



# Distribution, source identification, and health risk assessment of heavy metals in the soil-rice system of a farmland protection area in Hubei Province, Central China

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Received: 26 December 2020 / Accepted: 25 June 2021 / Published online: 19 July 2021

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## Abstract

Heavy metal contamination in farmland soil is of great concern due to the threat to food security arising from the bioaccumulation of heavy metals in crops planted in contaminated soil, such as rice, corn, and vegetables. Cd is the main contaminant in both paddy soils and rice. The purpose of this study was to reveal the spatial distribution of 8 heavy metals (Cd, Cr, Ni, Cu, Pb, Zn, As, and Hg) in the farmland protection areas in northwestern Hubei Province and to evaluate their pollution status, sources, and health risks. The total amounts of these 8 heavy metal elements in the samples were measured, and the health risk posed by their accumulation in rice was evaluated using the health risk evaluation model recommended by the US Environmental Protection Agency (US EPA). The mean contents of Cd, Ni, Cu, Zn, Hg, and As in soil exceeded the background levels (0.17, 37.3, 30.7, 83.6, 0.077, and 12.3 mg kg<sup>-1</sup>, respectively) of Hubei Province, and Cd showed the highest enrichment coefficient. The concentration of Cd in 89.1% of samples exceeded the limit values stipulated in the Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (Trial) (GB15618-2018). The contents of heavy metals showed dissimilar geographical distribution patterns. The principal component analysis (PCA) results indicated that Cd, Zn, Ni, As, and Cu mainly originated from the application of pesticides and fertilizers; Cr mainly originated from soil texture and pedogenesis; exhaust gas generated during transportation was the point pollution source of Pb; livestock wastewater, manure irrigation, and atmospheric deposition were the main pollution source of Hg. The contents of Ni and Cd in 52.2% and 58.7% of the rice samples, respectively, exceeded the limit values stipulated in the Food Safety National Standards for Contaminants in Foods (GB2762-2017), and the average effective Cd content accounted for 81.9% of the total Cd. The average bioconcentration factor of each heavy metal in rice followed the order Cd > Zn > Hg > As > Ni > Cr > Pb. Cd and As were the main noncarcinogenic contributing factors, accounting for 80.8% of the total noncarcinogenic risk. The carcinogenic risk indexes of Cd, As, and Cr exceeded the risk index threshold of 10<sup>-4</sup>, indicating a carcinogenic risk to the human body. The highest risks to local residents from heavy metals were found in rice. Cd and As were the main noncarcinogenic and carcinogenic factors and should receive greater attention in risk decision management.

**Keywords** Soil · Heavy metals · Rice · Sources · Risk assessment

Responsible Editor: Elena Maestri

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## Introduction

Farmland soil, as an indispensable natural resource for agricultural production, is China's largest carrier of rice crops. Its environmental quality determines the yield and quality of rice (Wu et al. 2019). With the development of the economy, improper disposal of wastewater and solid waste by humans, unreasonable use of chemical fertilizers and pesticides, exhaust emissions, etc. (Adimalla et al. 2019; Sun et al. 2019; Cao et al. 2010), pollutants enter farmland soil through different routes, causing paddy field pollution. Heavy metals are characterized by strong stability and nondegradability

(Huang et al. 2016), and they can enter the human body through direct ingestion, skin contact, and food chain transmission, causing harm to human health (Zhuang et al. 2009; Friedlingstein et al. 2014; Park et al. 2004). In a random inspection of harmful heavy metals in the soil of 300,000 hectares of basic farmland protection area in China, the Ministry of Environmental Protection found that the heavy metals in the soil had an overstandard rate of 12.1% (Chen et al. 2018). A survey by the Chinese Ministry of Environmental Protection shows that 16% of China's soils and 19% of agricultural soils are polluted (CCICED 2015), and the main pollutants are cadmium (Cd), nickel (Ni), copper (Cu), arsenic (As), mercury (Hg), lead (Pb), dichlorodiphenyltrichloroethane (DDT), and polycyclic aromatic hydrocarbons. A survey by the Ministry of Agriculture of 1.4 million hectares of sewage irrigation areas in China found that the heavy metal content exceeded the standard in 64.8% of the total area (Song et al. 2015). According to reports, the reduction in the country's annual output of food due to heavy metal pollution alone is more than 10 million tons. The amount of food contaminated by heavy metals also reaches 12 million tons (Lu 2016).

Rice is the grain crop with the largest planting area and highest yield in China, and it is also the bulk cereal crop with the strongest absorption of heavy metals. Rice is also the most widely consumed staple food, supporting a large part of the world's population, especially in Asia (Silva et al. 2015). However, it has been reported that eating food contaminated with heavy metals can cause more than 200 acute and chronic diseases resulting from gastrointestinal infections (World Health Organization 2015), such as cardiovascular, kidney, and neurological diseases (Ostrom 2009). Therefore, it is paramount to assess the risk to human health caused by heavy metals in soil and food crops (especially rice) in China.

In 1976, the US Environmental Protection Agency (US EPA) first published risk assessment guidelines for suspected carcinogens. In 1983, the National Academy of Sciences (US) established a basic framework for health risk assessment. On this basis, the US EPA compiled a series of technical documents and guidelines on health risk assessment (US EPA 1996, 2005). Among them, the evaluation model (US EPA 1989a, b) in "Superfund Site Risk Assessment Guidance: Human Health Risk Assessment," referred to as the US EPA model, has been widely used throughout the world (Ahmed et al. 2015; Giri and Singh 2015). In China, risk assessment research began in 1990. To date, the health risks for farmland (Zhao et al. 2015), mining areas (Zhuang et al. 2009), electronic dismantling plants (Wu et al. 2015; Zheng et al. 2007), and other sites with polluted soil have been evaluated using the US EPA model. However, studies have focused primarily on soil pollution, and there is relatively little research on the heavy metal pollution of food crops and its associated health risks.

