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Contamination characteristics, source analysis and health risk assessment of heavy metals in the soil in Shi River Basin in China based on high density sampling

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ABSTRACT

To explore the contamination of heavy metals in the Shi River Basin soil in China, a high density sampling of surface soil was conducted. In this study, an absolute principal component scores multiple linear regression model (APCS-MLR) was used to identify the sources of heavy metals in the soil and quantify their amounts. The methods to assess the heavy metals included a fuzzy synthetic evaluation, index and health risk assessment. The results show that heavy metals are relatively rich southwest of the study area. Their levels may be affected by natural sources, such as parent materials. The pollution caused by human factors cannot be ignored, and it is primarily influenced by traffic emissions and processing sources, which contribute 62.6%, followed by agricultural sources, such as pesticides and fertilizers, that contribute 21.1%. The risk assessment indicated that the study area was slightly to moderately polluted. All heavy metals pose higher carcinogenic and other health risks to children than adults, and ingestion is the main way that these pollutants enter the body. The carcinogenic risk of children owing to Cr from natural sources merits further study, while the carcinogenic risk to adults and the non-carcinogenic risk to both adults and children are at acceptable levels. Transportation and industrial processing sources are the main cause of the non-carcinogenic risk. The results could provide reference for reducing heavy metal pollution in the soil.

1. Introduction

Soil is an irreplaceable resource for human survival and development because it is the basis of all agricultural activities (Cao et al., 2010). With the rapid development of urbanization, industry and agriculture, the problem of heavy metal pollution in the soil has become increasingly prominent (Sarwar et al., 2017; Li et al., 2018a; Gabarrón et al., 2018; Liu et al., 2020). Pollution by heavy metals in the soil is closely related to crop quality and food safety. Heavy metals in the soil can be taken up by crops and then reach the human body through the food chain, directly threatening human health (Xu et al., 2015; Zhang et al., 2020). Therefore, it is critical to identify the sources of heavy metals in the soil and conduct comprehensive pollution and health risk assessments.

Many researchers have quantified the accumulation of heavy metals in the soil in typical regions around the world and found that the soils in most areas are contaminated by more than one type of heavy metal (Ciarkowska et al., 2019; Li et al., 2020a; Yang et al., 2020; Wu et al., 2021a). The distribution of heavy metals in the soil is primarily controlled by the sources of heavy metals. Therefore, it is necessary to identify these sources. Heavy metals accumulate in the soil through various sources, including industrial activities, coal burning, fertilization and irrigation, and the weathering of rocks (Luque-Espinar et al., 2018; Bi et al., 2020; Jiang et al., 2020). An absolute principal component scores multiple linear regression model (APCS-MLR) can quantify the contribution of pollution sources using multivariate statistical methods. This model was first applied to trace the source of heavy

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metals in atmospheric particulates. Since it does not require knowledge of the number of pollutants in advance, the conditions for realization are simple, and the results of source apportionment are objective and accurate. This model has also been widely used in field soils in recent years (Li et al., 2018b).

In China, human productive activities, such as excessive use of pesticides and fertilizers and unreasonable discharge of industrial waste water and garbage, lead to heavy metal pollution of the soil. Health risk and pollution assessments are of great importance, which are a prerequisite for remediation. Ingestion by mouth and hand, dermal contact and inhalation are generally considered to be the ways in which humans are exposed to heavy metals that migrate from the soil to human body. Based on these three pathways described above, the carcinogenic and non-carcinogenic risks of heavy metals to the human body can be evaluated (Cai et al., 2019; Jiang et al., 2020; Xu et al., 2021). Over the previous few decades, environmental scientists have developed many quantitative methods to assess heavy metal pollution. Classical evaluation indices include the single pollution index (P_i), geo-accumulation index (I_{geo}), ecological risks index of individual heavy metals (E_i) and comprehensive ecological risk index (RI) (Li et al., 2014; Hu et al., 2020a; Baruah et al., 2020; Gujre et al., 2021). Fuzzy synthetic evaluation based on entropy weight is a method combining qualitative and quantitative factors, which contains a lot of information. It is widely used in water, sediment and air quality evaluation, but seldom used in soil heavy metal pollution (Li et al., 2018c; Zhang et al., 2018; Yang et al., 2019; Wu et al., 2021b; Wang et al., 2021).

The Shi River Basin is a demonstration area for small watershed management in China. The ecological environment of barren hills has been improved by planting fruit trees. However, the problem of heavy metal pollution generated during the process of governance has not been addressed, and this may adversely affect the local environment and human health. Therefore, it is imperative to emphasize the importance of determining the status and sources of heavy metal pollution in the Shi River Basin. Most previous studies on the study area have focused on water and air pollution (Li et al., 2020a, 2020b; Wang et al., 2020a; Liu et al., 2021a, 2021b). Zhang et al. (2017) and Liu et al. (2016b) evaluated the environmental risk of heavy metals in the soil of Weifang city based on index methods. There are few studies on source apportionment and health risk assessment of heavy metals.

In general, results of soil pollution assessment and source apportionment become more accurate as the amount of soil quality data increases. Nevertheless, high-density sampling is difficult to conduct due to factors such as field conditions and testing costs. This study was conducted on the basis of high-density survey and soil sampling in the Shi River Basin. This study employs data from 667 soil samples and aims to (1) investigate the status of soil pollution by fuzzy synthetic evaluation and index methods; (2) identify the source and quantify its contribution to heavy metals in the soil using APCS-MLR; (3) conduct a health risk assessment to unravel the effects of pollution on human health. The results of this study can provide a theoretical and practical reference for the governance and prevention of pollution for the Shi River Basin. These findings can also be applied to the soil investigation and quality evaluation in other areas of the world that exhibit similar conditions.

2. Materials and methods

2.1. Description of the study area

The study area that is located in the central part of Shandong Province, China, is a demonstration area for small watershed management along the Shi River. It is located in the north temperate sub-humid continental monsoon climate zone, which is alternately affected by continental and maritime climates. The mean annual temperature is 12.7 ℃, and the mean annual precipitation is approximately 665 mm. The Shi River in the area flows into the Heihushan Reservoir from west to east (Fig. S1). The soil is primarily calcareous brown soil, which is very thick, moderately textured and good for cultivation. Since 1998, the study area has promoted the comprehensive management of small watersheds for soil and water conservation and planted fruits, such as hawthorn, persimmon, pear and apricot, to improve the ecological environment of barren mountains, as well as to revitalize the mountain economy. During the field work, it is found that processing and transportation are relatively developed in the study area, with dense distribution of factories for food, materials and accessories. Therefore, man-made pollutants, such as organic and inorganic pesticides and fertilizers and industrial wastes, that have emerged with the development of planting industry pose threats to the local environment.

2.2. Sampling and analytical methods

In October 2017, high density sampling of the surface soil was conducted in the Shi River Basin. A total of 667 sites that were laid out in grids were determined by GPS with a sampling density of 5.5 pieces/km^2. The sampling depth was 0–20 cm, and two to six subsamples were mixed in equal quantity to form one sample. The collected soil at each sample site was broken, and roots, straws, stones, insects and other debris were removed. After full mixing, 1–1.5 kg of soil was retained using the quartering method and placed in sample bags. The soil samples were naturally air-dried and passed through a 10-mesh nylon sieve.

