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Quantitative assessment of human health risks under different land uses based on soil heavy metal pollution sources

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ABSTRACT

Risk assessments and source analyses are important tools for the control of heavy metal soil pollution. In this study, the receptor model positive matrix factor method (PMF) and the health risk model are used to quantitatively evaluate the human health risks (carcinogenic and non-carcinogenic) of different pollution sources. The results showed that nickel, copper, and lead were significantly enriched due to human activities. The PMF model identified four pollution sources, among which agricultural activities contributed the most to soil heavy metal contamination (33.72%). Although the non-carcinogenic and carcinogenic risks to children were higher than those of adults, the health risks to both cohorts showed the same trend in the different land-use types. In terms of cancer risk, agricultural activities were the largest source of pollution, accounting for 37%, 41%, and 38% of the carcinogenic risk in construction, agricultural, and forest lands, respectively. Non-carcinogenic risks were primarily due to industrial emissions, and industrial activity was second only to agricultural activity in carcinogenic risk. This suggests that sources that contain dangerous heavy metals, such as Cr may lead to higher health risks. The results of this study provide a scientific basis for the quantitative assessment of health risks under different land-use.

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quantitative evaluation; source risk; land uses; heavy metal; health risks

Introduction

Soil is the foundational component of ecosystems, and its condition is essential for the survival of animals, plants, and humans (Mariagrazia et al. 2012). The problem of heavy metal soil pollution has gradually increased due to an increase in human activities and rapid societal development. Studies that examine heavy metal soil pollution are increasing, and many countries consider this a serious problem. The accumulation of heavy metals in soil affects the ecological environment and human health by transmission via the food

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chain (Rong et al. 2016; Xi et al. 2016). Therefore, an evaluation of the concentration distribution and environmental risks of heavy metals in soils and the identification the sources are essential for protecting the ecological environment and human health.

In recent years, China's environmental pollution problems have gradually surfaced, and the damage caused by heavy metals has had a significant impact (Chen et al. 2015). The pollution status of heavy metals in soil based on heavy metal soil background values or standard baselines has been extensively evaluated (Bednarova et al. 2013; Mamattursun et al. 2018). There are many reasons for heavy metal soil pollution, including both human and natural factors. For example, methods of land use can affect the degree of soil pollution, and heavy metals degrade agricultural soil functions and pollute agricultural products (Zhang et al. 2014). Different land uses, such as construction and forest lands, also have different pollution issues. Therefore, it is necessary to consider different types of land use when assessing health risks (Huang et al. 2018; Li et al. 2018). The analysis of the sources of soil heavy metal pollution has become a popular research topic. Most researchers have utilized receptor models to analyze the sources of heavy metals in soils, such as the positive matrix factorization (PMF) model, the principal component multiple linear regression model, and isotope labeling (Bai et al. 2016; Yang et al. 2017). The recent research has focused on assessing the risks of contamination of heavy metals in the soil, but ignoring the risks to the soil from its sources of pollution. Depending on the heavy metal pollution sources, land-use patterns, and other factors, the contents of heavy metals in soils differ, thereby affecting the heavy metal risk grade and distribution pattern (Chen and Lu 2018). The quantitative identification of soil heavy metal sources is extremely relevant for controlling key pollutants. The use of receptor models for source analyses and risk assessments is key to controlling pollution sources. The health and environmental risks due to heavy metal soil contamination need to be quantified in a manner that considers the different sources to more effectively control the sources of this pollution. In this manner, pollution sources can be prioritized to protect the environment and human health.

The Houzhai River Basin is located in Puding County of the Guizhou Province, China. It is a typical karst landform. The landforms in the area are complete, and the karst is developed. It is widely represented in the Yunnan-Guizhou Plateau (La et al. 2000). This location is a densely populated area in Puding County and a major grain producer. The health status of the soil greatly influences local food security and the health of the residents. Therefore, the environmental health risks of heavy metals in the local soils must be investigated.

In this study, the Houzhai River Basin was selected as the study area. The distribution of heavy metal concentrations in the surface soils from different land-use areas are analyzed. The sources of the heavy metals in the soil are then determined using a statistical analysis to assess the health risks due to the different sources. The identification of the heavy metal soil pollution sources provides certain suggestions and scientific basis for controlling and preventing this heavy metal pollution from the source.

Materials and methods

Study area

The study area (Houzhai River Basin) is located in Puding County, Guizhou Province, China. The geographical position is 105°40’ to 105°49’ E and 26°12’ to 26°18’N, at the