Based on this state of knowledge, this paper took a farmland protection area in northwestern Hubei as the research area; 46 pairs of soil and rice samples were collected from paddy fields, and the contents of eight heavy metals (Cr, Ni, Cu, Zn, Cd, Pb, As, Hg) were determined. The health risks caused by local cereal products in the study area to residents were assessed using the US EPA model. The results can provide a scientific basis for local environmental risk management and decision-making and offer a reference for the coordination of human health risk control and economic sustainable development.

## Materials and methods

### Study area

The research area is a farmland reserve in Zhuxi County, located in northwestern Hubei Province, China. The agricultural land area in Zhuxi County is 21,780 hectares, accounting for 6.6% of the county's total area. It has a subtropical monsoon climate with four distinct seasons and a mild climate. The average annual rainfall is approximately 1000 mm. The annual frost-free period is 238 days. The farmland protection area is flat, fertile, and rich in rice. It is the main production area and high-yield area of grain and oil crops in Hubei Province and the country as a whole; the rice produced in this area was given to the emperor in ancient times and was known as the "royal tribute." The sampling area encompasses a total of 148.4 hectares of farmland soil, taking the farmland reserve in Zhongfeng town as the core area.

Soils in the area are mainly paddy soil and yellow-brown soil, which are widely distributed in Central China. It has the characteristics of high mineral nutrient content, strong organic matter decomposition, and good fertilizer retention. Large amounts of chemical fertilizers and pesticides are applied in the area for better harvest, which leads to farmland soil pollution and pollutant enrichment in crops. Studies have shown that from 1995 to 2014, the intensity of agricultural chemical fertilizer application in Hubei Province showed an upward trend; meanwhile, pesticide consumption increased by 13.17% from 1.124 to 12.72 million tons (Hubei Province Statistical Yearbook Committee 2014). Aware of the problem of excessive use of fertilizers, livestock manure and wastewater are used as organic fertilizers.

### Sample collection and processing

The samples were collected in June 2016, and the sample points were set up according to the distribution of farmland in the study area, the avoidance of nearby pollution sources, and the relevant regulations of the "Soil Environmental Monitoring Technical Specification" (HJ/T166-2004).

During the field sampling phase, 46 sampling points shown in Fig. 1 were set according to the surrounding terrain while GPS positioning was used. Record and describe the coordinates of each sampling point and the environment around the point, recording relevant information such as soil type, color, etc. At each sampling point, adequate farmland surface soil (0–20 cm) was collected according to the five-point sampling method. The midpoint of the diagonal was taken as the center sampling point, and then four points on the diagonal that were equidistant from the center sample point were taken as the additional sample points. The samples from these five points were completely mixed; then, the quartering method was used to take approximately 1 kg of soil as the experimental sample. The detailed steps of the quartering method were as follows: the mixed samples were spread evenly into circles, and then divided into four equal parts. Soil from opposite quarters was removed, and the process was repeated until a 1 kg sample remained.

Each soil sample corresponds to a plant sample, and a total of 46 pairs of soil and rice grain samples were collected. The rice grain samples were also collected according to the five-point sampling method. The rice had reached maturity and was about to be harvested. The location map of the study area and sampling sites is shown in Fig. 1.

According to “Soil Environmental Monitoring Technical Specifications” (HJ/T166-2004) and other relevant requirements, the soil samples were naturally dried indoors to remove plant debris, gravel, and other debris. Then, they were sieved using 20- and 100-mesh nylon sieves after grinding, and finally, they were put into bags for use. Rice grain samples were

washed with tap water, rinsed with distilled water, air-dried, and shelled. The shelled rice was freeze-dried ( $-20^{\circ}\text{C}$ , 48 h) and ground into a fine powder with an agate mortar to obtain brown rice. The brown rice samples were sieved through a 60-mesh nylon sieve and then bagged and sealed (Satpathy et al. 2014).

### Sample analysis and testing

The pH of the soil was determined according to the standard method in “Determination of Soil” (NYT 1377-2007) using a 1:2.5 water: soil ratio for the leaching and glass electrode method. The determination of Cr, Ni, Cd, Cu, Pb, and Zn in the soil was performed according to the standard method in “Determination of 12 Metal Elements in Soil and Sediments: Aqua Regia Extraction-Inductively Coupled Plasma Mass Spectrometry” (HJ803-2016). A total of 0.1 g of sample was weighed into a digestion tube, 6 ml of aqua regia was added and mixed well, and then the sample was put in a microwave digestion apparatus for digestion. The digestion procedure was as follows: temperature rise to  $120^{\circ}\text{C}$  within 5 min, hold for 3 min; temperature rise to  $150^{\circ}\text{C}$  within 4 min, hold for 6 min; temperature rise to  $180^{\circ}\text{C}$  within 5 min, hold for 40 min. After the digestion was completed, the sample was cooled, filtered with slow quantitative filter paper, and brought to a volume of 50 ml with ultrapure water. The analyte contents were measured by inductively coupled plasma mass spectrometry (ICP-MS, iCAP Q, Thermo Scientific, USA). The extraction-based effective states of heavy metals in the soil were determined by diethylenetriamine pentaacetic acid

