

## Article

# Migration and Accumulation Mechanisms of Heavy Metals in Soil from Maoniuping Rare Earth Elements Mining, Southwest China

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**Abstract:** The Maoniuping Rare Earth Elements (REE) deposit, the second largest light REE deposit in the world, has been mined for decades, with serious impacts on the surrounding environment. However, the impact of mining on heavy metals in the downstream area (Nanhe River Basin) has not been systematically documented. To address this issue, this study explored the extent, transport, and accumulation of heavy metal contamination in the Nanhe River Basin through field surveys (2946 topsoil samples and four vertical soil sections) and regional geographic attributes (e.g., mining area, river, and elevation) combined with a variety of methods such as statistics, geostatistics, spatial analysis, geo-accumulation index, and potential ecological risk index. The results showed that soils in the Nanhe River Basin presented different degrees of heavy metal pollution, with Pb and Cd being the most abundant, and the soils as a whole showed moderate-heavy ecological risks. The spatial distribution and correlation of heavy metals exhibited similar distribution patterns and sources. Further analyses revealed that mining of REE in Maoniuping was the main source of heavy metal pollution in the Nanhe River Basin, with heavy metals entering the soil through runoffs. At the same time, mining activities led to the migration of heavy metals in different directions in the Nanhe watershed, i.e., about 1.3 km horizontally, 16 km longitudinally, and more than 1 m vertically. In addition, about 38.1 km<sup>2</sup> of the watershed is contaminated by mine wastes, which is 6.6 times the size of the mining area. In order to mitigate the threat of heavy metals, the local government has implemented water diversion projects and crop conversion in the Nanhe River Basin. This study provides a reference for research on the environmental problems caused by the exploitation of REE mines and other mineral resources.

**Keywords:** Maoniuping REE deposit; soil; heavy metals contamination; three-dimensional diffusion



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## 1. Introduction

China possesses significant strategic reserves of Rare Earth Elements (REE) ores, with key deposits located in Bayan Obo, Inner Mongolia; Ganzhou City, Jiang Xi; and Maoniuping, Sichuan Provinces [1–4]. The excavation or/and beneficiation of REE from

minerals present significant challenges, often requiring a large number of chemical reagents and complicated extraction processes [5,6]. These methods caused substantial negative environmental impacts, raising concerns in recent years.

Table 1 lists pollutants generated by REE mining and metallurgy, which have diverse environmental consequences. These include soil erosion and plant withering [7–10], dust pollution [11,12], nitrogen and ammonia eutrophication [13,14],  $SO_4^{2-}$  overdose [15], REE overdose [2,16–19], radioactive contamination [20,21], and heavy metals contamination [1–3,16,17,21,22]. Among these, heavy metal contamination constitutes a severe environmental concern.

**Table 1.** Environmental issues from REE mining.

| Deposits           | Locations                           | Pollutants                  | Environmental Consequences                                                                                                                                                 | References |
|--------------------|-------------------------------------|-----------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|
| Bayan Obo REE mine | Inner Mongolia, China.              | Polymetallic contamination. | Topsoil was affected by waste rock and tailings, with the Cd exhibiting 9.18 times higher than the control value.                                                          | [1]        |
| Bayan Obo REE mine | Inner Mongolia, China.              | REE, Cd, Pb, and Ag.        | Multiple heavy metals have reached concerning levels of pollution, with REE posing the most significant threat.                                                            | [2]        |
| Bayan Obo REE mine | Inner Mongolia, China.              | REE.                        | The elemental pollution level in dust has reached severe or extreme pollution.                                                                                             | [11,12]    |
| Bayan Obo REE mine | Inner Mongolia, China.              | REE, Mn, Zn, Co, and Ni.    | Groundwater pollution is mainly caused by high concentrations of heavy metals and REE, while surface water is polluted by high concentrations of REE and ammonia nitrogen. | [16]       |
| IAT-Res mine       | Jiangxi Province, China.            | Pb, Zn, Ni, Mn, and Co.     | Topsoil metals exceed the background reference for the province.                                                                                                           | [3]        |
| IAT-Res mine       | Northern Guangdong Province, China. | Mn, Zn, Cd, and Pb.         | Cd and Mn identified as the primary contaminants.                                                                                                                          | [23]       |
| IAT-Res mine       | Dingnan County, Ganzhou City.       | Ce and Eu.                  | Contamination mainly coming from abandoned tailings.                                                                                                                       | [17]       |
| IAT-Res mine       | Dingnan County, Ganzhou City.       | Pb.                         | The abandoned mine site and the surrounding soils are heavily contaminated with Pb.                                                                                        | [21]       |

Table 1. Cont.

| Deposits         | Locations                                 | Pollutants                        | Environmental Consequences                                                                                                                                                                                               | References |
|------------------|-------------------------------------------|-----------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|
| IAT-Res mine     | Dingnan and Longnan Counties, China.      | REE, ammonium, and sulfates, etc. | The lower concentrations of REE were dozens of times higher than control point.                                                                                                                                          | [15]       |
| IAT-Res mine     | Changting County, Fujian Province, China. | REE.                              | Mean value reaches 255.34 mg/kg, which accumulated from hilly areas to rice fields in the watershed.                                                                                                                     | [24].      |
| IAT-Res mine     | Changting County, Fujian Province, China. | Cd, Cu, and As.                   | The average value of Cd is 3.51, exceeding the standard by 10.7 times, and the soil environment is generally in the intensity of ecological risk.                                                                        | [22]       |
| IAT-Res mine     | Liancheng County, Fujian Province, China. | REE.                              | The range of REE of the Downstream soil was 727.96 mg/kg, which was 3.26 times of the background value of Fujian Province and posed a strong ecological risk.                                                            | [19]       |
| Xinlong REE mine | Anyuan County, Jiangxi Province, China.   | Total nitrogen, Mn, and REE.      | Water bodies exhibited elevated levels of total N, Mn, and REE. Mining caused 88% of pond and stream water unsuitable for agricultural purposes.                                                                         | [25]       |
| WCED-REO mine    | Longnan County, Jiangxi Province, China.  | Ammonia nitrogen pollution.       | Soil acidification is severe, and the ammonia–nitrogen content in deep soil is 12~40 times higher than the soil background level.                                                                                        | [14]       |
| Nuodong REE mine | Wuzhou City, Guangxi Province, China.     | Ammonia–nitrogen pollutant.       | The highest ammonia nitrogen pollutant in the monitoring area can reach 139.15 mg/L, with a cumulative pollution area of 1.95 km <sup>2</sup> . The water quality of streams and reservoirs has been seriously affected. | [26]       |

Table 1. *Cont.*

| Deposits               | Locations                                 | Pollutants                  | Environmental Consequences                                                                                                     | References |
|------------------------|-------------------------------------------|-----------------------------|--------------------------------------------------------------------------------------------------------------------------------|------------|
| Wenfang REE mine       | Liancheng County, Fujian Province, China. | Ammonia-nitrogen pollutant. | Significant increase in total nitrogen contents in the surface soil and water around the mining area, posing ecological risks. | [27]       |
| 815# REE mine          | Heping County, Guangdong Province, China. | Soil destruction and waste. | Exposed soil turned into a sandy landfill, causing soil destruction and waste, and preventing plants from growing.             | [8]        |
| -                      | Southern Jiangxi Province, China.         | Land degradation.           | Between 1988 and 2010, REE mining degraded 4281.8 hectares of land, with 2416.6 hectares severely impacted.                    | [28]       |
| REE mine               | Irma and Lovozero Rivers, Russia.         | REE.                        | Average values reached 561 and 736 mg/kg, respectively.                                                                        | [18]       |
| Mountain Pass REE mine | California, USA.                          | Radioactive contamination.  | Over 60 wastewater leakage incidents occurred between 1984 and 1998, releasing over 600,000 gallons of wastewater.             | [20]       |

Abbreviation: IAT-Res mine, ion adsorption type rare earth mines; Cd, Cadmium; Pb, Lead; Zn, Zinc; Ni, Nickel; Co, Cobalt; Mn, Manganese; Ag, Silver; Ce, Cerium; Eu, Europium; Cu, Copper; As, Arsenic.

The Maoniuping Rare Earth Element (REE) deposit, located in southwest Sichuan Province, is the world's second-largest light REE deposit [29]. Despite its economic significance, decades of intensive mining operations have led to severe environmental degradation in the region. The mining activities have left the landscape around the Maoniuping mine area heavily scarred, with vast expanses of farmland buried under tailings and slag piles. This has not only destroyed the natural geological landscape [10] but also increased the risk of landslides due to the instability of the accumulated tailings and waste rock [9]. Moreover, the improper disposal of non-standard tailings and untreated wastewater has resulted in the contamination of Nanhe River sediments with heavy metals and radioactive substances [4,30]. The Nanhe River Basin, which lies adjacent to the Maoniuping REE mine, serves as a critical agricultural zone and an important food production base for Mianning County. However, the mechanisms governing the migration and accumulation of heavy metals in the arable soils of the Nanhe Basin remain poorly understood. This knowledge gap hinders the development of effective strategies to mitigate the environmental and health risks posed by heavy metal contamination. Given the proximity of the basin to the mining area and its role in food production, there is an urgent need to investigate the pathways through which heavy metals are transported and accumulated in the soil. Such studies are essential to safeguard agricultural productivity, protect local ecosystems, and ensure the health and well-being of the communities reliant on this vital resource. In this regard, this study aims to investigate the spatial distribution characteristics of heavy metals

downstream of the Maoniuping REE deposit, employing geostatistical and multivariate statistical methods. Our objectives focus on the following: (1) assess the current status of soil heavy metal contamination in the Nanhe River Basin; (2) explore the sources of soil heavy metals; and (3) provide a preliminary assessment of heavy metal migration mechanisms and demarcate the extent of pollution. In this study, we reported the heavy metals released from the light REE mining in China, which provides a reference for other countries around the world (e.g., Australia, Ukraine, Russia, and the United States) to carry out mine waste prevention and control of REE mining.