To ensure strict quality monitoring and controlling, every 50 samples were inserted into four national first-level reference materials (GBW) for internal quality control and two external standard control samples to ensure external quality control, and their accuracy and precision were both shown to be 100%. Simultaneously, two replicate samples were established for every 50 samples to ensure that the results were repeatable. The pass rates of various analysis indicators were between 97.4% and 99.1% (> 90%), which complies with the quality inspection requirements of the "Specification of Land Quality Geochemical Assessment" (DZ/T 0295–2016). After sample analysis, the abnormal point repeatability test was carried out for the samples with extra high or extra low contents. Calculate the relative difference (RD) (RD is not more than 40% for qualified), test the unqualified samples at least 3 times, and report the two close data. The pass rate of all indicators in this abnormal point test is greater than 90%, which meets the standard requirements. All the samples were sent to the Experimental Testing Center of the Fourth Geological Brigade of Shandong Provincial Bureau of Geology and Mineral Resources for testing. Heavy metals were determined according to the Specification of Multi-Purpose Regional Geochemical Survey (1:250000) (DZ/T 0258–2014), Specification of Regional Ecogeochemistry Assessment (DZ/T 0289–2015), the Specification of Testing Quality Management for Geological Laboratories (DZ/T0130.4–2006), and Technical Requirements for Analysis of Ecological Geochemical Evaluation Samples (Trial). The levels of Cd were then determined using inductively coupled plasma mass spectrometry (ICP-MS). Cu, Pb, Zn, Ni and Cr were quantified using X-ray fluorescence spectrometry (XRF), while As and Hg were quantified using atomic fluorescence spectrometry (AFS).

2.3. Multivariate statistical analysis

In this study, a multivariate statistical analysis was conducted in SPSS 25.0 (IBM, Inc., Armonk, NY, USA). A Principal Component Analysis (PCA) is commonly used to reduce a multi-element heavy metal data set to three to four principal components. Potential sources of heavy metals are identified by exploring the source of each component. A PCA requires a strong correlation between the original variables, which is usually determined using the Kaiser-Meyer-Olkin (KMO) and Bartlett's tests. If the test coefficient of KMO is > 0.5, and the significance probability of Bartlett's Test is < 0.05, the data is applicable to a PCA (Zhao et al., 2019). R-type Hierarchical Clustering Analysis (HCA) is used to classify heavy metals, so that each category can represent certain characteristics, which can verify the results of PCA.

Table 1
Descriptive data of heavy metals in the Shi River Basin soil.

	Min	Max	Mean	SD	CV	Background value			K1	K2	K3
						Weifang	Shandong	China			
Cu	6.3	89.7	25.06	7.09	0.28	21.2	22.6	22.6	1.18	1.11	1.11
Pb	6.2	347.9	32.18	28.33	0.88	22.9	23.6	26	1.41	1.36	1.24
Zn	22.3	382	84.1	40.18	0.48	58.5	63.3	74.2	1.44	1.33	1.13
Ni	15.6	60.9	32.37	4.27	0.13	26.9	27.1	26.9	1.2	1.19	1.2
Cr	28.5	98.7	71.27	6.96	0.1	65.3	62	61	1.09	1.15	1.17
As	3.18	64.39	14.65	4.36	0.3	7.8	8.6	11.2	1.88	1.70	1.31
Cd	0.02	0.97	0.21	0.13	0.62	0.11	0.13	0.097	1.81	1.59	2.13
Hg	0.0098	1.33	0.05	0.08	1.57	0.032	0.03	0.065	1.63	1.74	0.8

The basic principle of APCS-MLR is to convert the principal component scores (PCS) obtained by PCA into absolute principal component scores (APCS) and then to construct a multiple linear regression model of the original concentration of heavy metals and APCS. A regression coefficient is used to calculate the contribution of pollution sources that correspond to each principal component. First, the heavy metal concentration data are standardized. GPCA/APCS can then be conducted as follows:

(1) An artificial sample with a concentration of 0 was introduced as follows:

$$Z_{i0} = \frac{0 - \overline{C_i}}{\sigma_i} = -\frac{\overline{C_i}}{\sigma_i} \tag{1}$$

(2) The APCS of each sample was obtained by subtracting the PCS of the 0 concentration sample that was obtained by PCA.
(3) APCS were taken as the independent variable and heavy metal contents as the dependent variable, and multiple linear regression was performed. The contribution of pollution sources is calculated as follows:

$$C_i = b_{i0} + \sum_{p=1}^{p} \left(b_{pi} \times APCS_p\right) \tag{2}$$

where b_{i0} is the artificial sample with a concentration of 0; $\overline{C_i}$ is the average concentration; σ_i is the standard deviation; b_{i0} is the constant term of multiple linear regression; b_{pi} is the regression coefficient; $APCS_p$ is $APCS$ of principal component p, and $b_{pi}\times APCS_p$ is the contribution of source p. The average of all samples $b_{pi}\times APCS_p$ is the average of source p (Cheng et al., 2020).

2.4. Fuzzy synthetic evaluation method

The concentrations of heavy metals were classified on five levels for fuzzy evaluation on the basis of the Environmental Quality Standard for Soil (GB15618–1995), the background values of soil and additional studies (Table S1) (Xie et al., 2005; Wang et al., 2010).

The calculation of heavy metal membership functions is shown below (Lourenço et al., 2010; Zhao et al., 2019):

If $j = 1$,

$$r_{ij}(C_i) = \begin{cases} 1 & C_i \le v_{ij} \\ \left(C_i - v_{ij} + 1\right) / \left(v_{ij} - v_{ij} + 1\right) & v_{ij} < C_i < v_{ij} + 1 \\ 0 & C_i \ge v_{ij} + 1 \end{cases} \tag{3}$$

If $j = 2,3$ and 4,

$$r_{ij}(C_i) = \begin{cases} 0 & C_i \le v_{ij} - 1 \\ \left(C_i - v_{ij} - 1\right) / \left(v_{ij} - v_{ij} - 1\right) & v_{ij} - 1 < C_i < v_{ij} \\ \left(C_i - v_{ij} + 1\right) / \left(v_{ij} - v_{ij} + 1\right) & v_{ij} < C_i < v_{ij} + 1 \end{cases} \tag{4}$$

If $j = 5$,

$$r_{ij}(C_i) = \begin{cases} 0 & C_i \le v_{ij} - 1 \\ \left(C_i - v_{ij} - 1\right) / \left(v_{ij} - v_{ij} - 1\right) & v_{ij} - 1 < C_i < v_{ij} \\ 1 & C_i \ge v_{ij} \end{cases} \tag{5}$$

Thus, the fuzzy matrix (R) can be obtained as follows:

$$R = \left(r_{ij}\right)_{n\times m} = \begin{pmatrix} r_{11} & r_{12} & \cdots & r_{1m} \\ r_{21} & r_{22} & \cdots & r_{2m} \\ \vdots & \vdots & \ddots & \vdots \\ r_{n1} & r_{n2} & \cdots & r_{nm} \end{pmatrix} \tag{6}$$

where C_i is the measured value; v_{ij} is the standard value; r_{ij} is the membership, and $r_{ij} \in [0,1]$. R is the fuzzy matrix.