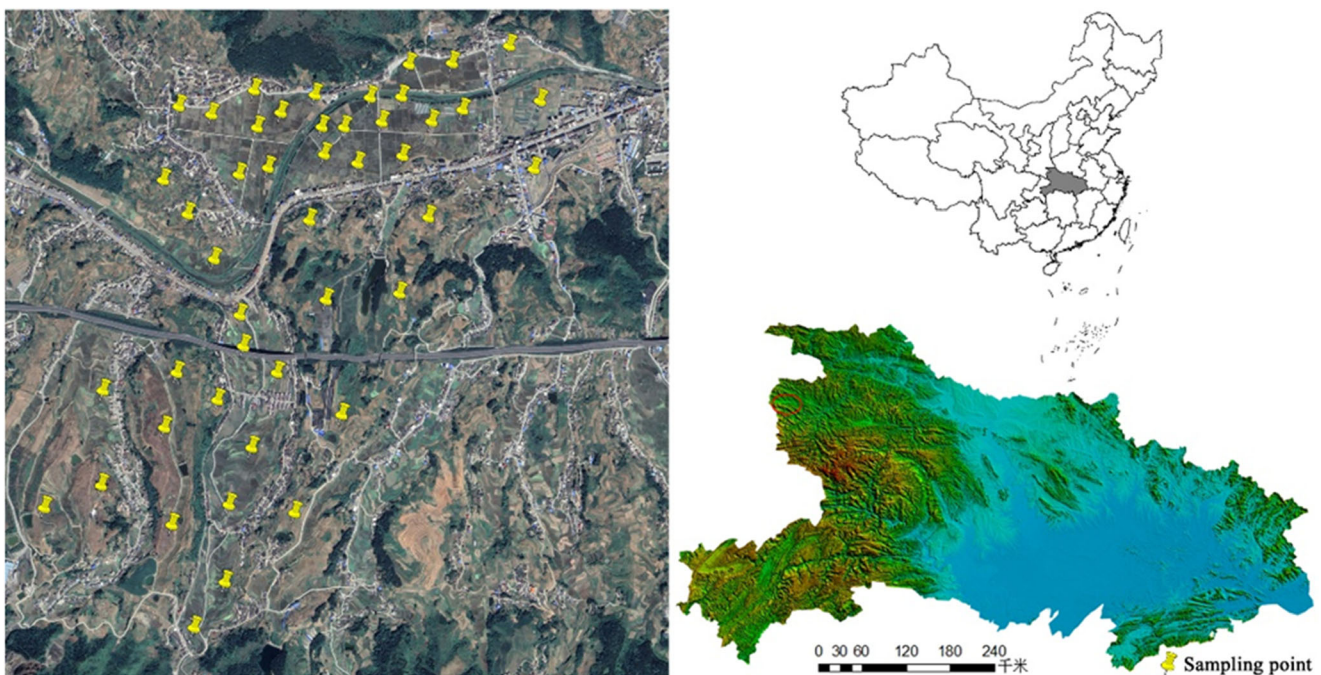


Fig. 1 Location map of the study area and sampling sites

(DTPA) leaching–inductively coupled plasma mass spectrometry (HJ804-2016). As and Hg in the soil were measured using HJ 680-2013 (“Determination of Mercury, Arsenic, Selenium, Bismuth and Antimony in Soil and Sediment by Microwave Digestion/Atomic Fluorescence Method”). The biologically effective states of As and Hg were extracted with  $\text{NaHCO}_3$  and  $\text{NH}_4\text{OAc}$ . A sample (10.0 g) was weighed and placed into a triangular flask, 20 ml of extraction solution was added, the sample was shaken at 170 r/min for 2 h at 20°C and then centrifuged for 10 min, and the supernatant was determined by atomic fluorescence spectrometry (AFS-8230, Jitian, Beijing) (Wang et al. 2011). Heavy metals in rice were digested with  $\text{HNO}_3\text{-H}_2\text{O}_2$  and determined by inductively coupled plasma mass spectrometry (ICP-MS, iCAP Q, Thermo Scientific, USA). A rice sample (0.2 g) was weighed into a digestion tube, 2 ml  $\text{HNO}_3$  was added for predigestion for 4 h, 1 ml  $\text{H}_2\text{O}_2$  was added, and the sample was digested in a microwave apparatus. The digestion procedure was as follows: heat to 120°C within 5 min, hold for 2 min, heat to 150°C within 4 min, hold for 6 min, heat to 180°C within 5 min, hold for 10 min, heat to 200°C for 6 min, hold for 25 min; acid digestion proceeded for approximately 4 h after the procedure was completed. The digestion extract was diluted to 50 ml with ultrapure water after cooling to room temperature, and then the heavy metal content was measured using ICP-MS.

Each soil and rice sample had three parallel samples, and a soil standard material (GBW07408), rice standard material (GBW08502), and blank samples were analyzed for quality control. After inspection, the sample repeat rate was 100%, and the pass rate was 100%. The analysis precision, report rate, detection limit, and related parameters all met the requirements of DZ/T0258-2015.

### Bioconcentration factor (BCF)

The bioconcentration factor (BCF) can reflect the ability of crops to absorb and concentrate heavy metals from soil. It is expressed as the ratio of the heavy metal content in a certain part of the crop to the corresponding heavy metal content in the soil (Satpathy et al. 2014). The expression is  $\text{BCF}=\text{CR}/\text{CS}$ , where BCF is the bioconcentration factor, CR is the heavy metal content in the crop (rice seeds in this study), and CS is the content of the same heavy metal in the corresponding soil. The larger the BCF is, the stronger the crop’s ability to concentrate heavy metals.

### Health risk assessment model and parameters

Health risk assessment is an evaluation method that links environmental pollution with human health. This method qualitatively and quantitatively analyzes the harmful factors in the environment that bring adverse health effects to the population

according to certain evaluation criteria and technical routes: the possibility and extent of physical and chemical factors causing damage to people in specific environments are evaluated (Hao et al. 2010). The human health risk assessment model (US EPA 1989a, b) proposed by the US EPA was used for health risk assessment. This method has been proven to be effective by a large number of studies and is widely used worldwide. According to the US EPA “Carcinogenic Chemical Classification Standards” (US EPA 2011), Cu, Zn, Ni, Hg, Cd, Cr, As, and Pb have chronic noncarcinogenic risks, among which Cd, Cr, As, and Pb also have carcinogenic risks. Therefore, when conducting health risk analysis in this study, it was necessary to consider both noncarcinogenic and carcinogenic risks.