## 2. Regional Setting and Method

### 2.1. Maoniuping REE Deposit and Mining History

The Maoniuping REE deposit, spanning  $\sim 5.80 \text{ km}^2$  in the upper Nanhe River, holds the distinction of being China's leading light REE deposit and the second largest REE deposit worldwide [29]. The deposit formed by late Oligocene hydrothermal veins and dominated by carbonatites. The primary ore minerals are bastnaesite, while vein minerals consist of nepheline, arfvedsonite, calcite, fluorite, quartz, biotite, K-feldspar, fluorophlogopite, as well as accompanying metal minerals such as galena [31,32].

The Maoniuping REE deposit was accidentally discovered during the galena exploration. Thereafter, to verify the reserve and mineralization mechanism of the deposit, detailed geological surveys were conducted spanning from 1985 to 1994, ultimately confirming the presence of a large light REE ore [33]. In the early stages of mining, REE resources were not subject to centralized control, with over 100 companies engaged in mineral extraction. This extensive mining resulted in the selective extraction of high-grade ores, the abandonment of lower-grade minerals, and a surge in illegal mining. These practices undoubtedly lead to resource wastage and environmental degradation [34]. In 2008, the mining administration imposed a one-mine-one-right policy to regulate the utilization of Maoniuping REE resources. Subsequently, the Sichuan Jiangtong REE Company successfully bid for the development rights to the mine. The company initiated efforts to arrange resources, manage mining operations, and implement environmental practices within the mine area. Over the next 15 years, formal mining operations commenced, yielding an annual output of  $\sim 30,000$  tons/year for REE concentrates, and a barite concentrate production rate of 140,000 tons. In a recent development in 2023, China Rare Earth Group Co., Ltd. (Ganzhou, China) (REGCC) has assumed control of the Maoniuping REE deposit [35].

### 2.2. Nanhe Basin

The Nanhe Basin, spanning approximately  $404.3 \text{ km}^2$  and extending 37 km, is composed of diluvial and alluvial deposits. The Nanhe River, originating from the eastern side of Maoniu Mountain, flows through the center of the basin before converging with the Anning River [36]. The basin's land is suitable for cultivating a diverse range of crops. The Nanhe Basin has an altitude of over 1500 m and exhibits a middle to high mountain landform. The climate in the Nanhe Basin belongs to the subtropical monsoon climate, featuring abundant sunshine. The precipitation exceeds 1000 mm, concentrating during summer and autumn [37].

Many north- and northeast-oriented faults and folds have been developed in the region, such as the Maoniuping dorsal slope, the Haha Fracture, and the Nanhe Fracture [38]. The stratigraphy is dominated by the Quaternary Pleistocene (Qp) and Holocene (Qh) sedimentary strata. These strata, composed primarily of granitic gravels, sands, and clays, are exposed in the western, central, and eastern portions of the basin, accounting for approximately 95% of the total basin (Figure 1). These strata exhibit a relatively loose structure.

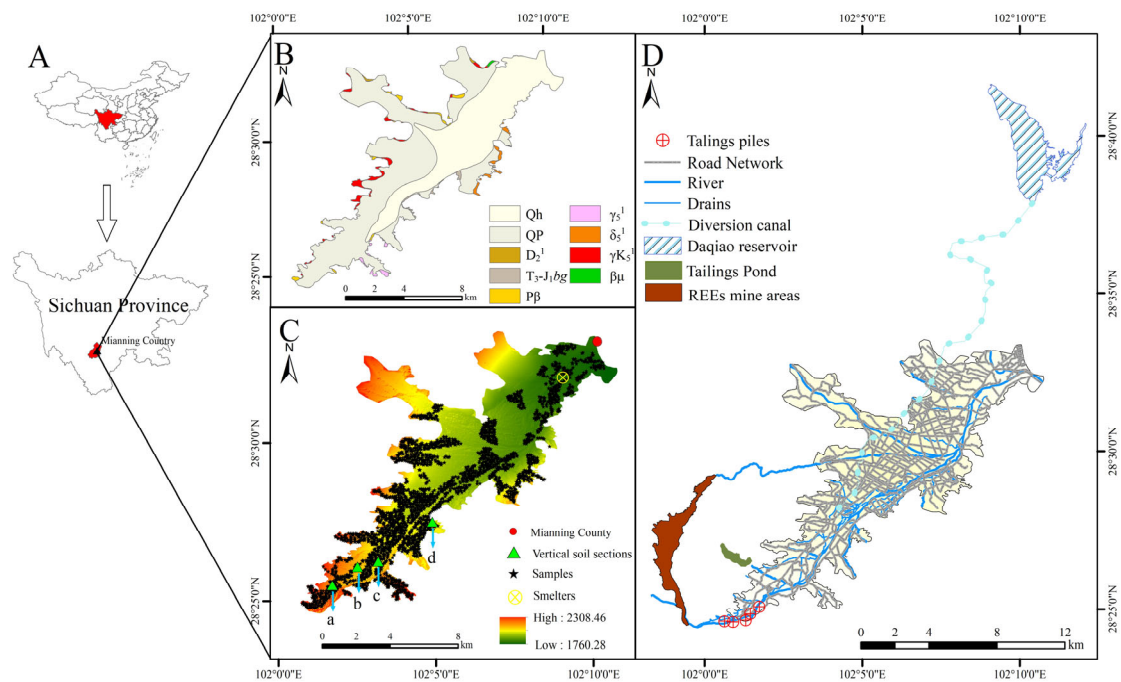


**Figure 1.** Regional maps of the Maoniuping REE deposit and the Nanhe Basin. (A) Location of the study area. (B) Regional geological background of the study area (Qh and Qp, Quaternary Holocene and Pleistocene, respectively;  $D_2^1$ , Lower Devonian;  $T_3-J_1bg$ , Upper Triassic-Lower Jurassic Baigowan Group;  $\gamma_5^1$ , Yanshanian fine-grained granite;  $\delta_5^1$ , Yanshanian fine-grained diorite;  $\gamma K_5^1$ , Yanshanian potassium diorite;  $P\beta$ , Permian basalt;  $\beta\mu$ , Diabase). (C) Area elevations, sampling points and vertical soil profiles (a, b, c, and d; green triangles). (D) Distribution of mining areas, tailings ponds, rivers, and roads.

### 2.3. Sample Collection and Analysis

A total of 2946 topsoil samples were collected in the Nanhe Basin (Figure 1). Sampling methodologies, including diagonal, plum blossom point, and checkerboard patterns, were employed and adapted to the specific characteristics of the terrain. The surface soil sampling method is based on the standard “Specification for Geochemical Evaluation of Soil Quality” (DZ/T 0295-2016) [39]. Specifically, a 20 m × 20 m grid was established at each designated sampling site. Before sample collection, surface debris, such as dead branches, leaves, animal and plant residues, and gravel, was removed using bamboo shovels and wooden chips. Subsequently, vertical soil columns were extracted from the center and corners of each grid to a depth of 0–20 cm using wooden shovels. Samples were combined to form composites weighing 1–1.5 kg, considering the gravel content and moisture conditions. After collection, the samples were placed in plastic bags, covered with cloth bags, and labeled.

The vertical soil section design considers factors including parent material, cultivation practices, planting structures, pollution source distribution, cultivated land concentration, and the spatial variability of geographical and geomorphological features. The vertical soil section samples were taken according to the standard “The Technical Specification for soil Environmental monitoring” (HJ/T 166-2004) [40]. When material composition remained consistent, samples were collected at intervals of approximately 10–20 cm along the vertical sampling point. Adjustments to the sampling interval were made to account for any observed anomalies. This approach allows for the investigation of vertical changes in the heavy metal pollution characteristics. A sampling depth of 1 m was used, with five soil samples collected at each vertical soil section: surface (0–20 cm), middle (20–40 cm, 40–60 cm), and deep (60–80 cm, 80–100 cm) samples (Table 2).

All samples were air-dried and smashed with a wooden hammer. The pulverulent samples were further processed in an agate ball mill or agate mortar, sieved through a 0.075 mm mesh size nylon sieve, and sealed into milled glass vials for further analysis. The total content of eight heavy metals (Cd, Pb, Cr, Cu, Ni, Zn, Hg, and As) was subjected to geochemical analysis. Using the method provided in the “Agricultural Industry Standard of the People’s Republic of China” (NY/T 1377-2007) [41], with distilled water as the leaching agent, the soil’s pH was determined potentiometrically in combination with a pH meter. The concentrations of Cd and Pb in the soil were quantified utilizing a graphite furnace atomic absorption spectrophotometer (PinAAcle 900, Perkin Elmer, Shelton, CT, USA) [42], whereas the levels of Cr, Cu, Ni, and Zn were ascertained through the application of a flame atomic absorption spectrophotometer (PinAAcle 900, Perkin Elmer, USA) [43]. Moreover, the determination of Hg and As within the soil was conducted via atomic fluorescence spectrophotometry (AFS, FL-100, LWL, Beijing, China) subsequent to microwave-assisted digestion [44]. The detection limits were 0.01, 0.1, 4, 1, 3, and 10 mg/kg for Cd, Pb, Cr, Cu, Ni, and Zn, respectively, and 0.002 and 0.01 mg/kg for Hg and As, respectively. Meanwhile, regional geographic datasets were collected to help explain the dispersal of heavy metals from the Maoniuping REE mine (Table 2).

