



# Source and risk assessment of heavy metals in mining-affected areas in Jiangxi Province, China, based on Monte Carlo simulation

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Received: 25 March 2023 / Accepted: 15 February 2024 / Published online: 23 February 2024  
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## Abstract

In recent years, heavy metal contamination of soils has become a major concern in China due to the potential risks involved. To assess environmental pollution and human health risks in a typical heavy metal polluted site in Jiangxi Province, a thorough evaluation of the distribution, pollution levels, and sources of heavy metals in soils of the Yangmeijiang River watershed was conducted in this study. Positive matrix factorization and Monte Carlo simulation were used to evaluate the ecological and human health risks of heavy metals. The research findings indicate that heavy metal pollution was the most severe at the depth of 20–40 cm in soils, with local heavy metal pollution resulting from mining and sewage irrigation. The high-risk area accounted for 91.11% of the total area. However, the pollution level decreased with time due to sampling effects, rainfall, and control measures. Leaf-vegetables and rice were primarily polluted by Cd and Pb. The main four sources of heavy metals in soils were traffic emission, metal smelting, agricultural activities and natural sources, mining extraction, and electroplating industries. Heavy metals with the highest ecological risk and health risk are Cd and As, respectively. The non-carcinogenic and carcinogenic risks of children were 7.0 and 1.7 times higher than those of adults, respectively. Therefore, children are more likely to be influenced by heavy metals compared to adults. The results obtained by the risk assessments may contribute to the identification of specific sources of heavy metals (e.g., traffic emissions, metal smelting, mining excavation, and electroplating industries). Additionally, the environmental impacts and biotoxicity associated with various heavy metals (e.g., Cd and As) can also be reflected. These outcomes may serve as a scientific basis for the pollution monitoring and remediation in the mining-affected areas.

**Keywords** Soil · Heavy metals · Pollution · Risk assessment · Positive matrix factorization · Monte Carlo simulation

## Introduction

Land is a crucial resource for human production and livelihood, and with advancements in agriculture and industrial technology, soil heavy metal pollution has become an increasingly significant issue (Hu et al. 2020). Soil heavy metal pollution is characterized by its wide extent of contamination, long duration, poor degradation, and strong concealment, causing serious harm to soil quality, crop yield, groundwater quality, and human health (Dong et al. 2017). Uncontrolled and unreasonable mining activities are the primary cause of soil heavy metal pollution in developing countries, leading to the encroachment of vast amounts of land resources and waste production, including dust and acidic wastewater, which increase soil heavy metal pollution (Xiao et al. 2017). Mining activities, particularly in industrial and agricultural areas, have been well-documented as

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Responsible Editor: Philippe Garrigues

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one of the primary sources of heavy metals in soil (Long et al. 2021).

Source analysis is an essential part of heavy metal pollution management, providing targeted advice (Dong et al. 2021). Previous studies conducted the source analysis of heavy metals through the sampling methods or multivariate statistical analyses (Liu et al. 2020; Qadeer et al. 2020). However, the sampling methods require large amounts of sample data and the accurate numerical models (Xu et al. 2022; Yang et al. 2023). This can be time-consuming and costly. Multivariate statistical analyses, including cluster analysis, principal component analysis, and positive matrix factorization (PMF), are frequently used to identify the sources of heavy metals (Ogundele et al. 2016). Among these methods, PMF is widely employed in source analysis due to its simplicity and robustness in dealing with missing or inaccurate data (Men et al. 2020; Sun et al. 2019). Unlike other methods, PMF uses least squares to identify the major source of pollution and their contributions, eliminating the need for source profiles (Demirsöz et al. 2022). For example, Huang et al. (2021) used PMF to identify that the heavy metals in the soil of a Shanghai urban park came from natural sources, agricultural activities, industrial production, and traffic emissions. Similarly, Zhao et al. (2022) used PMF to determine that agricultural activities were the main source of heavy metals in agricultural soils.

Soil heavy metal contamination has become a growing concern due to its potential impact on environmental pollution levels and human health (Fei et al. 2017; Wang et al. 2012). To evaluate heavy metal pollution, various indexes such as the single pollution index, Nemerow pollution index, geoaccumulation index, enrichment index, and potential ecological risk index are commonly used (Hakanson 1980; Yang et al. 2018). Non-carcinogenic and carcinogenic health risk indexes are also utilized to assess the effects of heavy metal pollution on human health (MohseniBandpi et al. 2018; Yang et al. 2023). Monte Carlo simulation is frequently employed in health risk assessments as it takes into account the uncertainty of heavy metal concentration and exposure parameters (Saha et al. 2017). For example, Jaafarzadeh et al. (2023) used the 95% percentile of the results from the Monte Carlo simulation to determine the health risk of heavy metals and found no significant non-carcinogenic health risks to humans. Yu et al. (2022) determined that the probability of a child's carcinogenic risk index exceeding the limited value was 30.75% based on Monte Carlo simulation and positive matrix factorization (PMF).

The other factors (e.g., the depth and spatial distribution of heavy metals and ecological risks in soils and plants) should also be taken into consideration in the heavy metal pollution management (Li et al. 2021; Liu et al. 2016; Wang et al. 2019). The previous studies of risk assessments mainly focused on determining the pollution sources or the

population groups that are most likely to be affected. For example, Wu et al. (2021) evaluated the health risks of soil heavy metals in an industrial city after the development of environmental protection strategies. Inaccurate conclusions about treatment effects may arise due to the absence of source analysis and ecological assessments. In consequence, a comprehensive analysis of heavy metal pollution in mining areas is required.

Chongyi County in Jiangxi Province is one of the major mining areas that has caused serious soil, water, and health impacts from heavy metal pollution. The objectives of this study in this area are to (1) provide ecological risk assessments and health risk assessments based on soil samples collected from the Yangmei River watershed; (2) examine the relationship between the spatial distribution of industrial, mining, and agricultural areas in the Yangmei River watershed and heavy metal pollution; (3) identify the sources of heavy metals in soil using PMF; and (4) determine the heavy metal that poses the highest health risk to humans.

## Materials and methods

### Study area

The Yangmei River Watershed, located in Chongyi County, southeastern China, covers an area of 281 km<sup>2</sup> and spans from 25°33'N to 25°45'N and 114°18'E to 114°38'E. The river stretches 57 km from west to east and receives an average annual rainfall of 1510 mm. The region experiences a dry period from October to February and a wet period from April to June. The middle and eastern portions of the watershed are rich in minerals such as tungsten, coal, rare earths, lead–zinc, and copper–zinc, with 13 key mining industries located in the upstream area (Fig. 1). Seven of these industries have been closed since 2016 (Fig. 1).

### Sampling and analysis

#### Soil sample analysis

In this study, 37 sampling points were selected along the mainstream of the river based on the field survey with consideration of the distribution of farmland and mining areas (Fig. 1). Soil samples were generally collected at depths of 0–20 or 20–40 cm, with the deepest sampling depth not exceeding 50 cm (Huang et al. 2018). Thus, soil samples were collected within 50 m of the riverbanks at depths of 0–20, 20–40, and 40–60 cm in this study. The samples collected in September 2018, December 2018, March 2019, and May 2019 were used to investigate the local soil pollution by heavy metals.



