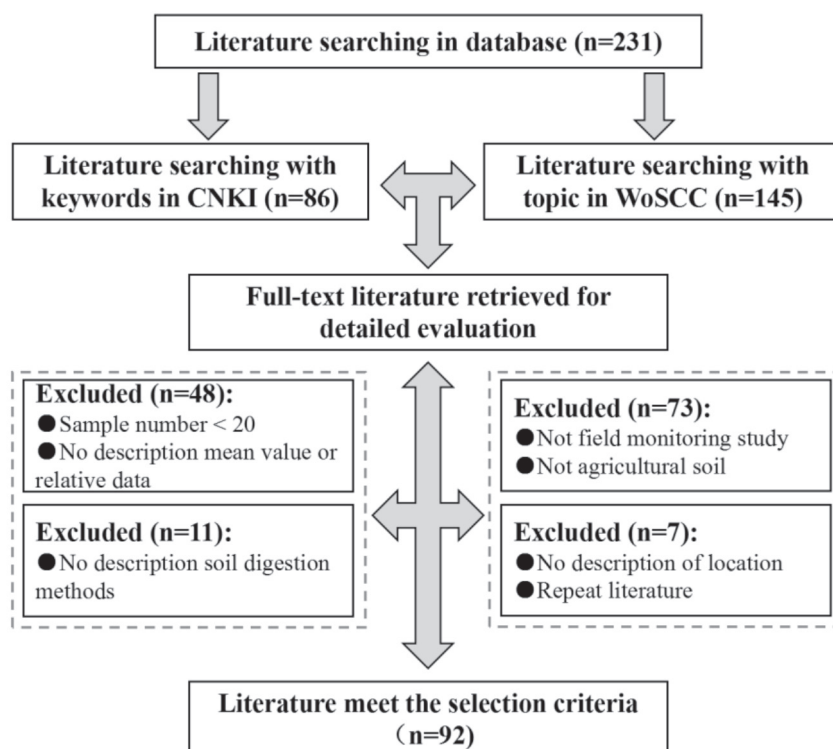


Fig. 1. Literature selection flow chart.

keywords that appeared in the topic of the publications to ensure the accuracy of data collected. The search terms included the following: “TS (Topic) = (heavy metals* OR cadmium OR Cd OR copper OR Cu OR lead OR Pb OR zinc OR Zn OR nickel OR Ni OR mercury OR Hg OR arsenic OR Chromium OR Cr) AND TS = (farmland soils* OR agriculture soils* OR cultivated soils*) AND TS = (mining area* OR smelting area*) AND TS = (China)”. A fuzzy search was performed with a “*” , and the “AND” and “OR” operators were used to make our search results inclusive and restrictive. Eventually, a total of 228 relevant literatures were searched and downloaded from the database of WoSCC and CNKI.

Literature Selection

To be included in unified analysis, the literature that did not meet the following mentioned criteria were excluded: (i) it must be field monitoring study, and topsoil samples were collected in agricultural soils in depth of 0-20 cm from mining-impacted communities; (ii) >20 soils samples were selected; (iii) it should obtain the mean values of HMs or relevant data, or it could be calculated with the sample-number-weighted mean when the mean value was not directly stated; (iv) soils should be digested with acceptable methods such as HCl-HNO₃-HF-HClO₄, HNO₃-HClO₄-HF, HNO₃-HClO₄-HCl, aqua regia digestion and other scientific digestion methods, and the samples were determined by atomic fluorescence spectrophotometry or inductively coupled plasma atomic emission spectrometry; (v) it must be remove duplicate publication literature. The criterion for the literature selection is shown in Fig. 1. Based on the eligibility criteria, 92 eligible were selected.

Data Extraction

We directly extracted data from each eligible literature included: (i) authors, title, and publishing year; (ii) administrative region, study area, and longitude and latitude; (iii) number of sampling sites; (iv) the types of the examined mining-impacted communities (coal mine, lead-zinc mine, multi-metal mine, etc.); (v) the mean value of HMs content. The relevant data from 76 mining areas and 12 smelting areas were extracted, and the cumulative quantity of obtained sampling sites of Cd, Cr, Hg, Pb, As, Cu, Zn and Ni was 8403, 5888, 4458, 7692, 5931, 7188, 7399 and 3592, respectively.

