

Article

Study on the Accumulation of Heavy Metals in Different Soil-Crop Systems and Ecological Risk Assessment: A Case Study of Jiao River Basin

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Abstract: The purpose of this study is to evaluate the bio-accumulation of different soil-crop systems (SCSs) for heavy metals (HMs) and the geo-accumulation of different agricultural growing regions. The ecological risk (ER) assessment was conducted to understand the impact of intensive agricultural production on the environment. To achieve this aim, four typical crops, wheat, corn, potatoes, and leeks grown in the Jiao River Basin (JRB), were selected as the research objects. The concentrations of eight HMs, including copper (Cu), lead (Pb), zinc (Zn), nickel (Ni), chromium (Cr), cadmium (Cd), arsenic (As), and mercury (Hg) in crop tissue and soil were detected. The statistical analysis, including the geo-accumulation index (I_{geo}), geostatistical analysis, correlation and cluster analysis were then used to evaluate soil contamination and determine the source types of HMs. The results show that the average concentrations of eight HMs in the soil follow the order: Zn > Cr > Ni > Pb > Cu > As > Cd > Hg and the calculated concentration coefficients (K) vary from 0.41–1.12, indicating relative scarcity in sources of HMs. All the I_{geo} values of HMs are less than 0 except the I_{geo} of Cr within potato-farmland is from 0 to 1, illustrating that the soil in JRB is uncontaminated. The correlation and cluster analysis reveal that Cu, Zn, and Cd have a strong relationship with each other and the relationship between Pb, Ni, and Cr is general. The content of eight HMs in different crops varies greatly and most of them are within the scope of National Food Safety Standards—Limit of Pollutants in food of China. The bioconcentration factors (BCF) indicate that wheat, corn, potato, and leek have strong bio-accumulation ability of Cu, Zn, and Cd. The ecological risk factor (Er) shows that JRB is in low risk of Cu, Pb, Zn, Ni, Cr, and As; however, the risk of Cr and Hg are mostly low, characterized by partially dotted moderate risk. The risk index (RI) is mainly moderate with partially low risk distributed in planar and high risk distributed in point.

Keywords: bio-accumulation; geo-accumulation index; ecological risk assessment; heavy metals; soil-crop systems; Jiao River Basin



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1. Introduction

Soil is the ultimate part of the ecological system, and it plays an essential role in the survival and development of human beings, especially for food supply [1,2]. However, with the rapid development of industry, the expansion of cities, and the continuous expansion of intensive agriculture, the high-quality land resources are gradually shrinking [3,4]. The accumulation of HMs in soil (geo-accumulation) often leads to the accumulation of HMs in plants (bio-accumulation) [5]. The geo-accumulation of HMs occurs through a variety of processes, posing a potential threat to soil biota and leading to many disorders within plants [6,7]. These soil HMs can enter other ecosystems by means of runoff, resulting in disadvantages for the environment and human health [8]. Soil HM pollution has attracted worldwide attention, particularly in agricultural production systems [9–11]. It was found

that the geo-accumulation of HMs is primarily affected by pollution sources and geological background, followed by land use [12–17]. For example, Li et al., through the study of the selected purple soil profile in clastic rock area, found that Pb, Zn, and Cd, were accumulated on the surface, and that the migration of HMs was affected by the combined factors such as foreign HMs, weathering degree, leaching and sedimentation, clay mineral adsorption, and atmospheric dust fall [15]. Li et al. found that the content of As and Hg in metamorphic sandstone are higher than those in slate, while the content of Mn, Pb, Zn, and Ni in slate are higher than those in metamorphic sandstone [13]. Xiao et al. pointed out that due to the different functions of land, the heavy metal pollution in soil varies greatly. Hence, it requires necessary research on the geo-accumulation patterns of HMs [16].

Previous studies have shown that when HMs enter the human body through SCSs, threats to human health are posed [18–20]. Cu and Zn are essential nutrients for crops [21]. However, the consumption of foods contaminated with other HMs (Pb, Ni, Cr, Cd, As, Hg) has been found to generate damage, for example, Cr, As, and Hg may cause cancer and genetic mutations [22–25]; Cd may damage due to a blood–brain barrier [26]; Pb may influence endocrine signaling and enzyme activity [27]; and Ni has been linked to allergic diseases, kidney, lung, and nasal cancer [28]. The bio-accumulation of HMs is widely found in various SCSs. Meanwhile, the bio-accumulation law varies in different SCSs and was influenced by complexity factors [29–31]. For instance, the pH and calcium content of soil are the main factors affecting the bio-accumulation of Cd [32]. Polyethylene facilitates the bio-accumulation of Cu and Pb, whereas they reduce the absorption of As and Cd [33]. In general, root vegetables accumulate more Cd than other types of vegetables [34]. Therefore, it is necessary to keep exploring the HMs' transformation rules for different SCSs with the same geological background and land type. Understanding the content of HMs in soil and their geo-accumulation patterns is the basis for ER assessment of the environment [35,36]. Meanwhile, it can provide inspiration for the removal of HMs in soil [37–40]. Furthermore, the exploration of the bio-accumulation ability of HMs in plants is beneficial for ensuring food safety and providing guidance for agricultural development.

Wheat is the basic nutrition for most of the human population and contributes 20% of the daily energy needed [41]. Corn is one of the most important crops and a major food source (>30% calories) for about 4.5 billion people spread over 94 developing countries [42]. Potatoes are grown for food and are the world's fourth most significant food crop, after rice, maize, and wheat [43]. Leeks belong to the crops with a long tradition of cultivation in China and Europe which provide pentanol, methyl furan, flavonoids, polysaccharides, glucosinolates, or organosulfur compounds for human beings [44–46]. The JRB is a typical agricultural growing region, in which wheat, corn, potatoes, and leeks are widely planted [47]. Due to the impact of long-term agricultural activities, some HMs continue to geo-accumulate, which may have adverse effects on the agricultural production and ecological environment [48]. Chemical fertilizers, pesticides, and wastewater irrigations constitute significant contributors to HM sources [35,49]. Most of the previous studies on JRB were focused on the characteristics of geochemical element assemblages in topsoil [47,50,51] and the relationship between geological environment and groundwater fluoride [52,53]. There are few studies on the geo-accumulation laws and bio-accumulation laws of HMs within different SCSs, resulting in a lack of understanding of the ER for the entire region. Therefore, in this paper, four crops (wheat, corn, potatoes, and leeks) and their root soil were selected to test and analyze the content of Cu, Pb, Zn, Ni, Cr, Cd, As, and Hg, aiming to study the accumulation laws of HMs for the JRB and evaluate the ER within the same watershed.