Fig. 1 Map of sampling sites and main mine factories in the Yangmei River

## Plant sample analysis

56 leaf-vegetable samples and 15 rice samples were collected from the farmland around the sampling points. The edible parts of leaf-vegetables and rice were washed and dried in an oven at 150 °C for half an hour. Then they were dried at 60 °C for 72 h until a constant weight was obtained. The leaf-vegetables and rice were ground pass through a 1.0-mm sieve after the drying process. The collected soil samples were air-dried, ground and passed through a 100-mesh nylon sieve. These soil samples were moistened with deionized water in the digestion tank. Crop samples were digested with a mixture of HNO<sub>3</sub>-H<sub>2</sub>O<sub>2</sub> in a closed PTFE (poly-tetrafluoroethylene) system at 120 °C for 10 h, and then they were analyzed together with soil samples. The soil samples were digested by adding a mixture of HNO<sub>3</sub>-HF in an acid-proof ventilated area. After microwave-assisted acid digestion, the concentration of the samples was determined by inductively coupled plasma mass spectrometry (Plasma MS). The control experiments used the same pretreatment method. Each sample was repeated 3 times.

## Spatial distribution analysis

Table S1 shows the test results of the normality of data. Inverse distance weighting (IDW) is adopted to investigate the spatial distribution of heavy metals as the heavy metals did not obey the normal distribution. Additionally, Spearman's correlation analysis was used to investigate patterns of similarity between the distribution of heavy metals and to determine the correlation among different heavy metals. The distributions of the original variables were not required as it is a non-parametric test (Wang et al. 2019).

## Ecological risk assessment

### Single pollution index and Nemerow pollution index

Two risk assessment indexes, namely, the single pollution index and the Nemerow pollution index, were used to evaluate the pollution level of heavy metals in the study area. The single pollution index is a widely adopted international index for assessing soil heavy metal pollution, which can be used to identify the primary sources of pollution (Gao et al. 2020). The Nemerow pollution index is usually used to investigate polymetallic pollution. These two indexes can be given by (Zhang et al. 2021).

$$P_i = C_i / B_i \quad (1)$$

$$P_n = \sqrt{(P_i^2 + P_{i_{max}}^2) / 2} \quad (2)$$

where  $P_i$  is the single pollution index of heavy metal  $i$ ;  $C_i$  is the measured concentration of heavy metal  $i$  ( $\text{mg} \cdot \text{kg}^{-1}$ ) and  $B_i$  is the background value (BV) of heavy metal  $i$ . The BVs are the heavy metal concentrations of the natural compound in soils, also known as the natural geographic concentrations (Duman et al. 2022). Additionally, the heavy metal BVs only depend on the soil substrates and forming processes, which are not related to human activities.  $P_n$  is the Nemerow pollution index;  $P_i$  is the average value of the single pollution index of each pollutant in soil, and  $P_{i_{max}}$  is the maximum value of the single pollution index. The classifications of contamination levels for  $P_i$  and  $P_n$  were referred to the study of Zhang et al. (2021).

There are two ways to estimate BV: geochemical method and statistical method (Santos-Francés et al. 2017). He et al. (2006) followed the geochemical method and collected over 700 soils samples. The effects of the natural factors such as the parent material, topography, climate, and hydrological conditions on BVs of 12 different heavy metals are highlighted. The BVs for different regions of the Jiangxi Province were evaluated. It should be noted that BVs is assumed to be the fixed values in this study as BVs do not change much within a county (e.g., < 10%). BVs of the main heavy metal pollutants Pb, Cr, Cd, As, Hg, Cu, Zn, and Ni in Chongyi County are 32.10, 48.00, 0.10, 10.40, 0.08, 20.80, 69.00, and 19.00 ( $\text{mg} \cdot \text{kg}^{-1}$ ), respectively.

### Geoaccumulation index

The geoaccumulation index  $I_{geo}$  was mainly used to study the damage to the water environment caused by pollutants in bottom sediments, which was later used in the evaluation of soil heavy metal pollution (Doabi et al. 2018). The effects of geological changes and human activities on heavy metal pollution in soils can be reflected by the value of  $I_{geo}$ . Additionally,  $I_{geo}$  is defined as follows (Muller 1969):

$$I_{geo} = \log_2 [C_i / (k \times B_i)] \quad (3)$$

where  $k$  is the correction coefficient that reflects the lithogenic effects. It is reported that the natural fluctuations in the concentrations of specific substances in the environment and anthropogenic impacts can be better reflected when  $k$  is equal to 1.5 (Duman et al. 2022; Santos et al. 2020). In addition, the classifications of the geoaccumulation index was based on the study of Doabi et al. (2018).

### Potential ecological risk index

The potential ecological risk index ( $RI$ ) is proposed to evaluate the pollution of soil heavy metals and their ecological hazards based on the environmental characteristics

(Hakanson 1980). The calculation equation of  $RI$  is given as follows (Kumar et al. 2021):

$$RI = \sum E_i = \sum T_i \times C_i / B_i \quad (4)$$

where  $E_i$  is the single potential ecological risk index for heavy metal  $i$  and  $T_i$  is the toxic response coefficient. The values of  $T_i$  were determined based on the study of Hakanson (1980). The classifications of levels for  $RI$  and  $E_i$  were referred to Kumar et al. (2021)

## Health risk assessment

The U.S. Environmental Protection Agency (EPA) model was used to investigate the health risks of soil heavy metal pollution. The calculation formulas of the three exposure pathways are as follows (USEPA 1997):

$$ADD_{ing} = (C_i \times IngR \times EF \times ED) / (BW \times AT) \times 10^{-6} \quad (5)$$

$$ADD_{inh} = (C_i \times InhR \times EF \times ED) / (PET \times BW \times AT) \quad (6)$$

$$ADD_{derm} = (C_i \times SA \times SL \times ABS \times EF \times ED) / (BW \times AT) \times 10^{-6} \quad (7)$$

where  $ADD_{ing}$ ,  $ADD_{inh}$ , and  $ADD_{derm}$  are the daily exposure dose to heavy metals via ingestion, inhalation and dermal contact, respectively ( $\text{mg} \cdot \text{kg}^{-1} \cdot \text{day}^{-1}$ ). Other parameters can be found in the relevant standards (USEPA 2011, USEPA 1997; USEPA 2009).

$$HI = \sum_{i=1}^n HQ_i = \sum_{i=1}^n (ADD_{ing} + ADD_{inh} + ADD_{derm}) / RfD_i \quad (8)$$

$$TCR = \sum_{i=1}^n CR_i = \sum_{i=1}^n (ADD_{ing} + ADD_{inh} + ADD_{derm}) \times SF_i \quad (9)$$

where  $HQ_i$  and  $CR_i$  are the non-carcinogenic and carcinogenic health risk indexes of heavy metal  $i$ , respectively,  $HI$  and  $TCR$  are the sum of  $HQ_i$  and  $CR_i$ , respectively, and  $RfD_i$  and  $SF_i$  are the chronic reference dose of toxicant and the carcinogenic slope factor, respectively.

According to the different classifications of chemical substances stipulated by the US EPA, the evaluation standards are different. For non-carcinogenic risk, if the values of  $HQ$  (of  $HI$ )  $> 1$ , it suggests a potential adverse health effect. As for carcinogenic risk, humans will be posed the risk of cancer when the values of  $CR$  (or  $TCR$ )  $> 10^{-4}$ . In contrast, a value of  $CR$  (or  $TCR$ ) below the acceptable threshold of  $10^{-6}$  means that heavy metals pose a negligible risk to humans (MohseniBandpi et al. 2018). Additionally, due to different living habits and body structures, different exposure

parameters were used in health risk assessments for adults and children, respectively (Fei et al. 2017).

## Source analysis

Positive matrix factorization (PMF) has been widely used to identify the sources of heavy metal pollution. The concentration of the sample can be decomposed by the following function based on PMF (Lv 2019):

$$C_i = \sum_{j=1}^n g_j f_{j,i} + e_i \quad (10)$$

where  $g_j$  is the contribution of source  $j$ ,  $n$  is the number of sources,  $f_{j,i}$  is the amount of heavy metal  $i$  produced by source  $j$ , and  $e_i$  is the residual error.