Geo-Accumulation Index

The Geo-accumulation index (I_{geo}) was developed by Muller [15] and has been widely applied in assessment soils contamination [16]. In this study, we used the I_{geo} to determine HMs contamination in soil. I_{geo} was calculated using the following formula:

$$I_{geo} = \log_2 [C_n / (1.5B_n)] \tag{1}$$

...where C_n stands for the measured concentration of the HMs in the examined soils (mg/kg) and B_n represents the geochemical background values of the HMs in the examined soils for different provinces. The Geo-accumulation index is categorized into seven classes based on the severity of contamination ranging from normal to heavy contamination (Table 1).

Health Risk Assessment

Considering the differences in behavioral and physiological, the present review divided the residents who lived in close proximity to the examined mining-impacted areas into three groups: adult males, adult females and children. Large numbers of researches indicated that incidental oral ingestion and dermal absorption through contact with soils particulates appeared to be the primary pathway among the potential exposure routes in the mining areas, but others indeed found that soils ingestion contributed the most to health risk of HMs exposure and played the most important roles among the exposure routes [17, 18]. Thus, this study only evaluated the health risk via oral ingestion of soils. The non-carcinogenic risk via soils ingestion was calculated as follows:

$$ADI_{ing} = C \times \frac{IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{2}$$

$$HQ = \frac{ADI_{ing}}{RfD} \tag{3}$$

$$HI = HQ_{Cd} + HQ_{Cr} + HQ_{Hg} + HQ_{As} + HQ_{Pb} + HQ_{Cu} + HQ_{Zn} + HQ_{Ni} \tag{4}$$

...where C is the HMs concentration in the soils (mg/kg), ADI_{ing} represents the estimated daily intake of the average daily intake of soils HMs by oral ingestion (mg/kg/day). HQ represents the target hazard quotients which is used to assess the potential non-carinogenic

Table 1. The criteria for grades of the geo-accumulation index.

Class	Value	Soil quality
0	$I_{geo} \leq 0$	Practically uncontaminated
1	$0 < I_{geo} \leq 1$	Uncontaminated to moderately contaminated
2	$1 < I_{geo} \leq 2$	Moderately contaminated
3	$2 < I_{geo} \leq 3$	Moderately to heavily contaminated
4	$3 < I_{geo} \leq 4$	Heavily contaminated
5	$4 < I_{geo} \leq 5$	Heavily to extremely contaminated
6	$5 < I_{geo}$	Extremely contaminated

Table 2. Parameter values in average daily dose calculation of HMs.

Parameter	Symbol	Unit	Value		
			Children	Adult males	Adults females
Ingestion rate	IngR	mg/day	LN (24±4)	LN (50±5)	LN (50±5)
Exposure frequency	EF	day/year	TRI (180, 345, 365)	TRI (180, 345, 365)	TRI (180, 345, 365)
Exposure duration	ED	years	POI (6)	POI (30)	POI (30)
Average body weight	BW	kg	TRI (5.25, 29.3, 56.8)	LN (67.55±8.72)	LN (57.59±8.03)
Averaging time for non-carcinogenic	AT	days	POI (365×ED)	POI (365×ED)	POI (365×ED)
Reference dose (<i>RfD</i>)	As	mg/kg/day	TRI (0.0003, 0.0003, 0.0008)		
	Cd		TRI (0.001, 0.001, 0.04)		
	Cr		TRI (0.003, 0.003, 2.5)		
	Ni		TRI (0.02, 0.02, 5)		
	Zn		TRI (0.3, 0.3, 0.91)		
	Hg		TRI (0.0003, 0.0003, 0.0008)		
	Pb		TRI (0.0035, 0.0035, 0.009)		
	Cu		TRI (0, 0.04, 0.04)		

LN, Lognormal distribution. TRI, Triangular distribution. POI, Point distribution.

risk of individual HMs, and HI represents the overall potential non-carcinogenic risk posed by multiple HMs in the study. The other input variables and values (i.e., IngR, EF, ED, BW, AT, and RfD) are given in Table 2. If the HQ or HI<1 indicates minimal health effects, whereas the HQ or HI>1 indicates a probability of adverse health effects.