**Table 2.** Description and acquisition methods of the geographical attributes of the Nanhe Basin.

| Attributes                            | Description                                                                                                                                                                                                                                                             | Sources                                                                                                                                                                                                                          |
|---------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Topsoil                               | Mixed soil samples (0~20 cm) of arable land in the Nanhe Basin were collected and analyzed for pH, Cd, Pb, Cr, Cu, Ni, Zn, Hg, and As, performing national standards [45]. Soil type is predominantly Anthrosols.                                                       | Field investigation in the Nanhe Basin.                                                                                                                                                                                          |
| Vertical soil section                 | Samples (vertical soil section a, b, c, and d) were collected from the bottom to the surface at a depth of ~1 m, 10–30 cm intervals. The four vertical soil sections are positioned at ~4.4, ~6.09, ~7.10, and ~10.79 km from the Maoniuping REE deposit, respectively. |                                                                                                                                                                                                                                  |
| Elevation, slope, and slope direction | The Digital Elevation Model (DEM) was derived from the aerial dataset. The slope and slope direction are extrapolated from the DEM dataset.                                                                                                                             | Shuttle Radar Topography Mission [46], the most complete and highest-resolution DEM. The 2000 Project was a joint endeavor of NASA, the National Geospatial-Intelligence Agency, and the German and Italian Space Agencies [47]. |

Table 2. Cont.

| Attributes     | Description                                                                                                                                                                              | Sources                                                                                                      |
|----------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------|
| River          | The Nanhe Basin consists of the main river channel, tributary streams, and both natural and artificial canals.                                                                           | Field survey and mapping from remote sensing images.                                                         |
| Mine area      | The land occupation of Maoniuping REE Mine (~5.80 km <sup>2</sup> ).                                                                                                                     |                                                                                                              |
| Tailings ponds | The mineral dressing residue in the Maoniuping REE mine comprises four non-standard waste dumps (28°24'36" N; 102°00'37" E) and one tailings pond (Figure 1, 28°26'36" N, 102°00'57" E). |                                                                                                              |
| REE smelter    | Metallurgy and ancillary facilities for concentrates of REE minerals (28°32'4" N, 102°9'0" E).                                                                                           |                                                                                                              |
| Stratum        | Pleistocene and Holocene sedimentary strata, intrusive rocks of the Yanshanian period in the Nanhe Basin                                                                                 | Manual extraction of 250,000 geological maps from the National Basic Geographic Information Database, China. |

#### 2.4. Kriging Interpolation

Kriging interpolation is a major part of the study of geostatistics, which essentially uses the structural characteristics of the raw data of regionalized variables and their corresponding variance functions to make linear unbiased predictions of the value of a regionalized variable at an unknown point location [48].

$$z^*(x_0) = \sum_{i=1}^n w_i z(x_i) \quad (1)$$

$$w_i = \frac{\frac{1}{d_i^2}}{\sum_{i=1}^n \frac{1}{d_i^2}} \quad (2)$$

where  $z^*(x_0)$  is an estimate of the sampling point at point  $X_0$ ,  $z(x_i)$  is the actual measurement at  $x_i$ ,  $w_i$  is the weight assignment of  $x_i$ ,  $n$  is the number of points in the neighborhood used to estimate the value of  $x_0$ , and  $d_i$  is the distance between the estimated point and the actual point.

#### 2.5. Geographically Weighted Regression

Geographically weighted regression (GWR) generates local regression parameters that reveal localized changes between variables.

$$y_i = \beta_0(u_i, v_i) + \sum_{k=1}^P \beta_k(u_i, v_i) x_{ik} + \varepsilon_i (k = 1, 2, 3 \dots) \quad (3)$$

where  $y_i$  represents the value of the dependent variable at location  $i$ ,  $x_{ik}$  denotes the value of the independent variable at position  $i$ ,  $(u_i, v_i)$  represents the geographic coordinates of the regression analysis points,  $\beta_k$  is the point coefficient in the regression model, and  $\beta_0$  is the intercept term.

## 2.6. Geo-Accumulation of Heavy Metals

The geo-accumulation index ( $I_{geo}$ ) reflects the natural variability of heavy metals and highlights the impact of anthropogenic input on the environment [49].

$$I_{geo} = \log_2(C_i/1.5B_i) \quad (4)$$

where  $C_i$  represents the measured value of soil,  $B_i$  denotes the background concentration, and 1.5 is a correction parameter commonly employed to discern between natural fluctuations or anthropogenic influences [50]. The  $I_{geo}$  is classified into seven categories: uncontaminated ( $\leq 0$ , Class I), uncontaminated to moderately contaminated (0~1, Class II), moderately contaminated (1~2, Class III), moderately to heavily contaminated (2~3, Class IV), heavily contaminated (3~4, Class V), heavily polluted to extremely polluted (4~5, Class VI), and extremely polluted ( $\geq 5$ , Class VII).

In the study, the Chinese environmental quality standards [45] were used as guiding values for heavy metal pollution. “Soil environmental quality risk control standard for soil contamination of agricultural land” (GB 15618-2018) [51] is a mandatory standard for heavy metal content in farmland soil issued by China. It specifies the risk screening and control values for soil heavy metal pollution.

## 2.7. Potential Ecological Risk Index

The potential ecological risk index (RI) takes into account the nature of heavy metals and biological sensitivity to heavy metal contamination, as well as differences in regional background values for heavy metals, effectively eliminating the effects of regional differences and heterogenic contamination [52,53]. Also, it takes into account the comprehensive impact of heavy metals on the environment [54].

$$C_f^i = C_s^i/B_i \quad (5)$$

$$E_r^i = T_r^i \times C_f^i \quad (6)$$

$$RI = \sum_{i=1}^n E_r^i \quad (7)$$

where  $E_r^i$  is the single coefficient of potential ecological hazards,  $T_r^i$  is the toxicity response coefficient of heavy metal  $i$ , which represents the toxicity level of heavy metals and the sensitivity of the environment to heavy metal contamination, and the toxicity corresponding coefficients of the eight soil heavy metals in this study were set to refer to the results of Hakanson and Zhang’s study ( $T_r^{Pb} = 5$ ,  $T_r^{Cd} = 30$ ,  $T_r^{Zn} = 1$ ,  $T_r^{Cu} = 5$ ,  $T_r^{Cr} = 2$ ,  $T_r^{Ni} = 5$ ,  $T_r^{Hg} = 40$ , and  $T_r^{As} = 10$ ) [53,55], and  $C_s^i$  is the measured concentration of heavy metal  $i$  (mg/kg). The grading criteria for the soil potential ecological risk index are shown in Table 3.

**Table 3.** Grading criteria for evaluating the potential ecological risk index.

| E <sub>i</sub> Classification Ranges | RI Classification Ranges | Pollution Level   |
|--------------------------------------|--------------------------|-------------------|
| $E_i < 40$                           | $RI < 150$               | Low risk          |
| $40 \leq E_i < 80$                   | $150 \leq RI < 300$      | Moderate risk     |
| $80 \leq E_i < 160$                  | $300 \leq RI < 600$      | Considerable risk |
| $160 \leq E_i < 320$                 | $600 \leq RI$            | High risk         |
| $320 \leq E_i$                       |                          | Very high risk    |

## 2.8. Multivariate and Geostatistical Analysis

The data processing was performed using Excel for descriptive statistics; data spatial interpolation methods and buffer analysis were performed using GIS technology to explore the spatial distribution and migration patterns of heavy metals. Among these, buffer

analysis refers to a spatial analysis method in GIS used to determine the influence range or adjacent areas of geographic features. Its core involves creating a buffer area of a certain width around specific geographic features and analyzing other features or attributes within that area. Correlation analysis and principal component analysis were performed on the data using SPSS 26 to determine the correlation of the data with geographic elements and sources of heavy metal pollution. Maps were prepared using Origin 2022.

### 3. Results

#### 3.1. Topsoil

The topsoil's pH in the Nanhe Basin, as presented in Table 4, ranged from 4.12 to 7.79 (arithmetic mean 5.62). Notably, acidic soils ( $4.5 \leq \text{pH} < 5.5$ ) accounted for 40.4%; weakly acidic soils ( $5.5 \leq \text{pH} < 6.5$ ) accounted for 56.4%. The remaining topsoil displayed a neutral to alkaline pH range (6.5 to 8.5), suggesting that a significant portion of the soil within the basin likely exhibited acidic characteristics. The contents of Pb, Cd, Cu, Zn, Ni, Cr, Hg, and As varied from 12 to 5180 mg/kg (arithmetic mean 338.8 mg/kg), 0.06 to 9.88 mg/kg (arithmetic mean 0.52 mg/kg), 3 to 179 mg/kg (arithmetic mean 39 mg/kg), 42 to 499 mg/kg (arithmetic mean 152 mg/kg), 4 to 177 mg/kg (arithmetic mean 38 mg/kg), 7 to 876 mg/kg (arithmetic mean 69 mg/kg), 0.01 to 4.81 mg/kg (arithmetic mean 0.13 mg/kg) and 0.12 to 35.20 mg/kg (arithmetic mean 3.45 mg/kg), respectively. According to the screening values in the Chinese soil quality standard (GB 15618-2018) [51], the exceedance rates of Cd, Pb, Cu, Zn, Ni, Cr, Hg, and As were 62.5%, 53.2%, 15.8%, 12.0%, 3.3%, 2.5%, 1.2%, and 0%, respectively, indicates soil heavy metals display different levels of contamination, with Cd and Pb being the most abundant contamination. Based on the exceedance rate found the pollution level follows this order: Cd > Pb > Cu > Zn > Ni > Cr > Hg > As (Table 4).