2. Materials and Methods

2.1. Research Background

The Jiao River, one of the three major water systems in Gaomi City, is known as the “the mother river” for Gaomi people. It runs from north to south, passing through Baicheng Town, Chaoyang Street, and Xiazhuang Town; and it is the main source of

irrigation for agriculture in the JRB [54]. The study area is located in the east of Shandong Peninsula, ranging from $36^{\circ}10' N$ to $36^{\circ}32' N$ and $119^{\circ}41' E$ to $120^{\circ}1' E$ (Figure 1a,b). It is a traditional agricultural planting area of the JRB, characterized by a monsoon warm-temperate continental semi-humid climate, in which summer is humid and rainy, while winter is dry and less snowy. The terrain of the study area is high in the south and low in the north (Figure 1c). There were four primary soil types in the study area, namely brown soil, cinnamon soil, moisture soil, and sandy turmeric soil, which are all suitable for farming (Figure 1d). According to the current situation of agricultural development in Gaomi City, potato cultivation is mainly concentrated in Baicheng Town, leek cultivation is mainly concentrated in Xiazhuang Town, and corn and wheat are rotated throughout the entire JRB [55,56] (Figure 1c). The local government has always advocated the concept of ecological low-carbon green and environmentally friendly development in agricultural. Therefore, “Jiaohe Potato” was recognized as a “pollution-free agricultural product of Shandong Province” by the Shandong Provincial Department of Agriculture and Rural Affairs in 2005 and was evaluated as a “national geographical indication protection product” by the General Administration of Quality Supervision, Inspection, and Quarantine of the People’s Republic of China in 2010 [55]. Moreover, “Gaomi Xiazhuang Dajingou Leek” was approved as a pollution-free agricultural product by the Agricultural Product Quality and Safety Center of the Ministry of Agriculture of China in 2011 [56].

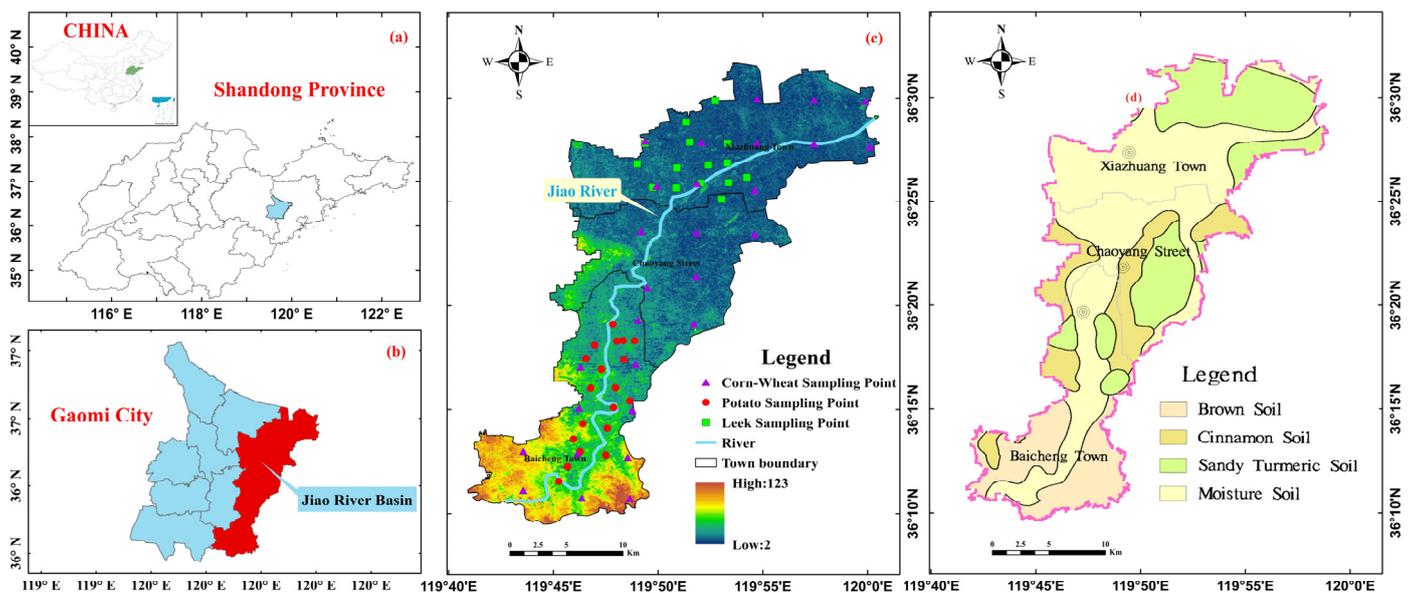


Figure 1. Map of JRB: (a) location of Shandong Province; (b) location of JRB; (c) location of sampling points; (d) soil type map.

2.2. Samples and Methods

Four types of crops, wheat, corn, potatoes, and leeks, which have wide planting areas in the JRB were selected during the peak harvest period for investigation, sampling, and analysis. During sampling, the edible part of each crop was picked and the supporting root soil was collected. An area of 0.2 hectares was set up as a collection unit and the chessboard or diagonal method was used to collect 20 sub samples within each unit, then equal parts were taken to form a mixed sample to make the single mixed wheat and corn sample weight 1kg (dry weight), respectively, and the single mixed potato and leek sample weight 2 kg (fresh weight), respectively. A total of 90 crop samples and 62 supporting root soil samples were collected, including 28 corn samples and 15 leek samples collected during September 2018, and 28 wheat samples and 19 potato samples collected during June 2019. Due to the rotation planting of corn and wheat in the JRB, corn and wheat were collected

at the same location (Figure 1c). The sampling depth of root soil was 0–20 cm, and two or three sub samples were equally combined to form one sample with 2 kg.

After the samples were collected, wheat and corn samples were dried, threshed and then their fruits were rinsed with distilled water once or twice. Finally, the fruits of wheat and corn were dried at room temperature and 0.5 kg were selected and sent to the laboratory. Leek and potato samples were washed at fresh to remove soil adhesion and pollution caused by fertilization or pesticide spraying. The samples were dried gently with clean gauze and 1 kg were taken in a polyethylene plastic bag immediately, the bag was tightly tied, and then were sent to the laboratory. Soil samples were placed in a clean and tidy indoor ventilated area for drying. Then, they were crushed with a wooden rod; small plant roots were removed with electrostatic adsorption method. Finally, 0.5 kg root soil samples were screened through a 2mm nylon sieve and then sent to the laboratory.

All sample testing was completed in the Experimental Testing Center of the Shandong Provincial NO.4 Institute of Geological and Mineral Survey. The quality monitoring of analysis and testing was carried out in accordance with the Technical Requirements for Analysis of Eco geochemical Evaluation Samples (Trial) (DD2005-03). All analysis and testing results meet the requirements, and the data quality is reliable. The testing methods and detection limits for Cu, Pb, Zn, Ni, Cr, Cd, As, and Hg indicators were shown in Table 1.

Table 1. Instrumental methods and detection limits for samples.

Indicators	Testing Methods	Detection Limits
Cu	X Ray Fluorescence (XRF)	1 mg/kg
Pb	X Ray Fluorescence (XRF)	2 mg/kg
Zn	X Ray Fluorescence (XRF)	4 mg/kg
Ni	X Ray Fluorescence (XRF)	2 mg/kg
Cr	X Ray Fluorescence (XRF)	5 mg/kg
Cd	Inductively coupled plasma-Mass Spectrometry (ICP-MS)	0.02 mg/kg
As	Atomic Fluorescence Spectroscopy (AFS)	0.5 mg/kg
Hg	Vapor Generation Cold Atomic Fluorescence Spectroscopy (AFS)	0.5 mg/kg

2.3. Quality Assurance and Quality Control

To ensure the accuracy of the experimental analysis results, the Chinese standard material (GBW) was added in each batch of experimental samples as the reference for quality control. The logarithmic difference between the measured values of GBW and the standard values was calculated to control batch deviations in daily analysis. The measurement results showed that both the accuracy and precision qualification rate requirement for each element analysis method were 98%, which complies with the “Specification of Land Quality Geochemical Assessment” (DZ/T 0295-2016) [57].