Uncertainty always exists in risk assessment process [19, 20], especially when the uncertainty arises owing to lack of precise knowledge, the variability of toxic response for individuals and the variability of environmental systems [21, 22]. The Monte Carlo simulation via Oracle Crystal ball software was employed in this study to minimize the uncertainties associated with health risk analysis process. The distribution of input variables for different population groups in the Monte Carlo simulation are shown in Table 2. To ensure the reliability of the results, the model was externally run 10,000 times in each simulation process. In addition, conduct cumulative probabilities and sensitivity were introduced to identify the variable that affected the distribution of risk outcome and calculate the contribution of each input variable to the probabilistic health risk.

Results and Discussion

Overall Status of HMs Concentrations in Agricultural Soils

The statistical information about the selected HMs concentrations in agricultural soils around the mining-

impacted communities is shown in Table 3. It is clear to see that the mean concentration of each metal exceeded the corresponding background value for soils in China, and concentrations of the collected samples for Cd, Hg and Cu nearly exceeded the corresponding reference concentration for 97.8%, 97.7% and 95.3%, respectively. The mean concentrations of Cd, Cr, Hg, As, Pb, Cu, Zn and Ni were about 15.66, 0.85, 0.55, 2.53, 2.98, 1.16, 2.19 and 0.82 times greater than the corresponding national standard values in agricultural soils (GB 15618-2018), respectively. The highest proportion of surpassing the national standard values was found in Cd (88.8%), indicating that mining activities introduced a considerable amount of Cd into agricultural soils, followed by Pb (50.6%), Zn (47.2%), As (39.7%), Cu (29.4%), Cr (13.2%), Hg (11.4%) and Ni (7.3%). According to the above analyses, agricultural soils near mining-impacted areas was contaminated the most by Cd. Previous reports have demonstrated that high Cd concentration in agricultural soils around mining areas was mainly stemmed from the mining activities, such as mineral excavation, ore transportation, tailing disposition, and smelting activities that took place in those areas were led Cd to flow and accumulate into soils [23-27]. Cadmium in soils can be taken up by the roots system of agricultural crops and transported to the above-ground organs. Consumption of agricultural crops grown in Cd-contaminated soils is an important Cd exposure route to human beings and caused extensive threats to human health. Therefore, a series of contamination control and remediation measures should be adopted in China to make significant efforts to address soils Cd contamination.

Table 3. The HMs concentrations (mg/kg) and the percentile concentrations (%) for the eight HMs from the examined the mining-impacted communities.

	Cd	Cr	Hg	As	Pb	Cu	Zn	Ni
Minimum value	0.03	3.07	0.04	4.31	5.95	9.30	22.42	4.55
Maximum value	68.64	3353.6	14.11	1051.44	3204.00	997.00	2576.00	1556.80
Mean value	4.70	170.09	1.33	76.00	357.11	115.65	547.99	82.44
BVSC ^a	0.097	61.00	0.065	11.20	26.00	22.60	74.20	26.90
SEQRCSA ^b	0.30	200.00	2.40	30.00	120.00	100.00	250.00	100.00
>SEQRCSA	88.8%	13.2%	11.4%	39.7%	50.6%	29.4%	47.2%	7.3%
BVSC~SEQRCSA	9.0%	60.3%	86.3%	49.2%	37.1%	65.9%	42.5%	63.4%
<BVSC	2.2%	26.4%	2.3%	11.1%	12.3%	4.7%	10.3%	29.3%

^a Background values for soils in China, it is derived from China National Environmental Monitoring Center.

^b Soils environmental quality risk control standard for soils contamination of agricultural land with $6.5 < pH \leq 7.5$ (GB 15618-2018).

Geo-Accumulation Classification of HMs

Table 4 is presented for the class distribution of geo-accumulation indices for the selected HMs. The results showed that about 85.07% and 92.5% of all the sampling sites for Cr and Ni were fallen below Class 2. However, the I_{geo} values for Cd and Pb ranged from Class 0 to Class 6, among which, 23.60% and 5.62% of the sampling sites for this two HMs belong to Class 6. Most of the I_{geo} values for other HMs were laid below Class 4, among which, >60% of the sampling sites for As and Cu were lower than Class 2, while 20.93% of the sampling sites for Hg belonged to Class 6. Based on the above analysis, although the selected HMs pollution levels varied among the examined sites, agricultural soils near the mining-impacted communities have been more severely polluted by Cd and Hg. This result was in the lines of a previous review by Yang et al. [28], which revealed that >50% of the soils sampling sites in agricultural regions were above Class 1 for Cd and Hg.

