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Spatial distribution, source and health risk assessment of soil heavy metals in typical agricultural areas of the red-bed intermountain basin, Sichuan-Yunnan ecological shield, China

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ABSTRACT

This study aimed to qualitatively and quantitatively apportion sources of soil heavy metals and assess associated health risks in a typical cultivated region within the red-bed intermontane basin of the Sichuan-Yunnan ecological barrier zone, thereby identifying priority control factors and providing scientific foundations for soil heavy metal pollution prevention. A total of 456 surface soil samples were collected from the Yao'an intermontane basin in central Yunnan Province. Geostatistical methods, the absolute principal component score-multiple linear regression (APCS-MLR) receptor model, and a health risk model based on Monte Carlo simulation were integrated to analyze the spatial distribution patterns, sources, and health risks of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in soils. The mean concentrations of Pb, Ni, Cd, and Cr in surface soils substantially exceeded the background values for Yunnan Province. Four

principal sources were identified: natural parent material (41.65%), agricultural irrigation (21.85%), agricultural fertilizers and pesticides (22.58%), and anthropogenic-related atmospheric deposition (13.92%). Monte Carlo-based health risk assessment indicated negligible non-carcinogenic risks for the exposed population; however, carcinogenic risks were evident, with children exhibiting significantly higher vulnerability than adults. Cr, Ni, and As were the predominant contributing factors. Integrating health risk assessment, sensitivity analysis, and source-oriented risk evaluation, the natural parent material source was identified as the dominant risk contributor, with Ni and Cr as the primary contributing elements. Despite their weak association with anthropogenic activities, continuous monitoring of Ni and Cr contamination remains warranted. The mixed agricultural source, encompassing agricultural fertilizers/pesticides and irrigation, was designated as the priority control source, with As and Cd identified as the priority control elements.

Keywords: Soil heavy metals; APCS-MLR receptor model; spatial distribution; source apportionment; health risk assessment; Sichuan-Yunnan ecological shield; Environmental geological survey engineering

Highlights: Source apportionment of heavy metals is a prerequisite for preventing and mitigating soil contamination. The intermontane basins in the red-bed region of southwestern China constitute critical functional zones for grain production within the Sichuan-Yunnan ecological barrier. This study employed the absolute principal component score-multiple linear regression (APCS-MLR) receptor model and a Monte Carlo simulation-based health risk assessment model to identify pollution sources of soil heavy metals, characterize health risks, and pinpoint priority control factors, thereby providing a scientific basis for soil heavy metal pollution prevention and control.

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1. Introduction

Soil is the basis of agricultural production and human survival. Soil heavy metal pollution not only affects the properties and functions of soil, reduces the yield and quality of agricultural products, but also can enter the food chain through crop accumulation, endangering human health (Wu J et al., 2016; Drobnik T et al., 2018; Chen L et al., 2021; Qin GW et al., 2021). Soil heavy metal pollution has attracted the attention and research of the whole society (Shi YH et al., 2017; Qin XY. et al., 2017).

In recent years, scholars at home and abroad have conducted extensive research on soil heavy metal pollution (Sedov S et al., 2023; Zhang CH et al., 2023). The results found that under the influence of different factors such as natural weathering, industrial and mining development and transportation, there are significant differences in surface soil heavy metal pollution (Wang GX et al., 2017; Dai LJ et al., 2019). To prevent or reduce soil heavy metal pollution, the most important and critical issue is to clarify the sources of heavy metals (Nicholson F A et al., 2003; Huang Y et al., 2015; Huo MZ et al., 2021). Soil heavy metal sources are mainly divided into natural sources and anthropogenic sources (Zhao XN et al., 2023; Xu L et al., 2024). Natural sources are mainly soil parent materials, and anthropogenic sources can be divided into industrial sources (mining, smelting, chemical industry, etc.), agricultural sources (fertilizers, pesticides, wastewater irrigation, etc.) and domestic sources (vehicle exhaust, household waste, domestic coal combustion, etc.). At present, source identification mainly applies receptor models. Through different computational techniques and methods, such as enrichment factor method, chemical element balance method, cluster analysis method, factor analysis method, principal component analysis and target transformation factor analysis and other multivariate analysis methods and type identification techniques, etc. (Liu Y et al., 2020; Wu J et al., 2020), the magnitude of contribution at an observation point (i.e., the receptor) can be obtained. The absolute principal component score/multiple linear regression model

(APCS-MLR) has been widely used in quantitative source apportionment of water bodies (Singh K P et al., 2005; Zhou F et al., 2007), sediments (Ateş A et al., 2020) and the atmosphere (Song Y et al., 2006; Luo XS et al., 2014). Because it does not require prior knowledge of the number of pollutants and takes the pollutants in the receptor as the research object, it is simple to implement and the source apportionment results are relatively objective and accurate. In recent years, it has also been widely used in the soil field (Huo MZ et al., 2021; Li J et al., 2018). Some scholars have used the APCS-MLR model to analyze soil heavy metal sources and demonstrated that the model's apportionment results are more intuitive and accurate (Huang Y et al., 2018; Jia ZY et al., 2020; Li YX et al., 2022).

The Yao'an intermountain basin in central Yunnan is located in the Sichuan-Yunnan ecological barrier area. It is a typical red-bed intermountain basin with concentrated cultivation in the southwest region and an important grain and vegetable production base in central Yunnan. However, research on the degree and source apportionment of soil heavy metal pollution has yet to be carried out. Secondly, in addition to natural sources from soil parent materials, soil heavy metals in this typical red-bed intermountain basin are usually superimposed with anthropogenic sources such as agricultural activities. Therefore, it is particularly important to distinguish the respective contributions of soil parent material and agricultural activities to soil heavy metals in the red-bed basin. At the same time, whether areas dominated by natural geogenic sources pose significant health risks to the local population is also a key issue that needs to be addressed urgently. Finally, formulating targeted prevention and control measures based on source apportionment results provides a scientific basis for precise zoning control and targeted remediation of local soil heavy metals, and also provides reference ideas and practical examples for the prevention and control of soil heavy metal pollution in similar red-bed intermountain basin areas in southwest China.

2. Materials and Methods

2.1. Overview of the Study Area

The research area is located in the intermountain basin of Dongchuan Town and Guanglu Town, Yao'an County, central Yunnan Province, China (Fig. 1a), and is situated in the Chuxiong Zhongshan Forest Ecological Area of the Sichuan-Yunnan Ecological Barrier Area. The research area covers an area of 65 km², with an average altitude of 1870 m. The climate is mild, rainfall is concentrated, the terrain is flat, the soil is fertile, the location advantage is obvious, and the natural conditions are superior. The Yao'an Intermountain Basin is a typical farming area in the Sichuan-Yunnan Ecological Barrier Area. It is an important grain and vegetable production base. It mainly grows rice, corn, vegetables, pomegranates, watermelons, edible roses and other crops. This area is an important part of the Central Yunnan Urban Agglomeration. The Qingling River flowing through the research area belongs to the upstream water system of the Jinsha River. It originates from the Taiping area in the south of the county. The Qingling River runs through the Yao'an Basin and flows north through Dayao and Yuanmou with a long runoff. It is the main irrigation river in the Yao'an Plain area and is known as the "mother river" of the Yao'an people. The agricultural irrigation water source in the area between the main stream and tributaries of the Qingling River in the research area and the surrounding area of the river is the Qingling River water; the agricultural irrigation water source in the central and western parts of the research area is the Yangpai Reservoir; and the agricultural irrigation water source in the central and eastern parts of the research area is a number of irrigation ponds represented by the Yizijiu Reservoir. Except for the water source of the Qingling River from the alkali-rich porphyry area in the Taiping area, the rest of the water sources basically come from the surrounding red-bed area. The transportation in the research area mainly includes the north-south national highway from Dongchuan Town to Guanglu Town, as well as the urban road network concentrated around the townships.