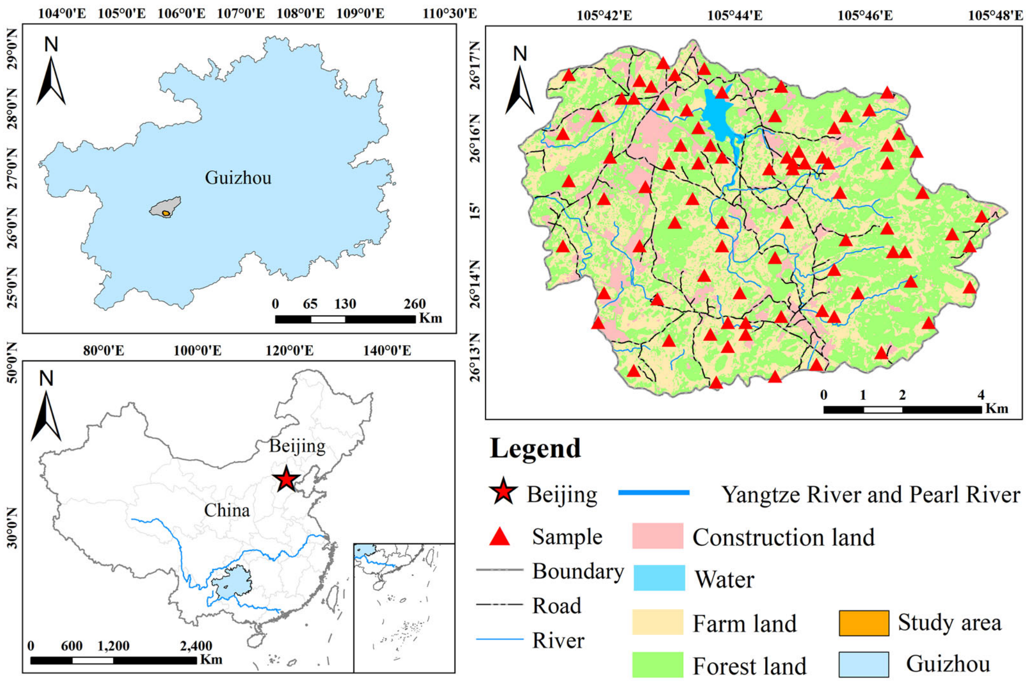

Figure 1. Location of soil samples in the study area.

junction of the Yangtze River Basin and the Pearl River Basin, with an altitude of 1 100 - 1 400 m. and the terrain is lower in the northwest and higher in the southeast. The area is about 81 km^2. The Houzhai River Basin is a typical karst landform, and the land use types are mainly agricultural land and forest land. The study area covers 13 administrative villages in the upper, middle and lower reaches of the basin. This area is the population gathering area of Puding County and the main grain producing area (Figure 1).

Sample collection and measurement

The 2015 Landsat8 remote sensing image is selected as the original data, and its resolution is 30 meters (the same as the sampling time). The software ENVI5.1 was used for image correction, cropping, classification and other processing on remote sensing images to extract the land use map of the study area. The 12, 29, and 41 samples sites were laid under the three land use types of construction, farm, and forest lands. The sampling depth was 20 cm divided into four layers. Approximately 100 g of soil was collected from each layer, and soil samples from the four layers were mixed to serve as the final soil sample. The samples were dried and filtered using a 200-mesh sieve. The soil samples were digested by the HNO_3-HCl-$HClO_4$-HF method, and six soil heavy metals were measured by inductively coupled plasma mass spectrometry (Thermo Electron, USA). The content includes Cr, Ni, Cu, Zn, Cd, and Pb.

During sample analysis, avoid contact with metal at every step to prevent cross-contamination of the sample. Use standard samples AGV-2, AMH-1, GBPG-1 for quality control. The recovery rate of standard samples is between 90% and 108%. The relative standard error (RSD) of the repeatability test is less than 10%. Results meet quality control requirements.

Risk analysis method

The enrichment factor (EF) statistical method was used to evaluate soil heavy metal enrichment level, and different land use types were divided and calculated (Reimann and Caritat 2005). Its formula is as follows:

$$\mathrm{EF} = \frac{(C/C_0)_S}{(C/C_0)_d} \tag{1}$$

$(C/C_0)_S$ is the measured concentration ratio of the calculated element to the reference element. $(C/C_0)_d$ is the soil background concentration ratio of the calculated element to the reference element. Common reference elements include Al, Cs, Nb, Y, Sc, Zr, Co, and Fe. (N'Guessan et al. 2009). Zr content was also measured in this study, because the coefficient of variation of the element Zr is small, and the average value is smaller than the background value for Guizhou Province. Zr was selected as the reference.

PMF model

The PMF model is a practical source analysis model that identifies and quantifies sources and calculates the source contribution of a sample point (Chen and Lu 2018; Mehr et al. 2017; Paatero 1999; USEPA E.P.A 2014). The model was developed using the software EPA PMF 5.0. The input file included the element raw concentration file and the uncertainty file. The uncertainty file was calculated based on the error in the experimental test and the method detection limit. The model solves the problem of chemical mass balance between metal concentration and source, including the number of factors and the contribution of each factor to the quality of each sample. The mathematical expression of PMF were described as Supplementary material.

The combination of the PMF model and the health risk assessment model can calculate the contribution rate of different heavy metal health risk sources and the proportion of risk values from different sources (Huang et al. 2018). Estimate the contribution concentration of each source of heavy metals in the soil at each sampling point, as shown in formula (2):

$$C_{nj}^{t} = C_{nj}^{t*} \times C_n \tag{2}$$

Where C_{nj}^{t} is the concentration contribution of the jth heavy metal element from the tth source in the nth sample (mg/kg); C_{nj}^{t*} is the calculated contribution rate of the jth element from the tth source in the nth sample, C_n is the concentration of elements in the soil in the nth sample (mg/kg).

Source-based health risk model

The cancer risk and non-cancer risk of children (under 12 years old) and adults were calculated using the risk model proposed by the Environmental Protection Agency (US

EPA S 1996). Heavy metals in soil endanger human health from three ways: (1) direct ingestion, (2) inhalation, and (3) dermal contact.

