SPATIAL IDENTIFICATION OF HEAVY METAL POLLUTION SOURCES IN THE ZHUXIANZHUANG MINING REGION, CHINA

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Abstract. The identification of heavy metal pollution sources and spatial differences in surface soil is the basis for soil recovery and pollution control. In order to achieve this, the concentrations of heavy metals (Cr, Zn, Pb, Fe, Mn, and Mo) in the Zhuxianzhuang mining region, China, were determined. The spatial distributions, sources, and pollution extent of the heavy metals were identified, visualized, and evaluated using a variation function, Kriging interpolation, hotspot analysis following multivariate statistical analysis, correlation analysis, principal component analysis, absolute principal component score-multiple linear regression, and geo-accumulation index. Most areas were moderately and moderately-heavily polluted by Mo, of which mean content was 26.39 times its background value. Zn, Pb, and Mo, with high-value clustering, loading coefficients of 0.748, 0.854, and 0.894, and contributions of 73.21%, 80.32%, and 93.68%, respectively, were accumulated mainly in sampling areas 2 and 4, where there were a lot of mining activities. Fe and Mn, with low contents and loading coefficients of 0.896 and 0.968, respectively, were found in sampling areas 4 and 5, affected mainly by industrial and agricultural production. Cr depended on multiple sources of pollution, derived mainly from parent material and mining activities, with contributions of 48.86% and 37.24%, respectively.
Keywords: *heavy metal pollution, absolute principal component-multiple regression analysis, geostatistical analysis, hot spot analysis, geo-accumulation index*

Introduction

Surface soil is an important part of the terrestrial ecosystem, which provides space and nutrients for the survival and growth of plants and animals. It is also the medium for the circulation and migration of heavy metals into the atmosphere, water courses, and organisms (Mirlean et al., 2005; Harvey et al., 2015). Heavy metals with high toxicity are easily accumulated in surface soil and, as a consequence, are difficult to be removed from the soil and degradation is slow as well (Kelepertzis, 2014; Huang et al., 2018). The toxicity of heavy metals poses a great risk to the survival and life processes of animals, plants, microorganisms, and humans, because of their continuous accumulation and biological amplification in the food chain (Zhang et al., 2012; Li et al., 2014). Identifying the sources of heavy metals in soil is of great importance for preventing and controlling soil pollution, protecting public health, preserving sustainable utilization of soil resources, and maintaining sustainable socio-economic development (Li et al., 2013).

Mining regions are generally polluted by heavy metals because large volumes of acid wastewater, harmful waste gases, and tailings are generated in the mining process which infiltrate into the soil through weathering, water dissolution, local atmospheric movements, and water circulation (Cai et al., 2019). The prevention and control of soil

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pollution by heavy metals have become a global challenge and important concerns for researchers and mining managers (Zhang, 2006; Huang et al., 2015).

Pollution by heavy metals generally Occurs for a variety of reasons, from natural factors to human activities (Soonthornnonda and Christensen, 2008; Cai et al., 2012). The natural factors include weathering of parent materials, geochemical processes such as magmatic activity, and atmospheric dust derived from volcanic eruptions, forest fires, sea splashing, vegetation discharge, and wind power (Ellam, 2010). Human activities include mining, mineral processing, metallurgy, electroplating, dyeing, textile manufacture, oil refining, combustion of fossil fuels, manufacture of pesticides and fertilizers, and sludge application and sewage irrigation (Hu et al., 2020; Wang et al., 2020).

The sources of heavy metals can be quantitatively identified using such methods as principal component analysis (PCA) (Pop et al., 2009), positive matrix factorization (PMF) (Shi et al., 2009; Lang et al., 2015; Cheng et al., 2020), absolute principal component score-multiple linear regression (APCS-MLR) (Zhang et al., 2018), random forest (RF) (Tan et al., 2020), generalized linear model (GLM) (Xu et al., 2021), decision trees (Zhong et al., 2014), support vector machine (SVM) (Chen et al., 2013; Hu and Cheng, 2013), stable isotope (Phillips and Gregg, 2003; Parnell et al., 2010), and geostatistics (Qu et al., 2013; Lv, 2019). Multivariate statistical analysis, including correlation analysis, cluster analysis, PCA, and APCS-MLR, can objectively identify the source of pollution, however, the results are easily biased because of the limited number of identification factors available. The PMF method can optimize data by means of standardization deviation and can process imprecise data to ensure their reliability. However, this method is only suitable for areas with simple hydrogeological conditions, large amount of observed data, and relatively few pollution sources and pollution types. The chemical mass balance (CMB) method is simple and easy to understand, with a low detection cost. However, the composition spectrum of the sources of pollution must first be clarified (Yang et al., 2013).

The methods for evaluating pollution by heavy metals include pollution index, geo-accumulation index (Haris et al., 2017), enrichment factor, and potential ecological hazard index. The pollution index method is composed of a single factor index and Nemerow multifactor indices to evaluate the degree of pollution by heavy metals (Cai et al., 2015). The geo-accumulation index method integrates geological processes and human activities to visually evaluate the pollution degree of heavy metals (Jiang et al., 2021). The enrichment factor method can quantitatively analyze the extent of human activity on enriching heavy metals in the soil environment (Thurston and Spengler, 1985). However, the reference elements of the method are generally subjectively selected, which reduces evaluation of the accuracy of pollution by heavy metals (Song et al., 2007). The ecological hazard index can reveal the degrees of migration, transformation, enrichment, pollution, and ecological hazard of heavy metals based on the sedimentology, biotoxicology, ecology, and environmental chemistry of the surface region (Sofowote et al., 2008).