The exposed strata in the research area are Quaternary alluvial sediments. The

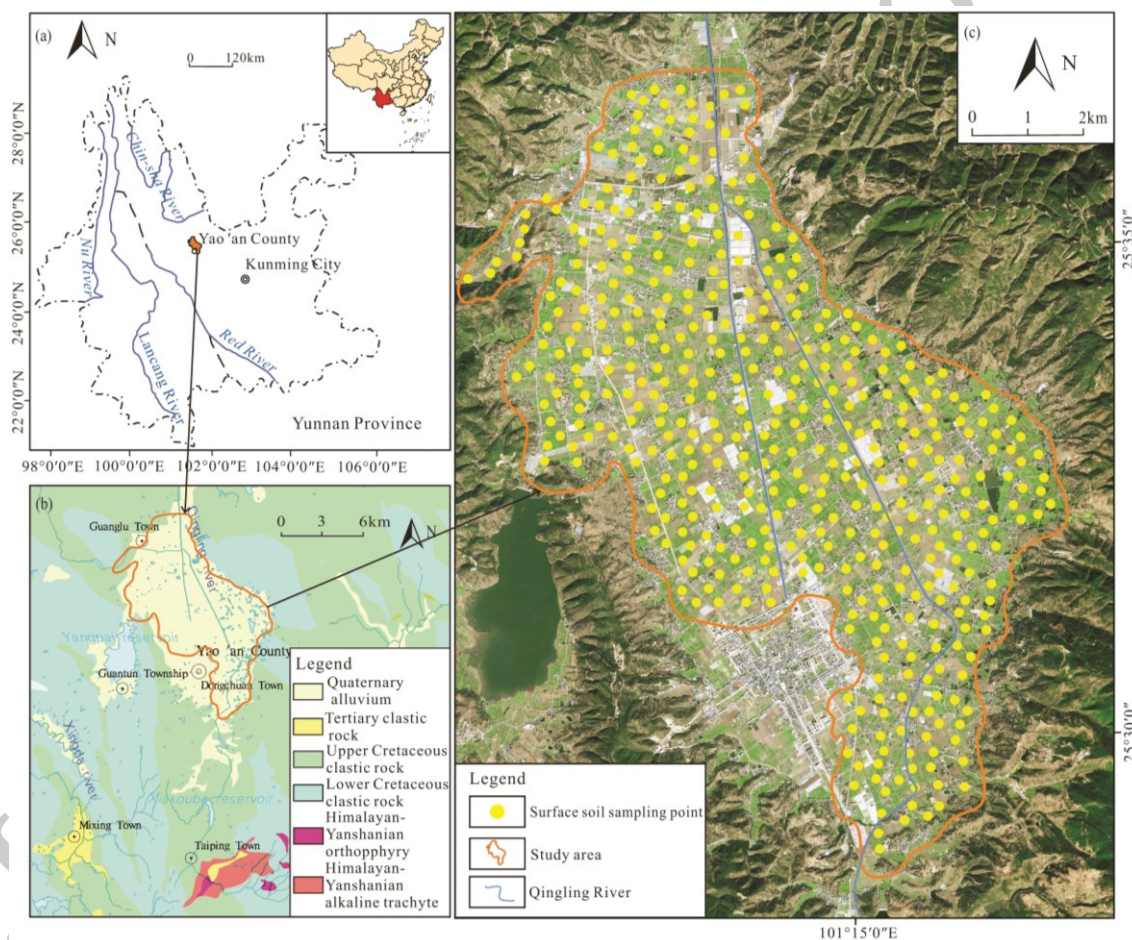
research area is surrounded by Cretaceous purple-red clastic rocks. The lithology is mainly purple-red sandstone, siltstone and mudstone. The alkaline trachyte and syenite porphyry of the Himalayan period are exposed in the Taiping area in the southern part of the research area (Fig. 1b). The source of alluvial materials in the research area is mainly Cretaceous clastic rocks and alkaline intrusive rocks. The alluviation of the Qingling River and the water system around the plain area provides a continuous supply of materials for the intermountain basin. It is worth noting that the alkaline intrusive rock area in the south of the research area is located in the middle segment of the Cenozoic high-potassium alkaline-rich intrusive rock belt distributed along the Jinsha River-Ailao Mountain-Honghe Deep and Large Fault Zone. The rock contact zone and surrounding rocks host Yao'an magmatic hydrothermal vein-type gold, copper, iron, lead, zinc and other polymetallic deposits. The representative deposits include the Laojiezi Pb-Ag deposit and the Baimaju Au (Cu) deposit. The parent material and soil in the alkaline intrusive rock area are

characterized by high background values of Zn, Cu, Pb, As, Ag, Hg and other elements.

Fig. 1. Geological background and sampling points of the study area

2.2. Sample Collection, Processing and Analysis

According to the *Land Quality Geochemical Evaluation Specification* (DZ/T 0295-2016), 456 surface soil samples were collected (Fig. 1c). The soil sample collection density is 7 points/km², and the sampling points are evenly distributed. The sampling points are mainly located on agricultural land, and the sampling depth is 0 to 20 cm. Each sample point consists of 4 to 5 sub-sample points, all of which are within



20 to 50 m of the central sample point and far from pollution sources. After the samples are naturally air-dried, they are passed through a 10-mesh (2 mm) nylon sieve, and 100 g of the sieved soil sample is taken into a clean polyethylene sample bottle and sent to the laboratory for further processing and analysis testing.

The surface soil sample analysis and testing was completed in the analysis and

testing laboratory of the Kunming Natural Resources Comprehensive Survey Center of the China Geological Survey, following the *Geological and Mineral Laboratory Test Quality Management Specification* (DZ/T 0130-2006) standard. Table 1 shows the relevant detection methods and detection limits of each elemental index of the sample. Password samples are inserted into the surface soil samples with 12 GBW standard substances for analysis, and the logarithmic error $\Delta \lg C$ (GBW) of the measured value and the standard value is calculated separately for each analysis result of each element to control the accuracy of the analysis. The accuracy and precision pass rate of this analysis test is greater than 98%, the accuracy pass rate is also greater than 98%, and the pass rate of repeatability test is greater than 92%. The reporting rate, detection limit, accuracy, precision and other indicators of the analysis test method meet the requirements of relevant specifications, and the quality of the analysis data is reliable.

Table 1. Element analysis methods and detection limits

Element/Indicator	Laboratory Detection Limit	Regulatory Detection Limit	Relative Standard Deviation / %	Analytical Method
Pb	1.00	2.00	1.13	Inductively Coupled Plasma Mass Spectrometry (ICP-MS)
Cu	0.46	1.00	1.23	
Cd	0.03	0.03	5.52	
Ni	1.00	2.00	1.33	Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES)
Zn	2.00	4.00	0.90	
As	0.63	1.00	1.60	Atomic Fluorescence Spectrometry (AFS)
Hg	0.003	0.005	2.65	
Cr	4.00	5.00	2.24	X-ray Fluorescence Spectrometry (XRF)
pH*	0.1	0.1	0.26	Ion Selective Electrode (ISE)

Note: * denotes dimensionless; the unit of measurement for all other items is $\text{mg} \cdot \text{kg}^{-1}$.

2.3. Analysis Method of Spatial Variation Structure

2.3.1. Moran's I Index

Moran's I index is used to characterize the correlation of the same variable at different spatial positions. It is a common method for spatial autocorrelation analysis. By detecting whether the variation at a position depends on the variation at adjacent positions, it can be judged whether the variation has spatial autocorrelation (Martin D, 1996; Zhang CS et al., 2008; Li ZF et al., 2021). The calculation formula of Moran's I

index is:

$$I = \frac{n}{\sum_{i=1}^n \sum_{j=1}^n \omega_{ij}} \times \frac{\sum_{i=1}^n \sum_{j=1}^n \omega_{ij} (x_i - \bar{x})(x_j - \bar{x})}{\sum_{i=1}^n (x_i - \bar{x})^2} \quad (1)$$

In formula (1): x_i is the actual value measured at location i of the heavy metal element x , $\bar{x}$ is the average value of element x , and ω_{ij} is the spatial weight function. The standard deviation z -score under the assumption of approximate normal distribution under random conditions is usually used to judge spatial correlation (Liu ZY et al., 2021). When $Z > 1.96$ and $p < 0.05$, the null hypothesis is rejected, and the spatial autocorrelation is positive, that is, similar observations tend to be spatially clustered; when $1.96 > Z > -1.96$, the null hypothesis is accepted, and the spatial distribution shows randomness; when $Z < -1.96$ and $p < 0.05$, the null hypothesis is rejected, and the spatial autocorrelation is negative, that is, similar observations tend to be spatially isolated (Liu Q et al., 2011).

2.3.2. Semivariance Function

The semivariance function is a key function in geostatistics to study soil variability. It can quantitatively describe the spatial variation law of soil properties. The calculation formula is:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i + h)]^2 \quad (2)$$

In formula (2), $\gamma(h)$ is the semivariance at sample point spacing h , $N(h)$ is the number of pairs with spacing h , $Z(x_i)$ is the content value of soil element i at x_i , and $Z(x_i + h)$ is the content value of soil element i at $x_i + h$. The theoretical variogram models mainly include exponential, Gaussian, spherical and linear models (Gan T et al., 2023). The main parameters include nugget constant (C_0), sill value ($C_0 + C$), range, coefficient of determination (R^2) and residual sum of squares (RSS), etc. (Franklin R B et al., 2023). The results of semivariance function analysis are mainly reflected by the nugget effect [$C_0/(C_0 + C)$]. Generally, the larger $C_0/(C_0 + C)$, the more dispersed the spatial distribution of the corresponding physical quantity and the weaker the correlation (Cambardella C A et al., 1994; Zhao KL et al., 2014; Yan WB et al., 2015; Xi XK et al., 2017).

2.4. Absolute Principal Component Score/Multiple Linear Regression Receptor Model

The basic principle of the absolute principal component score/multiple linear regression receptor model (APCS-MLR) is to convert the principal factor scores of factor analysis into absolute principal factor scores, and then perform multiple linear regression of each heavy metal content against all APCS. The regression coefficients are used to calculate the contribution rate of pollution sources corresponding to each principal factor to the content of a certain heavy metal at each sample point in the soil (Thurston G D et al., 1987; Cao JF et al., 2020; Yu F et al., 2023). The calculation steps are as follows: first, standardize all heavy metal contents; then introduce an artificial sample with zero concentration, calculate the factor scores of the zero-concentration artificial sample, and subtract the principal factor scores of the zero-concentration artificial sample from the element principal factor scores obtained from factor analysis to obtain the absolute principal component scores APCS; then, with APCS as independent variables, perform multiple linear regression on the content of each heavy metal, and use the regression coefficients to calculate the source contribution rate of each heavy metal (Cao JF et al., 2020; Qu MK et al., 2018; Zhang M et al., 2020; Shi WC et al., 2020). The calculation formula is:

$$Z_{i0} = \frac{0 - \bar{C}_i}{\sigma_i} = -\frac{\bar{C}_i}{\sigma_i} \quad (3)$$

$$C_i = b_{i0} + \sum_{p=1}^p (b_{pi} \times \text{APCS}_p) \quad (4)$$

In formula (3) and formula (4), Z_{i0} is the zero-concentration sample of element i , C_i is the average value of element i content, σ_i is the standard deviation of element i content, b_{i0} is the constant term of multiple linear regression, b_{pi} is the regression coefficient of multiple linear regression, and APCS_p is the content contribution of factor p to C_i . The average value of $b_{pi} \times \text{APCS}_p$ across all samples is the average absolute contribution of the pollution source corresponding to factor p . The pollution source contribution rate corresponding to factor p is the percentage of the average absolute contribution relative to the total contribution of all sources.