The object function  $Q$  is assumed to be the sum of the residual errors of all samples, and it can be calculated by the following function (Lv 2019):

$$Q = \sum_{i=1}^n \sum_{s=1}^m (e_{s,i} / u_{s,i}) \quad (11)$$

where  $e_{s,i}$  and  $u_{s,i}$  are the residual error and uncertainty of heavy metal  $i$  in sample  $s$ , respectively.

When the concentration of heavy metal is below or equal to the corresponding method detection limit ( $MDL$ ), the uncertainty is given by

$$Unc = \frac{5}{6} \times MDL \quad (12)$$

Otherwise, it can be given by

$$Unc = \sqrt{(\sigma \times c)^2 + MDL^2} \quad (13)$$

where  $\sigma$  is the relative standard deviation and  $c$  is the concentration of heavy metal.

The USEPA PMF 5.0 software was used for source analysis.

## Probability risk analysis

Monte Carlo simulation is an approach to characterize uncertainty and variability in health risk assessment through statistical sampling techniques (Kroese et al. 2014). The risk calculation was repeated 10,000 times for all the different random values entered using the Crystal Ball software in EXCEL (version 11.1). The distributions and values of Monte Carlo parameters (except for heavy metal concentrations) used in the health risk assessment can refer to Saha et al. (2017). 95th percentiles of  $HI$  and  $TCR$  were used as a reference point for human health risk, with  $HI$  and  $TCR$  allowed to have the values of 1.0 and  $10^{-6}$  as the final

judgment on the health risk of the heavy metals (Jaafarzadeh et al. 2023).

## Results and discussion

### Descriptive statistics of heavy metals in soils

To understand the basic situation of local heavy metal pollution, descriptive statistical analysis was conducted on the soil heavy metal concentrations determined at different times. According to Table 1, the mean concentrations of all heavy metals were higher than BVs. In other words, the study area had been affected by heavy metals. Additionally, the maximum values of all heavy metals were observed at the depths of 20–40 cm. It should be noted that the coefficients of variation for all heavy metals at the depth of 20–40 cm were larger than those at other depths. According to Liu et al. (2016), the coefficients of variation reflect the influence of human activities on the distribution of heavy metals and an increase in coefficients of variation indicates increased disturbance from humans. Therefore, it can be demonstrated that the heavy metals in soil at the depth of 20–40 cm were most likely to be influenced by human activities (Liu et al. 2016). However, it should be noted that the ecosystem and human beings are more likely to be influenced by the soil at the depth of 0–20 cm (Long et al. 2021). Additionally, it has been proved that people are mainly exposed to surface soil (Jiang et al. 2017). The ecological and health risks of heavy metals in soil were assessed based on the surface soil samples (Xiao et al. 2017). For example, Wu et al. (2020) assessed the health risks of agricultural soil in the industrial city based on the 28 soil samples collected at the depth of 0–20 cm. Thus, all of the assessments in this paper are based

on the heavy metal concentrations of the soil samples at depths of 0–20 cm.

As is shown in Table S2, the mean concentrations of Pb, Cr, Cd, As, Hg, Cu, Zn, and Ni in the first sampling were 86.96, 90.36, 4.66, 85.45, 0.071, 156.74, 282.05, and 36.71 (mg • kg<sup>-1</sup>). All of them except Hg were larger than BVs. The concentrations of Pb, Cr, Cd, As, Cu, Zn, and Ni were 2.71, 1.88, 46.60, 8.22, 7.54, 4.09, and 1.93 times bigger than that of their BVs, respectively. Only the concentration of Hg was 0.89 lower than BV. Compared to the first sampling, the mean concentrations of all the soil heavy metal concentrations in the second sampling except Cd and Hg were decreased by 2.39% to 38.06%. The Hg concentration remained relatively stable, while the Cd concentration exhibited a slight increase. It is noted that the concentrations of all heavy metals are larger than that of the background values. The mean concentrations of Cr, Cd, As, Cu, Zn, and Ni exceeded BVs in the third sampling campaign. The mean concentrations of Cd, As and Cu exceeded the soil BVs in the fourth sampling campaign.

It was noted that the mean concentrations of heavy metals decreased with time. For example, the mean concentration of Cd and As decreased from 4.66 and 85.45 (mg/kg) to 0.97 and 54.82 (mg/kg). It is noted that only a few representative points (e.g., near the source or town) were resampled during the third and fourth sampling events. Comparisons of, with the average heavy metal concentrations decreased by 10–20% when comparing the data from the same sampling points in the first and fourth samplings. This decreasing trend may be attributed to the control of wastewater discharge process and the amount of pesticide used on the farmland by the local government. For example, the emission of heavy metals in Chongyi County was decreased from 1568.84 kg in 2018 to 1470.18 kg in 2019. It is noted that the

**Table 1** Profiles of heavy metals at different depths in the study area (mg/kg)

Depth	Statistic	Cu	Pb	Zn	Cd	Ni	Cr	Hg	As
0–20 cm	Minimum	38.51	26.26	82.04	0.62	12.96	38.03	0.004	17.86
	Maximum	462.73	224.82	1029.29	21.62	81.7	162.04	0.203	405.87
	Mean	156.74	86.96	282.05	4.66	36.71	90.36	0.071	85.45
	SD	124.87	40.96	206.24	3.85	15.53	33.04	0.056	69.19
	CV (%)	79.67	47.11	73.12	82.62	42.3	36.57	78.15	80.98
20–40 cm	Minimum	25.6	18.88	55.74	0.63	16.15	45.81	0.002	17.91
	Maximum	701.65	401.19	1301.4	22.4	131.25	529.99	0.695	388.69
	Mean	164.26	90.01	306.57	5.21	38.94	99.96	0.082	113.27
	SD	179.29	74.01	316.83	5.28	19.04	71.49	0.107	86.74
	CV(%)	109.15	82.23	103.35	101.27	48.9	71.52	130	76.58
40–60 cm	Minimum	26.46	21.81	54.61	0.39	20.18	56.98	0.004	26.66
	Maximum	358.26	245.94	529.1	7.33	75.89	168.01	0.302	159.45
	Mean	122.1	71.04	202.62	2.86	36.93	90.07	0.071	89.15
	SD	113.55	55.73	144.33	1.75	15.15	34.32	0.077	37.61
	CV(%)	93	78.45	71.23	61.1	41.02	38.11	107.64	42.19

physical transportation and chemical treatment of the slag carried out by the local government effectively reduced the adverse effects. In addition, controlling the use of pesticides and recycling packaging wastes could also contribute to the heavy metal management. Zhou et al. (2020) reported that the use of fertilizers and pesticides might be the main source of heavy metals in agricultural land.

The decrease of soil heavy metal concentrations might be induced by the variation of rainfall amount as well. Slag was one of the main sources of soil heavy metals, and rainfall could accelerate the transport of pollutants in slag (Villanueva et al. 2016). The first sampling was at the end of the wet period with relatively abundant rainfall. The fourth sampling was at the end of the dry season with relatively little rainfall. The rainfall of the fourth sample decreased successively, which might be attributed to the reduction of the heavy metal pollution of soil. Zhang et al. (2018) found that heavy rainfall and long-lasting rainfall can cause soil heavy metal pollution aggravation. However, compared to human activities, their study showed that the impacts of rainfall were relatively small.