It is highly significant to obtain the weight of each heavy metal. The difference in the characteristics of heavy metals has a greater impact on the weight setting (Liu et al., 2016a). Nevertheless, certain elements with low concentrations but high toxicity are often overlooked (Zhang et al., 2014; Yang et al., 2016). Entropy weights and toxicity coefficients were introduced to calculate weights to scale the combined effect of enrichment and toxicity of heavy metals in this study.

$$H_i = -1 \Big/ \ln p \sum_{l=1}^{p} \left(C_{il} \Big/ \sum_{l=1}^{p} C_{il}\right) \times \ln\left(C_{il} \Big/ \sum_{l=1}^{p} C_{il}\right) \tag{7}$$

$$W_i = \left(1 - H_i\right) \Big/ \left(n - \sum_{i=1}^{n} H_i\right) \tag{8}$$

$$W_i' = T_i W_i \Big/ \sum_{i=1}^{n} T_i W_i \tag{9}$$

where C_{il} is the normalization value; p is the number of monitoring sites (667); W_i is the weight utilizing the entropy method; W_i' is the modified weight considering toxicity, and T_i is the toxic coefficient (2 for Cu, 5 for Pb, Zn and Hg, 1 for Ni, 10 for Cr, 30 for As, and 40 for Cd).

Thus, the weight matrix (W) was obtained as follows:

$$W = \{W_1', W_2', \cdots W_n'\} \tag{10}$$

The fuzzy synthetic matrix (B) can be obtained by multiplying W by R as shown below:

$$B = W \times R = \{b_1, b_2, \cdots b_m\} \tag{11}$$

In general, the max (b_j) is taken as quality class of the soil (Liu et al., 2016a), which may cause some loss of information. Thus, the following method is used based on the weighted average principle (Qiu, 1989; Qiu et al., 2017):

$$A = \sum_{j=1}^{m} \left(b_j{\cdot}j\right) \Big/ \sum_{j=1}^{m} b_j \tag{12}$$

where A is the grade, which is usually a non-integer.

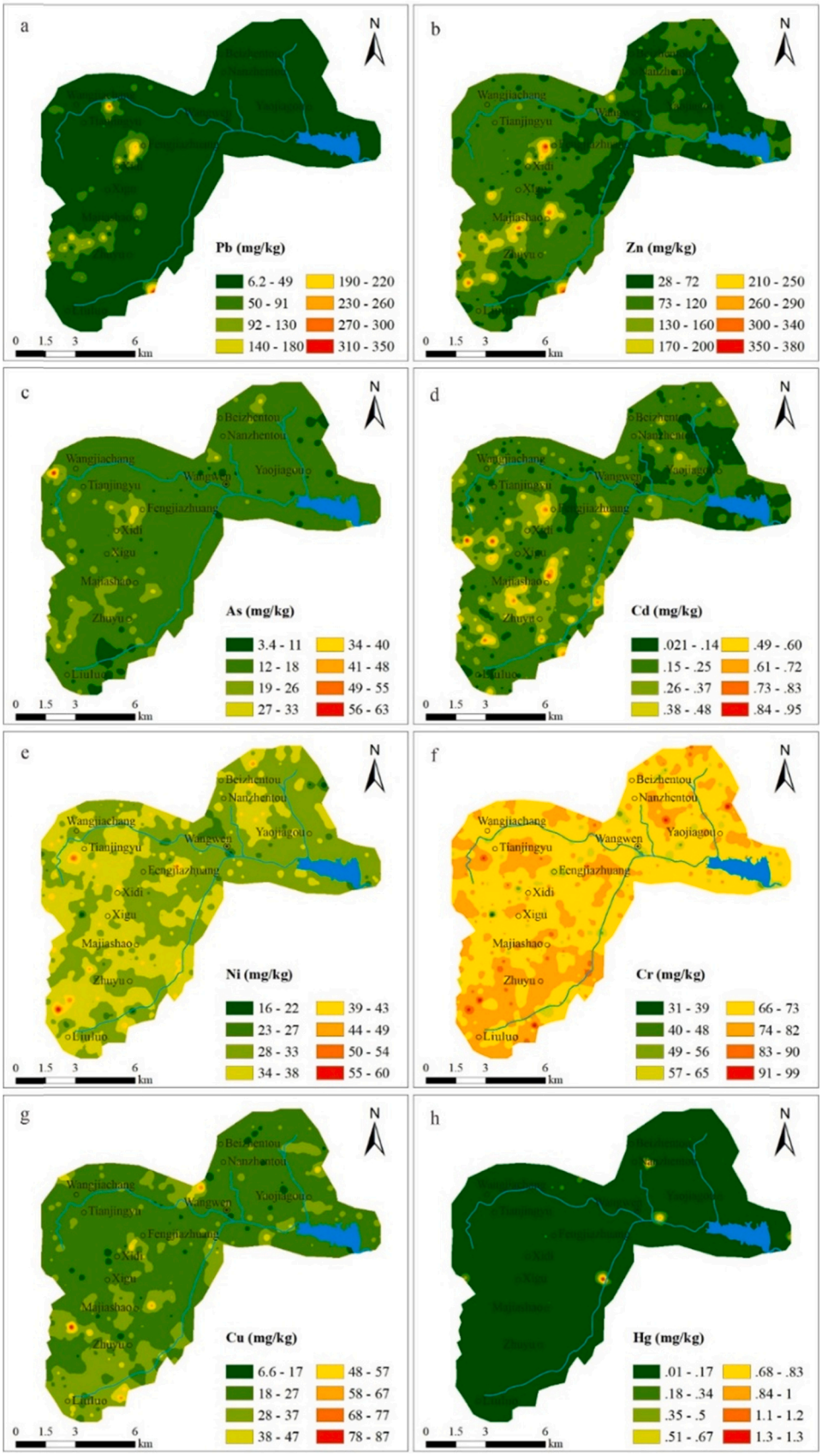

Fig. 1. Spatial distribution of heavy metals in surface soil in the Shi River Basin.

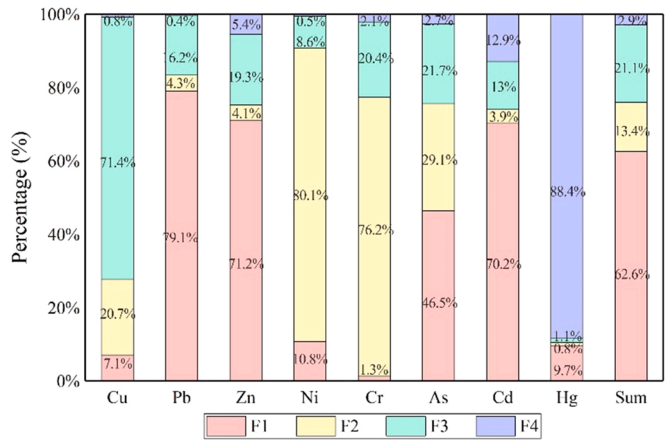

Fig. 2. Source contribution ratios of heavy metals in the Shi River Basin surface soil.