Numerous previous studies of soil heavy metal pollution have focused on the levels of heavy metals, source analysis, and ecological environmental risk assessment. However, there are few studies that have analyzed the sources of pollution by heavy metals in mining region from a spatial perspective. Therefore, the objectives of the study were: (1) to investigate the concentrations and spatial distribution of Cr, Zn, Pb, Fe, Mn, and Mo in the surface soil of Zhuxianzhuang mining region, Anhui Province, China; (2) to spatially identify their potential sources in mining topsoil with different land-use types;

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and (3) to evaluate the pollution degree of the mining region by the heavy metals. The novelty of the study is to spatially identify the distribution, sources, and pollution degree of heavy metals in mining soil with different land-use types using the hotspot analysis, PCA, APCS-MLR, and geo-accumulation index. The results of our study provide a reference for industrial transfer, soil recovery, pollution control, and land-use planning in the Zhuxianzhuang mining region and other regions that are heavily polluted by heavy metals.

Materials and Methods

Sample Collection and Processing

Zhuxianzhuang mining region is located to the southeast of Suzhou City, Anhui Province, China, with an area of approximately 26 km^2. The abundant coal mined there comprises coking coal and gas coal, with total annual output of 2.45 million tons. In recent years, a large number of coal mines in the region have caused serious environmental problems, putting great pressure on the environment and on human health.

The study area was divided into five sampling areas (*Fig. 1*). Sampling area 1 was located in the textile factory and on both sides of the road S306. Sampling area 2 was located in the subsidence area of the mining region. Sampling area 3 was located in the residential area and on both sides of the road S306. Sampling area 4 was located around the hospital, on farmland, and on both sides of the road S306. Sampling area 5 was located in the metal manufacturing industrial area.

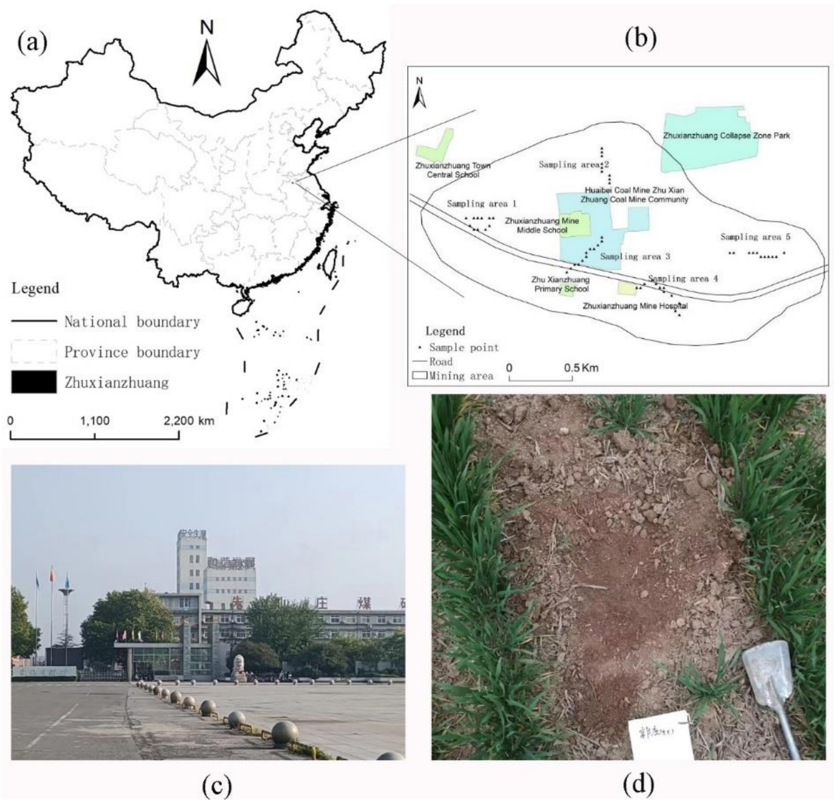

Figure 1. *Locations of study area in China (a) and sampling points (b) and photos about environment (c) and in situ sampling (d) in Zhuxianzhuang mine region, Yongqiao District, Suzhou City, Anhui Province, China*

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The longitude and latitude of each sample point in the five sampling areas were recorded using the Jisibao G120BD Beidou handheld outdoor GPS measuring instrument navigator locator (available from https://www.jishardware.com/Item/582756373159) in September 2020 (*Appendix Table 1*). Soil samples were collected from the four vertices and the center of a square dimensions of 1 m × 1 m around the sampling point and at a depth of 0–20 cm below the surface. Fifty-eight (58) soil samples were collected and placed in labeled polyethylene bags and sent to the laboratory for treatment and analysis (*Appendix Table 1*).

The soil samples were air-dried naturally. Debris was removed and the air-dried samples were ground and screened repeatedly through a stainless-steel nylon sieve (100 mesh). The Cr, Zn, Pb, Fe, Mn, and Mo contents were measured using an inductively coupled plasma mass spectrometer (ICP-MS, NexION 350X, SISG Group, Hanoi, Vietnam) after the 2 g sieved sample that had been randomly collected and was decomposed with a mixture of $HClO_4$-HNO_3-HF acids. The national soil primary standard material (GSS-1) was used for quality control. The recovery rate of all elements was 100 ± 10%.

Hotspot Analysis

Hot spot analysis was used to show the spatial aggregation morphology and clustering mode of the high and low contents of heavy metals by means of local spatial autocorrelation and Getis-Ord G_i* statistics in ArcGIS 10.1 software (https://doc.arcgis.com). The G_i* statistical index can be reflected by the standardized Z score (Getis and Ord, 2010). The greater the absolute Z value, the closer the cluster of high contents. A cluster is a hot spot if $Z > 0$. In contrast, a cluster is a cold pot if $Z < 0$. The G_i* statistical index and the Z score are calculated as follows:

$$G_i\left(d\right)=\frac{\sum_{i=1}^{n} w_{ij}(d)x_j}{\sum_{j=1}^{n} x_j} \quad \text{(Eq.1)}$$

$$Z\left(G_i\right)=\frac{\left[\left(G_i\left(d\right)\right)-E\left(G_i\right)\right]}{\sqrt{VAR\left(G_i\right)}} \quad \text{(Eq.2)}$$

where $G_i(d)$ is the statistic of the ith spatial unit based on the distance d between the ith and jth spatial units, x_j is the attribute value of the jth spatial unit, $w_{ij}(d)$ is the spatial adjacent weight matrix, $Z(G_i)$ is the statistical Z score in the ith spatial unit, $E(G_i)$ and $VAR(G_i)$ are the mathematical expectation and coefficient of variation (CV) of G_i, respectively.