2.4. Graphics and Data Processing

Excel2019 and IBM SPSS Statistics26 were selected for data statistics, correlation, and cluster analysis. Origin2021, ArcGis10.2, MapGis67, and CorelDraw2021 were used to process graphics.

The geo-accumulation index (I_{geo}) was originally defined by Müller for assessing HM accumulation in sediments [58] and a total of seven classes were classified (Table 2). The improved I_{geo} index was expressed as follows [59,60]:

$$I_{geo} = \log_2 \left(\frac{C_i}{1.5B_i} \right) \quad (1)$$

where C_i is the concentration of HM-i in the soil samples, mg/kg, and B_i is the geochemical background concentration of HM-i, mg/kg. The constant factor 1.5 is the background

matrix correction factor which was designed to analyze natural fluctuations in the content of a given substance in the environment and to detect very small anthropogenic influence.

Table 2. Grading standard of BCF and I_{geo} of HMs.

BCF (%)	Bioaccumulation Ability of Crop	I_{geo}	Contamination Degree of Soil
$0 < BCF \leq 1.5$	Low	$I_{geo} \leq 0$	Uncontaminated
		$0 < I_{geo} \leq 1$	Uncontaminated to moderately
$1.5 < BCF \leq 4.5$	Medium	$1 < I_{geo} \leq 2$	Moderately
		$2 < I_{geo} \leq 3$	Moderately to heavily
		$3 < I_{geo} \leq 4$	Heavily
$BCF \geq 1.5$	High	$4 < I_{geo} \leq 5$	Heavily to extremely
		$5 < I_{geo} \leq 6$	Extremely

The BCF was defined as the bio-accumulation ability of HMs from soil to crop and it was classified into three levels as shown in Table 2 [61]. It was calculated as below [6,62]:

$$BCF(\%) = \frac{C_{crop}^i}{C_{soil}^i} \times 100\% \tag{2}$$

where C_{soil}^i is the concentration of HM-i in soil, mg/kg, and C_{crop}^i is the concentration of HM-i in crop, mg/kg.

The formula for calculating potential E_r^i was introduced by Hakanson [63,64] and was calculated according to the following equation:

$$E_r^i = T_r^i \frac{C_{metal}^i}{C_{background}^i} \tag{3}$$

$$RI = \sum_{i=1}^8 E_r^i \tag{4}$$

where E_r^i is the single risk index for HM-i, T_r^i is the toxicity coefficient of HM-i, C_{metal}^i and $C_{background}^i$ are measured and the background values (BGV) for HM-i in environment, respectively. RI is the sum of E_r^i for eight HMs. The T_r^i of eight HMs according to Xu et al. [65] and the BGV of eight HMs according to Gao et al. [51] and Zhang et al. [3] were shown in Table 3. E_r and RI was classified into five classes according to Li et al. [66] and Junusbekov et al. [64], as shown in Table 4.

Table 3. The toxicity coefficient and background values of HMs.

HMs	T_r^i	BGV of JRB	BGV of Shandong mg/kg	BGV of China
Cu	5	17.7	21.2	22.6
Pb	5	23.3	22.9	23.6
Zn	1	45.3	58.5	63.3
Ni	5	21.9	26.9	27.1
Cr	2	59.6	65.3	62
Cd	30	0.1	0.11	0.13
As	10	8.75	7.8	8.6
Hg	40	0.027	0.032	0.03

Table 4. Grading standard of potential ER of HMs.

Grade	E_r^i	RI	Risk Status
I	$E_r^i < 40$	$RI < 55$	Low
II	$40 \leq E_r^i < 80$	$55 \leq RI < 110$	Moderate
III	$80 \leq E_r^i < 160$	$110 \leq RI < 220$	High
IV	$160 \leq E_r^i < 320$	$220 \leq RI < 440$	Very high
VI	$E_r^i \geq 320$	$RI \geq 440$	Extremely

3. Results and Discussion

3.1. Heavy Metal Concentrations in the JRB Soil

3.1.1. Statistical Analysis

The geochemical data of HMs in JRB soil are shown in Table 5. The concentration coefficient (K) is an important parameter that reflects the material source basis of HMs in the study area [67]. The concentration coefficient (K1, K2, K3) of most HMs in JBR was less than 1, except that of the K1, K2 and K3 of Pb, As, and Cd which were close to 1, respectively, and the K1 of As was more than 1. This indicates that compared to Weifang City, Shandong Province and China, the HMs present a low BGV distribution in the JRB with a relatively scarce degree of HMs source. The average concentrations of HMs in the soil followed the order: Zn > Cr > Ni > Pb > Cu > As > Cd > Hg, which is similar to the situation of other traditional agricultural growing regions of China and the BGV of China [3,68,69], illustrating that large-scale agriculture has subtle disturbance on the distribution of HMs under the condition of no external pollution. As shown in Table 5, the ratio of each heavy metal in the sample exceeding the BGV of JRB follows the order: Ni(48.4%) > Cu(40.3%) = Hg(40.3%) > Pb(37.1%) > As(32.3%) > Zn(30.6%) = Cd(30.6%) > Cr(4.8%). All the HMs have an average concentration within the optimal range except that the average concentration of Ni is higher than the BGV of JRB, indicating that among the eight HMs involved in this paper, only Ni shows geo-accumulation. It is noted that Cr may not start to geo-accumulate because its excessive rate is extremely low (Table 5). The coefficient of variance (CV), defined as the ratio of standard deviation to the average value, is a normalized measure of the dispersion of a statistical population and can reflect the uniformity of element distribution [70]. The CV values for Cu, Pb, Zn, Cr, Cd, and As were all less than 0.3, indicating a relatively consistent distribution. In contrast, the CV values for Ni, Cr, Hg were all greater than 0.3, revealing a heterogeneous distribution which may be influenced by human activities [68]. The maximum allowable limits of HMs in soils have been established by the World Health Organization (WHO) [71,72] and the Ministry of Ecology and Environment of the People's Republic of China (MEPC) [73], as shown in Table 5. According to Table 5, it was found that except for the maximum values of Ni which are slightly higher than the guideline of WHO, the maximum values of the other seven HMs in JRB were all within the allowable range, suggesting that the soil within the JRB is at a low risk of heavy metal pollution.

Table 5. Geochemical data and maximum allowable limits of HMs in soils from WHO and MEPC.

HMs	Range (mg/kg)	AVG ± SD	CV	REB	K1	K2	K3	Maximum Allowable Limits	
								WHO (mg/kg)	MEPC (mg/kg)
Cu	9.7–29.53	17.04 ± 4.94	0.29	0.40	0.83	0.78	0.78	100	100
Pb	14.9–50.2	22.73 ± 4.6	0.20	0.37	1.02	0.99	0.90	100	120
Zn	28.1–99.97	43.41 ± 11.21	0.26	0.31	0.77	0.72	0.61	300	250
Ni	10.2–57.80	28.17 ± 14.86	0.53	0.48	0.81	0.81	0.81	50	100
Cr	9.7–65.02	34.86 ± 14.67	0.42	0.05	0.91	0.96	0.98	100	200
Cd	0.04–0.16	0.09 ± 0.023	0.26	0.31	0.91	0.77	1.03	3	0.3
As	4.23–13.61	7.97 ± 2.11	0.26	0.32	1.12	1.02	0.78	20	30
Hg	0.009–0.054	0.025 ± 9.28	0.40	0.40	0.83	0.89	0.41	50	2.4

AVG. average; SD. Standard deviation; CV. Coefficient of variation; REB. Ratio exceeding-background value of JRB; K. concentration coefficient, K1 = JRB/BGV of Weifang, K2 = JRB/ BGV of Weifang; K3 = JRB/ BGV of Weifang.