Thus, the health risks of soil heavy metal source in different environmental contexts were quantitatively analyzed. The six metals studied have chronic non-cancer health risks. Cr, Ni, Cd, and Pb also have cancer risks. The non-carcinogenic risk and cancer risk (children and adults) were calculated using the formula of the average daily exposure dose of the *t*th source of the *j*th element in the *n*th sample, as shown in Eqs. 3-5 (Lei et al. 2012; US EPA S 1996).

$$ADD^{t}_{nj,\,ing} = \frac{C^{t}_{nj} \times R_{ing} \times EF \times ED \times CF}{BW \times AT} \tag{3}$$

$$ADD^{t}_{nj,\,inh} = \frac{C^{t}_{nj} \times R_{inh} \times ED \times EF}{PEF \times BW \times AT} \tag{4}$$

$$ADD^{t}_{nj,\,der} = \frac{C^{t}_{nj} \times SA \times AF \times ABS \times EF \times ED \times CF}{BW \times AT} \tag{5}$$

The values of the parameters in the above formula are shown in Table S1 (MEPPRC 2014; Peng et al., 2016; US EPA S 1996).

Non-cancer risk is the sum of risk factors (HQ) for different exposure pathways and is assessed using the hazard indices (HI). $HQ^{t}_{nj,\,i}$ is the risk factor of the ith exposure route of the *t*th source of the *j*th element in the *n*th sample. The formula for calculating the adult and child HI is shown in equation (6).

$$\mathrm{HI} = \sum HQ^{t}_{nj,\,i} = \sum \frac{ADD^{t}_{nj,\,i}}{RFD_{i}} \tag{6}$$

If the HI > value is 1, it indicates that there may be an adverse health effect; if HI < 1, it means the opposite (Qing et al. 2015). The cancer risk (CR) value of heavy metals is used to assess the risk of cancer. If the CR value is greater than 10^{-4}, it indicates a risk of cancer; if CR is less than 10^{-6}, it is the opposite (Wu et al. 2015). The total carcinogenic risk TCR for adults and children is calculated as (7).

$$TCR = \sum CR^{t}_{nj} = \sum_{1}^{i} \left(ADD^{t}_{nj,\,i} \times SF_{i}\right) \tag{7}$$

Where CR^{t}_{nj} is the carcinogenic risk of the jth heavy metal element in the nth sample from the tth source. The values of the parameters SF_i and RFD_i are shown in Table S2 (Ferreira-Baptista and Miguel 2005; Jiang et al. 2015; Li et al. 2017; MEPPRC 2014; Meixia et al. 2018).

Results

Statistical analysis of soil heavy metals

Figure 2a shows a statistical analysis of Cr, Ni, Cu, Zn, Cd, and Pb in six soils. The content distributions of the six heavy metals are the same under the three land-use types. ( ) The average concentrations of Cr, Ni, Cu, Zn, and Pb exceed the soil background values for the Guizhou Province, and the average value of Cd is only higher than the

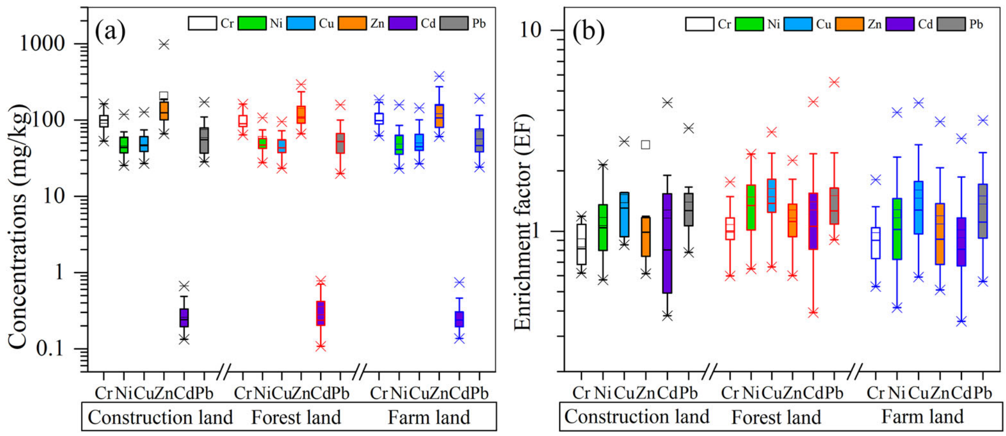

Figure 2. Statistical characteristics and enrichment of heavy metals in soils under different land uses.

background value in the forest land type. The maximum values of the six heavy metals were all found in agricultural land, so agricultural activities may be the primary source of heavy metals. When the soil background value of the Guizhou Province was used as the reference standard, the concentrations of Cr (66.7%), Ni (83.3%), Cu (100%), Zn (83.3%), Cd (66.7%), and Pb (91.7%) exceeded the corresponding background values among the samples from the construction land. The concentrations of Cr (69.0%), Ni (93.1%), Cu (96.6%), Zn (75.9%), Cd (65.5%), and Pb (86.2%) in the woodland soils exceeded the standard. For the agricultural land samples, the concentrations of Cr (82.9%), Ni (82.9%), Cu (97.6%), Zn (68.3%), Cd (65.9%), and Pb (92.7%) were higher than the background values. Cu and Pb pollution was concentrated in the construction and agricultural lands, whereas Cr and Ni pollution was concentrated in the agricultural and forest lands.