**Table 4.** Topsoil geochemical statistics in the Nanhe Basin.

| Soil                      | Parameters         | pH   | Heavy Metal Concentrations in the Soil (mg/kg) |       |      |      |      |      |       |      |
|---------------------------|--------------------|------|------------------------------------------------|-------|------|------|------|------|-------|------|
|                           |                    |      | Cd                                             | Pb    | Cu   | Zn   | Ni   | Cr   | Hg    | As   |
| Topsoil<br>(2946 samples) | Min                | 4.12 | 0.06                                           | 11    | 3    | 42   | 4    | 7    | 0.01  | 0.12 |
|                           | Max                | 7.79 | 9.88                                           | 5180  | 179  | 499  | 177  | 876  | 4.81  | 35.2 |
|                           | Mean               | 5.6  | 0.52                                           | 339   | 39   | 152  | 38   | 69   | 0.13  | 3.45 |
|                           | Median             | 5.6  | 0.4                                            | 117   | 37   | 139  | 36   | 63   | 0.09  | 2.79 |
|                           | CVs (%)            | 8.5  | 83.3                                           | 135.9 | 36.8 | 36.7 | 42.8 | 61.5 | 115.3 | 70.9 |
|                           | Exceeding rate (%) | /    | 62.5                                           | 53.2  | 15.8 | 12   | 3.3  | 2.5  | 1.2   | 0    |

Note: Exceeding rate (%): percentages of samples that exceed the screening value of the national arable soil standard, including the slightly contaminated and severely contaminated soil [45].

The coefficient of variation (CV) is a statistical measure used to quantify the degree of variability and is classified into three categories: low variation ( $\leq 15\%$ ), medium variation ( $15\sim 35\%$ ), and high variation ( $> 35\%$ ) [56]. The CV of pH in the topsoil equals 8.5%, representing a low variation. In contrast, the CVs for heavy metals were collectively higher than 35%, signifying a strong anthropogenic disturbance in the Nanhe Basin (Table 4).

Figure 2 illustrates the spatial distribution of heavy metals in the Nanhe Basin's topsoil, displaying distinct patterns. For example, overlapping areas of high Pb and Zn concentrations are observed near the upstream riverbanks. However, in the downstream, the Pb enrichment is particularly significant. Meanwhile, Pb, Cd, and Zn contents are relatively concentrated in the Nanhe River's main channel, with Pb showing the highest concentration and Zn exhibiting greater dispersion. This result suggests a potential connection between the distribution of heavy metals and the ongoing REE mining upstream. The

remaining heavy metals (Cu, Ni, Cr, Hg, and As) are relatively depleted near the river channel and enriched away from the river. Among them, the Ni levels are most negatively correlated with the river.

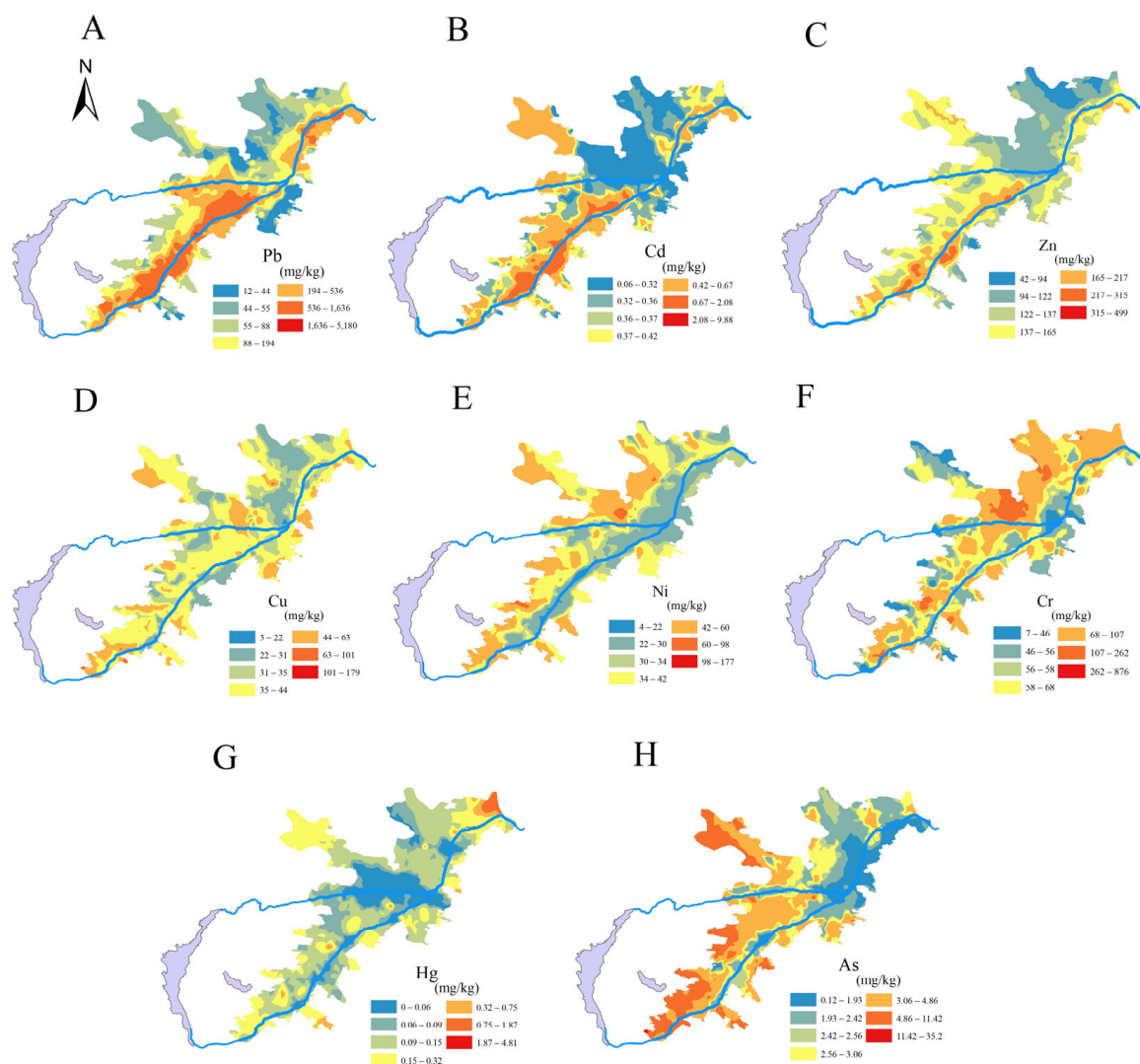


**Figure 2.** Spatial distributions of heavy metals in the Nanhe Basin. ((A) Pb, (B) Cd, (C) Zn, (D) Cu, (E) Ni, (F) Cr, (G) Hg, and (H) As; Blue line, Rivers).

### 3.2. Vertical Soil Sections

Deep soil demonstrates a higher degree of alkalinity compared to topsoil. The mean values of the vertical soil sections' soil are 6.21 (vertical section a), 6.01 (vertical section b), 7.04 (vertical section c), and 6.05 (vertical section d), and their CVs are 1.8% (vertical soil section a), 1.1% (vertical section b), 1.1% (vertical section c), and 2.4% (vertical section d). A lower CV level indicates minimal acid-base fluctuation in the soil vertical section. On the other hand, the levels of Pb and Cd in vertical section c and in the topsoil of vertical soil section a surpassed the risk screening values [45]. Zn contaminated the surface and middle layers of vertical section c, whereas Cu only exceeded the risk screening values in the topsoil of vertical section c. Other heavy metals in the vertical soil sections did not exceed the limits.

Additionally, heavy metals exhibited distinct characteristics in the soil vertical sections. In vertical section a, the Pb, Cd, and Hg levels decreased with depth, while other heavy metals showed minimal vertical variation. In vertical section b, apart from the insignificant

change in Pb levels, a notable enrichment of heavy metals was observed in the middle layer. In vertical section c, heavy metals displayed two or three distinct distribution patterns, primarily including Pb, Cd, Cu, Zn, and Hg, whose levels significantly decreased with depth but unexpectedly increased at a depth of 60–80 cm. In vertical section d, the Pb, Cd, Cu, Zn, and Ni levels remained relatively constant.

### 3.3. Risk Assessment of Heavy Metals

#### 3.3.1. Geo-Accumulation Index

In this study, vertical sections a and b, positioned on steep slopes and distant from mining activities, display moderate heavy metal levels. Vertical section c, situated in a low-elevation, gently sloping area near the Nanhe River, exhibited higher heavy metal concentrations in both topsoil and deeper soil layers (Figure 3). Vertical section c, situated on the right bank of the Nanhe River at a higher elevation, is irrigated by local creeks and is free from the impacts of mining or industrial activities. The average concentrations of Pb, Cd, Zn, Cu, Ni, Cr, Hg, and As, in vertical section d were determined as 32.4, 0.14, 77, 22, 39, 70.4, and 6.11 mg/kg, respectively, significantly lower than the clean soil [45]. As such, vertical section d can serve as a reference site for establishing baseline heavy metal levels within the Nanhe Basin.