3.1.2. The Geo-Accumulation Index of HMs

The calculated average values of I_{geo} according to Formula (1) are all nearly less than 0 (Figure 2), indicating that the soil in JRB was uncontaminated. The I_{geo} of Ni in the area where potatoes were grown ranges from 0 to 1, demonstrating that potato-growing region was uncontaminated to moderately contaminated with Ni. It was also noted that the I_{geo}

of Cr in the soil of the area where potatoes were grown is the lowest, illustrating that cultivation of potatoes lead no geo-accumulation of Cr to the soil.

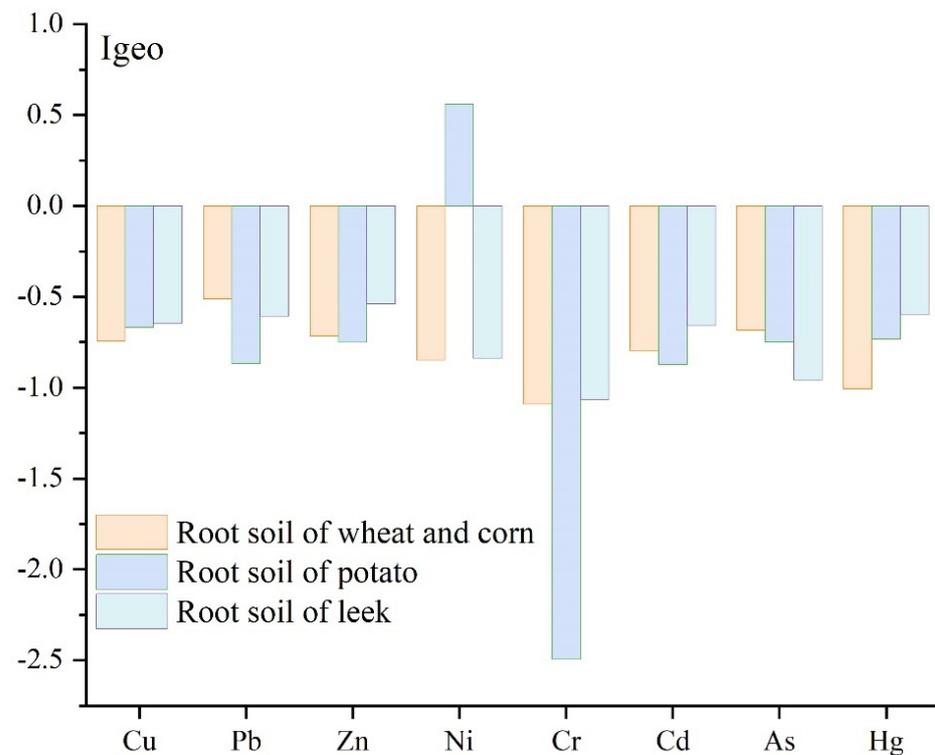


Figure 2. Geo-accumulation index of 8 HMs in JRB soil.

3.1.3. Geostatistical Analysis

Based on the geochemical data of HMs, the inverse distance weighting method was applied to obtain the distribution patterns of HMs in JRB (Figure 3). As shown in Figure 3, the heterogeneous distribution of HMs in the JRB is consistent with the analysis results of CV values mentioned above (Table 5), which is possibly due to differences in local characteristic planting industries (Figure 1c) and soil types (Figure 1d). Cr and Pb show an overall distribution trend of low in Baicheng town with brown soil and high in Xiazhuang town with sandy turmeric soil (Figure 3a,b), hinting that Cr and Pb may be homologous and the planting of leeks may cause their geo-accumulation. The distribution of Cu, Zn, and Cd are relatively close, displaying a trend of low in the middle (with cinnamon soil and moisture soil) and high on both sides (Figure 3d–f), which can be explained by Cu, Zn, and Cd having the same source; furthermore, compared to traditional agricultural production, vegetable cultivation has more significant impact on their geo-accumulation. Ni is mainly concentrated in Baicheng Town (Figure 3g), suggesting that potato cultivation may cause Ni geo-accumulation. The distribution of As is relatively uniform, dominated by low values (Figure 3c), which may be due to the lack of sources. Hg is relatively non-homogeneously distributed throughout the region with high values (Figure 3h), indicating that different agricultural activities have different impacts on Hg aggregation.

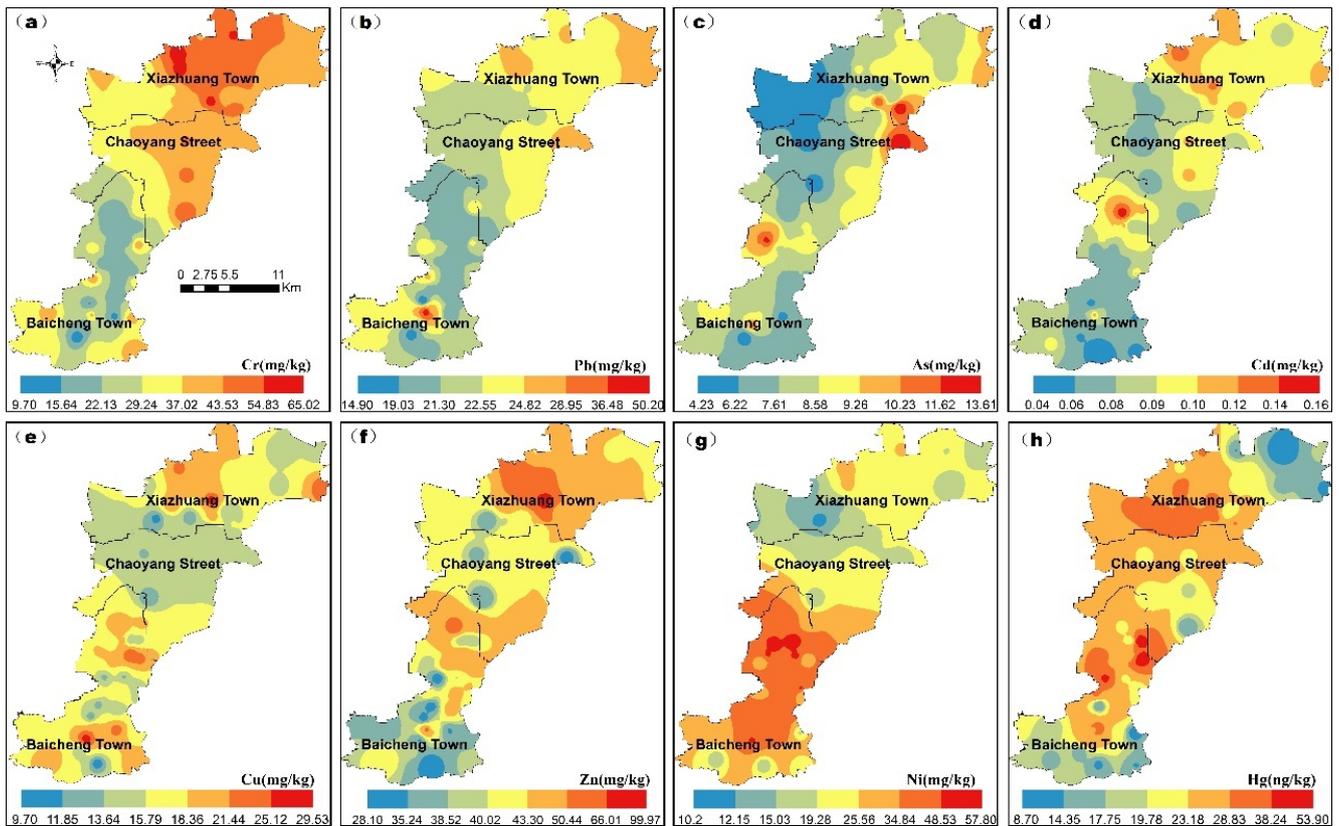


Figure 3. The distribution pattern maps of HMs in JRB soil: (a) Cr; (b) Pb; (c) As; (d) Cd; (e) Cu; (f) Zn; (g) Ni; (h) Hg.