To further understand the source of heavy metal soil pollution, the EF for six elements was calculated (Figure 2b). In the forest lands, the average EF of the six elements was greater than 1, whereas only the average EF of Cr was less than 1 in the construction and agricultural lands. It can be speculated that Cr is mainly enriched in forest lands. The EF average value of Zn was less than 1 in the construction lands and was greater in the agricultural lands, thereby indicating that the Zn may originate from agricultural activities. In forest, construction and agricultural land, the average EF values of the three heavy metals are Cu (1.55, 1.45 and 1.52), Ni (1.12, 1.41 and 1.23), Pb (1.46, 1.57 and 1.43). These results indicate that these three heavy metals were affected by human activities and natural factors, but the effect of human activities was lower.

Source analysis of soil heavy metals

Pearson correlation analysis

The correlation between the heavy metal elements was determined using the Pearson correlation results. Prior to performing the correlation analysis, a logarithmic transformation was applied to the data to give it a normal distribution. As shown in Table S3, in all of the samples, the correlation between each element is significant, and the correlation coefficient between Cr, Ni, Cu, and Pb is greater than 0.7. In particular, the correlation coefficient between Ni and Cu is above 0.9, thereby indicating a high probability that the two had the same source. A good correlation was obtained between Cr and Ni, Cr and Cu, Pb and Cr, Ni and Cu, Pb and Ni, and Cu and Pb in the construction lands ($P < 0.01$). A significant positive correlation was found between all the heavy metals in the farm lands ($P < 0.01$). However, no correlation was observed between Cd and the five other metals in the forest lands ($P < 0.01$), indicating that Cd had a separate source in the forest lands. A significant correlation between the heavy metals suggests that these metals may have the same source of contamination. Cu, Cd, and Ni in the farmlands were significantly correlated in this study, which was similar to previous findings (Pan et al. 2016). It was concluded that the type of land use affected the correlation between the heavy metals in the soil to some extent (Zhao et al. 2014).

PMF model analysis

The PMF5.0 model was used to identify the sources of heavy metals in the six soils. Figure S1 shows the results of the model run from four sources. The sources of six heavy metals were identified using the model. The input data for the model included six heavy metal concentrations (82 samples) and their uncertainty files. The number of factors in the model was set to 2, 3, and 4 to observe the results of model operation. When the Q value reached the minimum and was most stable, the number of source factors was set to 4. In the model fitting results, the R^2 of Cr, Ni, Cu, Zn, Cd, and Pb were 0.992, 0.951, 0.947, 0.995, 0.960, and 0.992, respectively. The R^2 value was greater than 0.8, indicating that the results of this model were very reliable. The source contribution of heavy metals is shown in Figure 3.

The first factor accounted for 33.72% of the total variance and was the largest of all. The heavy metals Ni, Cu, and Zn accounted for 55.1%, 54.0%, and 46.1%, respectively. According to Table S3, the correlation of Ni, Cu, and Zn in the farmlands was very high, and it was greatly affected by human activities. The distribution of heavy metals was influenced by land-use patterns and human activities. Studies have found that heavy metals in road dust are primarily caused by traffic related activities (Li et al. 2016). However, according to the land-use map based on remote sensing technology

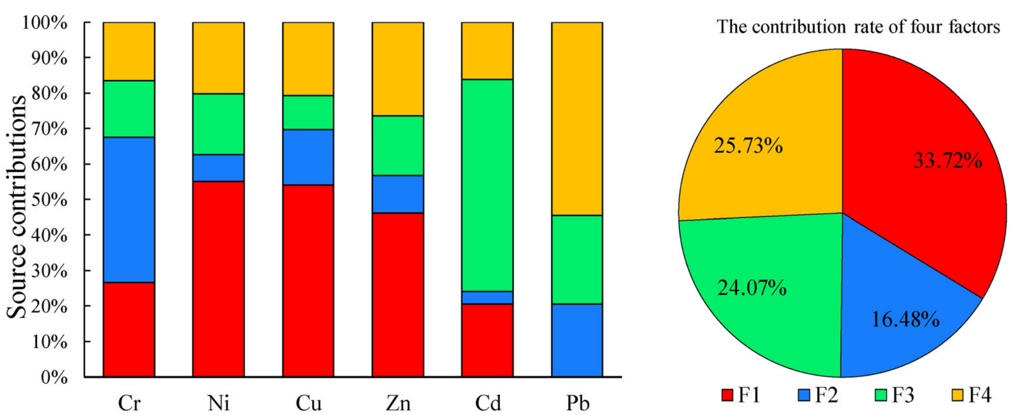

Figure 3. Source contribution of heavy metals (F1: agricultural activities, F2: industrial activities, F3: natural factors, F4: traffic activities).

(Figure 1), the study area is primarily dominated by agricultural area. The spatial distribution of heavy metals nickel, copper and zinc was similar (Figure S2), and the distribution of the three heavy metals was also similar to that in the farmlands. Studies have shown that annual use of copper-containing fertilizers and pesticides causes high copper pollution in Chinese farmlands (Liang et al. 2017). Cu enrichment in the sample was also reflected in the farmlands (Figure 2a). Studies have also shown that the primary cause of zinc and copper pollution is agricultural activities (Nicholson et al. 2006; Sun et al. 2013). According to the description of Ni, Cu, and Zn in Figure 2b, these metals were highly enriched in the farmlands. Therefore, F1 was identified as an agricultural activity.