2.5. Pollution and ecological assessment index

The P_i (Tomlinson et al., 1980), I_{geo} (Müller, 1980), and E_i and RI (Hakanson, 1980) are commonly used to assess levels of pollution (Tian et al., 2017; Men et al., 2018; Wang et al., 2020b). The equations and degrees of these indices are shown in Table S2.

2.6. Human health risk assessment

The EPA has established a set of health risk assessment methods to quantify the non-carcinogenic and carcinogenic risks. Heavy metals of soil can harm human health through the three main manners of exposure that include ingestion (ing), dermal contact (der) and inhalation (inh). The average daily intake (ADI) (mg/kg-day) should be calculated as follows:

$$ADI_{ing} = \frac{Ci \times R_{ing} \times EF \times ED \times CF}{BW \times AT} \tag{13}$$

$$ADI_{der} = \frac{C_i \times SA \times AF \times ABS \times EF \times ED \times CF}{BW \times AT} \tag{14}$$

$$ADI_{inh} = \frac{C_i \times R_{inh} \times EF \times ED}{PEF \times BW \times AT} \tag{15}$$

The hazard index (HI) indicates the non-carcinogenic risk owing to multiple pathways. The cancer risk (CR) is the probability that a person develops cancer from exposure to carcinogens. The calculation equations of HI and CR are shown as follows:

$$HI = \sum HI_i = \sum \left(\frac{ADI_{ing}}{RfD_{ing}} + \frac{ADI_{der}}{RfD_{der}} + \frac{ADI_{inh}}{RfD_{inh}} \right)_i \tag{16}$$

$$CR = \sum CR_i = \sum \left(ADI_{ing} \times SF_{ing} + ADI_{der} \times SF_{der} + ADI_{inh} \times SF_{inh} \right)_i \tag{17}$$

where HI_i is the non-carcinogenic risk for the *ith* heavy metal owing to all pathways, and CR_i is the cancer risk for the *ith* heavy metal owing to all the pathways. The definitions and values of the intake exposure parameters are shown in Tables S3 and S4. HI values lower than 1 indicate no adverse health risk, whereas an $HI > 1$ represents the occurrence of non-carcinogenic effects. CR values between 1×10^{-6} and 1×10^{-4} signify an acceptable carcinogenic risk, while values $> 1 \times 10^{-4}$ represent a significant health hazard (US EPA, 2011).

3. Results

3.1. Concentration of heavy metals in the Shi River Basin

Table 1 shows the descriptive data of eight heavy metals in the Shi River Basin soil. The average concentrations of heavy metals followed the order: Zn (84.1 mg/kg) > Cr (71.27 mg/kg) > Ni (32.37 mg/kg) > Pb (32.18 mg/kg) > Cu (25.06 mg/kg) > As(14.65 mg/kg) > Cd (0.21 mg/kg) > Hg(0.052 mg/kg). The coefficient of variance (CV) is an important parameter that reflects the uniformity of element distribution. The CV values of Cu, Ni, Cr and As ranged from 0.1 to 0.3, thus, indicating a relatively uniform distribution. In contrast, the CV value of Zn, Pb and Cd ranged from 0.48 to 0.88, reflecting an uneven distribution that may be affected by human activities. The CV value of Hg was 1.57 and reflects an extremely uneven distribution that was substantially affected by human activities. The risk control standard for the soil contamination of agricultural land (GB15618–2018) indicated that the average concentrations of all metals in the surface soil of study area were not more than their corresponding critical values, while 1.2%, 2.85%, 3.45%, 0.15%, 13.94% and 0.45% of Cu, Pb, Zn, As, Cd and Hg samples exceeded their critical values, respectively (Fig. S2). Compared with the reference background values of Weifang City, Shandong Province, China, the contents of all heavy metals in the study area were relatively high at 1.09–2.13-fold of the background values, with the exception of Hg, which was slightly lower than the background value in China. These results suggest that it is critical to analyze and evaluate the characteristics of heavy metal pollution of the surface soil in the study area.

3.2. Geostatistical analysis

The inverse distance weight was used to interpolate the heavy metal concentration data to obtain the spatial distribution of heavy metals in the Shi River Basin soil (Fig. 1). The spatial distribution of Pb, Zn, As and Cd was highly similar, indicating that these heavy metals may originate from the same source. These heavy metals were primarily distributed in the western part of the study area, particularly in the Fengjiazhuang-Majiashao-Chuyu area, where the concentrations were the highest observed in this study (Fig. 1a–d). Ni and Cr demonstrated similar distributions, with few low values in the study area, and high values occurred in western-southwestern Chuyu-Liuluo and northern Tianjiyu-Fengjiazhuang-Nanzhentou (Fig. 1e and f). The region with the highest amount of Cu was southwest of the study area and appeared sporadically north of Wangfen and Majiashao-Chuyu-Liuluo (Fig. 1g). Hg was evenly distributed, with low values in the whole area and high values that were scattered in Nanzhentou, Wangfen and eastern Xigu in the middle of the study area (Fig. 1h).

3.3. Multivariate statistical analysis

Identifying and quantifying the pollution of heavy metals in the soil is critical. The results of the KMO test (0.662) and Bartlett's test ($p = 0.000$) revealed that the variables were highly correlated. Thus, they are suitable for PCA. The original data were standardized. The results of PCA show that four principal components were extracted, with a cumulative rate of contribution reaching 80.45%, which could more effectively explain most of the information of heavy metals in the study area soil. As shown in Table S5, the first principal component (PC1) indicated that Pb (0.866), Zn (0.889), As (0.657) and Cd (0.74) exhibited higher factor loadings. Ni (0.898) and Cr (0.839) exhibited higher factor loadings in PC2. Cu (0.91) exhibited higher factor loadings in PC3, while Hg (0.988) exhibited higher factor loadings in PC4.

Similar results were obtained using an R-type HCA. As shown in Fig. S3, the eight heavy metals can be divided into four clusters at a rescaled distance of 15. Pb, Zn, As and Cd are members of cluster 1; Ni and Cr are members of cluster 2; Hg is a member of cluster 3, and Cu is a member of cluster 4.