2.5. Evaluation Method of Soil Heavy Metal Pollution

2.5.1. Geo-accumulation Index

The geo-accumulation index is often used to quantitatively evaluate the degree of heavy metal enrichment in sediments and other media (Tarun K D et al., 2004; Liu JY et al., 2010). It takes into account not only the influence of the geological background of the natural soil formation process, but also the impact of anthropogenic activities on heavy metal pollution. The calculation formula is as follows:

$$I_{\text{geo}} = \log_2 \frac{C_n}{KB_n} \quad (5)$$

In formula (5), I_{geo} is the geo-accumulation index, and C_n is the measured content of soil heavy metal n ; B_n is the background value of soil heavy metal n . This paper adopts the surface soil background value of Yunnan Province (Hou QY et al., 2020); K is the correction coefficient for the possible influence of diagenesis on the background value, and the correction coefficient k is taken as 1.5. The correspondence between I_{geo} and the degree of enrichment is: when $I_{\text{geo}} \leq 0$, no enrichment; when $0 < I_{\text{geo}} \leq 1$, slight enrichment; when $1 < I_{\text{geo}} \leq 2$, moderate enrichment; when $2 < I_{\text{geo}} \leq 3$, moderate to strong enrichment; when $3 < I_{\text{geo}} \leq 4$, strong enrichment; when $4 < I_{\text{geo}} \leq 5$, very strong enrichment; when $I_{\text{geo}} > 5$, extremely strong enrichment (Zhang Q et al., 2015).

2.5.2. Health Risk Assessment Based on Monte Carlo Simulation

This paper adopts the health risk assessment model based on Monte Carlo simulation released by the U.S. Environmental Protection Agency (USEPA) to quantitatively evaluate the human health risks caused by soil heavy metals. The main process covers the calculation of exposure doses and the description of risk characteristics (U.S. EPA, 1989). Heavy metals in soil usually enter the human body mainly through three pathways: hand-to-mouth ingestion, dermal contact and inhalation, thereby posing health risks to the human body. The calculation formula is:

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

$$\text{ADD}_{\text{inh}} = (C_i \times \text{InhR} \times \text{EF} \times \text{ED}) / (\text{PEF} \times \text{BW} \times \text{AT}) \quad (7)$$

$$ADD_{drem} = (C_i \times SA \times SL \times ABS \times EF \times ED \times 10^{-6}) / (BW \times AT) \quad (8)$$

In the formula, ADD_{ing} , ADD_{inh} and ADD_{drem} represent the average daily intake of soil heavy metals through hand-to-mouth ingestion, inhalation and dermal contact, respectively, where C_i indicates the actual content of soil heavy metal i . The remaining specific parameters are shown in Table 2 (Ma J et al., 2023).

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Table 2. Parameters of the Monte Carlo soil heavy metal health risk model

Parameter	Parameter Name	Unit	Distribution Type ¹⁾	Reference Value	
				Adult	Child
IngR	Daily soil intake	mg·d ⁻¹	Triangular	30(4 and 52)	103(66 and 161)
EF	Exposure frequency	d·a ⁻¹	Triangular	345(180 and 365)	
ED	Exposure duration	a	Single point	24	6
InhR	Daily air inhalation volume	m ³ ·d ⁻¹	Single point	19	8.6
BW	Average body weight	kg	Lognormal	57.03±1.18	16.68±1.48
AT	Average exposure time	d	Single point	365×76(carcinogenic)	365×ED(non-carcinogenic)
SA	Exposed skin area	m ²	Single point	0.54	0.23
SL	Skin adherence factor	mg·(cm ² ·d) ⁻¹	Lognormal	0.49±0.57	0.65±1.2
PEF	Surface dust emission factor	m ³ ·kg ⁻¹	Single point	1.36×10 ⁹	
ABF	Dermal absorption factor		Dimensionless	0.001(non-carcinogenic)	0.01(carcinogenic)

Note: Triangular: most probable value (minimum and maximum); Logarithmic: mean ± standard deviation

Non-carcinogenic risk (HI) and carcinogenic risk (TCR) are two key indicators for the health risk assessment of soil heavy metals. The calculation formulas are listed as follows:

$$HI = \sum HQ = \sum (ADD_{ij} / RfD_{ij}) \quad (9)$$

$$TCR = \sum CR = \sum (ADD_{ij} \times SF_{ij}) \quad (10)$$

In the formula, HI represents the comprehensive non-carcinogenic risk index, HQ represents the single non-carcinogenic risk index, TCR represents the comprehensive carcinogenic risk index, CR represents the single carcinogenic risk index, ADD_{ij} represents the non-carcinogenic average daily exposure of heavy metal i through different exposure routes j , RfD_{ij} indicates the non-carcinogenic risk reference dose of heavy metal i through different exposure routes j , SF_{ij} represents the carcinogenic slope factor of heavy metal i through different exposure routes j . The specific reference values of RfD and SF are detailed in Table 3 (Ma J et al., 2023).

When $HI (HQ) \leq 1$, the non-carcinogenic health risk of soil heavy metals to the human body is at an acceptable level and can be ignored; otherwise, there is a non-carcinogenic health risk. When $TCR (CR) \leq 1.00E-6$, the carcinogenic health risk of soil heavy metals to the human body is negligible. When $1.00E-6 < TCR (CR) \leq 1.00E-4$, it indicates that there is a tolerable carcinogenic health risk. When TCR

(CR) > 1.00E-4, it means that soil heavy metals pose an intolerable carcinogenic risk to the human body (Zhang C et al., 2024).

Table 3. Carcinogenic slope coefficients and reference doses for soil heavy metals under three exposure pathways

Element	Reference Dose (RfD) /mg·(kg·d) ⁻¹			Carcinogenic Slope Factor (SF)/kg·d·mg ⁻¹		
	Oral	Dermal	Inhalation	Oral	Dermal	Inhalation
As	3.00E-4	1.23E-4	3.00E-4	1.50E+0	3.66E+0	1.51E+1
Cd	1.00E-3	1.00E-5	1.00E-5	6.10E+0	6.30E+0	6.30E+0
Cr	3.00E-3	6.00E-5	2.86E-5	5.00E-1	2.00E+1	4.20E+1
Cu	4.00E-2	1.20E-2	4.02E-2	—	—	—
Hg	3.00E-4	2.10E-5	8.57E-5	—	—	—
Ni	2.00E-2	5.40E-3	9.00E-5	1.70E+0	4.25E+1	8.40E-1
Pb	3.50E-3	5.25E-4	3.52E-3	8.50E-3	—	6.30E+0
Zn	3.00E-1	3.00E-1	3.00E-1	—	—	—

1) Note: “—” indicates no data available.

2.6. Data Processing and Analysis

Soil indicator statistics, health risk assessment and multiple linear regression are completed with the help of Excel; K-S test and principal component analysis are completed with the help of SPSS 24.0; data visualization analysis (including correlation coefficient matrix, clustering dendrogram, box plot and Sankey diagram) is completed with the help of OriginLab 2021; the spatial distribution map is drawn with the help of ArcGIS 10.7; and the sensitivity analysis is completed using Oracle Crystal Ball 11.1.3.0.

3. Results and Discussions

3.1. Statistical Analysis of Heavy Metals in Soil

Table 4 shows the content statistics of 8 heavy metal elements in 456 surface soil samples in the research area. The soil pH ranges from 5.01 to 9.19, with an arithmetic mean of 7.29, and samples with pH greater than 7 account for 75.88%, indicating that the soil in the study area is mainly alkaline. The average contents of soil As, Cd, Cr, Cu, Hg, Ni, Pb and Zn are 9.17, 0.33, 102.11, 42.67, 0.05, 48.71, 58.81 and 105.40 mg·kg⁻¹, respectively. Compared with the background values of surface soil in Yunnan Province (Hou QY et al., 2020), the average contents of Cu and Zn are basically consistent with the background values, with ratios of 106.67% and 109.79%,

respectively; Pb, Ni, Cd and Cr are relatively enriched, with ratios of 150.80%, 128.20%, 123.16% and 112.21%, respectively; As and Hg show relative depletion, with ratios of 86.53% and 77.30%, respectively.

The coefficient of variation can reflect the uniformity and degree of variation of element distribution. Relevant studies (Wilding L P, 1985; Wollenhaupt N C et al., 1997; Ji ZH et al., 2021) show that a coefficient of variation greater than 36% indicates high variability, suggesting uneven spatial distribution of elements, possible point-source pollution, and input of external material sources. As (41.04%), Cd (46.13%), Hg (68.22%) and Pb (76.34%) in the surface soil of the study area are highly variable elements, indicating that their distribution in soil is uneven and greatly affected by external factors, which may be largely influenced by human activities.

Table 4. Summary of statistical result characteristics of heavy metals in surface soil in the study area (n=456)

Items	Elements(mg·kg ⁻¹)								pH
	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	
Minimum	2.97	0.10	62.40	27.00	0.02	19.10	17.40	41.70	5.01
Maximum	27.50	1.35	137.00	75.90	0.46	70.60	187.00	162.00	9.19
Mean	9.17	0.33	102.11	42.67	0.05	48.71	58.81	105.40	7.29
Median	7.86	0.30	103.00	42.30	0.05	49.35	29.80	100.00	7.43
SD	3.76	0.15	10.52	7.29	0.04	6.95	44.90	25.14	0.59
CV(%)	41.04	46.13	10.30	17.10	68.22	14.26	76.34	23.85	8.04
Background value of soil in study area(Hou et al., 2020)	10.60	0.27	91.00	40.00	0.07	38.00	39.00	96.00	6.14
K ^a (%)	86.53	123.16	112.21	106.67	77.30	128.20	150.80	109.79	118.72

Note: ^athe ratio of the average value of heavy metals in the study area to the background value.