#### 1. Calculation of exposure

The average daily intake (ADD) is a parameter used to quantify the oral exposure to heavy metals through hand-mouth intake of cereal products ( $\text{mg kg}^{-1} \text{d}^{-1}$ ). It is usually calculated using the following formula:

$$\text{ADD} = \frac{C_i \times \text{IR} \times \text{ED} \times \text{EF}}{\text{BW} \times \text{AT} \times 365} \quad (1)$$

$C_i$  indicates contaminant content in rice,  $\text{mg kg}^{-1}$ ; IR indicates daily intake content of rice,  $\text{kg d}^{-1}$  (children, 0.35; adults, 0.499); ED indicates exposure duration, a (children, 6; adults, 24); EF indicates exposure frequency,  $\text{d a}^{-1}$  (children, 365; adults, 365); BW indicates bodyweight, kg (children, 15.9; adults, 56.8); AT indicates averaging time, a (carcinogenic, 70; noncarcinogenic, 6 for children and 24 for adults).

#### 2. Noncarcinogenic health risk

$$\text{HQ} = \frac{\text{ADD}}{\text{RfD}} \quad (2)$$

$$\text{HI} = \sum \text{HQ}_i \quad (3)$$

The noncarcinogenic health risk (HQ) is computed as the ratio of ADD to RfD. RfD is the daily tolerance, and the RfDs of Cr, Ni, Cu, Zn Cd, Pb, As, and Hg are 0.003, 0.02, 0.04, 0.3, 0.001, 0.0035, 0.0003, and  $0.0003 \text{ mg (kg d)}^{-1}$ , respectively (US EPA 2014).

The risk index (HI) indicates the final assessment of overall noncarcinogenicity, expressed as the sum of the HQ values of 8 heavy metals.  $\text{HI} > 10$  means that there is a serious noncarcinogenic risk,  $\text{HI} > 1$  means that there is a noncarcinogenic risk, and  $\text{HI} \leq 1$  means that there is no noncarcinogenic risk (US EPA 1989a, b).

### 3. Carcinogenic health risk

$$CR = ADD \times SF \quad (4)$$

$$TCR = \sum CR \quad (5)$$

The carcinogenic health risk (CR) represents the incremental probability of an individual developing cancer over a lifetime of exposure to a potential carcinogen. TCR expresses the sum of the CR values. SF is the carcinogenic intensity coefficient ( $(\text{mg kg}^{-1} \text{d}^{-1})^{-1}$ ), and the SF values for Cd, Pb, As, and Cr via ingestion are 0.38, 0.0085, 1.5, and 0.5, respectively (US EPA 2016). Relevant expert research shows that the acceptable range of carcinogenic risk is  $10^{-6}$ – $10^{-4}$  (US EPA 1989a, b). The names and values of related parameters in formulas (1)–(5) are shown in Table S1 in the Supplementary Material.

### Statistical analysis

Statistical analyses of data were conducted using Microsoft Excel 2013. The normal distribution test, Pearson's correlation analysis, and principal component analysis (PCA) were performed using SPSS 17.0. The validity of PCA was verified by a p value lower than 0.001 in Bartlett sphericity tests and by a Kaiser-Meyer-Olkin (KMO) value of 0.611 ( $>0.5$ ). Ordinary kriging spatial interpolation analysis was carried out in ArcGIS 10.4 software.

## Results and discussion

### Spatial distribution and concentrations of heavy metals in soils

As seen from Table 1, in the study area, Cd, Ni, Zn, As, and Hg all had different overstandard levels according to the risk control values in the Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (Trial) (GB15618-2018). The Cd overstandard rate was the highest, reaching 89.1%, and the Ni, Zn, As, and Hg overstandard rates were 17.4%, 13.1%, 2.2%, and 2.2%, respectively; these results indicate that some paddy soil points have been polluted by heavy metals and that Cd pollution is the most serious. The overstandard multiple of Cd was 6 in soil with pH  $<5.5$  and 4.3, 2.5, and 1.6 in soil with pH 5.5–6.5, 6.5–7.5, and 7.5, respectively.

The average contents of heavy metals in the soil exceeded the environmental background values of Hubei, except for Cr and Pb, and the average contents of Cd, Ni, Cu, Zn, Hg, and As were 12.4, 1.6, 1.6, 2.1, 1.7, and 1.2 times the background values, respectively. This result indicates that the cumulative

effect of soil Cd in the study area is the most significant. The spatial distribution of heavy metals in soils of the investigated region reveals dissimilar geographical distribution patterns (Fig. 2). The spatial distribution map of Cd shows a decreasing trend from south to north, with very high concentrations appearing in the south-central area. The spatial distribution maps of Ni, Zn, and Cu show similar geographical trends, with decreases from southwest to northeast. Furthermore, high concentrations of As appeared in the southern part of the study area. Hg was relatively evenly distributed, and high-concentration points appeared in the northeast corner. Although the mean values of Cr and Pb were slightly lower than the background values, 30.4% and 32.6% of the samples exceeded the background value, respectively, indicating that Cr and Pb were enriched in some of the soil samples. The spatial distribution maps of Cr and Pb show that high concentrations of Cr were mainly located in the northeast and high concentrations of Pb were mainly located in the center.

The coefficient of variation (CV) reflects the average degree of variation of a heavy metal in the study area, which is computed as the ratio of the standard deviation (SD) to the mean. The greater the CV value is, the more uneven the distribution of heavy metal elements in the soil. According to the research of Zhao et al. (2020), the CV is less than 10%, indicating weak variation, which means that the content of heavy metals is mainly affected by natural sources; 10–90% indicates moderate variation; CV  $>90\%$  indicates strong variation, which means that the content of heavy metals is mainly affected by human factors. The CVs of the 8 heavy metal elements in the study area followed the order Cd  $>$  Hg  $>$  Ni  $>$  Cu  $>$  As  $>$  Zn  $>$  Cr  $>$  Pb; the CV of Cd was the largest, reaching 91.9%, and the CVs of Hg, Ni, and Cu were above 40%, indicating that Cd concentrations were highly influenced by anthropogenic sources, while Hg, Ni, and Cu concentrations were moderately affected by human factors.