Using the established baseline, the  $I_{geo}$  of heavy metals in the topsoil was determined (Figure 4). The analysis revealed that areas with high heavy metal enrichment in the soil are primarily concentrated near the Nanhe River, exhibiting a diffuse pattern extending outwards from the river channel. This observation suggests that heavy metal migration within the Nanhe Basin is predominantly water-mediated.

Pb enrichment is prevalent, with only 21.0% of the samples classified as uncontaminated. Moderate to severe contamination was found in 39.2% of samples, and 2.8% showed extreme enrichment. Approximately 80.1% of soil Cd pollution is classified below moderate ( $0 \leq I_{geo} \leq 2$ ), while 12.2% of Cd pollution falls within the moderate to extreme range. Only two sites exhibited extreme Cd pollution. The Hg is somewhat enriched in the study area, with more than average (58%) of the soil samples exhibiting Hg contamination, mainly in the form of no-moderate contamination. Hg enrichment is primarily concentrated in the vicinity of the urban and residential areas of Mianning County. The influence of human activity on soil Hg content in proximity to the urban area is significant, a finding corroborated by other studies [57]. The enrichment of Zn and Cu in the topsoil is limited, with 9.3% and 3.0% of the samples classified as moderately or highly contaminated, respectively. The mobility of these metals in acidic soils may explain these low levels. Contamination by Cr, Ni, and As is minimal in the topsoil, with 90% or more of samples being uncontaminated. The  $I_{geo}$  in the topsoil of Nanhe Basin follows the sequence of  $Pb > Cd > Hg > Zn > Cu > Cr > Ni > As$ .

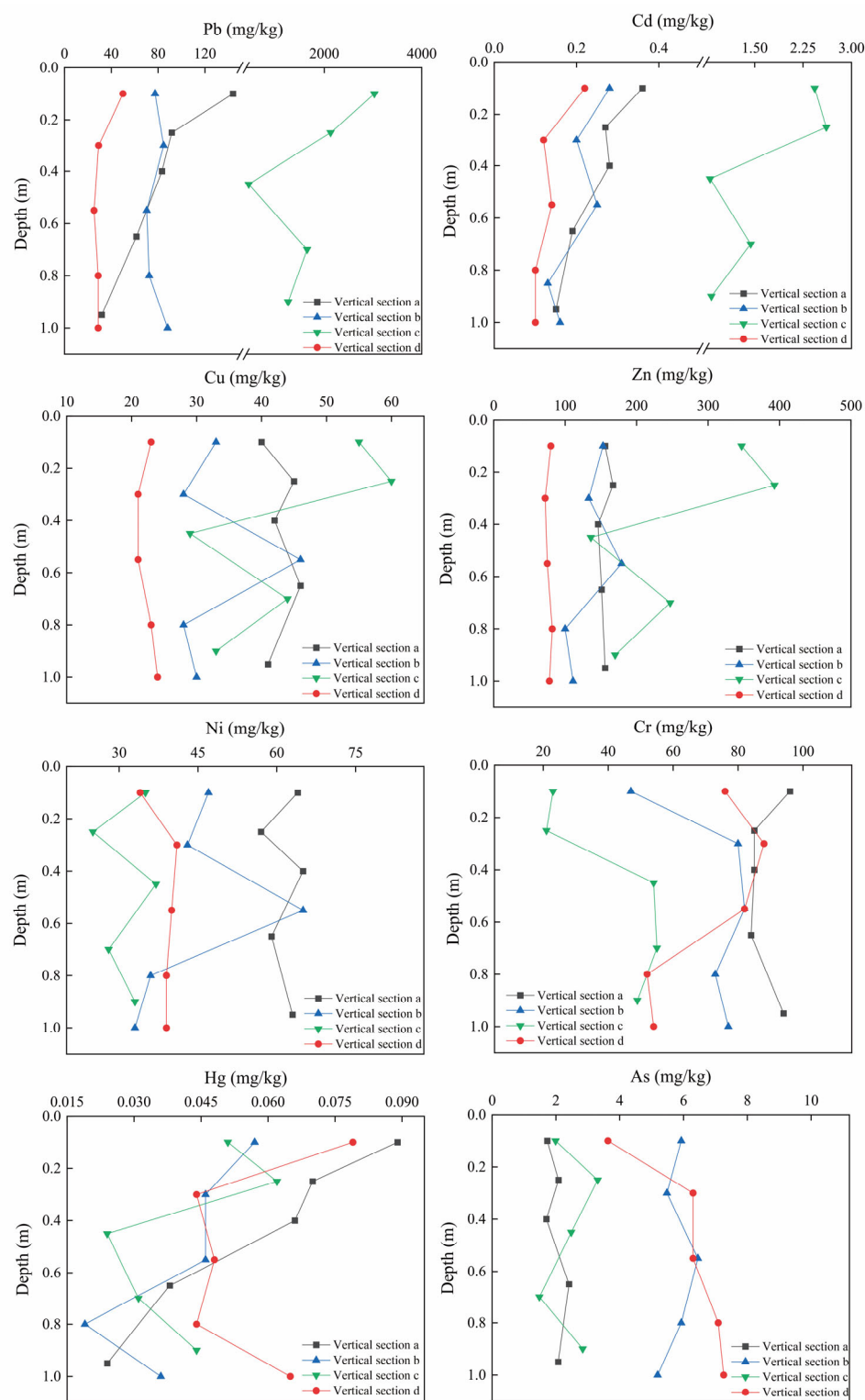


Figure 3. Vertical soil variation in heavy metals in the soil of Nanhe Basin.

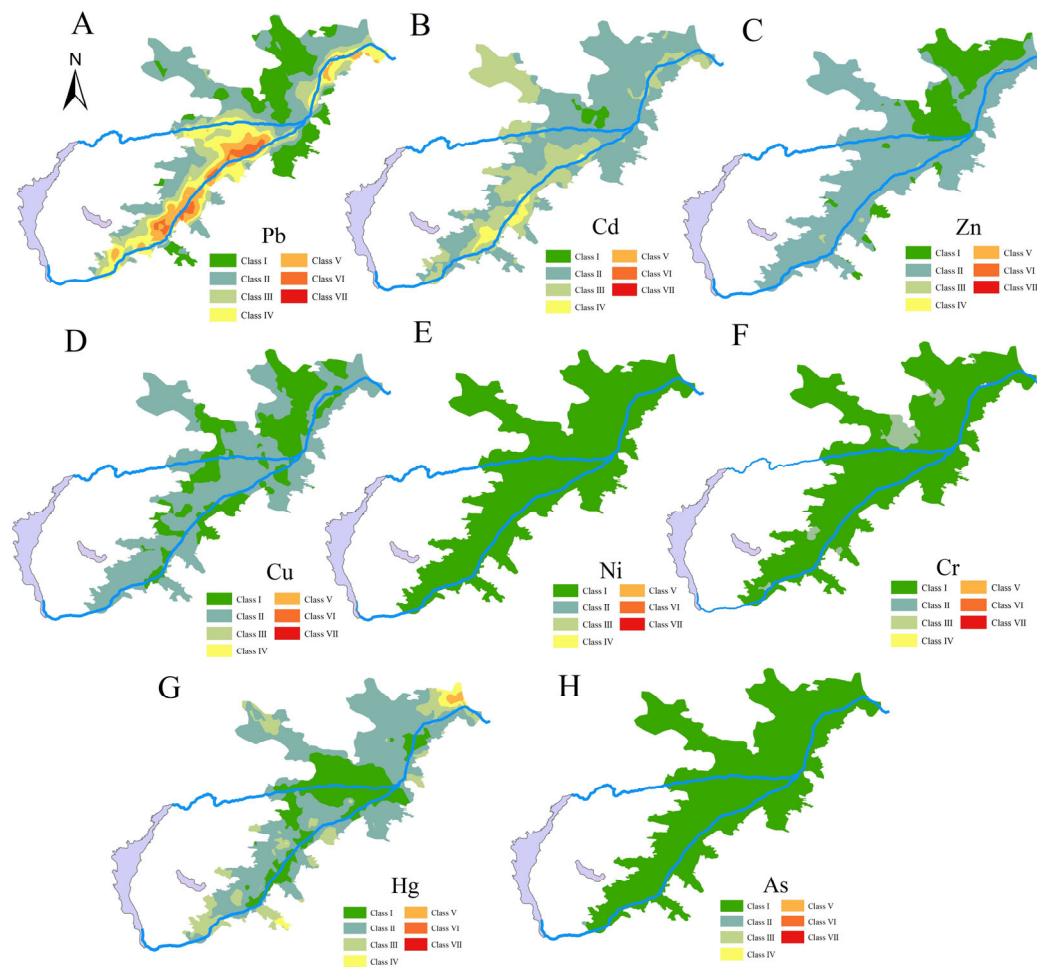


**Figure 4.** Geo-accumulation index ( $I_{geo}$ , see Equation (4) for details) of the distribution of heavy metals ((A)Pb, (B) Cd, (C) Zn, (D) Cu, (E) Ni, (F) Cr, (G) Hg and (H) As; Blue line, Rivers) in the Nanhe Basin.