A variation function can indicate the structure of the regionalized variables through a random field and a random process. It requires data to conform to the normal distribution. Kolmogorov–Smirnov (K–S) analysis was used to detect whether the measured contents of heavy metals conformed to the normal distribution. The heavy metal contents, which were all processed logarithmically to base 10, were normal distributed following K–S analysis (K–S> 0.05). The processed data were fitted to variation functions, including linear, spherical, Gaussian, and exponential models, to obtain the optimal variation

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function. The parameters of the variation functions included nugget value (Co), base station value (Co + C), range, residual error (RSS), determination coefficient (R^2), and nugget coefficient (Co / (Co + C)). A large value of R^2 and small values of Co and RSS indicated high fitting accuracy of a model. The nugget coefficient was used to indicate spatial autocorrelation to some extent, as well as the impact of natural and anthropogenic sources on the content and spatial distribution of the heavy metals. The variables of a variation function have a strong spatial autocorrelation if Co/ (Co + C) < 0.25, a moderate spatial autocorrelation if 0.25 ≤ Co/ (Co + C) < 0.75, and a poor spatial autocorrelation if Co/ (Co + C) ≥ 0.75.

Absolute Principal Component Score-Multiple Linear Regression (APCS-MLR)

The APCS-MLR model is derived from improved PCA in order to obtain the normalized factor fraction of heavy metal content and the contributions of different pollution sources to the same heavy metals by introducing a “zero” concentration factor (Thurston and Spengler, 1985; Qu et al., 2013; Luo et al., 2015).

$$Z_{ij} = \frac{C_{ij} - C_i^{'}}{\sigma_{ij}} \tag{Eq.3}$$

$$Z_{i0} = \frac{0 - C_i^{'}}{\sigma_i} \tag{Eq.4}$$

$$C_i = b_{i0} + \sum_{p=1}^{p} (b_{pi} \times APCS_p) \tag{Eq.5}$$

where Z_{ij} is the standardized content of the ith heavy metal in the jth sampling point, C_{ij} and $C_i^{'}$ are the measured and average contents of heavy metals (mg·kg^{-1}), σ_{ij} is the standard deviation of the ith heavy metal in the jth sampling point, Z_{i0} is the introduced “zero” concentration factor of the ith heavy metal, C_i is the value of the regression equation between the content of the ith heavy metal and the APCS, b_{i0} is the constant of the regression equation for the ith heavy metal, b_{pi} is the regression coefficient of the source p for the ith heavy metal, $APCS_p$ is the fraction of factor p, and $b_{pi} \times APCS_p$ is the contribution of source p to the ith heavy metal.

Geo-accumulation Index

The geo-accumulation index (I_{geo}) relates to the effect of geological processes on pollution by heavy metals, and which can evaluate the degree of pollution.

$$I_{geo} = \log_2\left(\frac{C_i}{K \times B_i}\right) \tag{Eq.6}$$

where I_{geo} is the geo-accumulation index of the heavy metals, C_i is the measured content of the ith heavy metal, B_i is the background value of the ith heavy metal, and K is the correction coefficient for eliminating the differences in parent materials between different regions; usually, $K = 1.5$.

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The background value of a heavy metal in the soil environment is the heavy metal content in the soil that has not been artificially polluted in a certain region, which is determined by using the methods of reference value, cumulative frequency distribution, and geostatistics. Reference value is obtained according to the existing relevant standards, soil background values in other areas, and the geographical, meteorological, and land-use factors in the study area. Cumulative frequency distribution is obtained using statistical methods according to the measured heavy metal concentration at each point in a specific region. Geostatistics is to describe the distribution of deviation value of the heavy metal concentration according to the relationship between high and low concentration, variance, and distance between sampling points.

The criteria for the geo-accumulation index are divided up as listed in *Table 1*.

Table 1. *Criteria for pollution levels of the geo-accumulation index*

Pollution-free	Mild pollution	Middle level of pollution	Moderate to high level of pollution	High level of pollution	High to extreme level of pollution	Extreme level of pollution
$I_{geo} \leq 0$	$0 < I_{geo} \leq 1$	$1 < I_{geo} \leq 2$	$2 < I_{geo} \leq 3$	$3 < I_{geo} \leq 4$	$4 < I_{geo} \leq 5$	$I_{geo} > 5$

Results

Characteristics of Heavy Metal Contents

The descriptive statistics of the heavy metal contents in the Zhuxianzhuang mining region are listed in *Table 2*. The average contents of Cr, Zn, Pb, Fe, Mn, and Mo were 65.67, 60.03, 23.29, 28,436.88, 482.34, and 12.67 mg·kg^{-1}, respectively. The average content of Mo was 26.39 times its background value, while those of the other heavy metals were less than their background values. The maximum contents of all heavy metals exceeded their respective background values. Background values of Cr, Zn, Pb, Fe, Mn, and Mo are 67.52 mg/kg, 62.00 mg/kg, 26.60 mg/kg, 31,400.00 mg/kg, 530.00 mg/kg, and 0.48 mg/kg, respectively. The number of non-polluted sampling control sites of Cr, Zn, Pb, Fe, Mn, and Mo are 34, 35, 48, 41, 39, and zero, respectively (*Appendix Table 2*).

Table 2. *Descriptive statistics of heavy metals in the surface soil of Zhuxianzhuang mining region*

Heavy metal	Mean value (mg/kg)	Background value (mg/kg)	Standard deviation	Skewness	Kurtosis	CV (%)
Cr	65.67	67.52	9.43	0.31	−0.47	14
Zn	60.03	62.00	22.12	3.33	17.92	37
Pb	23.29	26.60	4.85	1.53	4.13	21
Fe	28,436.88	31,400.00	5,426.49	0.23	−0.03	19
Mn	482.34	530.00	84.13	−0.11	0.14	17
Mo	12.67	0.48	4.77	1.29	1.89	38

The standard deviations of the six heavy metals were ordered as: Mo < Pb < Cr < Zn < Mn < Fe. According to their CV classification (Wilding et al., 1985), Cr (14%) had a low variation (CV ≤ 15%), while Pb (21%), Fe (19%), and Mn (17%) had moderate variations (15% < CV ≤ 35%), and Zn (37%) and Mo (38%) had high variations (CV >

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35%). The high CV variations of Zn and Mo indicate that they were not evenly distributed in the study area because of the great effect exerted by human activities.