### Source identification by principal component analysis and correlation analysis of metal concentrations between 8 heavy metals

Principal component analysis (PCA) and correlation analysis were used to investigate the causes of heavy metal pollution. The PCA results for the contents of the 8 heavy metal elements are shown in Table S2 in the Supplementary Material. The first principal component, PC1, showed high loadings of Ni, Cu, Zn, Cd, and As, with contribution rates of 98.1%, 94.9%, 94.3%, 92.6, and 71.8%, respectively, accounting for 54.12% of the total variance. Cd and Zn contents, as well the contents of Ni, Cu, in most of the surface soil of the study area, were significantly higher than the soil background values of Hubei Province. The correlation analysis between the concentrations of 8 heavy metal elements is shown in Table S3. Ni, Cu, Zn, Cd, and As showed significantly positive correlations

**Table 1** Analysis of the total contents of heavy metals in the soil in the study area ( $\text{mg kg}^{-1}$ )

Element	As	Hg	Cd	Cr	Ni	Cu	Pb	Zn	
Mean	14.2	0.13	2.1	78.8	58.6	49.8	26.2	178.6	
Standard deviation	4.5	0.09	1.9	16.9	27.8	20	2.8	46.4	
Range	9.4–33.3	0.07–0.69	0.5–9.1	43.1–122.7	28.4–161.6	28.4–122.2	19.7–31.7	126.5–343.2	
Coefficient of variation (%)	31.2	67.1	91.9	21.4	47.5	40.1	10.6	25.9	
Background value of Hubei	12.3	0.077	0.17	86	37.3	30.7	27.6	83.6	
Overstandard rate of the background value (%)	67.4	95.7	100	30.4	84.9	93.5	32.6	100	
Mean/background value	1.2	1.7	12.4	0.9	1.6	1.6	0.9	2.1	
Farmland soil pollution risk screening value	pH <5.5	30	0.5	0.3	250	60	150	80	200
	5.5 <pH <6.5	30	0.5	0.4	250	70	150	100	200
	6.5 <pH <7.5	25	0.6	0.6	300	100	200	140	250
	7.5 <pH	20	1	0.8	350	190	200	240	300
Sample excess rate (%)	2.2	2.2	89.1	0	17.4	0	0	13	

( $P < 0.01$ ) with each other ( $r = 0.805$  for Cd and Cu,  $r = 0.915$  for Cd and Ni,  $r = 0.866$  for Cd and Zn,  $r = 0.518$  for Cd and As,  $r = 0.929$  for Cu and Ni,  $r = 0.905$  for Cu and Zn,  $r = 0.717$  for Cu and As,  $r = 0.895$  for Zn and Ni,  $r = 0.526$  for Zn and As, and  $r = 0.736$  for Ni and As), which indicated that Ni, Cu, Zn, Cd, and As might come from common sources. The similar spatial distribution trends of Ni, Zn, and Cu supported this interpretation. Studies have shown that long-term and extensive use of pesticides in farmland may lead to the accumulation of heavy metals such as Cu, As, Zn, and Cd in soil (He et al. 2013; Huang et al. 2010). In addition, mining, smelting, and industrial activities can release heavy metals into the environment (Zhang et al. 2019). The research area is a farmland protection area where pesticides and inorganic fertilizers are applied year-round, and there are no industrial activities in the area. It can be considered that PC1 is a human active pollution component, and its contributing heavy metals are mainly derived from anthropogenic pollution related to agricultural activities. The second principal component explained 23.394% of the variance and had high factor loadings for Cr, Pb, and Hg; the contribution rates were 87.8%, 86.4%, and 33.9%, respectively. The average values of Cr and Pb did not exceed the background values, and only a few points exceeded the background values. Cr and Pb showed significantly positive correlations ( $P < 0.01$ ) with each other ( $r = 0.621$  for Cr and Pb). A previous study also showed that Cr was impacted by soil parent materials (Sajn 2003). Common sources of Pb in soils include car exhausts, manure, sewage sludge, and coal burning (Yang et al. 2014). There is no industrial pollution in the area, so Cr mainly originates from soil texture and pedogenesis. A highway crosses the farmland area, so exhaust gas generated during the transportation is the point pollution source of Pb. In addition, the Hg content exceeded the soil background value in Hubei Province. Studies have pointed

out that those sources of Hg pollution in soil include parent material, atmospheric deposition, coal burning, livestock wastewater irrigation, and livestock manure (Wang et al. 2020). The field investigation found that there was an obvious phenomenon of human and livestock wastewater irrigation in these areas, which inferred that the Hg pollution in the area might be caused by livestock wastewater and manure irrigation. Simultaneously, Hg pollution may originate from atmospheric deposition. Therefore, PC2 showed a complex source of components, including a mixture of natural and anthropogenic pollution (e.g., transportation and aquaculture wastewater and human waste).

### Contents of bioavailable heavy metals in soil

There are different forms of heavy metals in soil, and the forms that can be absorbed and utilized by plants are called bioavailable states (Chojnacka et al. 2005). The soil heavy metal enrichment ability of crops is directly related to the bioavailable heavy metal content. The average bioavailable content of Cd in the study area was  $1.72 \text{ mg kg}^{-1}$ , accounting for 81.9% of the total Cd (see Table S4 in the Supplementary Material). The content of bioavailable Cd in 67.4% of the samples exceeded the soil environmental quality risk control standards for agricultural land under different pH conditions, indicating that Cd pollution in the soil is serious. Lin et al. (2019) found that exogenous Cd entering the soil mainly exists in an exchangeable state, resulting in high Cd bioavailability. Some studies have shown that the use of fertilizers (such as ammonium nitrogen fertilizer) will increase the content of dissolved and exchangeable Cd in the soil (Wan et al. 2006). The effective contents of Cr, Ni, Zn, and As in the soil samples were not high and accounted for less than 10% of their respective total contents.