The second factor was the smallest source, accounting for 16.48% of the total variance. The primary elements that contributed to this value were Cr and Pb, and the contribution rate of Cr was greater than 40%. Cr and Pb in the construction land showed a significant positive correlation, indicating that they were affected by human activities. Many studies have shown that Cr is primarily derived from industrial activities (Liu et al. 2016; Zhao et al. 2015). Atmospheric deposition diffuses particulate Pb, which easily flows into the environment through waste water and waste gas (Chen et al. 2016; Zeng et al. 2009). Although no large industrial sites were present in the study area, certain waste and wastewater emissions were generated through the production activities of small enterprises. Moreover, the accumulation of garbage can lead to an accumulation of Cr and Pb. Therefore, F2 was identified as an industrial activity.

The third factor accounted for 24.07%. Ni, Cd, and Pb were the primary contributing elements, accounting for 17.2%, 59.7%, and 25%, respectively, of the total contributing source. The distribution of Cd and Ni showed that there was also a certain enrichment phenomenon in the upper reaches of the watershed, and this area was primarily dominated by forest land (Figure 1). Therefore, the enrichment of nickel and cadmium in the soil in the upstream region may be affected by natural factors. Studies have shown that limestone is widely distributed in karst areas, and Cd has a significant correlation with limestone (Borůvka et al. 2005; Chen et al. 2016). The average Cd concentration in this study was similar to the background value. Therefore, the enrichment of cadmium may be closely related to the geological background. F3 is considered a natural factor.

The fourth factor accounted for 25.73%, and the contribution of Pb was the largest, accounting for 54.5% of the total source, followed by Zn and Cu. Past research has shown that lead pollution is primarily caused by traffic activities, such as exhaust emissions and leaded gasoline use (Cechinel et al. 2016; Li et al. 2010). In addition, Zn is the primary additive in car tires, and car tire wear also increases Zn pollution (Duan and Tan 2013; Guan et al. 2018; Men et al. 2018). The literature has also shown that Zn and Cu are primarily derived from traffic emissions and subsidence (Li et al. 2016). Thus, F4 was identified as traffic emissions.

The health risks of soil heavy metals to adults and children

On the basis of the mass contribution of the sample and the quantitative characterization model of human health risk due to heavy metal exposure (Eqs. 3–7), the health risk values of multiple factors contributing to the heavy metals in each sample were

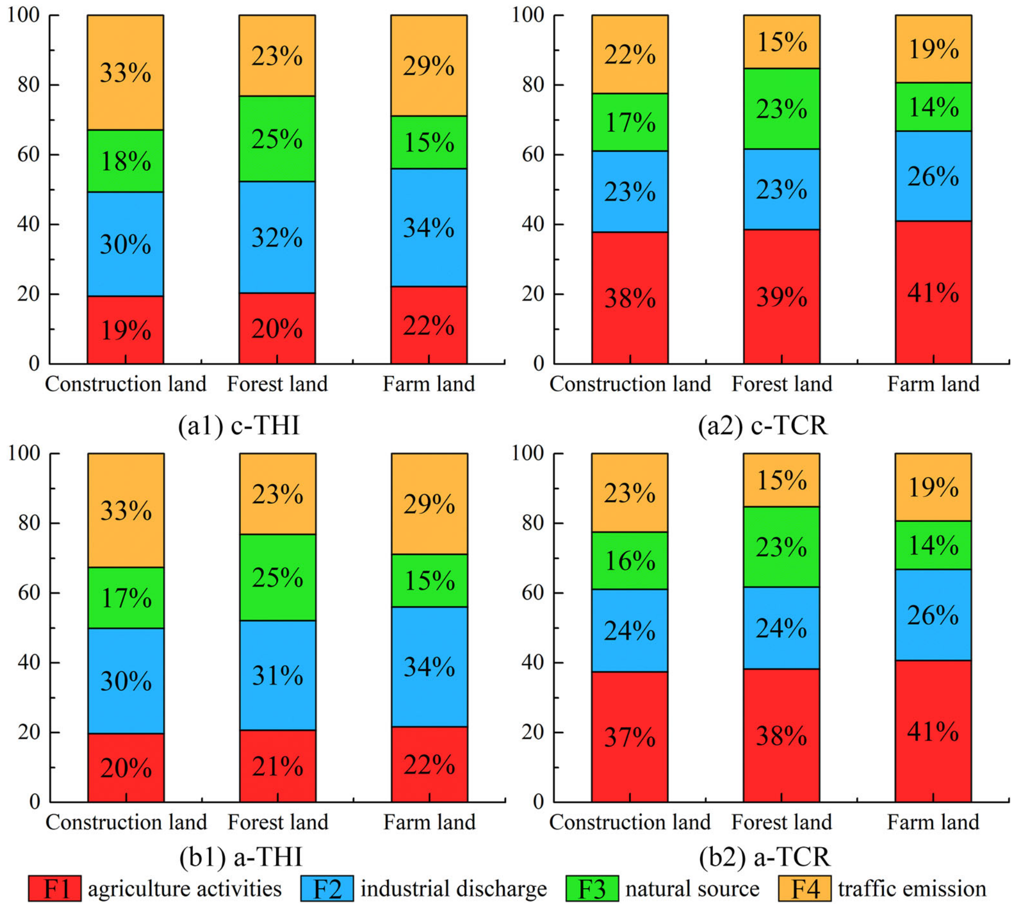

Figure 4. Percentage of total hazard indices (non- carcinogenic risks) and total carcinogenic risks for adult and children under different land use.

obtained. Given that two types of metals lack corresponding parameters, the cancer risk could not be calculated. Therefore, the carcinogenic risks mentioned below are the total cancer risks from Cr, Ni, Cd, and Pb.