### Spatial distribution characteristic analysis of heavy metals in soils

Figure 2 shows the spatial distribution maps of 8 heavy metals. The peak values of Cr and Ni were both located in the west side of the watershed, indicating similar distribution patterns. As, Cd, Cu, and Zn, on the other hand, had peak areas in the middle and west of the watershed, while Hg was concentrated in the south-middle and east. Pb was well dispersed throughout the studied areas, likely due to the high concentration of tungsten ore in Chongyi County, where Pb was found to be evenly distributed around the ore. Lei et al. (2022) noted that during the mineralization of tungsten ore, cations such as  $\text{Ca}^{2+}$ ,  $\text{Fe}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Pb}^{2+}$  could combine with  $\text{WO}_4^{2-}$ , leading to high concentrations of Pb around tungsten ore. This is likely one of the main reasons for the high background value (BV) of Pb observed in this study.

The correlation analysis results, presented in Fig. 3, showed high correlations between Cr and Ni, Cd and Cu, Cd and Zn, and Cu and Zn with correlation coefficients of 0.937, 0.837, 0.914, and 0.913, respectively. Moderate correlations were found between Pb and Cd, Pb and As, Pb and Cu, Pb and Zn, Cd and As, As and Cu, As and Zn, and Hg and Ni with correlation coefficients of 0.735, 0.609, 0.637, 0.711, 0.732, 0.757, 0.706, and 0.439. There were two main reasons for the correlation among the distribution of soil heavy metal content: (i) natural causes: Chongyi County was rich in mineral resources, and the distribution of minerals itself had some correlation. For example, the distribution of Cu and Zn, Pb and Zn itself was correlated as there were copper-zinc and lead-zinc mines in the area. The similar

correlation between Zn and Cu was reported by Jiang et al. (2021) who suggested that these two elements had a complex contamination relationship or homologous relationship with each other and might experience similar geochemical effects. (ii) Anthropogenic causes: it can be seen from Fig. 1 that the distribution of the industries was concentrated in the middle and west of the study area, which might contribute to the higher concentration of certain kinds of heavy metal pollution in the middle and west sides of the watershed. The human activities in the agricultural areas of the eastern and southern parts of the watershed led to the peak formation of heavy metals contained in the fertilizers into the soil. It was the main reason for the increase in the concentration of Hg in soils as the distribution of the peak concentration areas of Hg was the same as that of agricultural areas. In addition, the use of industrial wastewater for irrigation in agricultural areas had led to a significant increase in the concentration of heavy metals in the soil. The influence of land-use types on the distribution of heavy metals had also been proved by Dai et al. (2019). However, Yamaguchi et al. (2003) found that the main source of Hg was from the atmosphere rather than industrial and agricultural enterprises in the upper Thames, in contrast to the results of this paper. This was due to the small number of local polluting enterprises and limited use of fertilizers in their study areas. Dong et al. (2017) confirmed that sewage irrigation and the use of mercury-containing pesticides were the main sources of mercury contamination in soil.

### Ecological risk assessment

#### Ecological risk in soil

According to the results presented in Fig. 4 and Table S3, the degree of heavy metal pollution in the soil within the watershed was highest for Cd, followed by As, Cu, Zn, Pb, Ni, Cr, and Hg. The site was found to be seriously polluted by Cd and As, with pollution levels at stages III–IV. The mean pollution levels of the remaining heavy metals were at stages I–III, which exceeded the safety limit. These results indicate that there is still a certain level of pollution risk, and measures must be taken to reduce heavy metal pollution. Even for the fourth sampling, which had relatively low pollution, the average Nemerow pollution index was 7.08 (Fig. S1), which was much higher than the specified heavy pollution level of 3.00. The local soil heavy metal pollution was severe, with Cd and As having the highest  $P_i$  values and reaching the highest pollution levels due to their high concentrations in soils.

Fig. S2 shows that the  $I_{geo}$  results obtained by the same variation of  $P_i$  were similar. According to Fig. S1, the very high risk and high-risk areas accounted for more than 90% of the total area, indicating that the local potential ecological

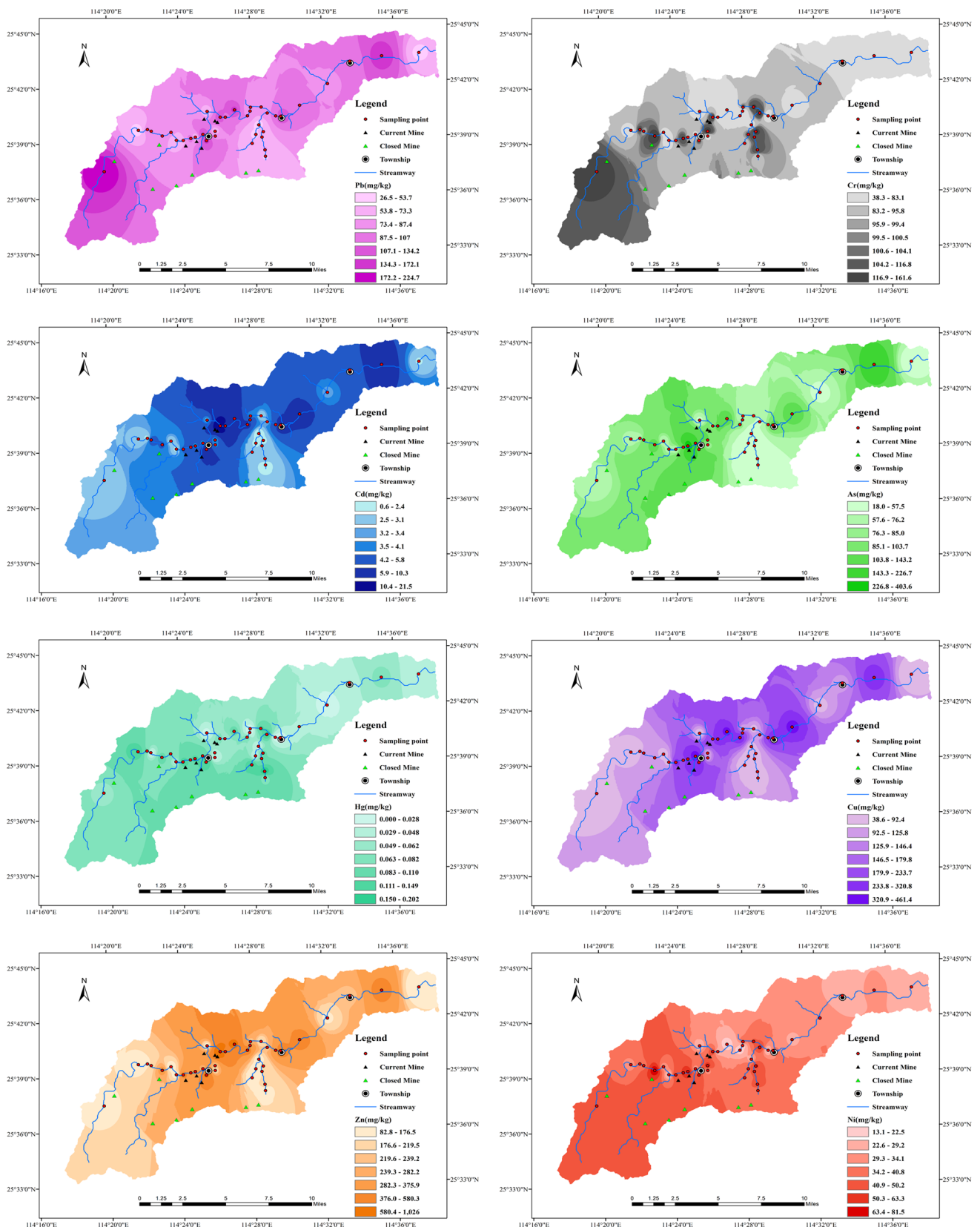
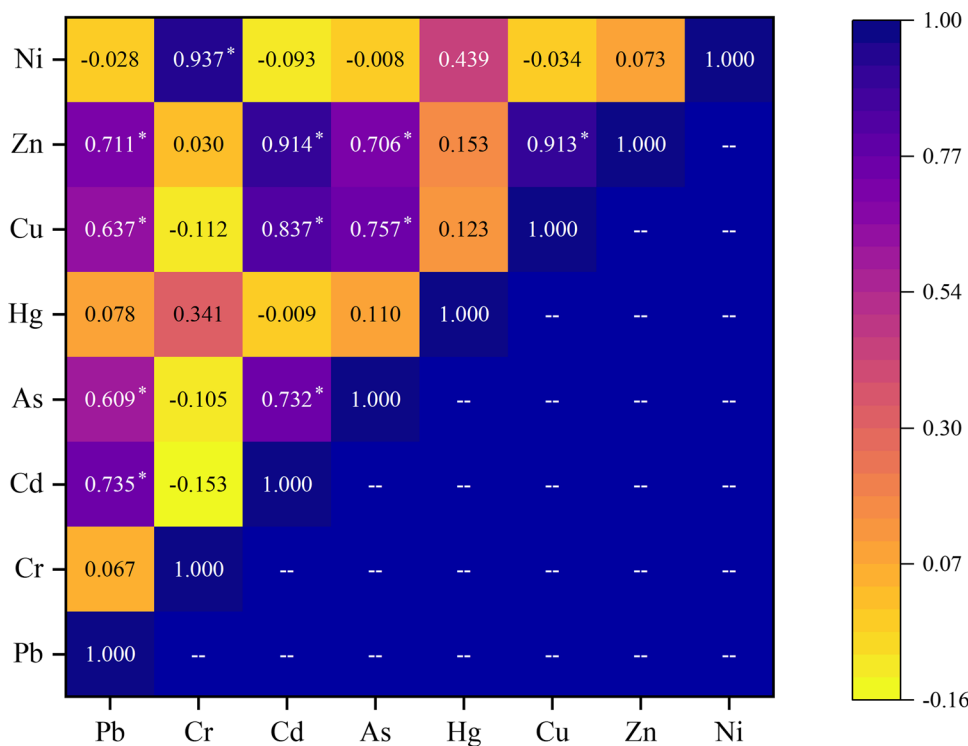