The spatial distribution of HMs which was based on the standard classes of I_{geo} is demonstrated in

Fig. 2. Higher I_{geo} values of Cd from the collected database were dispersed over a wide area, including Yunnan, Guangdong, Guangxi, Hunan, Henan, Sichuan, Liaoning, Gansu and Fujian provinces. Furthermore, as can be observed from the picture, Cd pollution in agricultural soils was more serious in the south than in north, especially pollution regions in the southwest and central-south were more extensive. Extremely contaminated for Pb ($I_{geo} > 5$) were mainly situated in the southwest China as well as the junction of Guangxi, Guangdong and Hunan three provinces. This may be caused by the fact that these provinces contained abundant with mineral resources, such as Hunan and Guangxi province are known as the homes of non-ferrous metals, Ganluo Pb-Zn mine in Sichuan province is the largest Pb-Zn reservoir in Western China, Hezhang in Guizhou province is a zinc smelting county with a history of over 100 years, Huize in Yunnan province is a typical representative of large Pb-Zn mine. Similarly, clear spatial distribution of Zn was apparent, with soils Zn I_{geo} values in south China nearly higher than other areas. However, the I_{geo} values of spatial distribution illustrated that the highest

Table 4. Class distribution of geo-accumulation index of HMs in agricultural soils near mining-impacted communities.

Class	Cd	Cr	Hg	As	Pb	Cu	Zn	Ni
0	4.49%	59.70%	11.63%	42.86%	24.72%	30.59%	25.29%	55.00%
1	12.36%	25.37%	11.63%	20.63%	16.85%	31.76%	21.84%	37.50%
2	22.47%	7.46%	23.26%	12.70%	19.10%	24.71%	19.54%	5.00%
3	10.11%	5.97%	23.26%	12.70%	19.10%	24.71%	19.54%	0.00%
4	11.24%	0.00%	9.30%	9.52%	13.48%	4.71%	17.24%	0.00%
5	15.73%	0.00%	0.00%	0.00%	4.49%	0.00%	4.60%	0.00%
6	23.60%	1.49%	20.93%	1.59%	5.62%	2.35%	0.00%	2.52%