Table S4 shows the health risk values for children and adults from the different land-use types (forest, construction, and agricultural lands). The non-cancer risk value for children and adults under the three land-use patterns was less than 1, indicating no health risks. However, the total non-carcinogenic risks for children was still greater than that for adults. The carcinogenic risk values for children and adults ranged from 10^{-5} to 10^{-4}. According to the carcinogenic risks assessment criteria, the four factors for adults and children had cancer risk values below the acceptable range. The total cancer risk to children exceeded the acceptable range (10^{-4}), thereby indicating a certain health impact from soil heavy metals to children.

Figure 4 (a1, b1) shows that the non-carcinogenic risk to children from the four sources was affected by the land-use types. Transportation activities (33%) and industrial sources (30%) were the primary sources in construction lands, while the main contribution from forest lands was due to industrial sources (32%). Industrial sources and transportation activities contributed the most in agricultural lands. Non-carcinogenic risks and carcinogenic risks from the four sources showed similar trends for both children and adults. Industrial sources were the major source of non-carcinogenic pollution risk. In addition, the harm from traffic sources in the construction and agricultural lands cannot be ignored. Figure 4 (a2, b2) shows that the contribution to cancer risk from agricultural activities under the three land-use patterns was greater than that of the other three types of sources. In agricultural, forest, and construction lands, agricultural sources accounted for 41%, 39%, and 38%, respectively, of the total cancer risk in children and 41%, 38%, and 38%, respectively, in adults. Therefore, agricultural sources contributed the most to cancer risk.

Table 1 shows the health risk values for children and adults under different exposure pathways according to the different land-use types (forest, construction, and agricultural lands). Direct intake had the greatest risk on health, followed by skin contact, with the lowest risk being inhalation by the nose and mouth. The carcinogenic and non-carcinogenic risks of children and adults showed the same trend under the different exposure pathways. In the two exposure routes of mouth and nose inhalation and skin contact, factor 2 (industrial sources) had a greater contribution to the cancer risk than factor 1 (agricultural activity). This result indicated that the effect of industrial activity was primarily on the risk of carcinogenicity through inhalation and skin contact, and non-carcinogenic risks were affected by three exposure routes. Factor 1 was a carcinogenic hazard to the human body, primarily through direct intake.

Discussion

The impact of the distribution of pollution sources contribution on health risks

The distribution of health risk sources for children and adults had the same trend. Therefore, the cancer risk and non-cancer risk to children was focused upon to discuss the relationship between source factors and human health risks. Figure 4 shows that the non-carcinogenic risk to children was primarily affected by industrial activities, and the leading factor in cancer risk was agricultural activities. Therefore, these two factors are discussed. Figure 5a and b shows the contribution of two factors; the contribution of industrial sources, F2, to children's non-cancer risk (Figure 5c) and the contribution of agricultural sources, F1, to carcinogenic risk (Figure 5d). These were obtained using ArcGIS and the ordinary Kriging interpolation.

As shown in Figure 5a and c, the influence of factor 2 was high in the southern and eastern portions of the study area, but low in the central and northwest. The sampling points with high non-cancer risk values for children were located in the south. In the eastern portion of the study area, factor 2 had a high contribution, and the land-use distribution map showed that the eastern portion of the study area was composed of forest land. Due to the small impact of natural accumulation on the accumulation of heavy metals in forest lands, most of them were affected by factors such as sedimentation of particulate matter produced by industrial emissions. Therefore, the heavy metals that accumulated in forest lands came from industrial activities. In the south, factor 2 and its contribution to non-cancer risk were equally high. This result indicated that the southern portion of the study area was susceptible to small industries. Figure 5b and d

Table 1. The carcinogenic risk values and non- carcinogenic risk values for different exposure pathway conditions.

		Total cancer risks of sources (TCR)					Total hazard indices of sources (THI)				
	source	F1	F2	F3	F4	Total	F1	F2	F3	F4	Total
Adults	ing	2.64E-05	1.55E-05	1.15E-05	1.21E-05	6.55E-05	2.07E-02	3.24E-02	1.91E-02	2.89E-02	1.01E-01
	inh	7.71E-08	1.17E-07	4.68E-08	4.96E-08	2.91E-07	1.84E-04	2.83E-04	1.11E-04	1.16E-04	6.94E-04
	der	1.26E-06	1.88E-06	7.74E-07	8.39E-07	4.75E-06	3.00E-03	4.66E-03	2.05E-03	2.26E-03	1.20E-02
	Total	2.77E-05	1.75E-05	1.23E-05	1.30E-05	7.05E-05	2.39E-02	3.73E-02	2.13E-02	3.13E-02	1.14E-01
Children	ing	4.71E-05	2.78E-05	2.05E-05	2.15E-05	1.17E-04	1.48E-01	2.32E-01	1.37E-01	2.07E-01	7.24E-01
	inh	3.56E-08	5.42E-08	2.16E-08	2.29E-08	1.34E-07	3.40E-04	5.24E-04	2.05E-04	2.14E-04	1.28E-03
	der	1.55E-06	2.32E-06	9.52E-07	1.03E-06	5.85E-06	1.48E-02	2.29E-02	1.01E-02	1.11E-02	5.89E-02
	Total	4.87E-05	3.02E-05	2.15E-05	2.26E-05	1.23E-04	1.63E-01	2.55E-01	1.47E-01	2.18E-01	7.84E-01

ing: ingestion; inh: inhalation; der: dermal contact. F1: agricultural activities; F2: industrial activities; F3: natural factors; F4: traffic activities.