Fig. 2 Spatial distribution of soil heavy metal concentrations at the depth of 0–20 cm



**Fig. 3** The result of Spearman’s correlation analysis



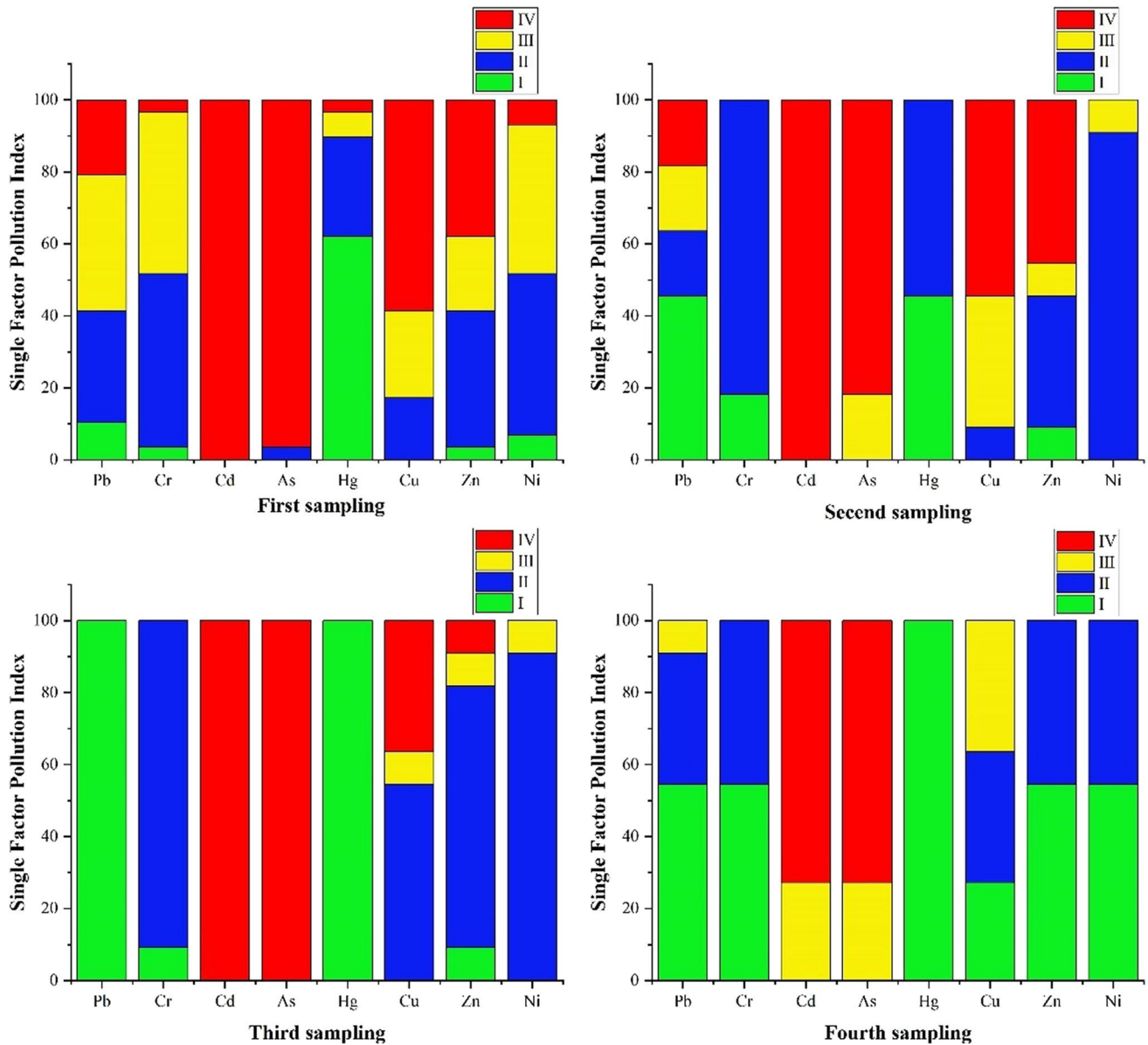
risks were very high, and the ecological pollution was severe. The peak areas of the ecological risk index were mainly distributed in the central, southwestern, and northeastern parts of the watershed, consistent with the peak areas of Cd (Fig. 2).  $E_i$  of Cd accounted for 87.54% of the overall  $RI$  (Table S4), indicating that the heavy metal with the highest ecological risk to the local area was Cd. The potential ecological risks of heavy metals were in the order of  $Cd > As > Hg > Cu > Pb > Ni > Zn > Cr$ . This can be explained by the fact that the mean concentration of Cd in soils was 46.60 times higher than BV. The high ecological risk of Cd might also be attributed to its large toxic response coefficient. It should be noted that similar phenomena have been reported in many industrial areas in China (Makokha et al. 2016; Rao et al. 2021). For instance, Rao et al. (2021) reported a serious ecological pollution caused by Cd in Houguan Lake, which was linked to the large discharge of pollutants from local industrial areas and the spraying of cadmium-containing pesticides during fish farming.