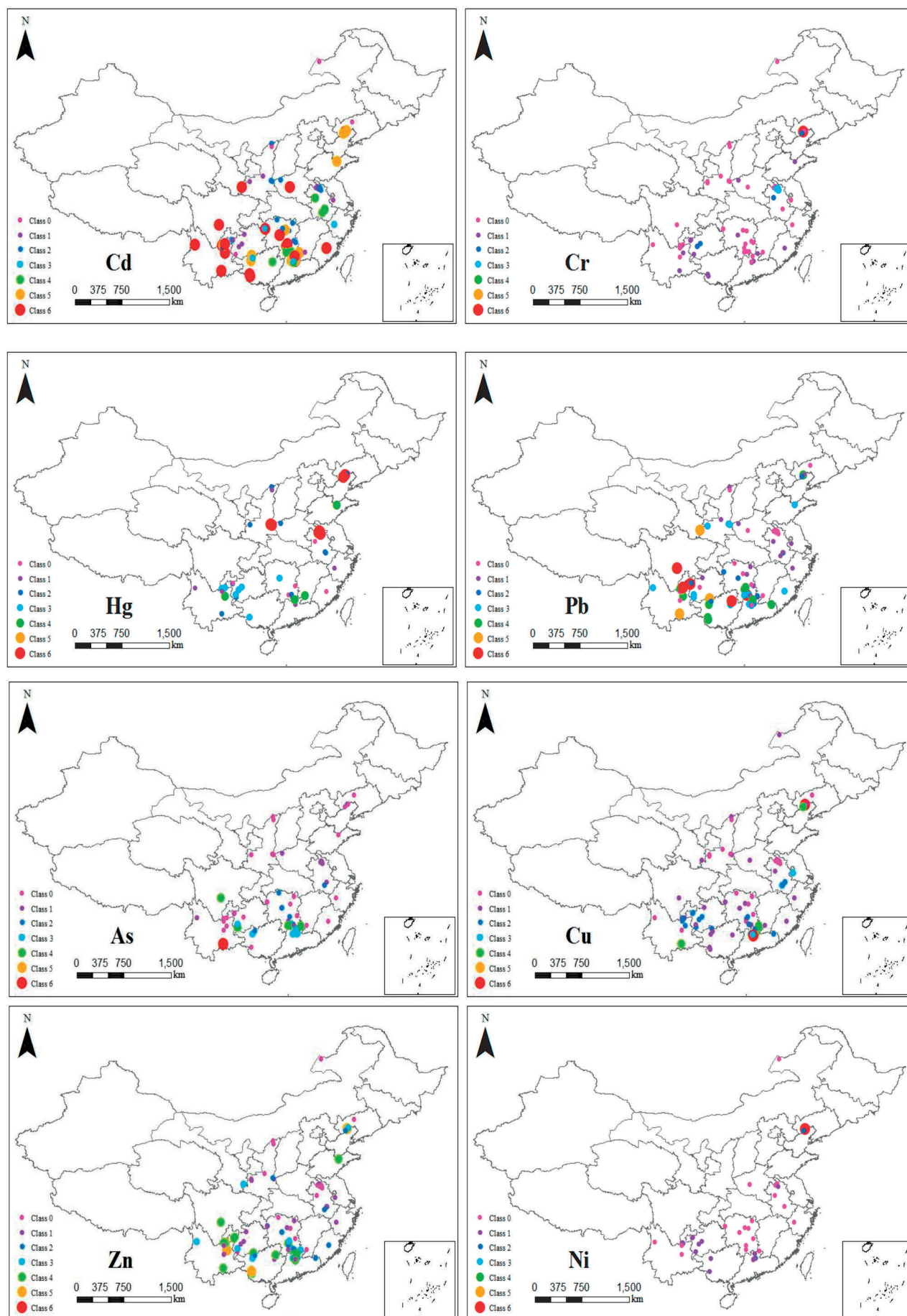


Fig. 2. Spatial distribution of HMs in agriculture soils near examined mining-impacted areas based on contamination classes.