3.2. Spatial Variation Structure of Soil Heavy Metals

The Moran's I index of spatial autocorrelation analysis for 8 heavy metals in the surface soil of the research area is shown in Table 5. According to the Z value, the two-sided test threshold of 1.96 for the 95% confidence interval of the normal distribution is used as the boundary to judge whether spatial autocorrelation is significant. The standardized Z values of the spatial autocorrelation index for As, Cr, Cu, Ni, Pb and Zn are all greater than 1.96, with $P < 0.05$, indicating that the above heavy metals have significant positive spatial autocorrelation, that is, their distribution has a good spatial structure and is mainly controlled by the soil parent material

(Quaternary alluvial sediments) in the study area. The Z values of Cd and Hg are between 0 and 1.96, with $P > 0.05$, indicating that the spatial autocorrelation of Cd and Hg is not significant and shows random spatial distribution. Hg is the most obvious, indicating that it is more affected by anthropogenic factors.

The semivariance function is a key function in geostatistics for model establishment and theoretical model fitting. It is also an effective method for analyzing influencing factors of spatial variation and can maximally preserve spatial variation information. Therefore, the semivariance function is further used to quantify the spatial relationships of heavy metals. After K-S test, the contents of 8 heavy metals in the surface soil of the study area all show skewed distribution. Cd, Cr, Cu, Hg and Ni follow normal distribution after Johnson transformation, while As, Pb and Zn still do not obey normal distribution after common transformation methods. Therefore, semivariance function fitting is performed on the transformed data of Cd, Cr, Cu, Hg and Ni, and the analysis results are shown in Table 6.

The theoretical semivariance function models for Cd and Hg conform to the Exponential model, while those for Cr, Cu and Ni conform to the Spherical model. The effective range of each heavy metal element is between 1620 and 3800 m, the coefficients of determination R^2 are all greater than 0.813, and the residual sums of squares are all less than 0.0184, indicating that the theoretical model fitting effect is good. The nugget-to-sill ratio $C_0/(C_0+C)$ can represent the spatial autocorrelation of parameters and reflect the roles of natural and anthropogenic factors. The $C_0/(C_0+C)$ values of Cd, Cr, Cu, Hg and Ni in the surface soil of the study area are 41.10%, 44.09%, 32.53%, 49.95% and 38.33%, respectively, ranging between 25% and 75%, showing moderate spatial correlation. This indicates that the spatial structure of these heavy metal elements may also be affected by human activities, such as traffic emissions, fertilization, industrial pollution and other factors, in addition to natural factors such as soil parent material. Range represents the variable of spatial correlation range; elements within the range have spatial correlation, while those beyond the range have no correlation. The ranges of Cr and Ni are relatively small, at 1890 and 1620 m respectively, indicating spatial correlation within a small range. In

addition to being affected by soil parent material, they may also be affected by random factors; the ranges of Cd, Cu and Hg are relatively large, between 2100 and 3800 m, indicating that Cd, Cu and Hg may be jointly affected by structural factors such as soil parent material and random factors such as human activities.

Table 5. Spatial autocorrelation analysis of soil heavy metals in the study area

Item	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Variance	0.04	0.04	0.04	0.04	0.03	0.04	0.04	0.04
Z	4.91	1.1	2.13	2.17	0.67	2.22	3.02	4.36
P	0.000	0.270	0.030	0.030	0.500	0.030	0.003	0.000

Table 6. Optimization model and fitting parameters of soil heavy metals concentration in the study area

Element	Theoretical model	Nugget (C_0)	Sill (C_0+C)	Range/m	Nugget/sill[$C_0/(C_0+C)$]/%	R^2	RSS
Cd	Exponential	0.402	0.978	2100	41.10	0.911	0.0122
Cr	Spherical	0.403	0.914	1890	44.09	0.959	0.0070
Cu	Spherical	0.010	0.030	3800	32.53	0.813	0.0001
Hg	Exponential	0.507	1.015	3000	49.95	0.935	0.0104
Ni	Spherical	0.358	0.934	1620	38.33	0.897	0.0184

3.3. Spatial Distribution Pattern of Soil Heavy Metals

In order to more intuitively analyze the spatial distribution and variation degree of heavy metal contents in the soil of the research area, the inverse distance weighted interpolation method was used with ArcGIS 10.8 software to draw the spatial distribution map of heavy metal contents in the surface soil of the research area (Fig. 2). It can be seen from the spatial distribution of pH that acidic soils are mainly distributed in the eastern and northwestern parts of the research area, and the rest of the areas are alkaline to weakly alkaline soils; the spatial distribution characteristics of As, Pb and Zn contents show strong consistency, with high-value areas mainly distributed between the Qingling River and its tributaries, and low-value areas mainly on the east side of the Qingling River and the west side of its tributaries; the spatial distribution characteristics of Cr and Ni contents also show strong consistency, with high-value areas mainly on both sides of the Qingling River and its tributaries, and low-value areas mainly in the southern part of the research area and between the Qingling River and its tributaries; the high-value areas of Cu are mainly distributed around the county town, most prominently in the northern part of the county; the high-value areas of Cd and Hg are unevenly distributed in patches in the research area,

and are present around the county town and around the market in Guanglu Town. The high spatial consistency of heavy metal elements indicates that they have the same material sources. Therefore, it can be preliminarily inferred that As, Pb and Zn have the same material sources, and so do Cr and Ni.

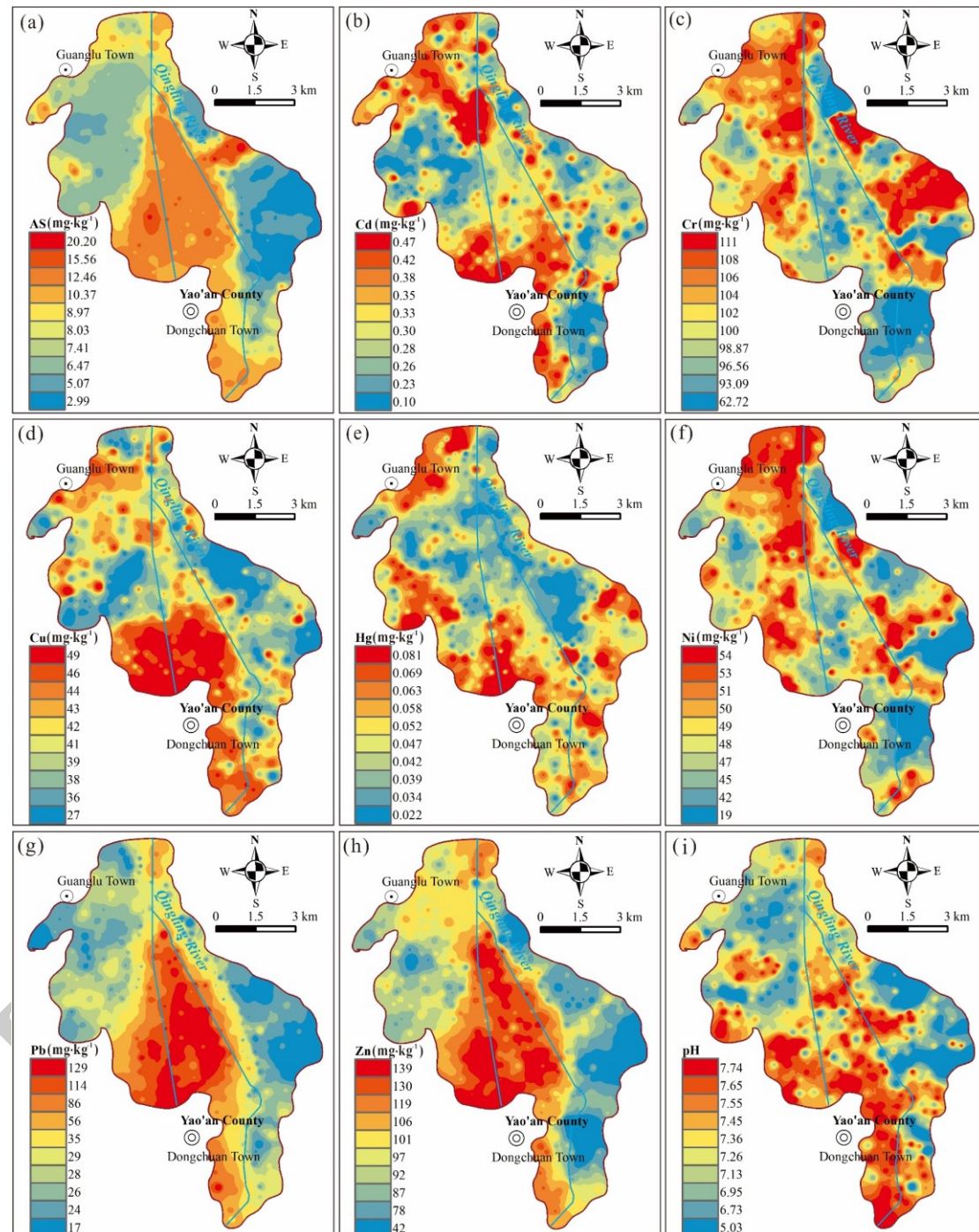


Fig. 2. Spatial distribution of heavy metals and pH in surface soil of the study area

3.4. Source Apportionment of Soil Heavy Metal Pollution

3.4.1. Correlation Analysis

Soil heavy metals mainly come from two pathways: natural factors and anthropogenic input. Correlation analysis is a commonly used analytical method for source analysis of soil heavy metals. Through the correlation between various heavy metals in soil, the similarity of source pathways of soil heavy metals can be inferred. The more similar the source pathways of heavy metals, the more significant the correlation between them (Guo GH et al., 2012). Figure 3a shows the Pearson correlation coefficient matrix of 8 heavy metals. The Pearson correlation coefficients between As, Pb and Zn range from 0.72 to 0.86, and the Pearson correlation coefficient between Cr and Ni reaches 0.85, both showing extremely significant positive correlations ($P < 0.05$), indicating that As, Pb and Zn, as well as Cr and Ni, have similar source pathways, which further explains the consistency of their spatial distribution; the correlation coefficients of Cu with Pb and Zn are 0.53 and 0.57 respectively, showing relatively significant positive correlations; the correlation coefficient between Hg and Cu is 0.23, showing a weak positive correlation; the correlation coefficients of Cd with the remaining 7 heavy metals range from 0.10 to 0.36, all showing weak positive correlations, indicating that Cu, Hg and Cd have multi-source composition characteristics. Cluster analysis was performed on 8 heavy metal elements in 456 samples (Fig. 3b). The 8 heavy metal elements can be divided into 4 categories: As, Cu, Pb and Zn as one category, Cr and Ni as one category, and Cd and Hg each as one category. The results of cluster analysis are basically consistent with the results of correlation analysis.