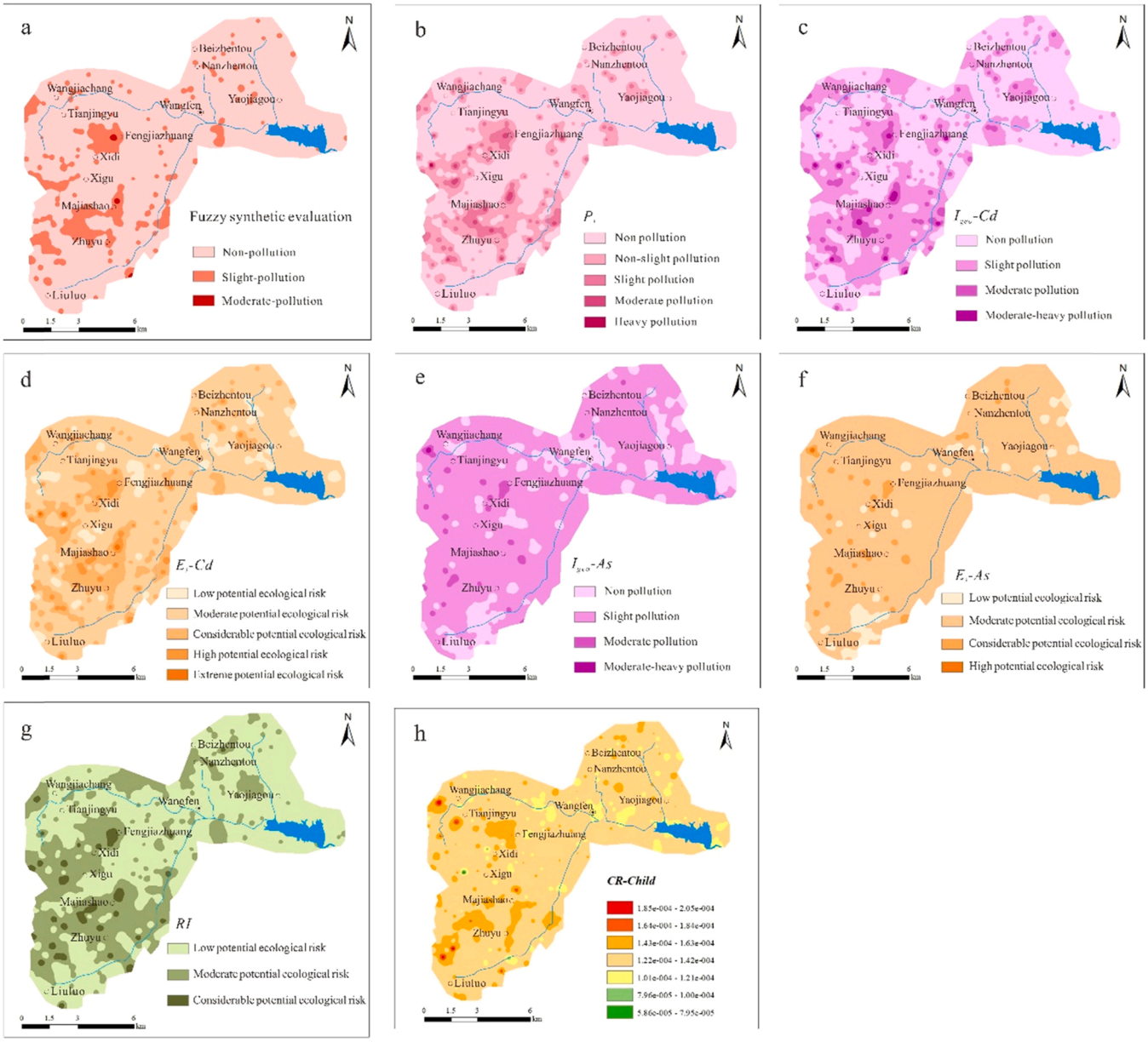

Fig. 3. Spatial distribution of heavy metal pollution, potential ecological risk and health risk levels in the Shi River Basin surface soil: a- pollution level based on fuzzy synthetic evaluation; b- pollution level based on P_i; c- pollution level of Cd based on I_{geo}; d- potential ecological risk level of Cd based on E_i; e- pollution level of As based on I_{geo}; f- potential ecological risk level of As based on E_i; g- potential ecological risk level based on *RI*; h- *CR* for the exposure of children to heavy metals.

APCS-MLR was used to quantify the contribution of four sources to the content of heavy metals in soil. The multiple linear regression fits the most closely when the measured value of heavy metal concentration is closer to 1 (Hingorani et al., 2020). As shown in Fig. S4, the ratios of fitting values to measured values are all close to 1. In addition, the fitting coefficients R2 of Cu, Pb, Zn, Ni and Hg are all greater than 0.8, and those of Cr, As and Cd are all greater than 0.6. This demonstrates that the APCS-MLR model effectively fit the heavy metals and provided suitable results from its calculations. As shown in Fig. 2, PC1 had higher relative contributions for Pb, Zn, and Cd, with contributions of 79.1%, 71.2% and 70.2%, respectively. PC2 contributed 80.1% and 76.2% for Ni and Cr, respectively. PC3 contributed predominately 71.4% of Cu. As was primarily affected by F1 with a contribution rate of 46.5%, followed by F2 and F3 that comprised 29.1% and 21.7%, respectively. PC4 was associated with Hg with a contribution of 88.4%.

3.4. Heavy metal pollution assessment

3.4.1. Fuzzy synthetic evaluation

The entropy values of Cu, Pb, Zn, Ni, Cr, As, Cd and Hg are 0.99, 0.956, 0.979, 0.995, 0.998, 0.991, 0.972 and 0.918, respectively. The weight of these eight indicators followed the order: Cd (0.513) > Hg (0.19) > As (0.127) > Pb (0.1) >Zn (0.048) > Cr (0.01) > Cu (0.009) > Ni (0.002). The main pollution factor in the surface soil of the study area was Cd, followed by Hg, As and Pb. The fuzzy synthetic evaluation results showed that 74.66% (498) of the samples were classified as level 1, which indicates that there is basically no heavy metal pollution in the soil of those sites. A total of 24.89% (166) of the samples were classified as level 2 indicating slight contamination. There were 0.45% (3) samples at level 3, indicating moderate contamination. The spatial interpolation results of soil fuzzy synthetic evaluation levels are shown in Fig. 3a. Consistent with the spatial distribution of Cd concentration in Fig. 1d, the slight to moderately polluted areas are primarily located in the west-