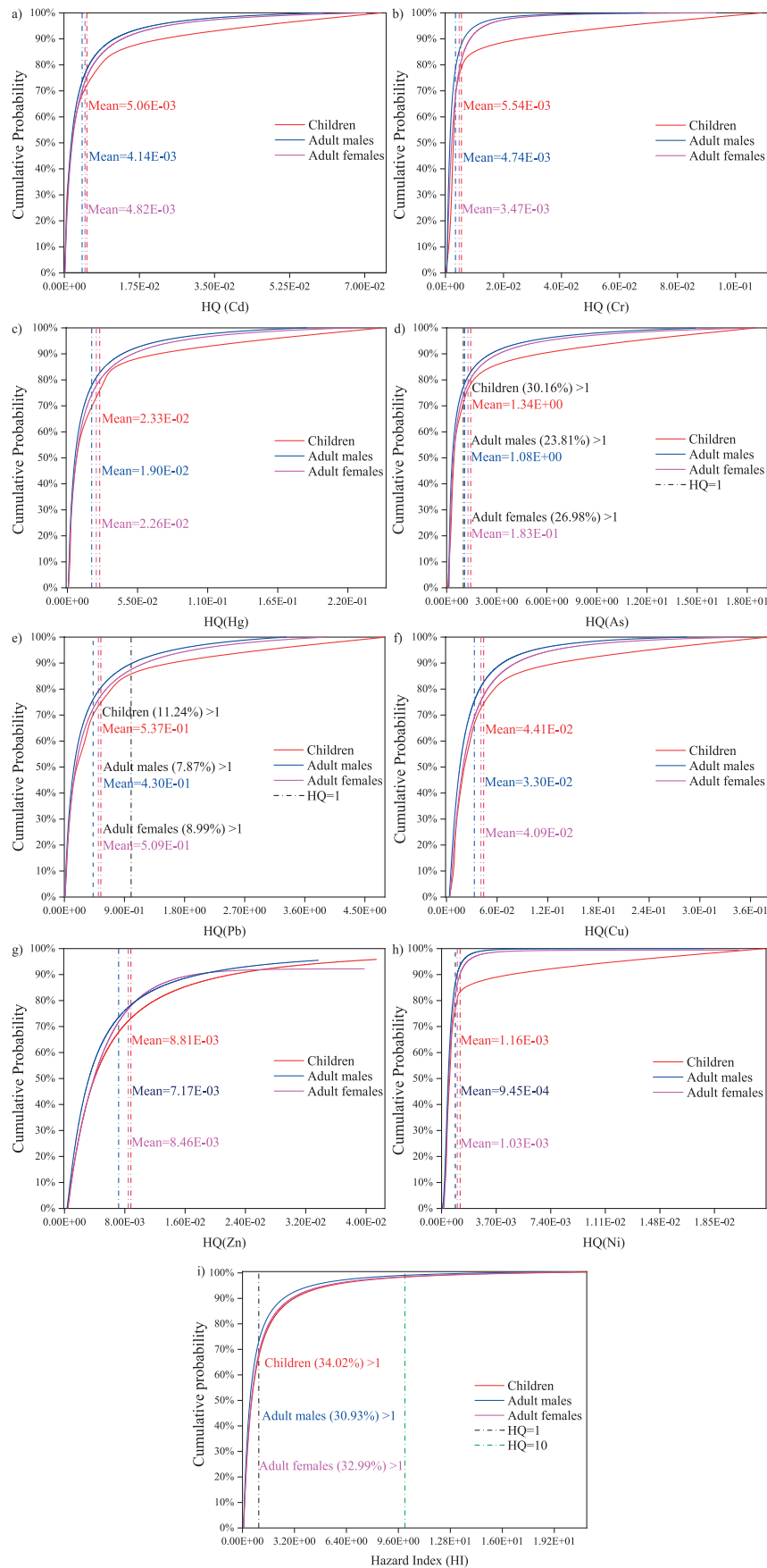


Fig. 3. Cumulative probability of hazard quotient (HQ) posed by a) Cd, b) Cr, c) Hg, d) As, e) Pb, f) Cu, g) Zn, h) Ni and cumulative probability of i) hazard index (HI) for the different population groups. (The red, blue and magenta solid vertical lines presented the mean values for children, adult males and females, respectively. The black and green dashed vertical lines presented the guideline values that of 1 and 10, respectively).

I_{geo} values of Cr, Ni and Cu were located in Huludao city in Liaoning province, the finding was relevant to the intensive metal-smelting industry [29]. In addition, another hotspot for Cu was observed in Dabaoshan mine in Guangzhou province, which is the largest multi-metal mine in southern China. For As, the highest I_{geo} value was located in Gejiu in Yunnan province due to the Sn-mineral exploitation and smelting activities. Because few sampling sites for Hg in examined mining-impacted communities, the higher I_{geo} values were only detected in Liaoning Huludao molybdenum mine, Shaanxi Xiaoqinling gold mine and Anhui Suzhou coal mining area. Based on the above analysis, higher I_{geo} values of the selected HMs except for Hg were mainly distributed in southern China, such as the Yunnan-Guizhou plateau, the karst regions in southern China and the southern part regions of the Yangtze River.

Non-Carcinogenic Risk Assessment

Non-carcinogenic health risk results of soil ingestion exposure to the selected HMs in agriculture soils around examined mining-impacted communities are presented in Fig. 3. It appeared from Fig. 3(a-g) that the mean HQ values of the investigated HMs for the three population groups all lower than 1, except for As. There is a clear trend of the mean HQ values decreased in the following order: As>Pb>Cu>Hg>Zn>Cr>Cu>Ni for the three population groups. Among the investigated HMs, human exposure to As and Pb were significantly higher than the other HMs due to their low RfD values (RfD is the reference oral dose available for HMs). In the meantime, from the data in Fig. 3(d-e), it is apparent that 30.16%, 26.98% and 23.81% of HQ outputs of As for children, adults females and males, respectively, exceeded 1, 11.24%, 8.99% and 7.87% of HQ outputs for children, adults females and males respectively exceeded 1 for Pb, suggesting that As and Pb contamination in part of Chinese agriculture soils around mining-impacted communities might pose a risk

for human health. Sensitivity analyses were adopted by applying the HQ values of As (the worst-case scenario) (Fig. 4), it is evident that the average body weight (BW) and ingestion rate (IngR) for children had relatively higher contribution than those for adults, with BW contributed the most. The result was in the lines of previous studies that children were likely to have an exceptionally sensitive of exposure to environmental contaminants per unit body weight due to unique physiological characteristics and behavioral [30, 31], such as hand-finger sucking, which is often considered to be the critical exposure routes of soils HMs might trigger adverse effects on health for children [32, 33]. Relying on the above analyses, it can be served as an evidence to explain the major cause why HQ outputs for children were far higher than those for adults.