3.1.4. Correlation and cluster analysis

We used Pearson correlation analysis to perform bivariate correlation analysis on data, and the results are shown in Table 6. The correlation between each variable can reflect their source information. Cu, Zn, and Cd express a strong relationship (Cu-Zn, $r = 0.642$; Zn-Cd, $r = 0.655$), indicating a common source of Cu, Zn, and Cd; whereas Pb, Cr, and Ni show a general correlation (Pb-Cr, $r = 0.565$, Cr-Ni, $r = -0.625$), suggesting a common source of Pb, Cr, and Ni. Arsenic and Hg have weak or no correlation with other HMs, revealing that the source of As and Hg may be different. In general, results of the correlation and cluster analysis are consistent with the conclusion of geostatistical analysis (Figure 3).

Table 6. Pearson Correlation matrix of HMs in soils.

	Cu	Pb	Zn	Ni	Cr	Cd	As	Hg
Cu	1	--	--	--	--	--	--	--
Pb	0.183	1	--	--	--	--	--	--
Zn	0.642 **	0.258 *	1	--	--	--	--	--
Ni	0.211	-0.344 **	0.040	1	--	--	--	--
Cr	0.166	0.565 **	0.384 **	-0.625 **	1	--	--	--
Cd	0.437 **	0.397 **	0.655 **	0.055	0.355 **	1	--	--
As	0.109	0.275 *	-0.016	0.222	0.206	0.222	1	--
Hg	0.144	-0.003	0.105	0.091	-0.063	0.089	0.255 *	1

** . Correlation is significant at the 0.01 level (2-tailed). * . Correlation is significant at the 0.05 level (2-tailed).

Applying system clustering analysis to HMs in the JRB, as shown in Figure 4, the HMs can be divided into two clusters at a rescaled distance of 25. Hg, As, and Ni were attached in one group, whereas Zn, Cd, Cu, Pb and Cr were cited in the other group (Figure 4). When the rescaled distance is 12.5 (shown in a red line in Figure 4), the HMs can

be further divided into five sub-clusters: Zn, Cd, and Cu are members of cluster 1; Pb and Cr are members of cluster 2; and As, Hg, and Ni belong to cluster 3, 4 and 5, respectively, suggesting the evident different anthropogenic sources obtained from the above mentioned statistical and spatial analyses (Table 5, Figure 3). Also, the data presented by hierarchical cluster analysis are consistent with the Pearson correlation (Table 6).

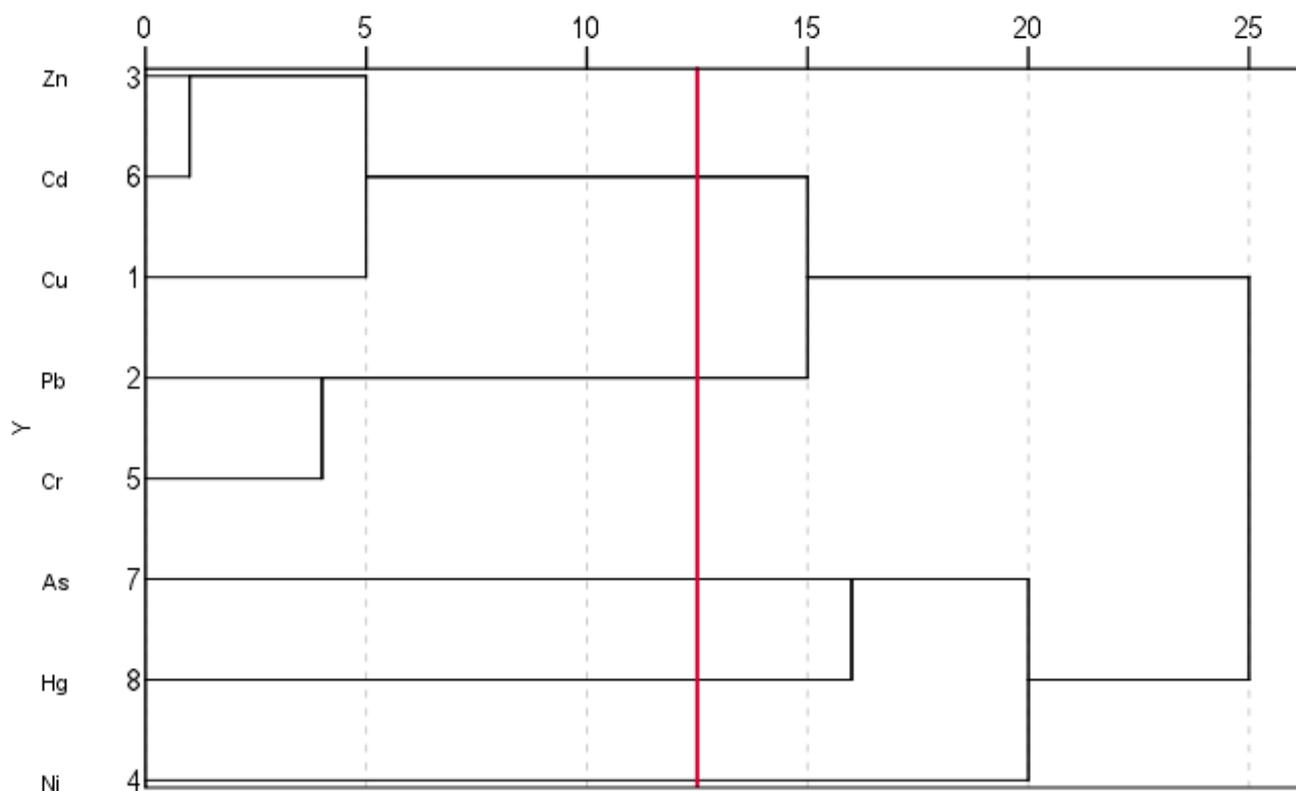


Figure 4. Cluster analysis dendrogram of eight HMs in JRB soil.