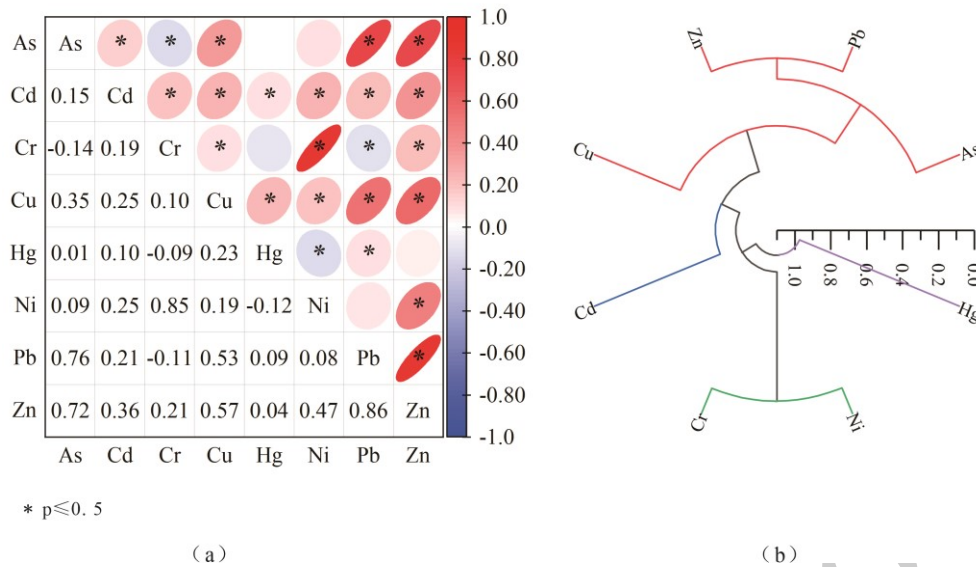


Fig. 3. Pearson correlation diagram (a) and cluster dendrogram (b) of heavy metals in surface soil

3.4.2. Source Apportionment by APCS-MLR Receptor Model

To further clarify the sources of soil heavy metals, the APCS-MLR receptor model was used for source apportionment. First, principal components were used to qualitatively analyze the sources of various heavy metals in the research area. The KMO and Bartlett's test was used to test the data (Comero S et al., 2014). The KMO calculation result is 0.618 (> 0.5), and the significance of Bartlett's test of sphericity is 0.00 (< 0.05), so the data can be subjected to principal component analysis, which is statistically significant. As shown in Table 7, the three principal components have good explanatory power for the variables, with a cumulative contribution rate of 77.89%, and eigenvalues of 3.19, 1.93 and 1.11 respectively, all greater than 1, which can represent the vast majority of information in the original data.

Among the first principal component, the elements with the largest Norm values are Pb and Zn (1.68), and the element whose Norm value differs from that of Pb and Zn within 10% (1.51–1.68) is As. Among the second principal component, the element with the largest Norm value is Ni (1.34), and the element whose Norm value differs from that of Ni within 10% (1.21–1.34) is Cr. The Norm value of Hg in the third principal component is 0.96. Therefore, the elements entering the minimum data set are As, Pb, Zn, Cr, Ni and Hg, representing the vast majority of information in the original data.

The first principal component F1 includes As, Cu, Pb and Zn, with a variance

contribution rate of 39.91%. The factor loadings of As, Pb and Zn are relatively large (> 0.89), and the factor loading of Cu is relatively small (0.56). According to semivariance function analysis, the contents of As, Pb and Zn and their skewness characteristics after transformation indicate a high degree of dispersion and are greatly affected by anthropogenic factors. The spatial distribution characteristics of As, Pb and Zn contents show strong consistency, and their spatial distribution is mainly controlled by the Qingling River. The high-value areas are mainly distributed between the main stream and tributaries of the Qingling River, and the low-value areas are mainly in the central-eastern and central-western parts of the research area. Field investigations show that the agricultural irrigation water source in the area between the main stream and tributaries of the Qingling River and the surrounding area of the river is Qingling River water; the agricultural irrigation water source in the central and western parts of the research area is the Yangpai Reservoir; and the agricultural irrigation water source in the central and eastern parts of the research area is a number of irrigation ponds represented by the Yizijiu Reservoir. The Qingling River originates from the alkali-rich porphyry area of Taiping Town south of the research area (Fig. 1). This area hosts multiple polymetallic deposits, including the Laojiezi Pb-Ag deposit and the Baimaju Au (Cu) deposit. Therefore, this area is characterized by high geological background of As, Pb, Zn and Cu. Natural weathering of rocks or mining activities bring large amounts of heavy metals into the Qingling River, so the water source of the Qingling River has relatively high contents of heavy metals such as As, Pb and Zn. Surface irrigation water sources such as Yangpai Reservoir and Yizijiu Reservoir mainly come from the Mesozoic red-bed area, with relatively low contents of heavy metals such as As, Pb and Zn. The first principal component F1 can be identified as an agricultural irrigation source.

The second principal component F2 includes Cr and Ni, with a variance contribution rate of 24.13%. The factor loadings of Cr and Ni are very high, both at 0.94. Cr and Ni are evenly distributed in the study area, with no obvious high-value and low-value areas, and their coefficients of variation are relatively small, at 10.30 and 14.26 respectively, indicating that they are less affected by human activities and

mainly influenced by natural factors. Relevant studies show that the contents of Cr and Ni in soil are similar to their contents in soil parent material and are related to lithogenic composition (Zhou F et al., 2007; Cheng GW et al., 2020). The surrounding strata rocks in the study area are mainly Cretaceous purple-red clastic rocks, with relatively single lithology. The soil parent material in the study area is mainly Quaternary alluvial sediments, that is, alluvial weathered materials from the surrounding Cretaceous clastic rocks. It is inferred that the second principal component F2 is mainly a natural parent material source.

The third principal component F3 is Hg, with a variance contribution rate of 13.85% and a factor loading of 0.88. However, the loadings of Cu and Cd on F3 are also relatively high, at 0.50 and 0.43 respectively, indicating that this source is also one of the sources of Cu and Cd. Moran's I index shows that the spatial autocorrelation of Cd and Hg is not significant and shows random spatial distribution. Hg is the most obvious, indicating that it is more affected by anthropogenic factors. Relevant studies show that atmospheric dry and wet deposition is an important source of soil Hg (Lv J.S et al., 2014). Anthropogenic Hg emissions account for 60%–80% of total global emissions, and coal mining and combustion are also major sources (Streets D G et al., 2005; Liu Y et al., 2016). Flue-cured tobacco is planted in the research area. Curing tobacco leaves requires large amounts of coal combustion, and Hg released by coal combustion enters the regional surface soil through atmospheric deposition. In addition, from the perspective of spatial distribution, the high-value areas of Hg, Cu and Cd are mainly distributed around the county town and township markets. Relevant studies show that the enrichment of Cu and Cd in soil is closely related to traffic (Wang WJ et al., 2022). Brake wear, tire wear and mechanical abrasion in traffic activities produce large amounts of Cu and Cd, which enter the soil through deposition and affect the accumulation of heavy metals in soil. F3 can be identified as an atmospheric deposition source greatly affected by anthropogenic factors.

Table 7. Principal component analysis matrix of heavy metals in soil

Item	Component	Group	Norm
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	F1	F2	F3		
As	0.89	-0.08	-0.06	1	1.60
Cd	0.23	0.36	0.43	3	0.80
Cr	-0.10	0.94	0.01	2	1.32
Cu	0.56	0.16	0.50	1	1.16
Hg	-0.05	-0.17	0.88	3	0.96
Ni	0.15	0.94	-0.04	2	1.34
Pb	0.94	-0.05	0.12	1	1.68
Zn	0.90	0.34	0.13	1	1.68
Eigenvalue	3.19	1.93	1.11		
Variance Contribution Rate / %	39.91	24.13	13.85		
Cumulative Contribution Rate / %	39.91	64.04	77.89		

Note: Bold elements indicate those included in the minimum data set (MDS).

After principal component analysis, multiple linear regression was performed on the contents of 8 heavy metals (Tab. 8). The predicted values of each element content can be obtained from the equation and compared with the measured values. If the ratio of predicted value to measured value is closer to 1, it indicates better fitting of multiple linear regression and more accurate prediction (Hingorani R et al., 2020). The results show that the R^2 values of linear fitting between predicted and measured values of As, Cr, Hg, Ni, Pb and Zn contents in the surface soil of the study area are all greater than 0.8, while the R^2 values of Cd and Cu are relatively low, at 0.369 and 0.587 respectively. This may be because the complexity of Cd and Cu sources is much higher than that of other heavy metals, which is also confirmed by the principal component analysis results. The P values of linear fitting for all 8 heavy metals are less than 0.05, and the ratios of predicted values to measured values are close to 1, indicating that the constructed APCS-MLR model is reliable and the calculation results are credible.