**Fig. 2** Spatial distribution of the concentrations of heavy metals in soils

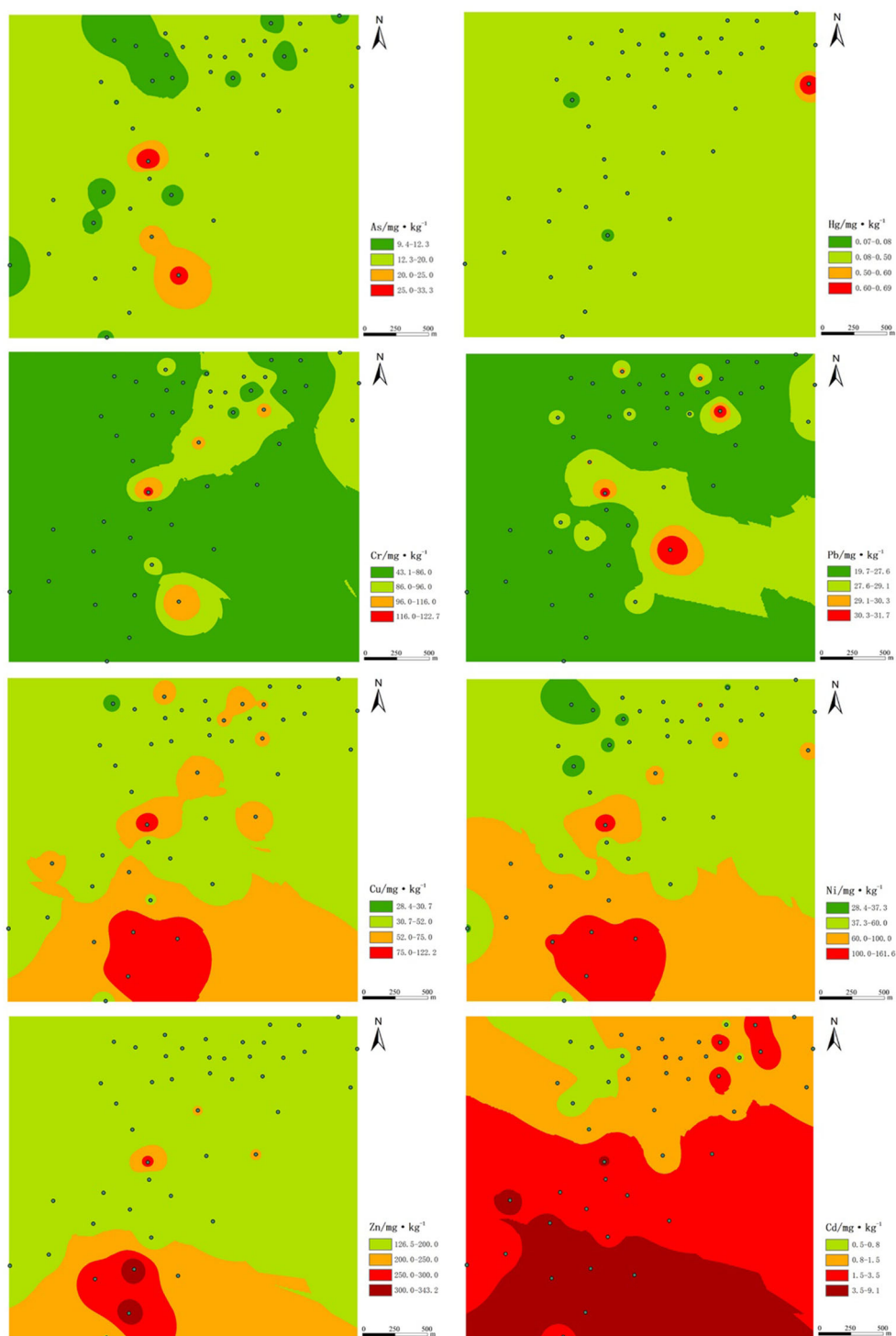


Table S5 in the Supplementary Material shows that, in addition to Hg, the effective contents of the 7 heavy metals Cd, Cu, Pb, Cr, Ni, Zn, and As were significantly negatively correlated with soil pH, and the correlation coefficients were 0.483, 0.537, 0.434, 0.509, 0.555, 0.552, and 0.536, respectively ( $P < 0.01$ ). The effective states of Cd and Cu were significantly positively correlated with soil organic matter,

and the correlation coefficients were 0.418 and 0.428 ( $P < 0.01$ ), respectively. The effective Hg content was significantly negatively correlated with soil organic matter, and the correlation coefficient was 0.381 ( $P < 0.01$ ). The correlations of the other five elements with organic matter were not significant. Moreover, the effective contents of Cd, Cu, Ni, Zn, and As in the soil were significantly positively correlated with each

other ( $P < 0.01$ ), indicating that the heavy metals Cd, Cu, Ni, Zn, and As have high homology. The effective Cu content in the soil was positively correlated with Cr with a correlation coefficient of 0.318 ( $P < 0.05$ ), and Zn was negatively correlated with Hg with a correlation coefficient of 0.291 ( $P < 0.05$ ).

### Contents of heavy metals in rice

Table 2 shows that the average contents of the 8 elements (As, Hg, Cd, Cr, Ni, Cu, Pb, and Zn) in edible rice in the study area were  $0.16 \text{ mg kg}^{-1}$ ,  $0.007 \text{ mg kg}^{-1}$ ,  $0.79 \text{ mg kg}^{-1}$ ,  $0.34 \text{ mg kg}^{-1}$ ,  $0.64 \text{ mg kg}^{-1}$ ,  $2.93 \text{ mg kg}^{-1}$ ,  $0.06 \text{ mg kg}^{-1}$ , and  $15.94 \text{ mg kg}^{-1}$ , respectively. The average contents of the heavy metals, except for Ni and Cd, were within standard limits<sup>(a,b)</sup>. Although the contents of Zn and Cu in rice were much higher than those of Cd, Zn and Cu are essential nutrients for rice growth and have low toxicity (Zheng et al. 2018). Cd is a nonessential element for rice and has strong physiological toxicity. The Cd overstandard rate in rice samples was 58.7%, and the Ni overstandard rate in rice reached 52.2%. It can be seen that Cd pollution in both soil and rice is relatively serious and needs attention. It is worth noting that although the average content of Ni in the soil did not exceed the standard, the average content of Ni in rice did exceed the standard.