The order of kurtosis was Cr < Fe < Mn < Mo < Pb < Zn, and the order of skewness was Mn < Fe < Cr < Mo < Pb < Zn. All of the heavy metals showed positive skewness, except for Mn. Zn and Mo had a large skewness, which indicates that the accumulations of Zn and Mo may have originated from human activities.

The descriptive statistics of the heavy metals in surface soil in the different sampling areas are given in *Table 3*. The average contents of Mo in the five sampling areas exceeded its background values, and their order was: sampling area 2 (16.3 $mg \cdot kg^{-1}$) > sampling area 4 (14.5 $mg \cdot kg^{-1}$) > sampling area 1 (11.92 $mg \cdot kg^{-1}$) > sampling area 3 (10.75 $mg \cdot kg^{-1}$) > sampling area 5 (10.5 $mg \cdot kg^{-1}$). The CVs of Zn (58%) and Mo (36%) in sampling area 2, Mn (41%) in sampling area 3, and Mo (41%) in sampling area 4 were greater than 35%, indicating that they were greatly affected by human activities. Ratios of mean concentration to background value of Mo in sampling sites 1, 2, 3, 4, and 5 were 24.83, 33.96, 22.4, 30.21, and 21.88, respectively, which shows severe pollution of Mo in these areas.

Table 3. *Descriptive statistics, including maximum, minimum, mean value, ratio of mean concentration to background value (ratio), standard deviation, skewness, kurtosis, and CV, of heavy metals in the surface soil of the different sampling sites in the Zhuxianzhuang mining region*

Sampling site	**Heavy metal**	**Content (mg/kg)**			**Morphology**				
		Maximum	Minimum	Mean value	Ratio	Standard deviation	Skewness	kurtosis	CV (%)
1	Cr	83	54	66.50	0.98	9.49	0.53	−1.09	14
	Zn	66	48	56.75	0.92	5.96	0.60	−0.69	10
	Pb	24	18	21.25	0.8	1.76	−0.34	−0.47	8
	Fe	32,403	23,362	27,931.58	0.89	3,214.79	−0.07	−1.63	12
	Mn	595	413	512.42	0.97	52.19	−0.31	−0.29	10
	Mo	15	6	11.92	24.83	2.75	−1.10	0.52	23
2	Cr	76	54	64.90	0.96	8.61	0.06	−1.73	13
	Zn	186	47	73.70	1.19	42.71	2.42	6.23	58
	Pb	43	20	26.70	1	7.53	1.39	1.31	28
	Fe	29,397	17,069	25,872.40	0.82	3,371.40	−2.23	6.16	13
	Mn	495	241	410.90	0.78	66.09	−2.04	5.79	16
	Mo	28	10	16.30	33.96	5.85	1.02	0.15	36
3	Cr	66	47	58.17	0.86	6.51	−0.63	−0.95	11
	Zn	82	32	43.42	0.7	14.02	2.12	5.33	32
	Pb	29	15	19.42	0.73	3.58	1.79	4.65	18
	Fe	31,989	18,962	23,317.08	0.74	4,089.73	0.86	0.09	18
	Mn	555	340	422.08	0.8	60.22	0.80	0.71	14
	Mo	22	5	10.75	22.4	4.41	1.45	3.38	41
4	Cr	86	53	70.08	1.04	11.56	0.04	−1.20	16
	Zn	84	41	66.92	1.08	15.84	−0.60	−1.26	24
	Pb	32	19	25.50	0.96	4.48	−0.02	−1.04	18
	Fe	40,690	22,205	32,821.75	1.05	6,645.33	−0.21	−1.45	20
	Mn	685	361	509.75	0.96	104.32	0.28	−1.05	20
	Mo	26	6	14.50	30.21	5.98	0.50	−0.34	41
5	Cr	81	58	68.58	1.02	6.14	0.27	−0.11	9
	Zn	70	53	61.67	0.99	4.52	−0.32	0.11	7
	Pb	26	22	24.17	0.91	1.28	−0.36	−0.58	5
	Fe	34,909	28,444	31,814.17	1.01	1,865.51	−0.21	−0.86	6
	Mn	596	483	544.67	1.03	33.53	−0.47	−0.34	6
	Mo	13	8	10.50	21.88	1.26	0.00	0.65	12

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Spatial Distribution of Heavy Metals

Spatial Variation of Heavy Metals

Table 4 lists the results for the six heavy metals in the surface soil of the Zhuxianzhuang mining region fitted by the Gaussian model, one of the variation functions. The range of variation of the Gaussian model for the six heavy metals was between 120.378 and 807.136. The values of R^2 of the heavy metals were all more than 0.75, except for that of Zn (0.576). The nugget coefficients of Cr and Mn were 0.735 and 0.740, respectively, which are moderate autocorrelations. The nugget coefficients of the other heavy metals were more than 0.75, showing a poor spatial autocorrelation and a large effect of human activities on the heavy metal contents.

Table 4. *Parameters of the theoretical model of the variation function for the six heavy metals in the surface soil of the Zhuxianzhuang mining region*

Heavy metal	Model	Co	Co+C	Range (km)	RSS	R^2	Co/(Co+C)
Cr	Gaussian	0.0,014	0.0,053	172.522	7.084E-07	0.949	0.735
Zn	Gaussian	0.0,079	0.0,413	807.136	5.958E-04	0.579	0.809
Pb	Gaussian	0.0,010	0.0,073	175.110	4.175E-06	0.889	0.867
Fe	Gaussian	0.0,007	0.0,073	176.842	7.260E-06	0.848	0.901
Mn	Gaussian	0.0,022	0.0,084	176.437	5.303E-06	0.895	0.740
Mo	Gaussian	0.0,010	0.0,044	120.378	5.797E-06	0.786	0.778

Spatial Distribution of Heavy Metal Contents

The heavy metal contents of the 58 sampling points were spatially interpolated using the method of ordinary Kriging, according to the optimal Gaussian variation function (*Fig. 2*). The six high-content centers of Cr in the surface soil were symmetrically distributed in a saddle-like pattern around the Zhuxianzhuang mining region. Cr had the smallest CV, which indicates the significant effect of parent material on its distribution.