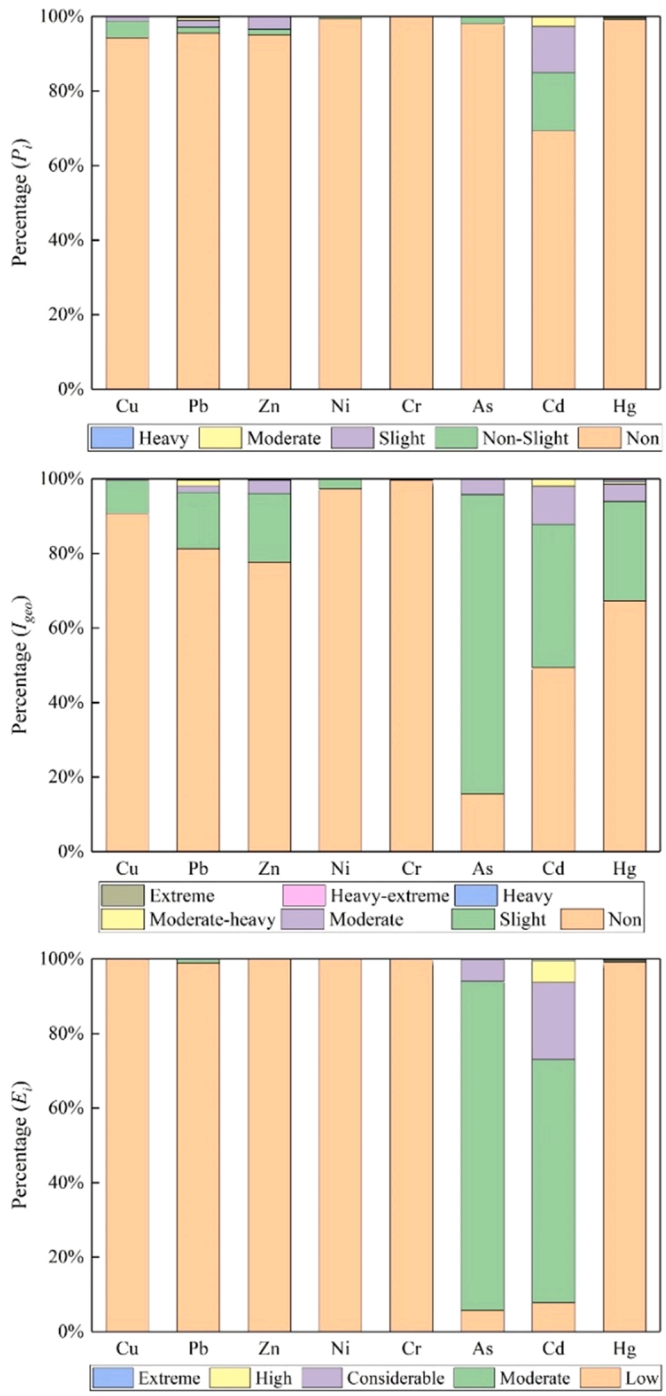

Fig. 4. Heavy metal pollution and potential ecological risk levels in the Shi River Basin surface soil based on index calculation.

southwest of the study area.

3.4.2. Calculation of pollution and ecological assessment index

Indices, such as P_i, I_{geo}, E_i and *RI*, were calculated to evaluate the degree of pollution and ecological risk of each heavy metal in the study area (Fig. 4). The results showed that the P_i values of all seven heavy metals with the exception of Cd were less than 0.7 in the surface soil of almost all the samples in study area. It is worth noting that 15.74% of the samples had negligible levels of Cd pollution; 12.29% were slightly polluted, and 2.7% were moderately or heavily polluted. The level of pollution obtained by the I_{geo} was more serious than that obtained by the P_i. Almost all of the Ni and Cr samples and most of the Hg, Zn, Pb and Cu samples were not contaminated. As pollution was the most serious; 80.36% of the samples were slightly polluted, and 4.2% of the samples were moderately or more seriously polluted. Followed by Cd, 38.53% of the samples were slightly polluted, and 12.14% of the samples were moderately or more seriously polluted. Similarly, the calculation results of the E_i for As and Cd do not bode well, with 88.31% and 65.22% of the samples of moderate ecological risk, respectively, and 6% and 26.99% of the samples of considerable ecological risk, respectively. The calculated *RI* value of 53.37% of the samples was less than 150, indicating a slight ecological risk. A total of 41.08% of the samples were more than 150 and less than 300, indicating a moderate ecological risk. In contrast, 5.55% of the samples were more than 300, indicating a considerable ecological risk. The spatial interpolation analysis of *RI* and P_i, I_{geo} and E_i of As and Cd with higher pollution levels was conducted. It can be observed that there were slight levels of pollution owing to As in the whole study area. Moderate or more serious pollution sporadically appeared in the west (Fig. 3d and f). The pollution owing to Cd was primarily concentrated in the west-southwest of the study area (Fig. 3c and e). The highest values for P_i as well as those for *RI*, appeared in the southwest (Fig. 3b and g).

On the whole, there were slight to moderate levels of heavy metal pollution and ecological risk in the study area. The contamination and risk caused by As and Cd are of particular note.

3.5. Health risk assessment

As shown in Fig. 5a and b, the *HI* values of heavy metals in all the samples for adults and children are < 1, indicating that the non-carcinogenic risk of heavy metals is negligible. The *HI* of heavy metals to children is more than 8-fold that of the adults, and the non-carcinogenic risks owing to As and Cr are higher (Fig. 6a). Fig. 5c shows that the *CR* values of heavy metals for adults in all the samples are $< 10^{-4}$, indicating that the carcinogenic risk of soil heavy metals to humans is at an acceptable level. However, Fig. 5d shows that higher CR_{Cr} values lead to a *CR* value $> 10^{-4}$, indicating that soil heavy metals result in a significant carcinogenic risk to children. With the exception of Ni, the *CR* values of other heavy metals to children were more than 8-fold that of adults (Fig. 6b). The contribution of ingestion, dermal contact and inhalation to the human health risk decreased in turn (Fig. 6c). A spatial interpolation analysis of the *CR* values of children showed that their *CR* values were only $< 10^{-4}$ in Xidi, the western part of Xigu and the southeastern part of Zhuyu. The highest *CR* values were obtained in the west near Wangjiachang, Tianjingyu and Liuluo, with a maximum value of 2.05×10^{-4} (Fig. 3h).

4. Discussion

4.1. Source analysis of heavy metals

The average concentrations of all heavy metals in the Shi River Basin was higher than the background values, with large CV values of Pb, Zn, Cd and Hg, indicating the influence owing to human activities. The results of an assessment of heavy metal pollution showed that the pollution and ecological risk level of the soil was slight to moderate, and the high values of heavy metals, particularly As and Cd, were primarily concentrated in the southwest. The heavy metals were divided into four groups based on their spatial distribution characteristics obtained by geostatistical analysis. Each component of a group may have the same source, namely (1) Pb, Zn, As and Cd; (2) Ni and Cr; (3) Cu; and (4) Hg. The fruit planting industry in the Shihe River Basin is relatively developed. Subsidiary industries, such as processing, manufacturing and transportation, are also developed, which lead to heavy metal pollution. In addition, the spatial distribution of heavy metals is affected by natural factors, such as the parent materials of soil. In conclusion, the distribution of heavy metals is determined by the comprehensive effects of agriculture, industry, transportation and natural factors.