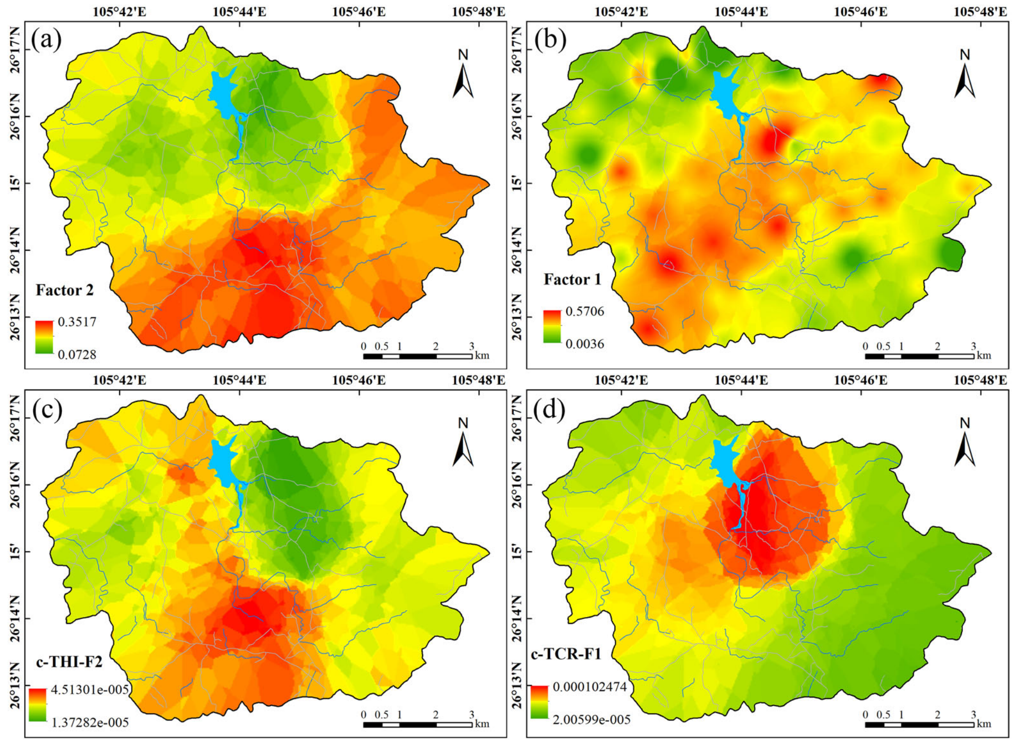

Figure 5. Spatial distribution of: (a) industrial activities; (b) agricultural activities; (c) the contribution of factor 2 to children's non-carcinogenic risk (THI); (d) the contribution of factor 1 to children's carcinogenic risk (TCR).

shows the distribution of factor 1 and its contribution to cancer risk in children. The change trend differed from the distribution of non-cancer risks. The distribution of factor 1 (Figure 5b) was similar to the distribution of agricultural land types, thereby indicating again that factor 1 was the source of agricultural activity. In a majority of the Houzhai River Basin, except for the central region, the cancer risk value was below the acceptable limit (10^{-5}). This result was due to the primary water system in the study area being located in the middle, thereby becoming the primary source of water for irrigation of the surrounding land. Therefore, agricultural activities are frequent, resulting in the enrichment of carcinogenic elements such as chromium, nickel, cadmium, and lead. People in the southwestern regions may also be affected by weak carcinogenic risks compared with other regions. The results showed that the health risks in the source must be quantified, and that the influence of the use of different land types must be considered to develop an accurate environmental pollution source management strategy.

Impact of pollution source contribution rate on health risks

Figure 4 shows that the health risks of children and adults had the same trend. Thus, this section only discusses the source contribution rates of the children's health risk effect. The human health risks from different sources were quantified using the PMF model and the health risk model. As shown in Figure 6 (a and b), the average non-