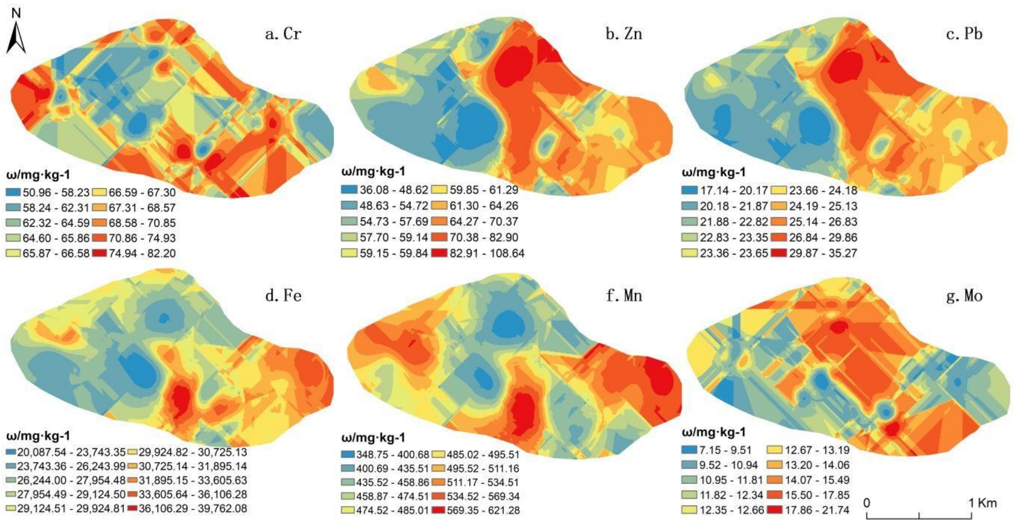

Figure 2. *Spatial distribution of heavy metals in the Zhuxianzhuang mining region*

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The spatial distributions of Zn and Pb were similar, with S-shaped boundaries between the high (right-hand side of the boundaries) and low (left-hand side of the boundaries) contents of Zn and Pb. The highest contents of Zn and Pb appeared in the subsidence area (now a park), the highway, and the farmland in sampling area 2. This indicates that the main sources of pollution by Zn and Pb may have come from human activities, such as mining, transportation, and the application of chemical fertilizers and pesticides. In addition, moderate variations in the Zn and Pb contents indicate that their sources of pollution are also derived mainly from human activities other than mining activities.

The results for the spatial distributions of Fe and Mn were similar, which were found to be mainly around the factories in sampling areas 4 and 5. However, the area of Mn with a high content, which was accumulated in sampling area 1, was larger than that of Fe. Fe and Mn, with low contents, were distributed in sampling areas 2 and 3. Mo was mainly distributed in sampling area 4 and the mining subsidence area (now a park) of sampling area 3, indicating that mining has had a great impact on its content and distribution.

Analysis of Heavy Metal Hot Spots

The spatial distribution of heavy metal hots pots in the surface soil of the Zhuxianzhuang mining region was obtained using local spatial autocorrelation and the Gi^* hotspot in ArcGIS 10.1 (*Fig. 3*). Mn in sampling area 1 showed a hot spot (high-value clustering) and a nonsignificant distribution, indicated a severe pollution of Mn in sampling area 1. Pb in sampling area 1 showed a cold spot distribution (low-value clustering), while the other heavy metals were not significant.

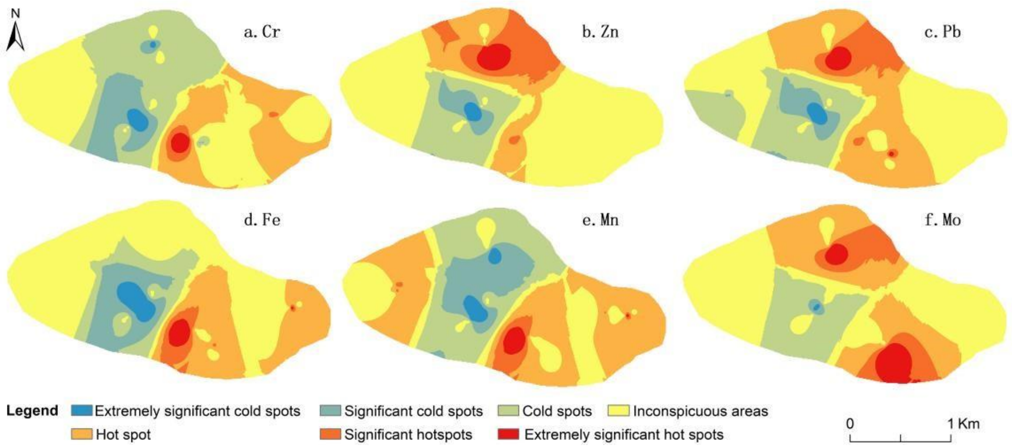

***Figure 3.** Distribution of heavy metal cold and hot spots in the surface soil of the Zhuxianzhuang mining region*

Sampling area 2 was affected mainly by Zn, Pb, and Mo because of high-value clustering (hot spots, significant hot spots, and extremely significant hot spots) of Zn, Pb, and Mo in sampling area 2. However, there was low-value clustering (cold spots and significant cold spots) of Cr and Mn and no significance of Fe in sampling area 2.