### 3.3.2. Potential Ecological Risk Assessment

The RI of soil heavy metals in the studied area was systematically calculated, with the comprehensive statistical results presented in Table 5. The analysis demonstrated varying degrees of potential ecological risks associated with heavy metal contamination. Based on the individual ecological risk index ( $E_i$ ) values, the average  $E_i$  followed a descending order: Cd > Hg > Pb > Cu > As > Ni > Cr > Zn. Notably, only Cd, Pb, and Hg exhibited average  $E_i$  values exceeding 40, indicating significantly elevated ecological risks. In contrast, Cu, As, Ni, Cr, and Zn generally posed low ecological risks across most sampling sites. Specifically, Cd demonstrated the highest ecological risk with an average  $E_i$  of 114, peaking at an alarming 2179. This substantial value indicates that Cd contamination predominantly reached medium to heavy risk levels, with its average potential risk classified as heavy. Pb showed an average  $E_i$  of 52.3, with a maximum value of 799, suggesting that while its potential risk was primarily low, the average risk level reached moderate status. Hg presented an average  $E_i$  of 92, with an exceptionally high maximum value of 3436, reaching severe risk levels. The risk level distribution of Hg paralleled that of Cd, predominantly ranging from medium to heavy, with an average risk classified as medium. The pronounced risk levels associated with Hg may be attributed to two key factors: the region's naturally low geological background values and the inherently high toxicity coefficient of Hg.

**Table 5.** Evaluation results of potential ecological risk index.

|    | Parameters |        |       | Potential Risk Index Rating (%) |               |                   |           |                |
|----|------------|--------|-------|---------------------------------|---------------|-------------------|-----------|----------------|
|    | Min        | Max    | Mean  | Low Risk                        | Moderate Risk | Considerable Risk | High Risk | Very High Risk |
| Cd | 13.2       | 2179.4 | 114.6 | 5.5                             | 41.3          | 41.5              | 13.6      | 3.6            |
| Pb | 1.8        | 798.9  | 52.3  | 65.4                            | 11.4          | 13.4              | 9.1       | 0.6            |
| Cu | 0.7        | 40.7   | 8.8   | 99.9                            | 0.1           | 0.0               | 0.0       | 0.0            |
| Ni | 0.5        | 22.7   | 4.8   | 100.0                           | 0.0           | 0.0               | 0.0       | 0.0            |
| Zn | 0.5        | 6.4    | 2.0   | 100.0                           | 0.0           | 0.0               | 0.0       | 0.0            |
| Cr | 0.2        | 25.0   | 2.0   | 100.0                           | 0.0           | 0.0               | 0.0       | 0.0            |
| Hg | 6.4        | 3435.7 | 91.5  | 19.7                            | 40.3          | 27.5              | 10.9      | 1.6            |
| As | 0.2        | 57.6   | 5.6   | 99.8                            | 0.2           | 0.0               | 0.0       | 0.0            |
| RI | 56.7       | 3673.0 | 281.6 | 14.0                            | 53.2          | 28.3              | 4.5       | -              |

An analysis of the RI characteristics in the study area revealed that the RI values ranged from 57 to 3673, with a mean value of 282. The risk levels were distributed as follows: 14.0% of the soils were classified as low risk, 53.2% as moderate risk, 28.3% as heavy risk, and 4.4% as severe risk. This indicates that the majority of the soils in the study area (85.6%) exhibited medium to high ecological risk levels, highlighting a significant degree of environmental risk.

### 3.4. Provenance of Heavy Metals

The observed correlation revealed intrinsic connections and potential synergistic effects between heavy metals. Strong correlations generally suggest a shared origin, while weak correlations indicate distinct sources [58]. Concerning the non-normal distribution of heavy metal contents in the topsoil, Spearman's correlation coefficient ( $r$ ) was utilized to assess their interrelationship [59]. Figure 5 illustrates positive correlations between Pb and Zn ( $r_{Pb-Zn} = 0.60$ ,  $p < 0.01$ ), Pb and Cd ( $r_{Pb-Cd} = 0.58$ ,  $p < 0.01$ ), and Zn and Cd ( $r_{Zn-Cd} = 0.69$ ,  $p < 0.01$ ), suggesting a shared sources for these metals. Conversely, Pb shows a weak negative correlation with Ni ( $r_{Pb-Ni} = -0.35$ ,  $p < 0.01$ ) and Hg ( $r_{Pb-Hg} = -0.23$ ,  $p < 0.01$ ). Cu is only positively correlated with Zn ( $r_{Cu-Zn} = 0.48$ ,  $p < 0.01$ ) and Ni ( $r_{Cu-Ni} = 0.35$ ,  $p < 0.01$ ) and has weak or no correlation with other heavy metals. Ni has weak correlations with Cr, Hg ( $r_{Ni-Hg} = 0.35$ ,  $p < 0.01$ ), and As ( $r_{Ni-As} = 0.35$ ,  $p < 0.01$ ). The correlations show that soil heavy metals have multiple sources.

PCA is an algorithm that achieves dimensionality reduction by mapping high-dimensional features to lower dimensions. Also, it is often used to analyze soil heavy metal sources [60,61]. Its purpose is to focus single or multiple variables with similar characteristics into a common factor from the variation in the original variable that is explained through fewer common factors [62]. The Kaiser–Meyer–Olkin (KMO = 0.64) and Bartlett sphericity tests ( $p < 0.01$ ) confirmed the suitability of topsoil's data for PCA. On this basis, the results of PCA showed three principal components (Factor 1–3) with eigenvalues greater than 1.0 and that explained 70.2% of the total variance (Table 6). The table shows that PCA classifies soil heavy metals into three fractions, i.e., three different sources. More specifically, the first principal component (Factor 1) was mainly composed of Pb, Cd, and Zn. The Cu, Ni, and Cr were classified as the second principal component (Factor 2). The final principal component (Factor 3) contains Hg and As.



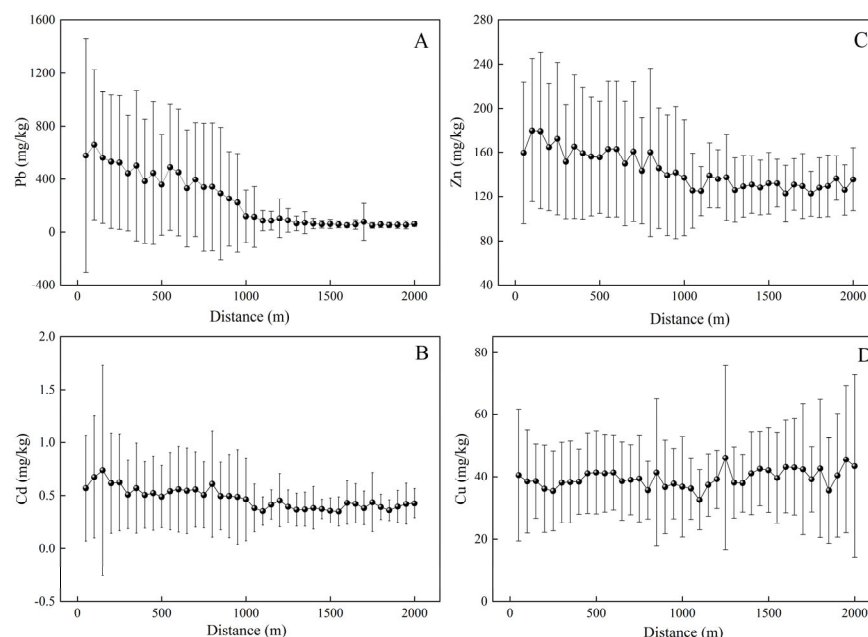


**Figure 6.** Error bar chart of heavy metals in the lateral migration of the Nanhe River ((A) Pb, (B) Cd, (C) Zn, (D) Cu). A 50 m interval buffer distance (perpendicular distance to river). The nodes are the statistics of the samples within the buffer area (a new area formed when the channel of the Nanhe River is extended outwards for a certain distance). The error bar represents the CVs, and the solid circles represent the average contents.

The lateral migration of other heavy metals varies. For instance, Cu concentrations remained relatively stable between 50 and 1050 m from the river (Figure 6D). Beyond this range, Cu concentrations increased, suggesting that while the Nanhe River may not contribute to Cu enrichment, it could facilitate its migration. Consequently, the influence of Maoniuping REE mining on the Nanhe Basin is more pronounced at ~1.35 km from the main channel.

### 3.5.2. Longitudinal Migration of Heavy Metals

We hypothesize that the impact of the Maoniuping REE mining extended up to ~16 km into the Nanhe Basin. Figure 7 demonstrates similar spatial variation patterns for Pb, Cd, and Zn along the Nanhe River. The concentrations of these metals initially increased between 3 and 7 km along the main channel, then decreased. A secondary increase was observed between 12 and 15 km, followed by a rapid decline and stabilization beyond 16 km. This fluctuation is likely due to a tributary on the left bank connected to the Maoniuping REE mine (Figure 1).

An upward trend in heavy metal concentrations was observed at distances of 18~20 km along the Nanhe River main channel, potentially linked to the REE smelter located on the left bank in this region (Table 2, Figure 8). The evidence is as follows:

(1) The left bank of the Nanhe River, near the REE smelter, exhibits elevated heavy metal levels in the topsoil, especially concerning Pb concentrations. REE smelting, an energy-intensive process, involves complex chemical reactions, wastewater, and incineration gas release, which contain high levels of heavy metals [63].

(2) The Maoniuping REE mining utilizes fluorocarbon cerium mineral, employing an oxidation roasting–sulfuric acid leaching process [64]. While a significant portion of heavy metals, like Pb, are recovered and recycled into smelter slag during chemical processing [65], many residues are released into the environment via wastewater and gas, posing a significant environmental risk [66].

(3) The Pb, Cd, and Zn concentrations are elevated in the southern part of the smelter, likely due to the influence of prevailing north- and northeast-oriented winds in the basin [67]. In the southwest direction, these metals exhibit increasing concentrations with distance.