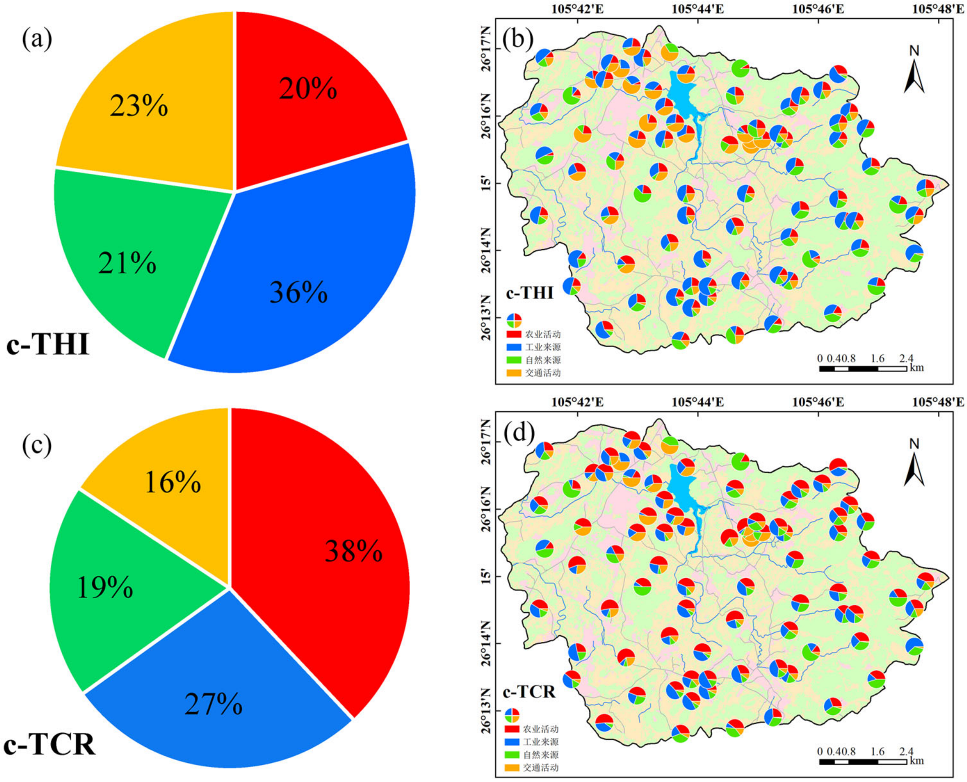

Figure 6. a. The percentage contribution of c-THI values, b. source contribution for c-THI of heavy metals, c. percentage contribution of c-TCR values, d. source contribution for c-TCR of heavy metals.

carcinogenic risk (c-THI) among children in the four source categories was dominated by industrial activity (36%), followed by traffic activities (23%). The mean non-cancer risk of both industrial sources and traffic activities did not exceed unacceptable levels (= 1). From the perspective of the spatial distribution, the southern portion of the study area was primarily affected by industrial-related activities, whereas the central portion was affected by traffic activities. There were more construction land and highways in the central portion, which was the most frequent area of human activities. In addition, small factories, such as fertilizer plants, were present in the southern region. As shown in Figure 6 (c and d), in contrast with the results of c-THI, the contribution of sources decreased in the following order: agricultural activities > industrial activities > natural activities. The average carcinogenic risk from agricultural activities was 4.76×10^{-5}, which was higher than that from other sources. However, all of the four sources were above the acceptable carcinogenic risk level (10^{-6}). The total area of agricultural land was large in the study area, and the central water system was rich and primarily based on agricultural activities. Therefore, the risk of cancer to children was affected by agricultural activities. Second, the results were the same as for the non-carcinogenic risks. The central portion was influenced by traffic activities, and the risk in the east was mostly derived from natural factors. Therefore, during agricultural production, given the rational application of chemical fertilizers, strict control of the quality of irrigation water is important for the prevention of human cancer risk. In particular, industrial activities contributed only 16.48% to the heavy metal content, but its contribution to the non-cancer risk was greater than 36%. In addition, it was also another major factor that contributed to the cancer risk (27%). The results suggest that sources that contain dangerous heavy metals, such as Cr, may easily lead to higher health risks.

Health risk assessments that consider the source distribution are more meaningful than those that merely use health risk thresholds. Nevertheless, this study does contain some shortcomings, given that only four sources were considered during the study. Furthermore, only four heavy metals were considered in the cancer risk assessment. Thus, the total cancer risk may be underestimated. Unutilized species sensitivity distribution models and heavy metal elements should be included in future studies to assess risks and improve understanding regarding the health risks of other pollutants from different sources in the study area.

Conclusions

This study quantified the health risks of soil heavy metal sources in the Houzhai River Basin under different land-use types. The average EF of Ni, Cu, and Pb in the soil in the study area exceeded 1, and the average concentrations of Cu, Pb, Cr, and Ni in agricultural soils were higher. Both of these results indicate that the risks were primarily due to human activities, especially agricultural activities. Using the receptor model PMF, four heavy metal sources consisting of agricultural activities (F1), industrial activities (F2), natural sources (F3), and transportation activities (F4) were identified, and their contributions were quantified. These results were combined with the health risk assessment model to calculate the carcinogenic risk and non-carcinogenic risk of four pollution sources.

Direct intake was found to be the primary exposure route for non-carcinogenic and carcinogenic risks. Only children had a carcinogenic risk beyond acceptable limits. In terms of carcinogenic risk, agricultural activities had a greater contribution to adult cancer risk, accounting for 37%, 38%, 41% from construction lands, forest lands, and agricultural lands, respectively. Hence, agricultural activities were considered the number one source of carcinogenic risk. In terms of non-carcinogenic risks, industrial emissions were the primary source for adult risks from agricultural lands (34%), and transportation activities are the primary source for adult risks from construction lands (33%). The health risk trends for children were the same as those of adults. The spatial distribution of the source factors indicated that the sources from agricultural activities were concentrated in the central and southwestern portions of the study area, and the carcinogenic risk contribution for adults and children was primarily concentrated in the central and northern portions. Industrial activities in the southern region contributed the most to both the non-carcinogenic and carcinogenic risks of adults, suggesting that adults living in the south are primarily affected by industrial-led health risks. It was also found that industrial activities were the largest source of non-carcinogenic risks and the second largest source of carcinogenic risks after agricultural activities. This result suggested that sources containing dangerous heavy metals (Cr) may lead to higher health risks. This study provides an effective way to control and manage soil heavy metal pollution from the sources to manage and prevent human health risks.

Disclosure statement

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