**Ecological risk in plant**

According to Fig. S3, the average concentrations of Pb, Cr, Cd, As, and Hg in leaf-vegetables were found to be 3.90, 3.89, 6.44, 2.31, and 1.73 times higher than their maximum allowable concentrations (MAC) in China (MEP 2014). Similarly, the mean concentrations of Pb, Cd, and As in rice were 3.60, 5.84, and 1.90 times higher than MAC, respectively. In contrast, the average concentrations of Cr

and Hg were only 0.89 and 0.21 times higher than MAC. While heavy metals are known to be easily and largely accumulated in rice due to their absorption capacity and metal bioavailability, we found that heavy metal pollution was more serious in leaf-vegetables. This difference could be due to the basic physical and chemical properties of upland and paddy that affect the chemical transformation and bioavailability of heavy metals. For instance, plants in paddy fields are more likely to be polluted by Cd and Pb under wet conditions (Ok et al. 2011). The higher foliar deposition of heavy metals is the main cause of more serious pollution in leaf-vegetables. Xu et al. (2022) found that foliar deposition was the primary way for plants to absorb heavy metals from nature, and leaf-vegetables with larger leaves were more prone to induce foliar deposition than rice. Additionally, Tu et al. (2020) reported that leaf-vegetables were more likely to absorb heavy metals from the soil and air due to their larger leaves, faster growth rates, and transpiration. It should be noted that the anthropogenic activities (e.g., the mining excavation and traffic emissions) can significantly influence the foliar deposition (Adhikari et al. 2022).

The concentrations of Cd and Pb in both leaf-vegetables and rice were found to be much higher than MAC, which is consistent with the findings of Li et al. (2021) that plants are more likely to be polluted by Cd and Pb. Typically, the concentration of Pb accumulated in plants is low due to the chemisorption process in the soil (Chen et al. 2008). However, our study showed that the mean concentrations of Pb in leaf-vegetables and rice were 3.89 and 3.60 times higher



**Fig. 4** The proportion of single pollution indices in each degree for the four sampling times

than MAC. This could be attributed to the high mean concentration of Pb observed in the first sampling, which was 86.96 times larger than the background value. Cd, on the other hand, is easily absorbed by plant roots due to its highly transportable capacities (Tu et al. 2020), and Cd in the air produced by mining is also an important source for plants to absorb by foliar deposition.

#### Comparison of ecological risks in soil and plant

The average ecological risks for the five heavy metals in plants were as follows: Cd > Pb > Cr > As > Hg, based on the average pollution of all the plants. The ecological

risks for the heavy metals in soil were in the order of Cd > As > Pb > Cr > Hg. As described in the previous section, Cd and Pb had high ecological risks in plants due to their high accumulation and transportable capacities. In contrast, Cr and As were weakly translocated in soil according to Rahman et al. (2021), with Cr being more translocated than As. This resulted in a higher ecological risk of As in soil compared to Cr, while the ecological risk of As in plants was lower than Cr. Mercury was found to accumulate mostly on the soil surface and is easily taken up by plants, as it is adsorbed by clay minerals and organic matter in the soil, according to Pei et al. (2022). However, the ecological risk of Hg in plants was

not high, possibly because of the low concentration of Hg in the soil.

### Heavy metal source analysis

Concentration data from 78 soil samples, including 8 heavy metals, were used to identify the possible sources of heavy metals in soils based on PMF, along with uncertainty data. It should be noted that the numbers of soil samples collected in September 2018, December 2018, March 2019, and May 2019 are 41, 19, 11, and 11, respectively. The value of the rotation parameter was chosen to be 0.1 to improve the slope edge. Five factors were identified, and their contributions are presented in Fig. 5.

The first factor (F1) consisted predominantly of Pb (58.34%) and Ni (18.63%) and explained 20.3% of the total variances. Previous studies have shown that lead in soil can be attributed to traffic emissions (Huang et al. 2021), and the large amount of Ni in F1 can also be attributed to traffic emissions since it is reported that the main source of Ni was the gas emissions produced by cars (Jiang et al. 2017). Therefore, F1 can be defined as traffic emissions.

The second factor (F2) was predominated by Ni (61.52%) and Cr (59.20%) and explained 17.2% of the total variances. Numerous studies have proven that Ni and Cr contaminations are elements that occur in metal smelting wastes (Moreira et al. 2020). For example, Ni is mainly used in the manufacture of stainless steel and other anticorrosive alloys

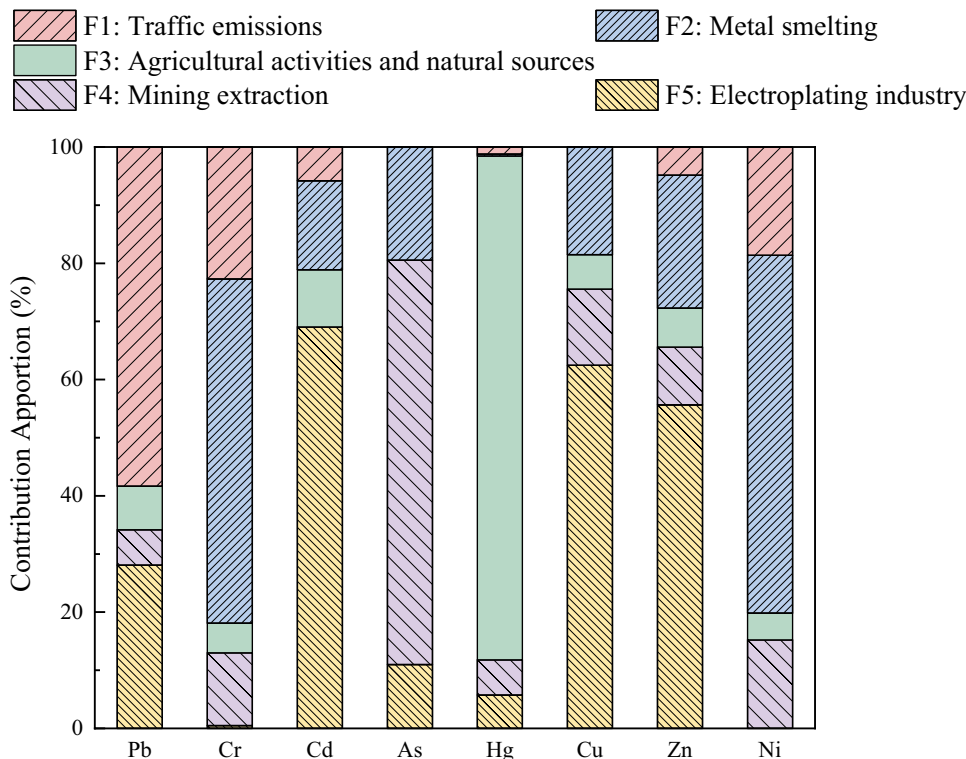
(Demirsöz et al. 2022). Cr may originate from the metallurgical industry (Dong et al. 2021). The concentrations of Ni and Cr in the soil were 1.93 and 1.88 times larger than BV, respectively, indicating that the Ni and Cr in the soil were affected by human activities. This is also illustrated by the high value of CV. Thus, F2 can be defined as metal smelting.

The third factor (F3) was predominated by Hg (86.66%), explaining 18.2% of the total variances. It was reported that the main unnatural source of Hg in the soil was the consumption of pesticides and fertilizers containing mercury (Cheng et al. 2020). Therefore, the high CV value of Hg induced by human activities could be specified by agricultural activities. However, the average concentration of Hg in the soil was 0.89 times lower than BV, indicating the influence of natural factors. As a result, F3 can be defined as the combined effects of agricultural activities and natural sources.

The fourth factor (F4) was predominated by As (69.54%) and explained 24.8% of the total variances. As mainly comes from the sloppy and irregular production processes of some mining and metallurgical enterprises (Alloway 2013). Although there were also studies indicating that As is likely to come from the parent rock material, As in the study area highly exceeds BV (Liu et al. 2020). Therefore, F4 can be defined as mining extraction.