Combined with the actual situation, the sources of heavy metal

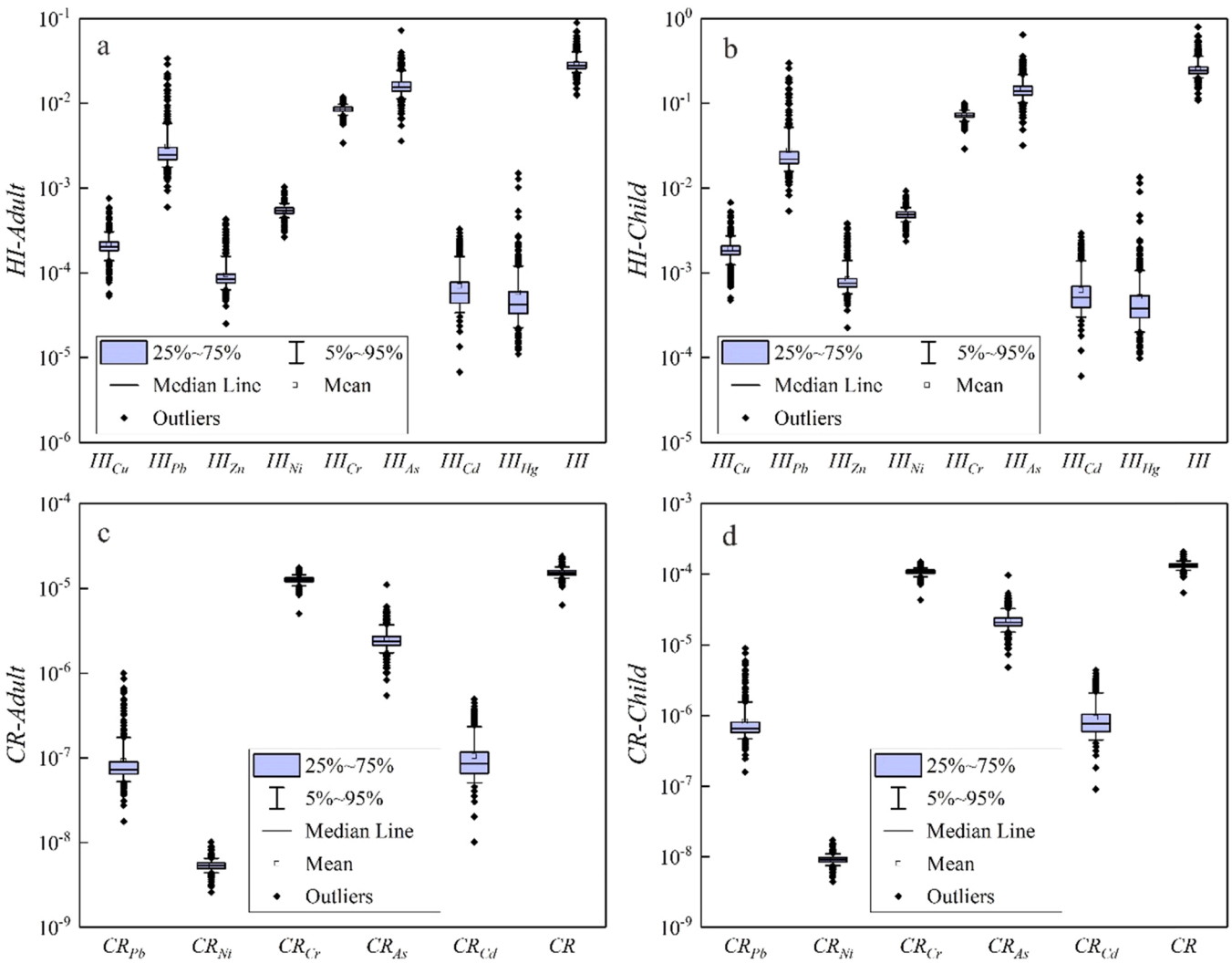

Fig. 5. Boxplot of *HI* and *CR* for adults and children following exposure to heavy metals in Shi River Basin.

pollution in the study area were further refined. The heavy metals of PC1 present in large amounts included Pb, Zn, As and Cd. Vehicle exhaust is an important source of Pb in the soil. Tetraethyl lead, an antiknock agent, is generally added to gasoline to reduce engine knock, resulting in particles from vehicle exhaust including lead that enter the soil (Lee et al., 2006). Zinc is an important component of tires. Zn-containing particles enter the soil with the frequent wear of vehicular parts (Zheng et al., 2013; Trujillo-González et al., 2016; Yadav et al., 2019). The processes of producing glass, pigments, and paper that use Cd and As or their compounds as raw materials can lead to soil contamination by particles that contain As and Cd (Zhao et al., 2014; Hu et al., 2020b). As shown by the spatial distribution of Pb, Zn, As and Cd, the areas with large amounts of them were concentrated in forest areas in which various fruit trees, such as hawthorn, persimmon, and pear, were planted. Owing to the advantages of fruit cultivation, a series of food company and packaging materials processing plants were developed, which require long-distance transportation. The combined effect of vehicles and factories that surround the planting regions contributed to the presence of these four heavy metals. It could be summarized that PC1 is owing to the mixed sources of processing and manufacturing, as well as traffic emissions.

The primary heavy metals of PC2 were Ni and Cr. A large number of studies have proved that Ni and Cr pollution comes from industrial activities such as smelting and coal burning (Chen et al., 2016). However, most of the industrial activities in the study area are materials and food processing. And the Ni and Cr contents of all samples were lower than their critical values in GB15618–2018 (Fig. S2), with low pollution level shown in Fig. 4. Thus, industry was not the main source of Ni and Cr. The small CV values (0.1–0.13) indicated that their accumulation in the soil was not apparent, suggesting their levels were less owing to human activities. Previous studies have shown that Ni and Cr in the soil are greatly related to the weathering and pedogenesis of parent material (Ma et al., 2018; Lv, 2019). Therefore, PC2 could be considered as a natural source.

The heavy metal with the highest load in PC3 was Cu. Agricultural activities are usually the main source of Cu. Cu is usually utilized as a feed additive to prevent diseases of livestock and poultry (Gupta et al., 2014). Cu enters the soil through manure and its compost (Belon et al., 2012; Hu et al., 2018; Qian et al., 2018). Cu is also often derived from pesticides and fertilizers, which are widely used in agriculture, leading to a significant increase in the Cu content in farmland (Qu et al., 2018). Combined with the spatial distribution of Cu (Fig. 1g), high values only appear in the large-scale crop growing area in the study area. Therefore, PC3 can be identified as an agricultural source.

The high load heavy metal of PC4 was Hg. The Hg pollution in the soil in the study area is distributed sporadically, and the high value area of Hg content exceeds that of other areas by more than 10 times. Industrial wastes are the main source of Hg (Hu et al., 2018; Fei et al., 2020). In the field investigation, it was found that sewage irrigation was mostly used in the study area to relieve the pressure of farmland water.