Sampling area 3 was less disturbed by heavy metals because the six heavy metals showed low-value clustering and nonsignificant distributions in sampling area 3. Sampling area 4 was affected by the six heavy metals because Cr, Pb, Fe, Mn, and Mo showed extremely significant hot spots, while Zn showed high-value clustering but a

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nonsignificant distribution, especially Mo, which showed the largest extremely significant hot spot by area.

Sampling area 5 was affected mainly by Cr, Fe, and Mn because of high-value agglomeration (hot spots and significant hot spots) of Cr, Fe, and Mn, while no significance distributions of Zn, Pb, or Mo were shown in sampling area 5. Therefore, the heavy metals were more heavily accumulated in sampling areas 2, 4, and 5, and less in sampling areas 1 and 3.

Source Identification of Heavy Metals

Correlation Analysis of Heavy Metal Contents

The analysis of correlation between the different heavy metals is one of the important methods for identifying the sources of pollution by heavy metals. *Table 5* presents the results of Pearson correlation analysis of the heavy metals in the surface soil of Zhuxianzhuang mining region. The correlation coefficients for Cr-Zn, Cr-Pb, Cr-Fe, and Cr-Mn were 0.408, 0.474, 0.526, and 0.348, respectively, which passed the correlation test of $P < 0.01$. Significant positive correlations of Cr with Zn, Pb, Fe, and Mn indicated that Cr may have been affected by multiple sources of pollution. Zn may have been affected by two or more pollution sources because Zn had a significant positive correlation with Pb, Fe, and Mo ($0.391 \leq r \leq 0.821$ and $p < 0.01$), and a significant positive correlation with Mn ($r = 0.31$ and $p < 0.05$). The correlation coefficients of Pb with Fe and Mo were 0.441 and 0.661, respectively, which passed the correlation test at the 0.01 level. Mn had the same sources as Fe because the correlation coefficient between Mn and Fe was 0.856, which passed the correlation test at the 0.01 level.

Table 5. *Correlation between the heavy metal contents in the surface soil of the Zhuxianzhuang mining region*

	Cr	**Zn**	**Pb**	**Fe**	**Mn**	**Mo**
Cr	1					
Zn	0.408**	1				
Pb	0.474**	0.821**	1			
Fe	0.526**	0.440**	0.441**	1		
Mn	0.348**	0.310*	0.212	0.856**	1	
Mo	0.21	0.391**	0.661**	0.073	−0.155	1

PCA of Heavy Metal Contents

PCA can identify the sources of heavy metals in surface soil effectively. The heavy metal contents were processed logarithmically to base 10 and tested with Kaiser–Meyer–Olkin (KMO) and Bartlett spheres. The values of the KMO and Bartlett spheres were 0.667 and 0, respectively, which met the PCA requirements.

The number of principal factors was set to three according to the correlation analysis. The loading coefficients obtained following rotation are listed in *Table 6*. The cumulative contribution of the three principal components was 90.88%. The contribution of variance to the first principal component (PC1) was 36.475%. Zn, Pb, and Mo had large loads on PC1, with loading coefficients of 0.748, 0.854, and 0.894, respectively, following the maximum orthogonal rotation of variances between the heavy metals. The contribution of variance to the second principal component (PC2) was 34.533%. Fe and Mn had large

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loads on PC2, with loading coefficients of 0.896 and 0.968, respectively, following the maximum orthogonal rotation of variance between the heavy metals. The contribution of variance to the third principal component (PC3) was 19.871%. The loading coefficient of Fe on PC3 was 0.946, indicating that Fe may have been greatly affected by PC3. The loading coefficients of Zn following maximum orthogonal rotation of the variances in PC2 and PC3 were 0.442 and 0.329, respectively, indicating that the Zn content was affected by PC1, PC2, and PC3 simultaneously.

Table 6. *Loading coefficients following orthogonal rotation of heavy metals in the surface soil of the Zhuxianzhuang mining region*

Heavy metal	Loading coefficient after rotation		
	PC1	PC2	PC3
Cr	0.209	0.200	0.946
Zn	0.748	0.442	0.329
Pb	0.854	0.271	0.33
Fe	0.236	0.896	0.271
Mn	−0.027	0.968	0.083
Mo	0.894	−0.15	0.002
Contribution rate of variance (%)	36.475	34.533	19.871
Accumulated contribution rate of variance (%)	36.475	71.009	90.88

APCS-MLR of Heavy Metal Contents

The APCS-MLR model was used to analyze and verify the sources of heavy metals (*Fig. 4*). The values of R^2 of Cr, Zn, Pb, Fe, Mn, and Mo were 0.978, 0.863, 0.912, 0.932, 0.945, and 0.823, respectively, which are all more than 0.8. The ratios of the predicted to measured contents of heavy metals were between 0.999 and 1, which validates the effectiveness and credibility of the APCS-MLR model for identifying the sources of pollution by heavy metals.

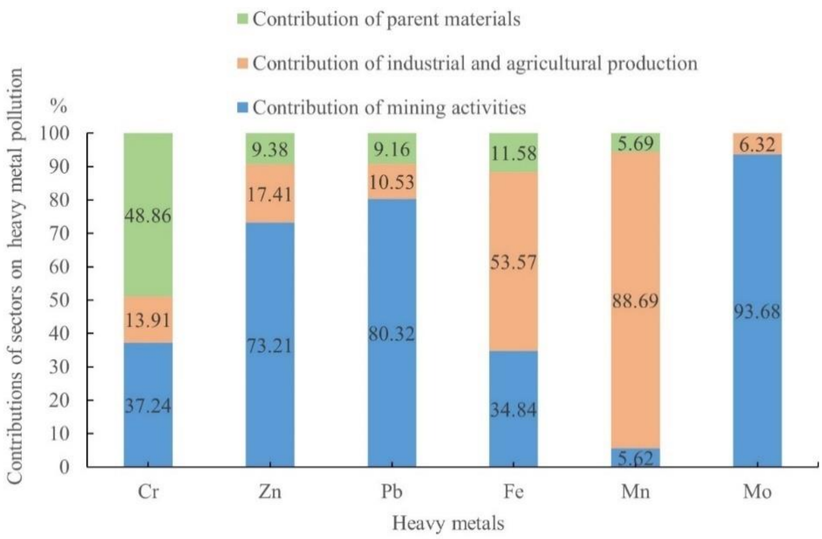

Figure 4. *Cumulative percentages of contributions of the sectors on the heavy metal pollution in the Zhuxianzhuang mining region*