Table 8. Soil heavy metal APCS-MLR receptor model

Receptor model	R^2	Sig.	Predicted value/ Observed value
$C_{As}=3.360 \times APCS_{F1} - 0.320 \times APCS_{F2} - 0.229 \times APCS_{F3} + 5.509$	0.805	0.000	1.03
$C_{Cd}=0.036 \times APCS_{F1} + 0.055 \times APCS_{F2} + 0.067 \times APCS_{F3} - 0.329$	0.369	0.000	1.08
$C_{Cr}=-1.105 \times APCS_{F1} + 9.938 \times APCS_{F2} + 0.059 \times APCS_{F3} + 28.047$	0.901	0.000	1.00
$C_{Cu}=4.110 \times APCS_{F1} + 1.164 \times APCS_{F2} + 3.634 \times APCS_{F3} + 16.568$	0.587	0.000	1.01
$C_{Hg}=-0.002 \times APCS_{F1} - 0.006 \times APCS_{F2} + 0.033 \times APCS_{F3} + 0.025$	0.809	0.000	1.09
$C_{Ni}=1.065 \times APCS_{F1} + 6.564 \times APCS_{F2} - 0.249 \times APCS_{F3} - 3.058$	0.915	0.000	1.00
$C_{Pb}=42.122 \times APCS_{F1} - 2.394 \times APCS_{F2} + 5.467 \times APCS_{F3} - 20.186$	0.895	0.000	1.04

$$C_{Zn}=22.614 \times APCS_{F1} + 8.471 \times APCS_{F2} + 3.322 \times APCS_{F3} - 12.699 \quad 0.938 \quad 0.000 \quad 1.00$$

The source composition based on the constructed APCS-MLR model analysis is shown in Figure 4a. F1 has the greatest impact on As, Pb and Zn, with contribution rates of 43.94%, 61.67% and 34.39% respectively. Combined with the principal component analysis results, F1 represents the agricultural irrigation source, and As, Pb and Zn are greatly affected by agricultural irrigation, with contribution rates of 43.94%, 61.67% and 34.39% respectively. At the same time, the agricultural irrigation source also has a relatively large contribution rate to Cu, reaching 19.16%. Based on the analysis of spatial distribution characteristics of soil heavy metals, Qingling River irrigation water provides large amounts of As, Pb, Zn and Cu elements to the soil in the irrigated area, while other irrigation sources have relatively low contributions to soil heavy metals. As a natural parent material source, F2 has the greatest impact on Cr and Ni, with contribution rates of 71.46% and 89.67% respectively. At the same time, it also has a relatively large impact on Zn and Cd, with contribution rates of 49.60% and 42.64% respectively, and has certain impacts on Cu, As and Pb, with contribution rates of 20.89%, 16.11% and 13.50% respectively. F3 is an atmospheric deposition source closely related to anthropogenic factors. It has the greatest impact on Hg, with a contribution rate of 52.19%, and also has certain impacts on Cu and Cd, with contribution rates of 21.11% and 16.81% respectively.

F4 is another source, which has relatively large impacts on Cu, As and Cd, with contribution rates of 36.22%, 33.30% and 38.83% respectively. It also has certain impacts on Cr, Hg and Pb, with contribution rates of 26.33%, 15.95% and 14.86% respectively. Relevant studies have shown that excessive use of pesticides and herbicides may lead to the enrichment of As and Cu in surface soil. As is commonly found in feed, and Cd is also commonly found in organic fertilizers (Shomar B H et al., 2006; Qu MK et al., 2018). Long-term application of livestock manure and commercial organic fertilizers will lead to the accumulation of elements such as As, Cd and Cu in soil. In the past decade, the land in the research area has mainly been planted with corn, flue-cured tobacco and vegetables. Investigations have found that large amounts of pesticides and herbicides are used in the research area, and large

amounts of livestock manure and commercial organic fertilizers are applied. It can be inferred that the other source F4 is an agricultural fertilizer and pesticide source.

From the perspective of the contribution rate of various sources to soil heavy metals (Fig. 4b), the natural parent material source has the highest contribution rate to heavy metals, reaching 41.65%, indicating that heavy metals mainly come from natural parent material, and Quaternary alluvial sediments provide a continuous supply of materials to the soil in the research area. The sum of contribution rates of agricultural activity sources represented by agricultural irrigation source and agricultural fertilizer and pesticide source is 44.43%, indicating that agricultural activity sources are also the main source of soil heavy metals. The contribution rate of atmospheric deposition source, which is closely related to anthropogenic factors, is 13.92%, and this source is also closely related to human activities. In summary, part of the heavy metals in the surface soil of the study area comes from natural sources, and the remaining sources are largely closely related to anthropogenic factors, especially the contribution of agricultural activities.

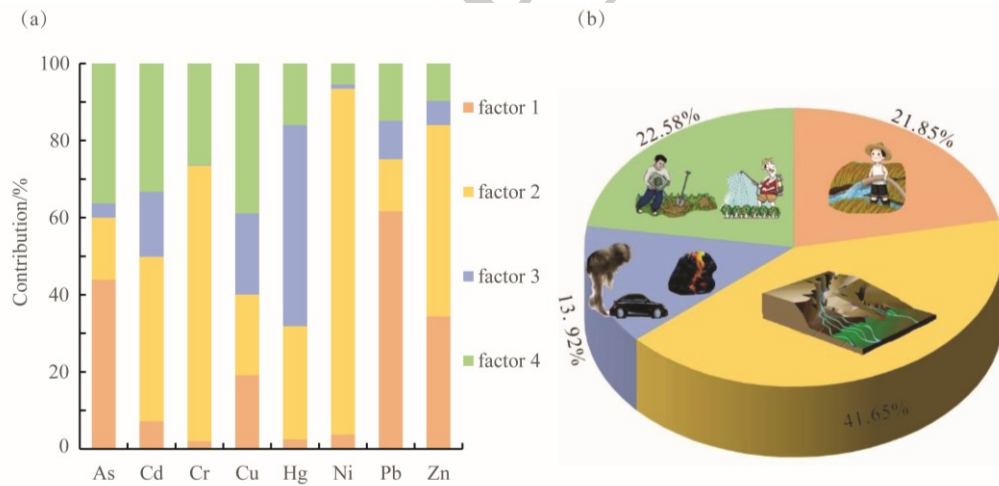


Fig. 4. APCS-MLR source apportionment: component composition (a) and contribution rate (b)

3.5. Geo-accumulation Index Analysis

The I_{geo} evaluation results of heavy metal elements in surface soil of the research area show that the I_{geo} values of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn range from -2.42 to 0.79, -2.05 to 1.74, -1.13 to 0.01, -1.15 to 0.34, -2.54 to 2.13, -1.58 to 0.31, -1.75 to 1.68 and -1.79 to 0.17, respectively (Fig. 5). Although the average I_{geo} values (-1.13 to

-0.24) of the 8 heavy metal elements are all less than 0, all elements have enrichment sites at different degrees. Among them, Cd, Hg and Pb elements have the highest enrichment degree. The proportions of sampling points at moderate to moderately strong enrichment level ($1 < I_{geo} \leq 3$) are 1.97%, 1.10% and 18.20%, respectively, and the proportions of sampling points at slight enrichment level ($0 < I_{geo} \leq 1$) are 19.30%, 4.17% and 16.45%, respectively. The enrichment degrees of As, Cr, Cu, Ni and Zn elements are relatively low, and the proportions of sampling points at slight enrichment level are 3.51%, 0.22%, 3.07%, 8.55% and 7.68%, respectively. Some sites of Cd, Hg and Pb elements are affected by obvious exogenous factors, especially Pb. From the perspective of spatial distribution characteristics, the enriched sampling points of Cd, Hg and Pb elements are all distributed around the county town or township markets, that is, in areas with intensive human activities, indicating that the exogenous sources of Cd, Hg and Pb elements are closely related to anthropogenic factors to a large extent, which is consistent with the principal component analysis results.

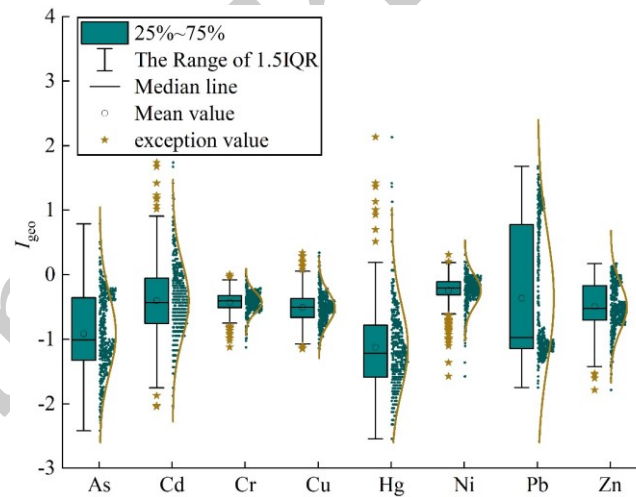


Fig. 5. Distribution box diagram of geo-accumulation index of eight heavy metals in surface soil

3.6. Health Risk Assessment

3.6.1. Non-carcinogenic Health Risk Assessment

As shown in Table 9, the non-carcinogenic health risk index (HQ) of 8 heavy metal elements in the soil of the study area for the population (adults and children) does not exceed 1. The average HQ values are ranked as follows: $Cr > As > Pb > Ni > Cu > Zn > Cd > Hg$. It can be seen that the primary heavy metal elements causing

non-carcinogenic risk to the population in the study area are Cr and As. The average values of the comprehensive non-carcinogenic health risk index HI for adults and children are $4.36\text{E-}2$ and $5.02\text{E-}1$, respectively, and the 95th percentile values are $6.40\text{E-}2$ and $7.41\text{E-}1$, respectively (Fig. 6), both of which are less than the non-carcinogenic health risk threshold of 1, indicating that there is no non-carcinogenic health risk for the population in the study area, which can be ignored.