Based on the cumulative probabilities of hazard index (HI) of the exposed three populations groups, Fig. 3i) shown that the percentage of the examined mining-impacted areas for adult males whose HI values was higher than the guideline value of 1, between 0 and 1, and between 1 and 10 were about 30.93%, 69.07% and 29.90%, respectively. For adult females, approximately 32.99%, 67.01%, 30.93% were the percentage of the examined mining-impacted areas whose HI values were higher than 1, between 0 and 1, and between 1 and 10. However, HI value for above 1 accounted for 34.02% for children, 65.98% laid between 0 and 1, and 31.96% laid between 1 and 10. On the basis of the above analyses, it can be concluded that part of agricultural soils near the mining-impacted areas posed potentially non-carcinogenic risk to the public. In comparison to the other two groups, children were more susceptible to the investigated HMs in soils than adults. Similar findings were consistent with previous studies by Man et al. [34] and Chen et al. [35]. Thus, the potential health risk to young children who living in the vicinity of mining-impacted areas resulted from exposure to soils via ingestion cannot be neglected.

Limitations and Uncertainties

Firstly, the distribution of the examined mining-impacted communities in this study cannot fully represent the overall pollution situation of HMs in agriculture soils around the mining-impacted communities across China due to the lack of relevant data in other provinces. Besides, owing to most previous studies tend to conduct in more polluted sites, which may influence the pollution levels and spatial distribution of the selected heavy metals. Secondly, some discrepancies in both digestion methods and analytical techniques from the eligible literature may impact the consistency of HMs pollution levels and the assessment results. However, these discrepancies are not be big enough to influence the general research results because the experimental methods are in accordance with the corresponding specifications and national standards and are all widely accepted by the scientific community.

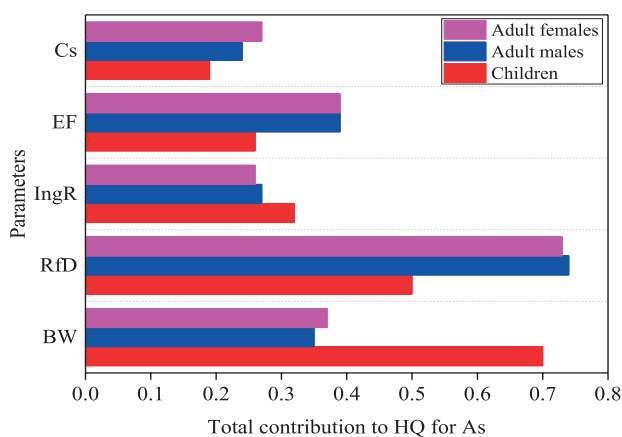


Fig. 4. Sensitivity ratio of average body weight (BW), reference oral dose (RfD), ingestion rate (IngR), exposure frequency (EF) and concentrations (C) for hazard quotient (HQ).

Moreover, the choice of exposure parameters control the reliability of the assessment results. In order to more accurately and realistically estimate the health risks for the different population groups, some existing parameters (e.g. BW, IngR, EF and ED) in present study were derived from relevant regulations and literature conducted within China, while the toxicity parameters mainly cited foreign sources. Ever so, we cannot fully eliminate the uncertainty of the assessment results caused by exposure parameters. Lastly, agricultural soils properties (e.g. pH, Eh, CEC, and SOM) affect HMs absorption and concentrations in soils. It should be noted that the above chemical properties of soils cannot all obtained in the selected literature, thus, we have not been discussed in this study because of the limited data sources.

Conclusions

This study gives a description of the overall contamination of HMs in agriculture soils around the mining-impacted communities across China and their ecological and health risk assessment. Based on the results of this study, it is apparent that the selected HMs pollution levels varied among the examined sites. In particular, agriculture soils are seriously polluted by Cd Pb and Hg. Additionally, the spatial distribution characteristics suggested that HMs pollution except for Hg were more serious in southern China than northern regions. Moreover, agriculture soils contamination by HMs emitted from mining activities continues to pose health risk to children and adults, especially to As and Pb should be paying more attention and regulation owing to their high non-carcinogenic health risk.

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Conflict of Interest

The authors declare no conflict of interest.

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