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Zn, Pb, and Mo were mainly affected by PC1, with contributions of 73.21%, 80.32%, and 93.68%, respectively. They had significant positive correlations, which passed the correlation test at the 0.01 level, indicating that they may have been from the same pollution source. The Zn, Pb, and Mo contents in sampling area 2 exceeded their background values. The Zn and Mo contents in sampling area 4 exceeded their background values, which were spatially accumulated around the mining region. PC1 was inferred to be generated by mining activity because a large volume of acid wastewater produced by mining, along with accumulation and leaching of tailings, caused the heavy metals to infiltrate into the surface soil in the form of metal sulfates.

Fe and Mn were affected mainly by PC2, with contributions of 53.57% and 88.69%, respectively. Fe and Mn should come from the same source because they belong to the iron family of elements in period (row) 4 of the periodic table and had a strong positive correlation (coefficient of 0.856). Fe and Mn were distributed mainly in the farmland of sampling area 4, because Fe and Mn, important components of fertilizers, entered surface soil through the application of chemical fertilizers and pesticides. In addition, Fe and Mn were distributed in the industrial zone of sampling area 5 because of their wide use as deoxidants and desulfurizers in industrial production. Therefore, PC2 is a result of industrial and agricultural production activities.

The contribution of Cr was less than 12%, which indicates that Cr was less affected by PC3. Cr is a moderately incompatible element similar to copper, with an average content of 1.1 $mg \cdot kg^{-1}$ in the surface layer. However, the minimum content of Cr in the study area is five $mg \cdot kg^{-1}$, and the average Cr content was 26.4 times its background value. In contrast, the average Cr content in each sampling area was higher than its background values. PC3 is speculated to be derived from parent material because Cr is generated mainly from parent material.

Evaluation of Heavy Metal Pollution

The results for the evaluation of the geo-accumulation indices of heavy metals in the surface soil of the Zhuxianzhuang mining region are shown in *Fig. 5*. The average geo-accumulation indices for Cr, Zn, Pb, Fe, Mn, and Mo were −0.640, −0.701, −0.804, −0.754, −0.744, and 1.988, respectively. All the sample points were not polluted by Cr, Fe, and Mn, which indicates that they presented a small threat to human health and the surrounding environment. The number of sampling points with the geo-accumulation indices of Zn and Pb of $I_{geo} \in (0,1]$, which were mildly polluted, accounted for 3.448% and 1.695%, respectively, of the total number of sampling points in the study area. Mo had 6.897% of the total number of sampling points with a mild level of pollution, 50% of the total number of sampling points with a moderate level of pollution, 39.655% of the total number of sampling points with a moderately high level of pollution, and 3.448% of the total number of sampling points with a high level of pollution. Therefore, Mo contributed the largest risk of pollution to human health and the environment surrounding the mining region. Mo comes mainly from mining activities, so mining operations should be rigorously standardized and supervised, and chemical and ecological restoration should be carried out in a timely manner following mining activities.

The spatial distribution of Mo pollution levels, which heavily polluted the study area, is shown in *Fig. 6*. A small area of sampling areas 2 and 4 was heavily polluted by Mo. The east and west of the study area (sampling areas 1, 2, and 4) were moderately to heavily polluted by Mo, which further verifies that mining activities had a great impact

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on the distribution, content, and degree of pollution by Mo. The north and south of the study area (sampling areas 1, 3, and 5) were moderately highly polluted by Mo.

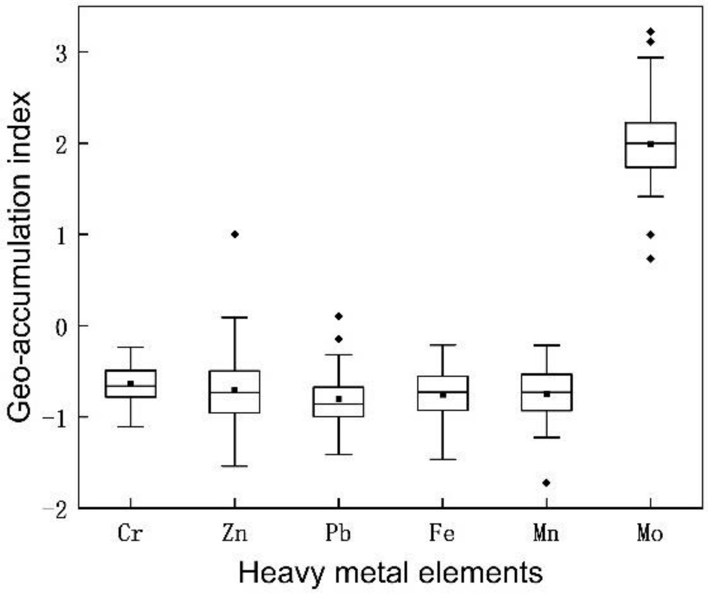

Figure 5. *Boxplot of geo-accumulation indices of heavy metals in the surface soil of the Zhuxianzhuang mining region*