The crop enrichment factor is the ratio of the heavy metal content of edible parts of crops to the content of the same heavy metal in the soil. It can reflect the migration and accumulation intensity of heavy metals from soil to rice (Lokeshwari and Chandrappa 2006). The migration and enrichment of heavy metals in soil to crops is a key process by which humans contact heavy metals through the food chain (Friedlingstein et al. 2014). The average BCFs of heavy metals in rice seeds in the study area were 0.005 (Cr), 0.011 (Ni), 0.350 (Cd), 0.065 (Cu), 0.002 (Pb), 0.093 (Zn), 0.012 (As), and 0.064 (Hg) (Table 2). The enrichment coefficients were  $\text{Cd} > \text{Zn} > \text{Hg} > \text{Cu} > \text{As} > \text{Ni} > \text{Cr} > \text{Pb}$ . The enrichment capacity of rice for Cd was 159.0 times that of Pb, 71 and 30

times those of Cr and Ni, and 5.4 and 3.8 times those of Cu and Zn. This result shows that Cd has the strongest enrichment ability, followed by Zn. This finding is consistent with previous studies (Lokeshwari and Chandrappa 2006).

Comparing the heavy metal contents of rice in this study with results for other regions (Table 3) reveals that the Cd content of rice in the study area was higher than those for basic farmland in the Yangtze River Delta (Xiao et al. 2010), Jiaozuo (Ma et al. 2014), and Cixi (Shen et al. 2013) and higher than those around electronic dismantling plants (Yin et al. 2018, Gao 2015), but lower than those in mining areas in Shaoguan (Zheng et al. 2018) and Xiangbei (Jiang et al. 2017). There was little difference in the contents of Cr, Ni, Zn, Pb, As, and Hg in rice between the study area and basic farmland (Xiao et al. 2010., Xiao et al. 2010; Ma et al. 2014; Shen et al. 2013). The Cu content was slightly lower than that of basic farmland (Xiao et al. 2010; Shen et al. 2013). The Ni, Pb, Cu, and Zn contents of rice in this study were lower than those in mining areas (Zheng et al. 2018; Jiang et al. 2017) and electronic dismantling plants (Yin et al. 2018; Gao 2015). However, the contents of As and Hg were higher than those of electronic dismantling plants (Yin et al. 2018).

The results of the correlation analysis of the heavy metal contents in rice and the available heavy metal contents in the soil are shown in Table S6 in the Supplementary Material. The contents of Ni, Cd, and As in rice were significantly related to the effective contents of Ni, Cd, and As in the soil, and the correlation coefficients were 0.427, 0.590, and  $-0.421$  ( $P < 0.01$ ). The content of Zn in rice was related to the content of Zn in soil with a correlation coefficient of 0.343 ( $P < 0.05$ ), and the correlation between other heavy metal elements was not significant. Scientific control of the effective contents of Ni and Cd in the soil can reduce the contents of Ni and Cd in rice to a certain extent (Yang et al. 2009).

**Table 2** Characteristics of heavy metal elements in rice ( $\text{mg kg}^{-1}$ )

Elements	AS	Hg	Cd	Cr	Ni	Cu	Pb	Zn
Mean	0.16	0.007	0.79	0.34	0.64	2.93	0.06	15.94
Standard deviation	0.04	0.002	0.95	0.17	0.51	0.96	0.02	2.12
Range	0.07–0.27	0.003–0.012	0.01–3.13	0.17–1.73	0.08–2.01	0.80–6.12	0.0–0.15	11.56–21.12
Standard limit	0.7 <sup>b</sup>	0.02 <sup>a</sup>	0.2 <sup>a</sup>	1.0 <sup>a</sup>	0.4 (Fu and Hu 1999)	10.0 <sup>b</sup>	0.2 <sup>a</sup>	50.0 <sup>b</sup>
Overstandard rate %	0	0	58.7	0	52.2	0	0	0
BCF	0.012	0.064	0.350	0.005	0.011	0.065	0.002	0.093

<sup>a</sup>“National Food Safety Standard Limits of Contaminants in Food” (GB2762-2017)

<sup>b</sup>“Limit of eight elements such as lead, cadmium, chromium, mercury, selenium, arsenic, copper and zinc in grains (including grains, beans and potatoes) and products” (NY 861-2004)



**Table 3** Comparison of heavy metal contents in rice from different areas (mg kg<sup>-1</sup>)

Areas	Cr	Ni	Cu	Zn	Cd	Pb	As	Hg	References
Farmland in Jiaozuo	0.40	0.40	-	-	0.00	0.03	-	-	Ma et al. 2014
Farmland in the Yangtze River Delta	0.19	-	5.20	22.73	0.07	0.10	0.13	0.007	Xiao et al. 2010
Farmland in Hubei	0.34	0.64	2.93	15.94	0.79	0.06	0.16	0.007	The study area
Farmland in Cixi	0.13	-	6.80	-	0.06	0.14	0.10	0.006	Shen et al. 2013
Mining area in Shaoguan	-	-	5.10	32.40	1.40	2.30	-	-	Zheng et al. 2018
Mining area in Xiangbei	0.30	1.40	8.10	20.90	1.20	0.10	-	-	Jiang et al. 2017
Electronic dismantling plant in Guiyu	0.21	1.37	3.00	17.32	0.07	0.13	0.07	0.001	Yin et al. 2018
Electronic dismantling plant in Wenling	-	0.30	5.30	28.00	0.09	-	-	-	Gao 2015

### Human health risk evaluation of heavy metals in rice

The evaluation results of the noncarcinogenic hazards of rice to humans (Table 4) show that the HQs of Ni, Pb, and Hg for both adults and children were <1. For Cr, Cu, and Zn, the HQ for adults was <1, but the HQ for children was >1, indicating that Ni, Pb, and Hg have no noncarcinogenic risk to adults and children, while Cr, Cu, and Zn have noncarcinogenic risks to children. The HQ range of Cd for adults was 0.1~22.08, with a mean value of 5.58, and that for children was 0.33~68.84, with a mean value of 17.41. The HQ range of As for adults was 1.65~6.28, with a mean value of 3.76, and that for children was 5.14~19.59, with a mean value of 11.72. The HQ values of Cd and As were higher than 1 for adults and more than 10 for children, indicating that Cd and As pose noncarcinogenic risks to adults and serious noncarcinogenic risks to children. The HI for adults was 11.55, while that for children was 36.03, 3.12 times greater than that for adults. The contribution rate of HQ<sub>Cd</sub> to HI was 48.3%, and the contribution rate of HQ<sub>As</sub> to HI was 32.5%. Therefore, Cd and As are the main noncarcinogenic contributing factors.