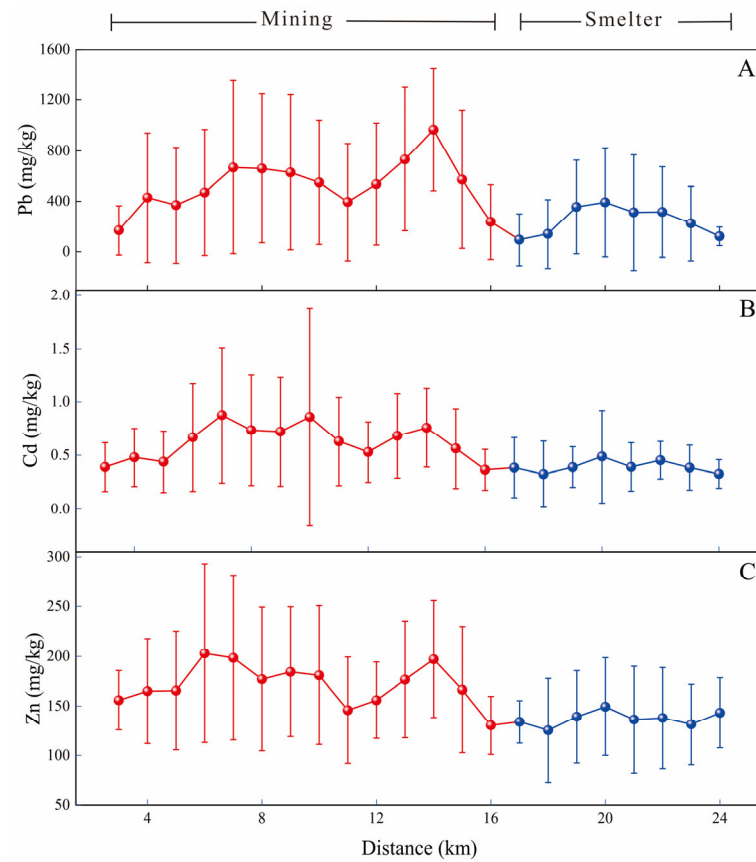


**Figure 7.** Effect of migration distance from the mining area on heavy metals along the Nanhe River channel ((A) Pb, (B) Cd, (C) Zn). Statistics of nodes are the samples within the buffer area (a new area based on the mining area as a baseline that extends outward a certain distance). Error bar represents the CVs, and solid circles represent the average contents.

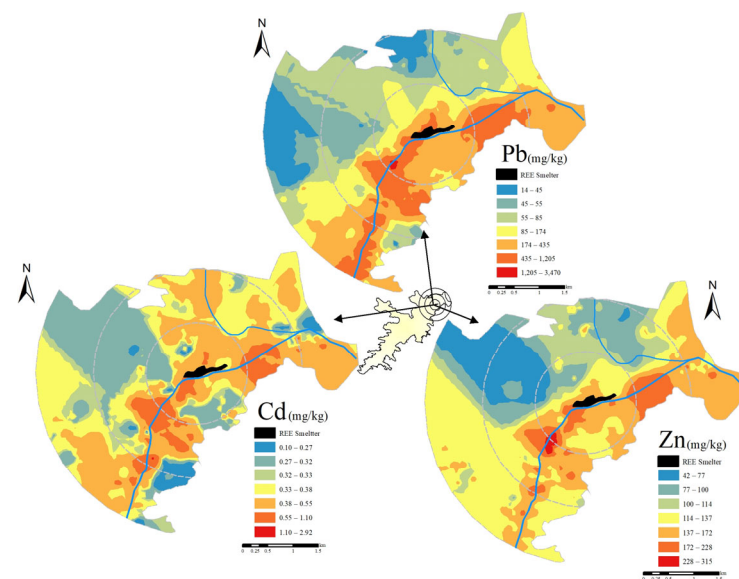


**Figure 8.** Impact of REE smelter on topsoil heavy metal concentrations downstream in the Nanhe River (dotted line indicates the distance to the smelter: 1~3 km; Blue line, Rivers).

## 4. Discussion

### 4.1. Heavy Metal Pollution and Sources

The results of our study show that agricultural soils in the studied area contain high levels of heavy metals and some degree of contamination compared to soil quality standards. The variation in heavy metals in the vertical direction of the soil also indicates their high accumulation in the surface soil. The content of heavy metals was characterized similarly to the study of heavy metals in soils downstream of Pb and Zn mines by Lu et al. [68]. Therefore, this result prompts us to consider some sources of unnatural factors. The high variability of the heavy metal content also indicates that the heavy metals have been influenced by activities related to anthropogenic activities [69].

The geostatistical results show a more significant variability in the spatial distribution of soil heavy metals in the studied area, and at the same time this provides us with the direction to look for anthropogenic sources [70]. The spatial distribution of heavy metals revealed a distinct aggregation pattern near the riverbank, indicating a strong association of Pb, Cd, and Zn with the river, suggesting they share similar sources. Additionally, Hg exhibited significant enrichment in residential and urban areas, reflecting its strong influence by anthropogenic activities. Correlation analysis further supported these findings, showing positive relationships among the heavy metals. Notably, the high correlation coefficients between Pb, Cd, and Zn confirmed that these metals likely originate from common sources.

The results of PCA with the type of correlation analysis indicated the possible sources of the three heavy metals. Specifically, Factor 1 accounted for 30.8% of the total variance, with the main loadings being Pb (0.83), Cd (0.83), and Zn (0.93). The spatial distribution of high concentrations of these metals in the vicinity of the Nanhe River (Figure 2) suggests that the river has a strong influence on the enrichment of these metals. In addition, studies have shown that Maoniuping REE mining has a significant impact on the downstream river [4]. Mining activities are an important source of accumulation of Pb, Cd, and Zn [71,72]. Therefore, Factor 1 is mainly attributed to the sources related to the mining activities of the Maoniuping REE mine (Table 6).

In the PCA model, Factor 2 accounted for 23.7% of the total variance, exhibiting strong loadings for Cu (0.74), Ni (0.84), and Cr (0.78). Notably, these elements (Cu, Ni, and Cr) demonstrated significantly lower coefficient of variation (CV) values compared to other heavy metals in the study area (Table 3). The study revealed that the presence of Cu, Cr, and Ni is predominantly attributed to lithogenic sources, originating from the weathering of parent rocks [73]. Therefore, Factor 2 can be considered as a geogenic source.

Factor 3 accounted for 15.7% of the total variance, demonstrating significant factor loadings for Hg (0.65) and As (0.80). The elevated coefficient of variation (CV) values for Hg and As in the Nanhe Basin suggest substantial anthropogenic influences. Spatial analysis revealed that Hg and As are significantly enriched in suburban areas and regions with high population density, where elevated levels of vehicle emissions and coal combustion are prevalent (Figure 2). Furthermore, studies have confirmed that atmospheric dust deposition serves as a significant contributor to the accumulation of Hg and As in agricultural soils [74,75]. Therefore, Factor 3 is determined to represent anthropogenic input.

### 4.2. Factors Affecting Heavy Metal Migration

#### 4.2.1. pH

Figure 5 illustrates that the  $r_{pH-heavy\ metal}$  is below 0.3, suggesting a weak or negligible correlation between pH and heavy metals. That is, pH exerts a limited effect on the mobility and enrichment of heavy metals in the Nanhe Basin. Similar results were reported by Eisa et al. [76].

#### 4.2.2. Distance to River

In Section 3.4, the river runoff is identified as a key factor in the long-distance transport of heavy metals, particularly Pb, Cd, and Zn. Specifically, the Pb shows a negative correlation with the distance from rivers, with  $r_{Pb-Distance\ to\ river}$  of  $-0.54$  ( $p < 0.01$ , Figure 5), indicating that rivers have a significant impact on soil Pb. The Cd and Zn also exhibit a negative correlation ( $r_{Cd-Distance\ to\ river} = -0.17$ ,  $r_{Zn-Distance\ to\ river} = -0.24$ ), but a lower  $|r|$  indicates a smaller relationship between the metals and river distance. In Figure 5, although Cd and Zn decrease with increasing river distance, the magnitude of this decrease is not statistically significant. Conversely, a moderate positive correlation between Ni ( $r_{Ni-Distance\ to\ river} = 0.39$ ,  $p < 0.01$ ), Hg ( $r_{Hg-Distance\ to\ river} = 0.30$ ,  $p < 0.01$ ), and As ( $r_{As-Distance\ to\ river} = 0.32$ ,  $p < 0.01$ ), shows the river's influence on Ni, Hg, and As migration. Therefore, differential migration patterns are observed on the lateral sides of the Nanhe River. Pb, Cd, and Zn are transported to both banks of the river, while Ni, Hg, and As exhibit a tendency to migrate towards the river channel. This is consistent with the findings of Liu et al. [77]. Their study demonstrated that the use of contaminated water for irrigation results in abnormal enrichment of heavy metals in the near-shore soils along the riverbank. Additionally, the level of heavy metal contamination exhibits a gradual decline as the distance from the river increases. This observation further corroborates that the pollution of Pb, Cd, and Zn is predominantly derived from mining activities.