The fifth factor (F5) was predominated by Cd (69.06%), Cu (62.45%), Zn (55.65%), and Pb (28.08%) and explained 19.5% of the total variances. Previous studies have shown

Fig. 5 Contribution of sources to each heavy metal



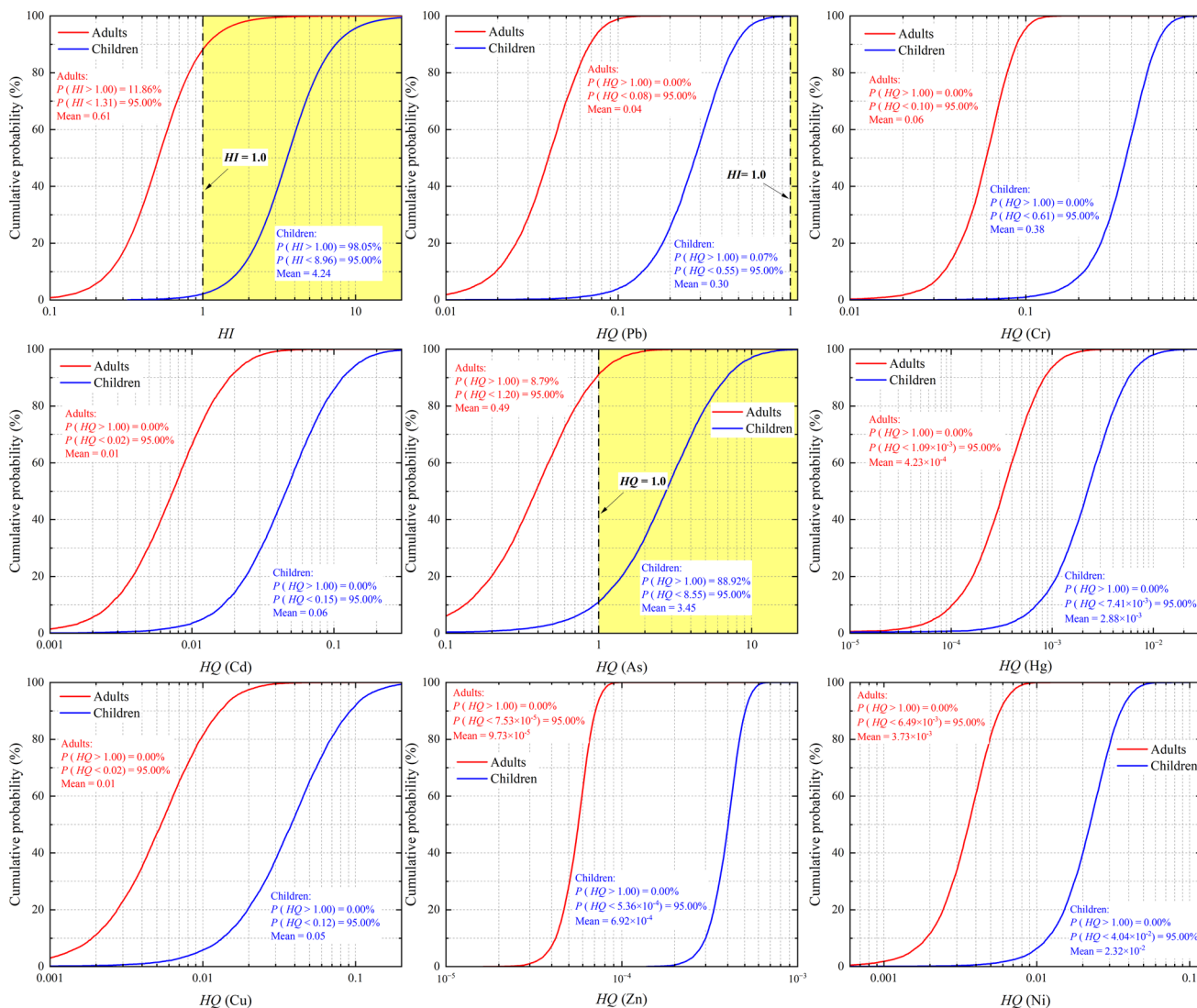
that the typical pollutants of electroplating factories are Cd and Cu, and some Zn and Pb are also produced (Martinková et al. 2016). There were several electroplating factories distributed in the Yangmei River watershed. Therefore, we can identify the source of Factor 5 as the electroplating industry (Fig. 5).

### Health risk assessment

#### Non-carcinogenic health risk assessment

A Monte Carlo simulation was used to estimate the probabilities of carcinogenic and non-carcinogenic risks associated with exposure to heavy metals. The resulting probability distributions of health quotient (*HQ*) and hazard index (*HI*) for both adults and children were presented in Fig. 6.

In addition, Table S5 and Table S6 showed the health risk indexes of heavy metals in soil samples without using Monte Carlo simulation. The heavy metals were ranked in order of  $As > Cr > Pb > Cd > Cu > Ni > Zn > Hg$ , with As being the most hazardous metal. The *HQ* values of all heavy metals, except for As, were below 1.0. Only 8.79% of simulation results for As were greater than 1.0 for adults, while for children, 88.92% and 0.07% of simulation results for As and Pb, respectively, were greater than 1.0. These results suggest that As is the heavy metal that should be prioritized for treatment and control. According to the study of He et al. (2018), Cr posed the highest risk to human health in Jiangxi Province. The main reason for this difference is that there are a large number of heavy metal smelters in the area where He et al. (2018) collected samples. This can contribute to the high concentrations of Cr in their samples. However, the mining



**Fig. 6** Probability distribution of total non-carcinogenic risk (*HI*) and non-carcinogenic risk (*HQ*) index (the yellow region means that the risk is out of the acceptable range)

extraction is the main industrial activity in Chongyi County, resulting in the higher As concentrations in soils. This can also be concluded from the “Heavy metal source analysis” section that the F4 (mining extraction) has a more significant influence on the heavy metal concentration in soil than the F2 (metal smelting). This indicates that the different industrial activities can lead to the different types of pollution. The impacts of the industrial activities should be subdivided.

The non-carcinogenic risk of heavy metals in soils was assessed based on the HI values. For adults and children, 11.86% and 98.05% of simulation results were greater than 1.0 for HI, respectively. The mean and 95th percentile levels of HI were 0.61 and 1.31 for adults and 4.24 and 8.96 for children, respectively. This indicates that heavy metals in soils pose a non-carcinogenic health risk to humans. Moreover, the HI value for children was 6.85 times higher than that for adults, indicating that children are more susceptible to the harmful effects of exposure to heavy metal pollution due to their physical habits and lifestyle. The results also show that ingestion is the primary route of exposure for non-carcinogenic risks, with  $HQ_{ingest} > HQ_{inhale} > HQ_{dermal}$  for both adults and children.