The evaluation results of the carcinogenic hazards of rice to humans (Table 5) show that the carcinogenic risk indexes of Cd in rice were 7.27E-04 (adults) and 5.67 E-04 (children), those of As were 5.8 E-04 (adults) and 4.52 E-04 (children), those of Cr were 4.17 E-04 (adults) and 3.25 E-04 (children), and those of Pb were 1.17 E-06 (adults) and 9.13 E-07

(children). Only the carcinogenic risk of Pb was within the acceptable range of 10<sup>-6</sup>~10<sup>-4</sup>, and the carcinogenic risks of Cd, Cr, and As for both adults and children exceeded the risk threshold of 10<sup>-4</sup>, indicating that Cd, Cr, and As have a certain carcinogenic risk. Cd had the highest carcinogenic risk, accounting for 42.1% of TCR, and Cr and As accounted for 24.2% and 33.6% of TCR, respectively.

### Conclusion

The average values of Cd, Ni, Cu, Zn, Hg, and As in the soil of the study area exceeded the background values by 12.4, 1.6, 1.6, 2.1, 1.7, and 1.2 times, respectively. The contents of Cd, Ni, Zn, As, and Hg exceeded the limits in the “Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (Trial)” (GB15618-2018), where the Cd content exceeded the standard value by 89.1%, indicating that Cd has significant accumulation in the farmland protection area and that Cd pollution is serious. The available Cd in the soil accounted for 81.9% of the total Cd in the study area.

The heavy metals in the study area showed dissimilar geographical distribution patterns. Cd appeared to decrease from south to north. Ni, Zn, and Cu showed similar geographical trends, and high concentrations of As appeared in the southern part of the study area. High concentrations of Cr were mainly

**Table 4** Indexes of noncarcinogenic risk for adults and children

	HQ	Cr	Ni	Cd	Cu	Pb	Zn	As	Hg	HI
Adults	Minmum	0.40	0.03	0.10	0.14	0.07	0.27	1.65	0.08	11.55
	Maximum	4.07	0.71	22.08	1.08	0.29	0.50	6.28	0.29	
	Mean	0.81	0.23	5.58	0.52	0.11	0.38	3.76	0.17	
Children	Minmum	1.25	0.08	0.33	0.44	0.23	0.85	5.14	0.24	36.03
	Maximum	12.68	2.21	68.84	3.37	0.91	1.55	19.59	0.89	
	Mean	2.53	0.70	17.41	1.61	0.36	1.17	11.72	0.53	

**Table 5** Indexes of carcinogenic risk for adults and children

	CR	Cr	Cd	Pb	As	TCR
Adults	Minimum	2.07E-04	1.36E-05	7.41E-07	2.54E-04	
	Maximum	2.09E-03	2.88E-03	2.98E-06	9.69E-04	
	Mean	4.17E-04	7.27E-04	1.17E-06	5.80E-04	1.73E-03
Children	Minimum	1.61E-04	1.06E-05	5.77E-07	1.98E-04	
	Maximum	1.63E-03	2.24E-03	2.33E-06	7.56E-04	
	Mean	3.25E-04	5.67E-04	9.13E-07	4.52E-04	1.35E-03

located in the northeast, and Pb was mainly located in the center. Hg was relatively evenly distributed. The PCA results showed that the main sources of Cd, Zn, Ni, As, and Cu in the surface soil of the study area are anthropogenic pollution related to agricultural activities; the main sources of Cr are soil-forming parent materials; there is anthropogenic Pb pollution near highways; Hg pollution in the area might be caused not only by livestock wastewater and manure irrigation but also atmospheric deposition. Therefore, Hg is a complex source, including a mixture of natural and anthropogenic pollution.

The content of available Cd was significantly positively correlated with soil organic matter content and significantly negatively correlated with soil pH. The average contents of Ni and Cd in rice in the study area exceeded the national food hygiene standard. The Cd overstandard rate of rice samples was 58.7%, and the Ni overstandard rate of rice reached 52.2%. The contents of other heavy metals in the rice did not exceed the standard. The health risk assessment results show that Cd and As are the main noncarcinogenic contributing factors and that heavy metal pollutants in rice have a higher noncarcinogenic risk for children than adults. Cd, Cr, and As in rice in the study area have a certain carcinogenic risk to adults and children. Cd has the highest carcinogenic risk, followed by As and Cr.

**Supplementary Information** The online version contains supplementary material available at <https://doi.org/10.1007/s11356-021-15213-6>.

**Author contributions** Ting Huang wrote the paper, analyzed the data, and created the figures. Xianglian Wang conceptualized the study, analyzed the data, and revised the paper. Shan Huang designed the experiments. Yuxia Deng and Xiaolong Zhang collected samples and performed the experiments. Daishe Wu organized resources. All the authors read and contributed to the submitted version of the manuscript.

**Funding** This work was financially supported by the Science and Technology Project of the Jiangxi Provincial Department of Education (GJJ190976), the Natural Science Foundation of Jiangxi Province (2018BAB213017), and the National Natural Science Foundation of China (41402312).

**Data Availability** All relevant data are within the manuscript and available from the corresponding author upon request.

## Declarations

**Consent to participate** All authors participated in this work.

**Consent for publication** All authors agree to publish.

**Conflict of interest** The authors declare no conflict of interest.

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