3.2. The Bio-Accumulation of HMs in Different Soil-Crop Systems

The content of HMs in different crops is given in Table 7. According to Table 7, the content of HMs varies greatly in different crops. Among all the samples tested in JRB, only two corn samples and two leek samples exceed the limit of Cr and Pb, respectively, based on National Food Safety Standards-Limit of Pollutants in Food of China (GB2762-2022) [74]. The detection values of HMs in other samples were all in accordance with food safety standards provided by GB2762-2022. The average content of each heavy metal was far less than the limit values. In conclusion, the agricultural products in JRB are all safe and the risk of heavy metal pollution is low. Hg was only detected in wheat with a detection rate of 60.7%, which may be ascribed to the low content of Hg in the soil of JRB, suggesting that wheat is more sensitive to Hg than corn [75]. The average content order of HMs in wheat is as follows: Zn(21.443) > Cu(4.711) > Ni(2.297) > Cr(0.442) > Pb(0.078) > As(0.024) > Cd(0.017) > Hg(0.006); which is different from typical sewage irrigation areas of Longkou City, Shandong Province (Zn(30.945) > Cu(4.622) > Ni(1.463) > Pb(1.081) > Cr(0.782) > As(0.311) > Cd(0.051) > Hg(0.01)) [68], indicating that the heavy metal content of crops in polluted areas is generally higher than that in traditional agricultural areas. The average content order of HMs in corn is as follows: Zn(18.457) > Cu(1.21) > Ni(0.402) > Pb(0.084) > Cr(0.079) > As(0.021) > Cd(0.011), which is similar to Zn(24.73) > Cu(2.86) > Ni(0.61) > Cr(0.56) > Pb(0.0664) > As(0.0167) > Cd(0.0026) from a study on HM content in corns near coal mines with high soil HM content [76], suggesting that corn is not sensitive to changes in HM content in the soil. The average content order of HMs in potatoes is as follows: Zn(1.883) > Cu(0.581) > Ni(0.28) > Cd(0.017) > Cr(0.058) > As(0.01) > Pb(0.04), which is

significantly different from potatoes irrigated with acid mine drainage, characterized as: Pb(20.65) > Ni(14.56) > Cu(9.00) > Cd(4.35) [71]. The average content order of HMs in leeks is as follows: Zn(2.92) > Cu(1.212) > Cr(0.696) > Ni(0.47) > Pb(0.096) > As(0.038) > Cd(0.015); which is different from the same study within Slovakia, (Zn(2.32) > Cu(0.62) > Pb(0.283) > Ni(0.24) > Cr(0.192) > Cd(0.102)) [77], showing that the content of HMs in leeks is different under different background values. From the above four sets of data, it can be seen that the content of Cu and Zn were higher than other HMs and the content of Cu and Zn in wheat and corn is about 10 times higher than those in potatoes and leeks. The content of Cd in the four crops is similar. Ni has the highest content in wheat and the lowest content in potatoes with a detection rate of 47.36%. Cr, Pb, and As content are the highest in leeks with a detection rate of 100% and lowest in potatoes with a detection rate of 63.16%, 5.26%, and 84.21%, respectively, indicating that a certain correlation may exist among these three HMs. Based on the entirety of Table 7, a pattern is found wherein the higher the detection rate is, the higher the HMs content is, suggesting that types of plant with high HMs contents and high detection rates are more likely to bio-accumulate HMs. In conclusion, the agricultural products in JRB are all safe and the risk of heavy metal pollution is low.

Table 7. Content of HMs in different crops.

Crop (NS)		Cu	Pb	Zn	Ni	Cr	Cd	As	Hg
Wheat (28)	max	9.06	0.17	29.28	6.32	0.92	0.099	0.14	0.009
	min	1.36	0.02	14.56	1.17	0.13	0.006	0.01	0.005
	mean	4.711	0.0778	21.443	2.296	0.442	0.017	0.0237	0.006
	DR	100%	100%	100%	100%	100%	100%	96.4%	60.7%
	GB2762-2022	--	0.2	--	--	1.0	0.1	0.5	0.02
	NES	0	0	0	0	0	0	0	0
Corn (28)	max	1.96	0.22	26.7	1	0.25	0.02	0.04	--
	min	1.18	0.09	20.4	0.45	0.07	0.01	0.01	--
	mean	1.21	0.084	18.457	0.402	0.079	0.011	0.021	--
	DR	100%	100%	100%	67.9%	82.1%	46.4%	35.7%	0%
	GB2762-2022	--	0.2	--	--	1.0	0.1	0.5	0.02
	NES	0	0	0	0	2	0	0	0
Potato (19)	max	0.75	0.04	2.41	0.36	0.08	0.029	0.01	--
	min	0.37	0.04	1.35	0.2	0.05	0.007	0.01	--
	mean	0.581	0.04	1.88	0.28	0.058	0.017	0.01	--
	DR	100%	5.26%	100%	47.36%	63.16%	89.47%	84.21%	0%
	GB2762-2022	--	0.2	--	--	0.5	0.1	0.5	0.01
	NES	0	0	0	0	0	0	0	0
Leek (15)	max	2.35	0.19	3.94	0.82	1.55	0.02	0.08	--
	min	0.29	0.04	1.41	0.22	0.25	0.01	0.02	--
	mean	1.212	0.096	2.92	0.47	0.696	0.015	0.038	--
	DR	93.3%	100%	100%	93.3%	100%	73.3%	100%	0%
	GB2762-2022	--	0.3	--	--	0.5	0.2	0.5	0.01
	NES	0	2	0	0	0	0	0	0

NS. number of samples; DR: detection rate; NES. Number of exceeding standards. --. not detected or not specified in GB2762-2022; unit: mg/kg.

The BCF of the eight HMs in different SCSs were calculated according to Formula (2) and the bio-accumulation ability was evaluated based on Table 2, these results are shown in Table 8. The CVs of the BCF of eight HMs in different crops are relatively large (Table 8), which may be ascribed to the small number of samples collected or the bio-accumulation of HMs in crops is easily affected by external factors. The bio-accumulation of Cu, Zn, and Cd in the four SCSs is high, while the accumulation of Pb and Cr is low. The BCF of Ni, Hg, and As vary greatly among different SCSs, reflecting significant bio-accumulation differences in different HMs within different SCSs (e.g., the BCF of Ni for wheat is high, for potato is low, and for corn and leek is medium). Most BCFs in Table 8 are in accordance

with the content in Table 7, showing a positive relationship between BCF values and the detection values and rates. However, the detection values and rates of some specified HMs (e.g., Cr and Pb in leeks) are relatively high with a low BCF, which is ascribed to the high background values as shown in Figure 2.

Table 8. The BCF of HMs in different SCSs.

Crop (NS)		Cu	Pb	Zn	Ni	Cr	Cd	As	Hg
Wheat (28)	max%	61.91	0.78	82.51	36.32	2.66	99.00	1.03	70.31
	min%	12.12	0.09	33.59	4.64	0.24	6.36	0	0.00
	mean%	30.59	0.32	52.26	13.22	1.07	19.85	0.26	13.09
	CV%	41.51	61.16	22.02	56.87	54.75	90.23	86.85	153.21
	BA	high	low	high	high	low	high	low	high
Corn (28)	max%	12.78	1.05	72.81	6.82	0.55	22.22	0.80	0
	min%	1.58	0.12	6.80	0	0	0	0	0
	mean%	7.77	0.35	45.08	1.74	0.15	5.41	0	0
	CV%	34.24	68.75	26.22	114.02	82.02	128.55	235.83	0
	BA	high	low	high	medium	low	high	low	low
Potato (19)	max%	5.54	0.27%	7.07	0.75	0.56	48.33	0.22	0
	min%	2.28	0	2.7	0	0	0	0	0
	mean%	3.55	0.01	4.64	0.28	0.23	17.20	0.11	0
	CV%	30.11	435.89	22.52	111.76	86.68	66.18	54.18	0
	BA	medium	low	high	low	low	high	low	low
Leek (15)	max%	15.91	0.89	10.86	5.56	4.25	33.33	33.33	0
	min%	1.93	0.19	3.14	0	0.40	0	0	0
	mean%	5.48	0.34	5.62	1.99	1.13	10.47	10.47	0
	CV%	77.84	53.26	39.50	74.54	89.49	88.79	88.79	0
	BA	high	low	high	medium	low	high	high	low

NS number of samples; BA: bio-accumulation ability.