#### 4.2.3. Impact of Geographical Attributes

Topographic heterogeneity plays a significant role in the transport, transformation, and deposition of heavy metals in soil [78,79]. Studies have implicated gasoline additives and automobile exhaust as the causes of Pb release [18]. In the Nanhe Basin, the weak correlation between Pb and road network density ( $r_{Pb-road} = 0.21$ ) indicates that automobile exhaust emissions exert a low contribution to soil Pb accumulation (Figure 5). Alternatively, the geographic slope ( $r_{Pb-slope} = -0.36$ ,  $p < 0.01$ ) and slope direction ( $r_{Pb-slope\ direction} = -0.24$ ,  $p < 0.01$ ) were negatively correlated with Pb levels and exhibited minimal correlation with Cd and Zn ( $r_{Cd-slope} = -0.07$ ,  $r_{Cd-slope\ direction} = -0.14$ ,  $p < 0.01$ ) and ( $r_{Zn-slope} = 0.01$ ,  $r_{Zn-slope\ direction} = -0.11$ ,  $p < 0.01$ ), respectively. These correlations suggest that slope gradient and aspect may inhibit the migration and enrichment of Pb within the Nanhe Basin. Nevertheless, their impact on Cd and Zn seems to be less pronounced. The reason can be attributed to the fact that the Nanhe River and its banks are mainly low-lying and flat areas in the main river valley, and Pb migration is greatly influenced by the river, with the enrichment areas mainly concentrated near the coast. Cd and Zn exhibit different behavior, potentially due to their distinct properties (see Section 4.2.4).

The terrain attributes exhibit a certain positive impact on Ni, Hg, and As, with moderate positive correlations with Ni ( $r_{Ni-elevation} = 0.35$ ,  $r_{Ni-slope} = 0.40$ ,  $p < 0.01$ ), Hg ( $r_{Hg-elevation} = 0.24$ ,  $r_{Hg-slope} = 0.33$ ,  $p < 0.01$ ), and As ( $r_{As-elevation} = 0.54$ ,  $r_{As-slope} = 0.42$ ,  $p < 0.01$ ). The studied area has a steep terrain with high altitudes, low mining and human impact, and the increased slope easily forms the exposure and accumulation of geogenic heavy metals [80].

#### 4.2.4. Differences in Heavy Metal Migration Capabilities

Figure 9 shows the spatial variation in the correlation between Pb, Cd, and Zn through  $\beta_k$ . As a result, Cd and Zn showed both local positive and negative correlations in geographic space, with more positive correlation between Cd and Zn in the river channel. Cd and Zn have similar ionic radii and electronegativity, which enable them to exhibit comparable geochemical behavior and migration in environmental systems, eventually

affecting their mobility and reactivity [81,82]. Thus, they exhibited the same enrichment performance in the region.

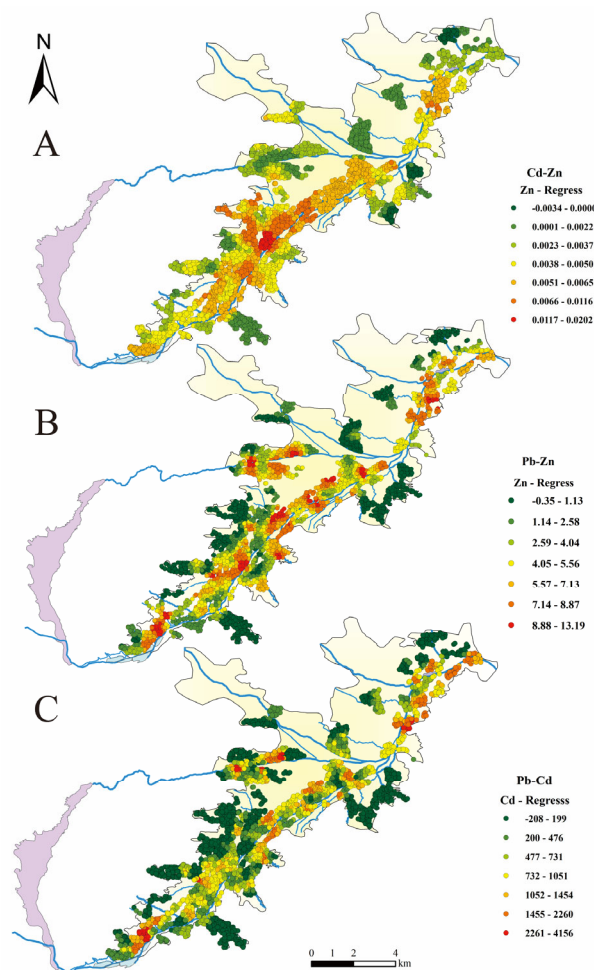


**Figure 9.** Spatial coefficients ( $\beta_k$ ) of heavy metals through a geographically weighted regression model (see Equation (3) for details; (A) Cd-Zn, (B) Pb-Zn, (C) Pb-Zn; Blue line, Rivers and Drains).

The  $\beta_k$  of Pb with Cd and Zn near the riverbank are larger, indicating the precipitate of Pb is more pronounced. The migration and transformation of elements are usually influenced by environmental physics, chemistry, and biology [83], and its intrinsic hydrolysis equilibrium constant ( $K_h$ ), ion radius, atomic weight, and Misono's softness scale [84]. In comparison to Cd and Zn, Pb displays a larger hydrolysis equilibrium constant ( $P_{kh}$  values of Pb, Cd, and Zn are 7.8, 10.1, and 9.0, respectively), higher atomic weight, larger ion radius ( $Pb^{2+} = 0.119$  nm,  $Cd^{2+} = 0.095$  nm,  $Zn^{2+} = 0.074$  nm), and relatively higher Misono's softness scale ( $Pb^{2+} = 3.58$ ,  $Cd^{2+} = 3.04$ ,  $Zn^{2+} = 2.34$ ) [85]. Likewise, the distribution coefficient ( $K_d$ ) of the element reflects the migration ability between the solid and soil solution [86]. Elements with a larger  $K_d$  values exhibit an affinity for soil adsorption, consequently restricting metal mobility within soil solution. Pb consistently demonstrates higher  $K_d$  value in soils [87]. These properties contribute to Pb's greater affinity for adsorption and retention by soil organic components and are easily enriched in the soil environment [88,89].

#### 4.3. Heavy Metals Accumulation

While the total global area dedicated to mining is approximately 66,000 km<sup>2</sup> [90], the potential environmental influence of these activities spans a considerably larger area of 50 million km<sup>2</sup> [91]. This discrepancy underscores the extensive and often indirect impacts of mining operations. For instance, in the open-cut pit Grasberg gold/copper mine in West

New Guinea, forest losses larger than 42 times greater than the mine area [92]. Artisanal gold mining in the Peruvian Amazon was identified as the cause of nearly 1000 km<sup>2</sup> of deforestation [93]. The footprint of small-scale gold mines is also potentially seven-fold greater than that of industrial mines [90,94].

The topographic characteristics of the valley in which the Maoniuping REE mine is situated presented minimal barriers to the dispersion of mine wastes [95]. As mentioned for Pb, which exhibits the most extensive distribution due to fluvial transport, it served as an indicator of contamination extent (Figure 4). Specifically,  $I_{geo-Pb}$  value exceeding Class III (surpassing clean arable soil standards) was employed to delineate the area of mining's influence. Spatial partitioning calculations indicate that a minimum of 38.1 km<sup>2</sup> of land ( $I_{geo-Pb} \geq \text{Class III}$ ) is likely relevant to the impact of Maoniuping REE mining.

In response to concerns regarding both water scarcity and potential pollution from mining activities within the Nanhe Basin, the local government has implemented a water diversion project sourced from the pristine Daqiao Reservoir (102°11'28" E, 28°40'35" N, Figure 1), situated upstream along the Anning River. The construction is currently in progress and is anticipated to be finalized in 2025 [96]. Meanwhile, to facilitate heavy metal pollution, local communities have implemented a proactive strategy by transitioning from paddy/corn to tobacco farming, a crop with lower heavy metal uptake.

## 5. Conclusions

This study systematically investigated the extent of heavy metal pollution in the study area through the application of the geo-accumulation index and potential ecological risk index. Furthermore, the migration and accumulation sources of heavy metals in the soils of the Nanhe River Basin were comprehensively analyzed using a multidisciplinary approach, including geostatistical analysis, correlation analysis, and principal component analysis. The research yielded the following key conclusions:

(1) According to the Soil Environmental Quality Standard (GB 15618-2018) [51], there are different degrees of heavy metal pollution in the surface soil of the Nanhe River Basin, of which Cd (62.5%) and Pb (53.2%) are the most seriously polluted, with more than 50% of the samples exceeding the standard. Heavy metal contents showed spatial differences at moderate and above levels. Geospatial mapping indicated a significant accumulation of heavy metals with higher concentration levels in the southwestern sector of the investigated region. The assessment of the geographical accumulation index based on the regional background values showed that Pb, Cd, and Hg were significantly enriched in the soil, with some Pb (2.8%) even reaching extremely polluted levels. Similarly, the evaluation of potential ecological risks indicates that Cd, Pb, and Hg present moderate to severe ecological risks, with Cd demonstrating the highest ecological risk level among them. Moreover, the overall soil in the study area exhibits moderate to severe levels of potential ecological risk.

(2) The elevated levels of heavy metals in the soil of the Nanhe River Basin are primarily attributed to the combined effects of mining activities, geological background, and other anthropogenic sources. Specifically, mining activities are identified as the dominant source of Pb, Cd, and Zn pollution. In contrast, Cu, Ni, and Cr are largely derived from the geological background, while As and Hg are mainly associated with other human activities. Furthermore, the Nanhe River serves as a key pathway for the dispersion of heavy metal pollution originating from mining operations.

(3) Heavy metals have a certain ability to migrate in the Nanhe River Basin, mainly causing serious pollution within ~1.35 km vertically of the Nanhe River. Mining has affected the soil in the downstream area of ~16 km. Mining and smelting activities have seriously affected the soil safety of the surrounding area of nearly 38.1 km<sup>2</sup>.

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