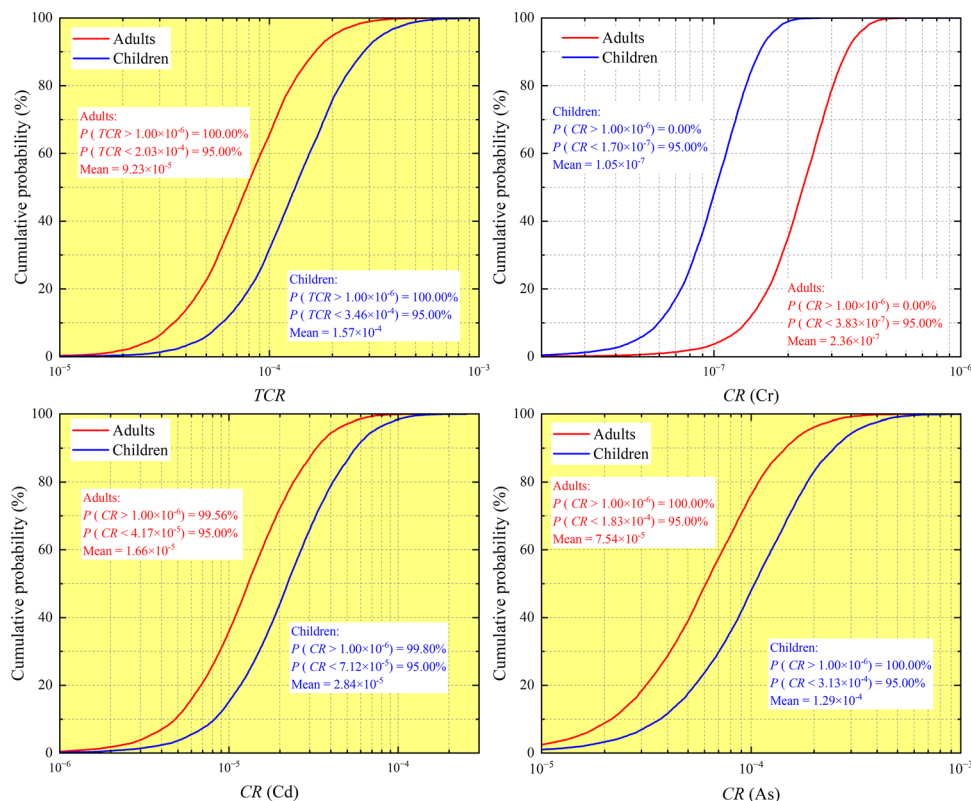
### Carcinogenic health risk assessment

The probability distributions of carcinogenic risk (CR) and total carcinogenic risk (TCR) for both adults and children are

presented in Fig. 7. Heavy metal concentrations were found to follow the order of  $As > Cd > Cr$ , with As being the main contributor to the carcinogenic risk to human health. The mean CR values of As for adults and children were 75.42 and 129.21 times larger than the limited value, respectively, indicating a significant risk. These findings are consistent with those of Cui et al. (2020). For Cd, 99.56% and 99.80% of simulation results were above the limited value for adults and children, respectively. The mean and 95th percentile levels of Cd for adults and children were also above the limited value. According to the study of Teng et al. (2015), Cd always poses the insignificant carcinogenic health risk to human due to its low carcinogenic slope factor. However, it is noted that the concentration of Cd in the study area was 46.60 times higher than the background value (BV). The extremely high concentration can result in the potential risk to human health.

For adults and children, all of the simulation results are larger than  $1.0 \times 10^{-6}$  for TCR, and respective mean and 95th percentile levels of TCR were  $9.23 \times 10^{-5}$  and  $2.03 \times 10^{-4}$  for adults and  $1.57 \times 10^{-4}$  and  $3.46 \times 10^{-4}$  for children. This indicates that the heavy metals in soils posed very high carcinogenic health risks to humans. The TCR value for children was 1.70 times higher than that for adults and the carcinogenic risk of adults and children for different exposure pathways are in the following order:  $CR_{ingest} > CR_{inhale} > CR_{dermal}$ . The same conclusion was also found by Wu et al. (2021).

**Fig. 7** Probability distribution of total carcinogenic risk (TCR) and carcinogenic risk (CR) index



## Conclusions

In conclusion, a systematic evaluation of soil pollution in the Yangmei River watershed of Jiangxi Province was conducted using soil samples. The distribution and correlation of heavy metals were analyzed based on IDW, and source analysis was conducted based on PMF. Additionally, ecological risk assessment and health risk assessment based on Monte Carlo simulation were conducted to provide scientific guidance for soil heavy metal pollution prevention and control and ecological environmental protection.

1. The concentrations of Pb, Cr, Cd, As, Hg, Cu, Zn, and Ni in soils along the Yangmeijiang River were found to be 2.71, 1.88, 46.60, 8.22, 0.89, 7.54, 4.09, and 1.93 times higher than background values in the first sampling. The degree of soil heavy metal pollution within the watershed was in the order of  $Cd > As > Cu > Zn > Pb > Ni > Cr > Hg$ . This was mainly due to the large number of tungsten-tin mines in Chongyi, which would produce a large amount of Cd and As during mining. Additionally, the maximum values of all heavy metals were observed at the depths of 20–40 cm.
2. Spearman's correlation analysis indicated that Cr and Ni, Cd and Cu, Cd and Zn, and Cu and Zn were highly correlated. Cr and Ni in soils were influenced by common minerals and therefore the spatial structure and distribution of the two were similar. There were copper-zinc and lead-zinc mines in the area, so the distributions of Cd, Cu, and Zn were similar. Also, the distribution of industries was concentrated in the middle and west of the study area, which may be attributed to the higher concentration of certain kinds of heavy metals.
3. The high ecological risk area accounted for 91.11% of the total area, and Cd was found to be the heavy metal that posed the highest ecological risk to the soils in the watershed. The potential ecological risk parameter of Cd accounted for 87.54% of the overall *RI*. The main reason for this was that the concentration of Cd was 46.60 times higher than the background value. The larger toxic response coefficient of Cd may also contribute to the high potential ecological risk of Cd.
4. The mean concentrations of Cd and Pb in leafy vegetables were 3.90 and 6.44 times higher than the maximum allowable concentration (MAC), while those in rice were 3.60 and 5.84 times higher than MAC. Leafy vegetables were likely to be more polluted due to their larger leaves, faster growth rates, and transpiration. The high concentration of Pb in plants was attributed to the large mean concentrations of Pb observed in soils, while the high concentration of Cd was induced by its high transportable capacity.

5. Mining extraction, traffic emissions, electroplating industry, agricultural activities and natural sources, and metal smelting were identified as the main sources of soil heavy metals in the study area, accounting for 24.76%, 20.25%, 19.58%, 18.23%, and 17.18% of the variances, respectively. Additionally, the main source of Pb is traffic emissions; the main sources of Ni and Cr are metal smelting; the main sources of Hg are agricultural activities and natural sources; the main source of As is mining extraction; and Cd, Cu, and Zn are mainly from electroplating industries.
6. The average *HQ* values rankings of heavy metals from high to low were  $As > Cr > Pb > Cd > Cu > Ni > Zn > Hg$ . The average *CR* values of heavy metals were in the order of  $As > Cd > Cr$ , as was the heavy metal that posed the greatest risk to human health, which was suggested to be controlled first. The average non-carcinogenic and carcinogenic risks of children were 6.85 and 1.70 times higher than those of adults, respectively.

**Supplementary Information** The online version contains supplementary material available at <https://doi.org/10.1007/s11356-024-32554-0>.

**Acknowledgements** We appreciate the efforts of all the researchers whose articles were included in this study.

**Author contribution** Haijian Xie: conceptualization, funding acquisition, project administration, resources, supervision, writing—original draft, and writing—review and editing. Yanghui Shi: conceptualization, investigation, methodology, validation, visualization, writing—original draft, and writing—review and editing. Liang Wang: formal analysis and review and editing. Huaxiang Yan: conceptualization, resources, supervision, writing—original draft, and writing—review and editing. Manting Ci, Ziheng Wang, and Yun Chen: review and editing.

**Funding** The financial supports provided by the Pioneer and Leading Goose R&D Program of Zhejiang (2022C03051), the National Natural Science Foundation of China (Grant Nos. 41977223 and 41931289), the National Key R & D Program of China (Grant Nos. 2018YFC1802303 and 2019YFC1806005), and the Natural Science Foundation of Zhejiang province (Grant No. LR20E080002) are greatly appreciated.

**Data availability** Data are available from the authors upon reasonable request.

## Declarations

**Ethics approval and consent to participate** Not application.

**Consent for publication** Not application.

**Competing interests** The authors declare no competing interests.

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