Table 9 shows the correlation of BCF in different SCSs. Correlation analysis was conducted on 28 wheat samples. It was found that Cu and Pb ($r = 0.773$), as well as Hg and Cr ($r = 0.662$), are strongly correlated, whereas Ni and Cu ($r = 0.576$), Ni and Pb ($r = 0.548$), Ni and Cr ($r = 0.445$), and Cd and Zn ($r = 0.573$) are generally correlated. Correlation analysis of HMs in 28 samples of corn show that Zn and Cu ($r = 0.591$), Cr and Pb ($r = 0.474$), and Hg and Cd ($r = 0.556$) are generally correlated. The correlation between HMs in 19 potato samples is strong for As and Ni ($r = -0.672$), Cr and Ni ($r = -0.826$), and Cd and Zn ($r = 0.691$), respectively, and generally correlated for As and Cr ($r = 0.549$). Based on correlation analysis, HMs in 15 leek samples can be divided into two categories: Ni, Cr, and Pb, which are strongly correlated with each other; and Cu, Zn, Cd, and As which are generally correlated with each other. It can be inferred that HMs exhibit different synergistic effects during the bio-accumulation process from root soil to crops.

3.3. ER Assessment

The E_r and RI for different HMs were calculated based on Equations (3) and (4), respectively. The spatial distribution of the E_r and RI are presented in Figure 5. Combining Figure 5 with Table 4, it was found that ERs of Cu, Pb, Zn, Ni, Cr, and As are low ($E_r < 40$) (Figure 5a–f). A large portion of Hg and Cd in JRB were in a low ER, with moderate ER distributed sporadically in a dotted pattern (Figure 5g,h). The RI was obtained by summing the E_r values of eight HMs (Figure 5i) varying from 50 to 140. The RI for JRB is mainly moderate ($55 \leq RI < 110$), with both low ($RI < 55$) and high ($110 \leq RI < 220$) ER areas distributed in a dotted pattern. This illustrates that although the E_r value of individual HM is low, the RI value may be high.

Table 9. Pearson Correlation matrix of HMs in different crops.

Correlation in wheat	Cu	Pb	Zn	Ni	Cr	Cd	As	Hg
Cu	1	--	--	--	--	--	--	--
Pb	0.773 **	1	--	--	--	--	--	--
Zn	0.267	-0.072	1	--	--	--	--	--
Ni	0.576 **	0.548 **	0.325	1	--	--	--	--
Cr	-0.011	-0.052	0.235	0.445 *	1	--	--	--
Cd	-0.109	-0.2	0.573 **	-0.002	0.258	1	--	--
As	-0.145	-0.143	-0.271	-0.239	-0.223	-0.184	1	--
Hg	-0.105	-0.026	-0.063	0.299	0.662 *	0.037	0.251	1
Correlation in corn	Cu	Pb	Zn	Ni	Cr	Cd	As	Hg
Cu	1	--	--	--	--	--	--	--
Pb	0.118	1	--	--	--	--	--	--
Zn	0.591 **	0.062	1	--	--	--	--	--
Ni	-0.082	0.195	-0.32	1	--	--	--	--
Cr	-0.087	0.474 *	-0.314	0.228	1	--	--	--
Cd	0.183	0.321	0.181	0.121	0.019	1	--	--
As	0.014	0.354	-0.061	0.135	0.314	0.242	1	--
Hg	-0.164	0.396	-0.208	0.345	0.389	0.556	0.09	1
Correlation in potato	Cu	Pb	Zn	Ni	Cr	Cd	As	Hg
Cu	1	--	--	--	--	--	--	--
Pb	a	a	--	--	--	--	--	--
Zn	0.275	a	1	--	--	--	--	--
Ni	-0.377	a	0.39	1	--	--	--	--
Cr	-0.053	a	0.162	-0.826	1	--	--	--
Cd	-0.055	a	0.691 **	0.579	0.192	1	--	--
As	-0.072	a	0.023	-0.672	0.549	0.045	1	--
Hg	a	a	a	a	a	a	a	a
Correlation in leek	Cu	Pb	Zn	Ni	Cr	Cd	As	Hg
Cu	1	--	--	--	--	--	--	--
Pb	0.558 *	1	--	--	--	-1	--	--
Zn	0.488	0.353	1	--	--	--	--	--
Ni	0.6 *	0.761 **	0.525	1	--	--	--	--
Cr	0.629 *	0.903 **	0.439	0.93 **	1	--	--	--
Cd	0.543	-0.01	0.613 *	0.373	0.141	1	--	--
As	0.543	-0.01	0.613 *	0.373	0.141	1 **	1	--
Hg	a	a	a	a	a	a	a	a

** . Correlation is significant at the 0.01 level (2-tailed). * . Correlation is significant at the 0.05 level (2-tailed). a. Not detected, unable to calculate.

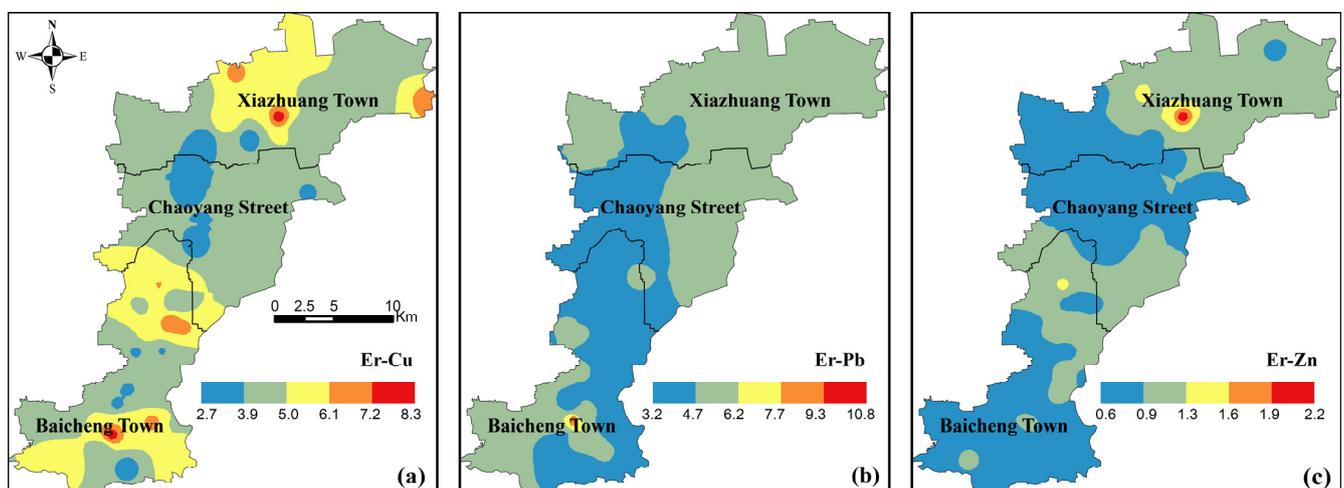


Figure 5. Cont.