Table 9. Non-carcinogenic health risk index of heavy metals in soil for different population groups in the study area

Element	Adult				Child			
	Hand-to-Mouth	Inhalation	Dermal	HQ _i	Hand-to-Mouth	Inhalation	Dermal	HQ
As	1.52E-02	7.08E-06	3.27E-07	1.53E-02	1.78E-01	1.10E-05	6.32E-07	1.78E-01
Cd	1.65E-04	7.70E-06	1.45E-07	1.74E-04	1.94E-03	1.19E-05	2.81E-07	1.95E-03
Cr	1.69E-02	8.27E-04	7.46E-06	1.78E-02	1.99E-01	1.28E-03	1.44E-05	2.00E-01
Cu	5.30E-04	2.46E-07	1.56E-08	5.31E-04	6.23E-03	3.80E-07	3.02E-08	6.23E-03
Hg	8.97E-05	1.46E-07	1.12E-08	9.02E-05	1.05E-03	2.26E-07	2.16E-08	1.05E-03
Ni	1.21E-03	1.25E-04	3.95E-08	1.34E-03	1.42E-02	1.94E-04	7.63E-08	1.44E-02
Pb	8.36E-03	3.87E-06	4.98E-07	8.41E-03	9.81E-02	5.99E-06	9.61E-07	9.81E-02
Zn	1.75E-04	8.13E-08	1.54E-09	1.75E-04	2.05E-03	1.26E-07	2.98E-09	2.05E-03
HI	4.36E-02				5.02E-01			

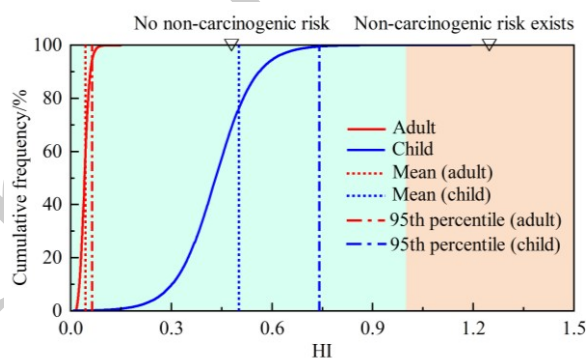


Fig.6. Probability distribution of non-carcinogenic risk from heavy metals in study area soils for different population groups

3.6.2. Carcinogenic Health Risk Assessment

The average carcinogenic health risk index (CR) of soil heavy metals in the study area is ranked as follows: Ni > Cr > As > Cd > Pb (Fig. 7). The average CR values of Ni, Cr and As for adults are $1.30\text{E-}5$, $8.36\text{E-}6$ and $2.17\text{E-}6$, respectively, and the 95th percentile values are $1.56\text{E-}5$, $9.60\text{E-}6$ and $3.51\text{E-}6$, respectively. The average CR values of Ni, Cr and As for children are $3.82\text{E-}5$, $2.37\text{E-}5$ and $6.34\text{E-}6$,

respectively, and the 95th percentile values are $4.57\text{E}-5$, $2.72\text{E}-5$ and $1.03\text{E}-5$, respectively. All significantly exceed the threshold of $1.00\text{E}-6$ for tolerable carcinogenic health risk, indicating that Ni, Cr and As elements pose tolerable carcinogenic risks to the population in the study area, and the risk for children is significantly higher than that for adults. The 95th percentile value of CR for Cd in children reaches $1.67\text{E}-6$, and 33.11% of the sampling points exceed $1.00\text{E}-6$, indicating that Cd is also an important elemental factor causing carcinogenic health risks for children in the study area. Both the average and 95th percentile values of CR for Pb do not exceed $1.00\text{E}-6$, indicating that the carcinogenic health risk of Pb to the population can be ignored. The average values of the comprehensive carcinogenic health risk index (TCR) for adults and children are $2.40\text{E}-5$ and $6.94\text{E}-5$, respectively, and the 95th percentile values are $2.78\text{E}-5$ and $8.08\text{E}-5$, respectively, indicating that soil heavy metals pose tolerable carcinogenic risks to the population, and the carcinogenic factors are most likely Ni, Cr and As.

The carcinogenic health risk index of heavy metals for children is greater than that for adults, indicating that soil heavy metals in the study area pose a greater carcinogenic health threat to children. This may be related to children's lower body weight, frequent hand-to-mouth behavior, higher respiration rate and other behavioral habits. On the other hand, children's physiological development is incomplete; with long-term exposure to polluted environments, they are more sensitive to the toxicity of heavy metals, resulting in higher carcinogenic risks caused by soil heavy metals for children (Cui YX et al., 2021). Therefore, children's exposure behavior management and environmental health control should be strengthened, and maintaining hand and mouth cleanliness is particularly important.

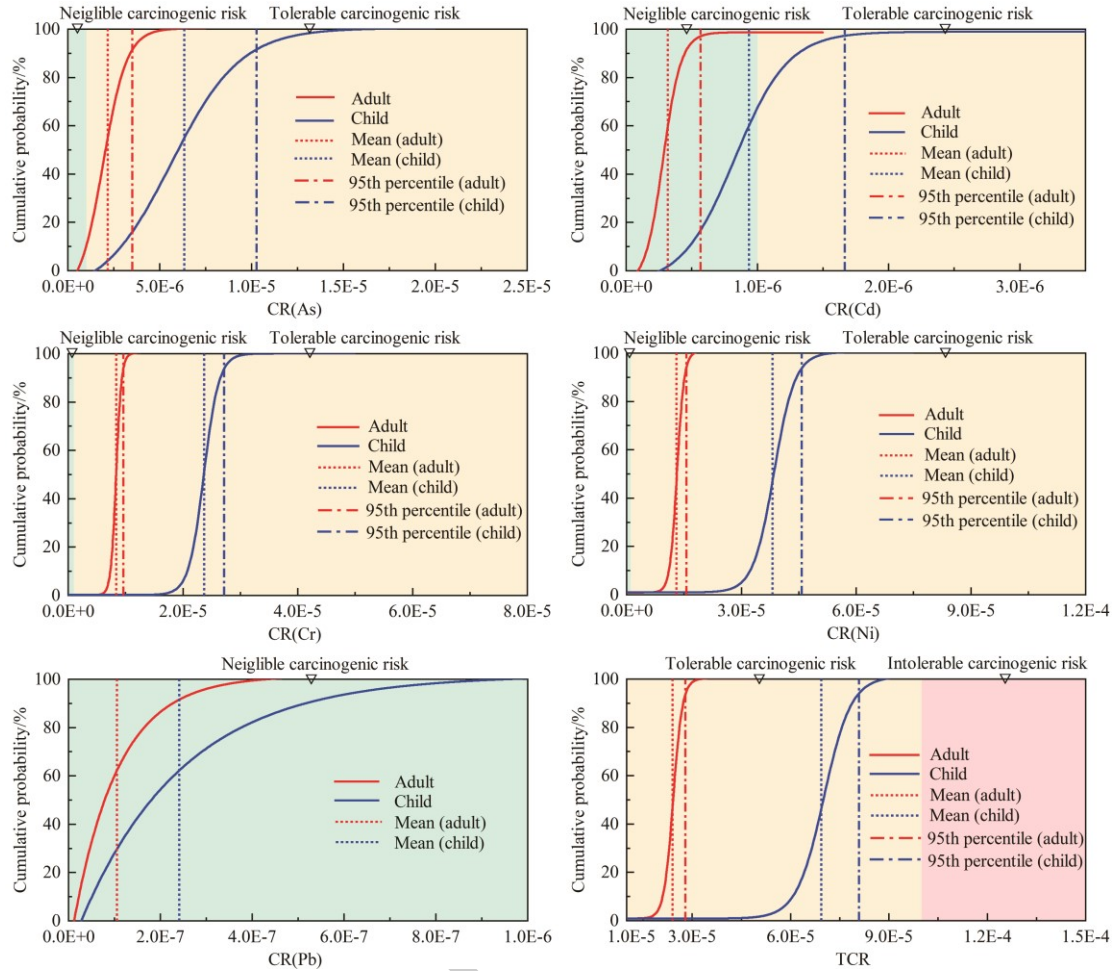


Fig.7. Probability distribution of carcinogenic risk from heavy metals in study area soils for population groups

3.6.3. Sensitivity Analysis

Sensitivity analysis is used to reveal the degree of influence of different parameters on health risk assessment results. The degree of influence on health risk results is positively correlated with the sensitivity value, and the absolute value of the sensitivity value indicates the degree of influence (Bao LR et al., 2020; Ma J et al., 2023; Zheng LL et al., 2025). The analysis results are shown in Figure 8.