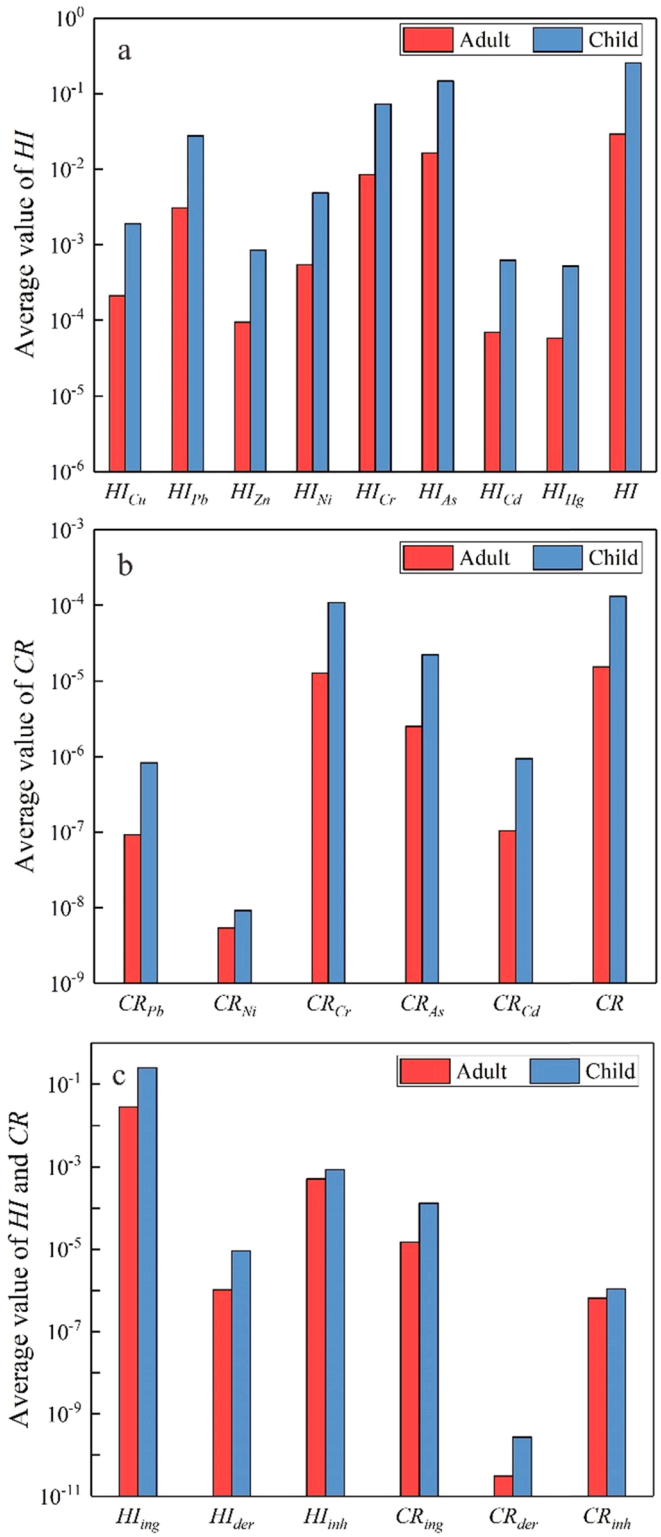

Fig. 6. Average values of *HI* and *CR* for adults and children: a- *HI* values following exposure to heavy metals; b- *CR* values following exposure to heavy metals; c- *HI* and *CR* values by three pathways of exposure.

The discharge of domestic sewage and waste water from factories and other enterprises leads to the pollution of the trunk canal. There is a risk of heavy metal enrichment in the soil after irrigation with trunk canal water. Weifang, the city to which the research area belongs, has reported excessive Hg in irrigation water. It is deduced that Hg originated from sewage irrigation caused by industrial wastewater. Therefore, PC4 could be identified as an irrigation source.

With the spatial changes of human activities in different regions, the sources and rates of contribution of heavy metals in the soil differ. For the whole study area, traffic emissions, as well as processing and manufacturing, contributed the most to soil heavy metals, followed by agricultural inputs.

4.2. Health risk assessment

Ingestion, dermal contact, and inhalation are the three main ways that people are exposed to heavy metals. As shown in Fig. 6c, ingestion is the most important route in this study. The carcinogenic and non-carcinogenic risks of all heavy metals to children are higher than those of adults, with a consistent variation tendency (Fig. 6a and b). The activities of children, such as their large numbers of hand to mouth and object to mouth contacts in concert with their low tolerance to toxicants, increase their risk of being exposed to carcinogens (Zhao et al., 2019; Jiang et al., 2020). Thus, children should be guided to pay attention to personal hygiene, particularly frequent hand washing, to reduce potential health risks. As was the most likely to result in non-carcinogenic risk, followed by Cr. The carcinogenic risk owing to Cr was the highest, followed by As. This study suggests that transportation, manufacturing and natural sources dominate the occurrence of health risks. It is worth noting that although natural sources are not the pollution source with the highest contribution to heavy metals, the risk of carcinogenesis to children caused by Cr, which is primarily derived from the parent material of soil, cannot be ignored throughout the whole region. Studies have shown that long-term exposure of the human body to low concentrations of Cr will produce toxic effects that lead to cancer and deformities (Khan et al., 2015; Zhao et al., 2019). The study suggested that heavy metals that contribute the most to pollution levels do not necessarily contribute the most to health risks. This can be attributed to the greater risk posed by the nature of the harmful heavy metals themselves (Wang et al., 2020b).

5. Conclusions

The average concentrations of heavy metals in the soil followed the order: Zn > Cr > Ni > Pb > Cu > As > Cd > Hg. The average concentrations of all heavy metals was lower than their corresponding threshold values but higher than their background values. Among them, the CV values of Pb, Zn, Cd and Hg were large, indicating that they are affected by human activities. Four heavy metals source factors in the study area were apportioned through a combination of APCS-MLR model and geostatistical analysis. PC1 represents a mixed source of transportation and industry; PC2 represents the natural source; PC3 represents the agricultural source, and PC4 represents the irrigation source. For the whole study area, transportation, processing and manufacturing have the largest impact, contributing 62.6%. The second is agricultural inputs, which contributes 21.1%. Combining fuzzy synthetic evaluation based on entropy weight with index methods was beneficial to determine soil pollution level from both qualitative and quantitative perspectives. The degrees of heavy metal pollution and ecological risk were both slight to moderate in the soil in study area. The pollution was primarily concentrated in the southwest, which was mainly caused by As and Cd. The major heavy metals that cause pollution do not necessarily pose the greatest health risks. The health risk assessment determined that the non-carcinogenic risk primarily caused by As was at an acceptable level. However, the carcinogenic risk to children caused by Cr in the study area should be the focus of more research. All heavy metals pose a higher carcinogenic and non-carcinogenic risk to children than to adults. In conclusion, human activities have adversely affected soil quality in the Shi River Basin, and mixed sources of transportation and processing should be subjected to priority management. Fuzzy synthetic evaluation based on entropy weight can increase the accuracy of pollution evaluation results and

provide reference for soil pollution evaluation in other areas. APCS-MLR method was used to quantitatively apportion soil heavy metals, and the subsequent work can also be combined with a variety of receptor models to test the results.

CRediT authorship contribution statement

Yuqi Zhang: Conceptualization, Methodology, Software, Formal analysis, Data curation, Writing – original draft, Writing – review & editing, Project administration. **Songtao Wang:** Conceptualization, Investigation, Writing – review & editing, Project administration. **Zongjun Gao:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Project administration. **Hairui Zhang:** Investigation. **Zihui Zhu:** Investigation. **Bing Jiang:** Investigation. **Jiutan Liu:** Data curation. **Hongzhi Dong:** Data curation. All authors have read and agreed to the published version of the manuscript.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2021.112926.

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