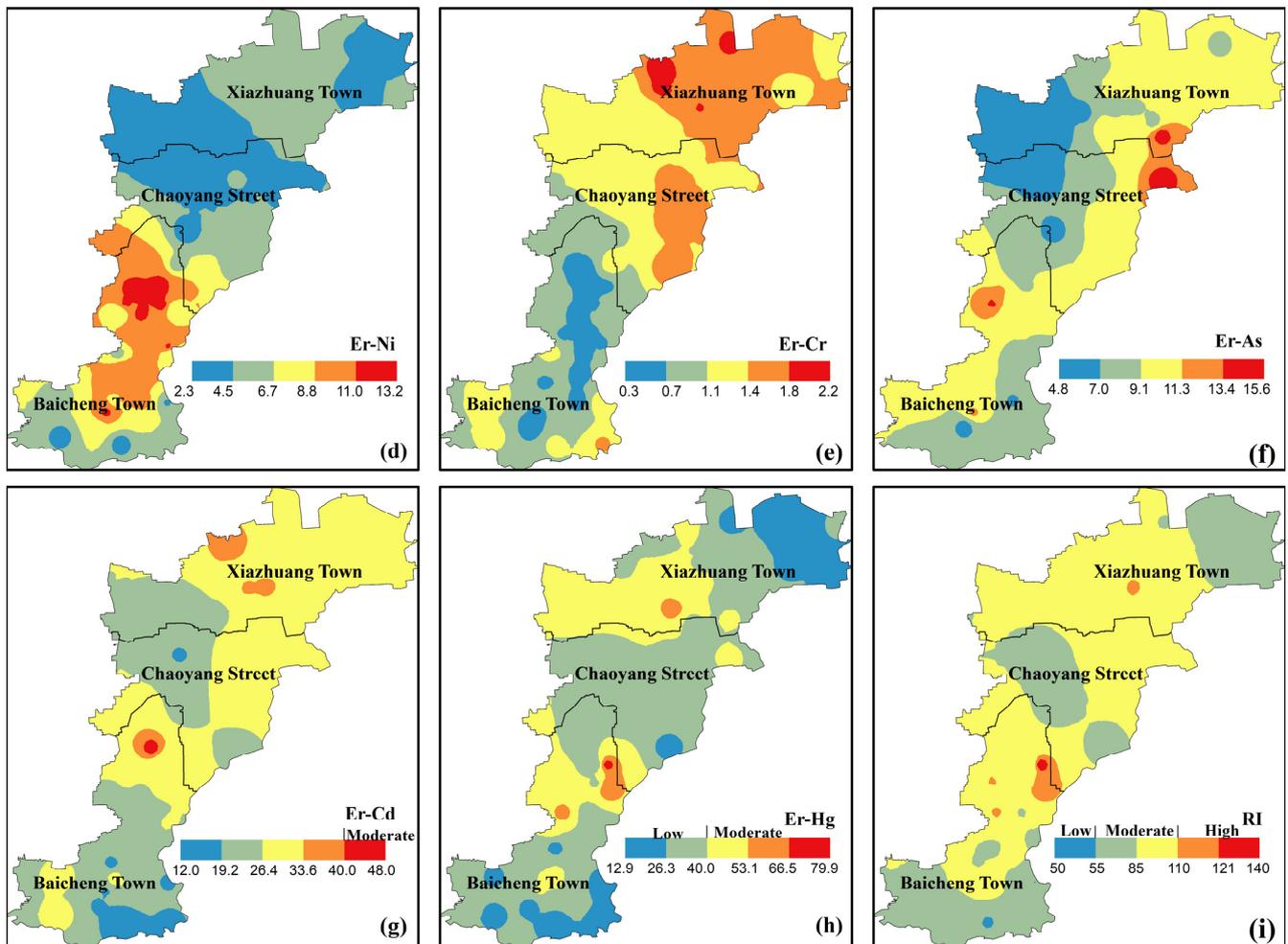


Figure 5. Spatial distribution of Er(a–h) and RI (i).

4. Conclusions

Based on our study, certain conclusions were drawn as follows:

1. Soils in JRB are short of HM sources. The average concentrations of HMs in the soil followed the order: $Zn > Cr > Ni > Pb > Cu > As > Cd > Hg$, similar to that of the other traditional agricultural growing regions in China and BGV, indicating that the disturbance of large-scale agricultural production on soil HMs is acceptable without external pollution sources. Both the CV and the distribution patterns reveal that distributions of Cu, Pb, Zn, Cd, and As are relatively homogeneous, whereas distributions of Ni, Cr, and Hg are heterogeneous, which may be explained by the cultivation of different crops with inconsistent migration and transformation abilities for HMs.
2. The maximum values of Zn, Cr, Pb, Cu, As, Cd, and Hg in JRB are all below WHO guidelines and national standards of China, and their I_{geo} values are less than 0. Whilst the maximum value of Ni exceeds the WHO guideline and the national standards of China, and it has an I_{geo} value greater than 0. Data analysis and accumulation analysis jointly show that there is only Ni accumulation concentrated in the potato growing region in JRB, which may be explained by the bio-accumulation ability of Ni within potatoes, which is lower than those of corn, wheat, and leek.
3. Results of geostatistical analysis, correlation, and cluster analysis are similar. Cu, Zn, and Cd are strongly correlated; Pb and Cr are generally correlated; and Hg, As, and Ni are poorly correlated with each other, suggesting that Cu, Zn, and Cd are homologous; Pb and Cr are homologous; whereas Hg, As, and Ni are from different sources.

4. There were very few crop samples taken in JRB exceeding the National Food Safety Standards-Limit of Pollutants in Food of China, reflecting the good safety of agricultural products and a low risk of heavy metal pollution in JRB. The rankings of the average content of HMs in wheat, corn, potatoes, and leeks is different, in addition to different HMs which were characterized by different BCFs within different SCSs. Cu and Zn are the dominate elements in the four crops, with an order of magnitude greater than other elements; the content of Cd in four crops is approximately equal to each other. Moreover, the BCFs of Cu, Zn, and Cd for the four crops are high, whereas the BCFs of Pb and Cr of the four crops are low. A pattern is found that the higher the detection rate is, the higher the content is, suggesting a high bio-accumulation of HMs for plants. The correlation of BCF exhibits a different mechanism for HMs during the bio-accumulation process from root soil to crops.
5. ER assessment results in JRB show that the ER of Cu, Pb, Zn, Ni, Cr, and As are low. Hg and Cd are mainly in the low risk category, whereas a fraction in moderate risk were distributed in pairs of spots. RI reveals a medium risk with a few low-risk and high-risk areas distributed in a point like manner.

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Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

soil-crop systems (SCSs); heavy metals (HMs); ecological risk (ER); Jiao River Basin (JRB); copper (Cu); lead (Pb); zinc (Zn); nickel (Ni); chromium (Cr); cadmium (Cd); arsenic (As); mercury (Hg); geo-accumulation index (I_{geo}); concentration coefficients (K); bioconcentration factors (BCF); ecological risk factor (Er); risk index (RI).

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