For the non-carcinogenic health risk assessment results of adults (Fig. 8a), IngR has the highest sensitivity, explaining 66.71% of the sensitivity, followed by Pb, As and EF, explaining 11.87%, 10.56% and 9.85%, respectively. For the non-carcinogenic health risk assessment results of children (Fig. 8b), SL has the highest sensitivity, explaining 32.30% of the sensitivity, followed by IngR, EF, Pb and As, explaining 17.53%, 15.80%, 14.41% and 11.65%, respectively. For the

carcinogenic health risk assessment results of adults (Fig. 8c), IngR has the highest sensitivity, explaining 81.30% of the sensitivity, followed by EF, Ni, As and Cr, explaining 12.87%, 3.86%, 0.90% and 0.72%, respectively. For the carcinogenic health risk assessment results of children (Fig. 8d), SL has the highest sensitivity, explaining 91.93% of the sensitivity, followed by EF, Ni, IngR and Cr, explaining 3.76%, 1.21%, 1.11% and 0.37%, respectively. BW also contributes to a certain extent to the sensitivity of population health risk assessment results. The sensitivity value is negative, indicating that weight gain helps reduce health risks, especially for children.

In summary, IngR is the primary parameter factor affecting adult health risk, and SL is the primary parameter factor affecting children's health risk. Therefore, reducing adult intake frequency and lowering children's skin adherence factor can effectively reduce the health risks of the population in the study area. For example, people in the study area should pay attention to food hygiene and personal hygiene practices. Pb and As are the primary elemental factors affecting non-carcinogenic health risks of the population. Ni, As and Cr are the primary elemental factors affecting carcinogenic health risks of the population, which is related to the relatively high contents of Pb, Cr and Ni in the soil of the study area. Although the content of As in the soil of the study area is low, its sensitivity is high due to its large carcinogenic slope factor (SF) and strong toxicity (Huang JL et al., 2021).

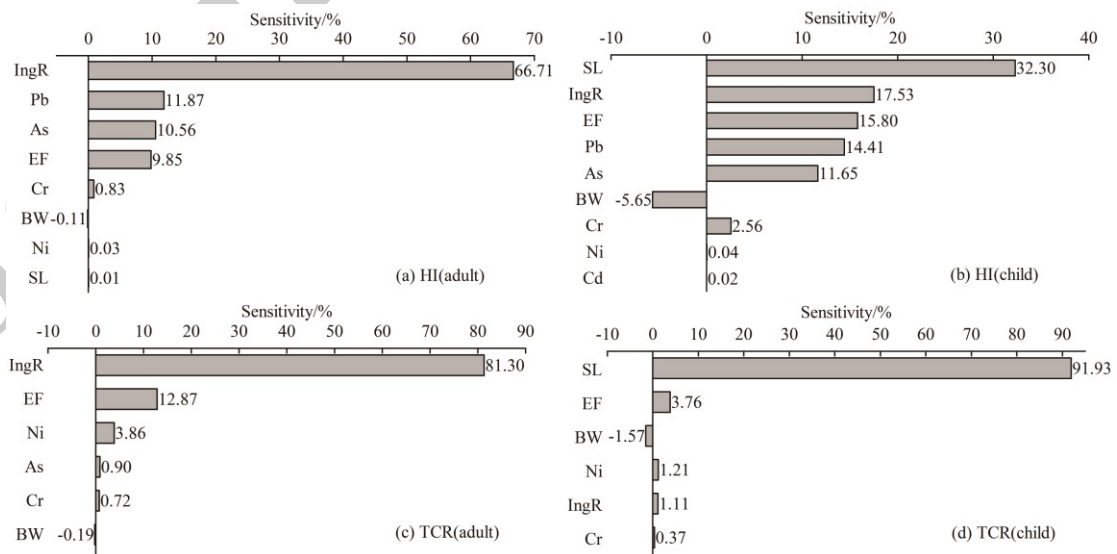


Fig.8. Distribution characteristics of non-carcinogenic and carcinogenic health risk sensitivity among the study population

3.6.4. Priority Control Factor Analysis

Given that the health risk index of children in the study area is significantly higher than that of adults, taking children's health risk as an example, the main control factors of soil in the study area are analyzed in combination with the APCS-MLR model analysis results. Figure 9 reflects the relationship between soil heavy metals, sources and children's health risks in the study area. The order of contribution of the four sources of soil heavy metals to HI in the study area is: F2 (46.32%) > F4 (26.85%) > F1 (23.65%) > F3 (3.19%), and the order of contribution to TCR is: F2 (79.23%) > F4 (15.96%) > F1 (4.03%) > F3 (0.78%).

The contribution of the four sources of soil heavy metals to children's health risks in the study area is positively correlated with the corresponding source contributions. F2 (natural parent material source) is not only the main source of soil heavy metals in the study area, but also the primary factor affecting children's health risks. The contribution rates of F2 to children's HI and TCR are 46.32% and 79.23%, respectively. In addition to F2, F4 (agricultural fertilizer and pesticide source), F1 (agricultural irrigation source) and F3 (atmospheric deposition source closely related to anthropogenic factors) are all closely related to human activities and can be combined to represent anthropogenic sources. The contribution of anthropogenic sources accounts for 58.35%, and their contributions to HI and TCR are 53.68% and 20.77%, respectively. Therefore, the carcinogenic and non-carcinogenic health risks caused by anthropogenic sources cannot be ignored.

Combined with health risk assessment results, sensitivity analysis and source-oriented health risk assessment, natural parent material sources are the main contributing sources of heavy metal risks in the study area, and Ni and Cr are the main contributing elements. Although they are not closely related to human activities, it is still necessary to strengthen the monitoring of soil Ni and Cr pollution in the study area. The mixed sources of agricultural activities represented by agricultural fertilizer and pesticide sources and agricultural irrigation sources are the priority control sources of soil heavy metal risks in the study area, and As and Cd are the priority control elemental factors.

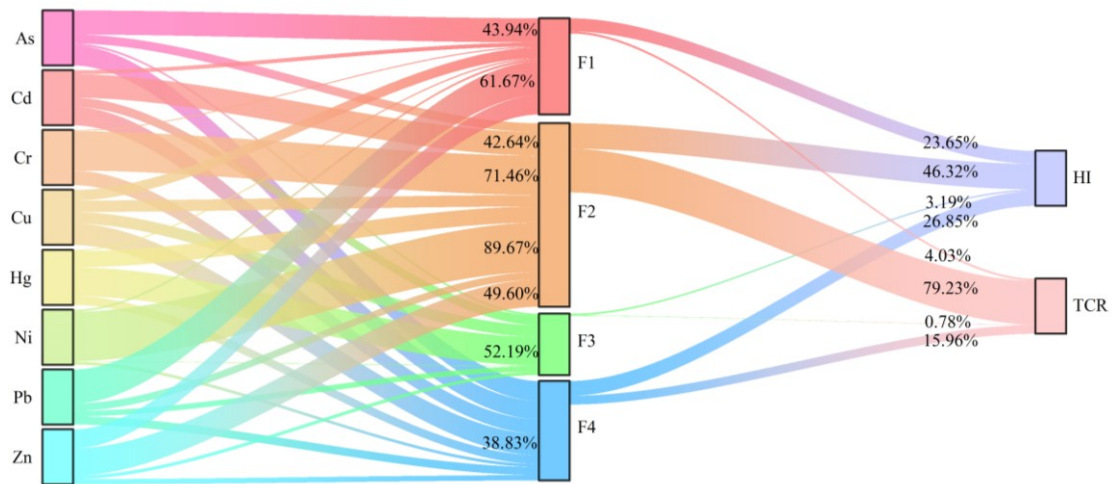


Fig. 9. Sankey map of health risks of heavy metals in soil

4. Conclusion

(i) The average contents of Pb, Ni, Cd and Cr in the surface soil of the research area are higher than the background values of surface soil in Yunnan Province, among which the enrichment of Pb is more prominent. Spatial variation structure analysis shows that Cd and Hg are randomly distributed and greatly affected by anthropogenic factors. As, Cr, Cu, Ni, Pb and Zn have significant positive spatial autocorrelation, mainly controlled by soil parent material, but also affected by human activities.

(ii) The main sources of soil heavy metals in the research area are natural parent material sources, agricultural irrigation sources, agricultural fertilizer and pesticide sources, and atmospheric deposition closely related to anthropogenic factors, with contribution rates of 41.65%, 21.85%, 22.58% and 13.92%, respectively. Part of the soil heavy metals comes from natural sources, and the remaining sources are largely closely related to anthropogenic factors, especially the contribution of agricultural activities.

(iii) The geo-accumulation index shows that the enrichment degrees of Cd, Hg and Pb are the highest, while the enrichment degrees of other heavy metal elements are relatively low. The evaluation results of the health risk assessment model based on Monte Carlo simulation show that soil heavy metals in the study area pose no non-carcinogenic health risk to the population, but pose a certain degree of carcinogenic health risk, and the risk for children is significantly higher than that for adults. The main contributing factors to carcinogenic health risks are Cr, Ni and As.

Combined with health risk assessment results, sensitivity analysis and source-oriented health risk assessment, natural parent material sources are the main contributing sources of heavy metal risks in the study area, and Ni and Cr are the main contributing elements. Although they are not closely related to human activities, it is still necessary to strengthen the monitoring of soil Ni and Cr pollution in the study area. The mixed sources of agricultural activities represented by agricultural fertilizer and pesticide sources and agricultural irrigation sources are the priority control sources of soil heavy metal risks in the study area, and As and Cd are the priority control elemental factors.

CRedit authorship contribution statement

Lei Xu conceptualized the entire paper and prepared the manuscript. Yi-fan Yu, Jun-xiang Zhang, Yong-jun Yang and Zi-jian Wang collected soil samples. Tai-tang Guo, Meng-sheng Zhao, and Ming-long Yang conducted the preliminary processing, shipping, and quality control of the samples. Xiao Jiang, Zhi-wei Li, Jiang-tao Pan and Yan-xun Cheng processed the geochemical data and plotted the illustrations. Chang-wei Linghu supervised the work and contributed new findings. All authors discussed the results, drew conclusions, and contributed to the manuscript.

Declaration of competing interest

The authors declare no conflict to interest.

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