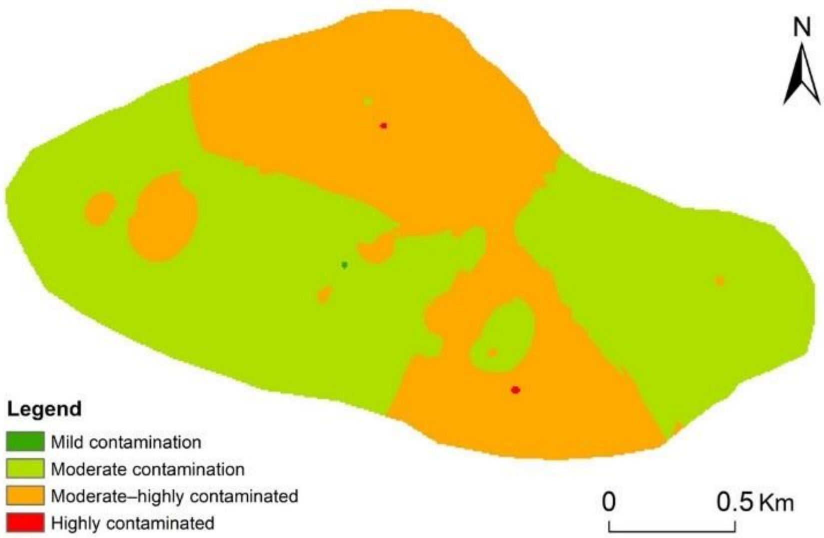

Figure 6. *Distribution of Mo pollution levels in the surface soil of the Zhuxianzhuang mining region*

The distributions of Cr and Mo strip regions with well-defined boundaries were interpolated using the Kriging method with Gaussian and semi-variance models. Further methods of interpolation, such as the inverse distance interpolation method, need to be used to find the optimal distribution of heavy metal contents.

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Discussion

The integration of source identification, spatial distribution, and pollution extent of heavy metals in this study provides a visual and identification methodology of heavy metal sources. The method can contribute to ecological restoration, agricultural production, industrial activity, and health risk evaluation in an overexploited mining area.

PCA can statistically identify anthropogenic from natural sources of heavy metals. According to the results obtained from Cai et al. (2012), Cu, Cr, Ni, and partially Pb and Zn had strongly positive correlations with parent materials according to the method of PCA. As, Cd, and partially Zn showed a weak correlation between soil properties and anthropogenic metals. Hg and partially Pb showed a low relationship with soil properties. In Cai et al. (2015), PC1 mainly included Co, Cr, and Ni. PC2 mainly included Cd and As. PC3 mainly included Hg. In Cai et al. (2019), PC1 mainly included Ni, Cu, Cr and As. PC2 was loaded by Zn, Pb, and Cd. PC3 was dominated by Hg. In our study, Zn, Pb, and Mo had large loads on PC1. Fe and Mn had large loads on PC2. Fe had large loads on PC3. Zn content was affected by PC1, PC2, and PC3 simultaneously.

Heavy metal pollution sources in different areas could be different because of their different natural endowments and human activities (*Table 7*). Coal mining, coal transportation, and coal combustion process contribute a lot to the pollution sources of heavy metals in this study. Fe and Mn came from coal mining and agricultural activities. Zn, Pb, and Mo mainly originated from industrial activities. Cr mainly derived from parent materials.

Table 7. *Comparison of heavy metal pollution sources identified from different study areas*

Study area	Vehicle emission	Agricultural activity	Industrial activity	Parent material	Reference
Puning City	Pb, Zn, Cu	As	Hg, Cd	Cr, Ni, V, Ti	Wang et al. (2019)
Lianyuan City	Pb, Sb, Hg, As	Zn, Cu, Cd	Hg, Cd, M, V	Mn, C, Fe	Liang et al. (2017)
Huilai County	Cd, Zn, Pb	As, Cu	Hg, Cd	Ni, Cr	Jiang et al. (2021)
Huizhou City	Hg, Pb	Cd, As	Hg, Pb	Cr, Ni, Cu	Cai et al. (2012)
Shunde City	Hg	Cd, As	Hg	Co, Cr, Ni	Cai et al. (2015)
Gaogang Town	Pb, Zn, Cd	Pb, Zn, Cd	Hg	Cu, Ni, As, Cr	Cai et al. (2019)
Zhuxianzhuang mining region	NA	Fe, Mn	Zn, Pb, Mo, Fe, Mn	Cr	Ours

NA: not available

In Wang et al. (2019), Puning City has well developed industries, such as clothing and medicine, and agriculture, such as tobacco, tea, and fruit. Therefore, Pb, Zn, and Cu mainly came from vehicle emission. As mainly originated from agricultural activities. Hg and Cd mainly derived from industrial practices. Cr, Ni, V, and Ti mainly rooted in parent material.

In Liang et al. (2017), main industries in Lianyuan City are coal mining, building material production, and non-ferrous metal mining. Pb, Sb, Hg, and As originated from coal combustion and vehicle emission. Hg, Cd, Mo, and V mainly derived from industrial

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practices. In addition, Zn, Cu, and Cd were mainly associated with agricultural activities. Mn, Cr, and Fe came from parent materials.

In Jiang et al. (2021), developed transportation, industry, and agriculture in Huilai County resulted in Cd, Zn, and Pb from vehicle emission, As and Cu from agricultural activities, Hg and Cd from industrial activities, and Ni and Cr from parent materials.

In Cai et al. (2012), Huizhou City is one of the most developed industrial regions in China. Soil parent materials includes river alluvial deposit, marine deposit, granite, sandstone, and shale. Hg and Pb in Huizhou City came from vehicle emission. Cd and As originated from agricultural activities. Hg and Pb derived from industrial activities. Cr, Ni, and Cu rooted in parent materials.

In Cai et al. (2015), Shunde City is also one of the most developed industrial regions in China. The soil types mainly include stacked soil, paddy soil, and lateritic red soil. Hg in Shunde City mainly came from vehicle emission and industrial activities. Cd and As originated from agricultural activities. Co, Cr, and Ni derived from parent materials.

In Cai et al. (2019), Gaogang Town has rich mineral resources, such as white stone, fluorite, rare earth, and porcelain clay, and developed heavy industries such as equipment manufacturing and precision instrument. Pb, Zn, and Cd came from vehicle emission and agricultural activities. Hg originated from industrial activities. Cu, Ni, As, and Cr derived from parent materials.

Therefore, the main pollution sources (Zn, Pb, and Mo) in this study generally originated from coal mining activity. However, other study areas in *Table 7* were generally well developed in industry, traffic, and agriculture, resulting in the main pollution sources from industrial and agricultural practices. In addition, Cr came from the parent materials in our study, which is in agreement with other results obtained from Cai et al. (2012, 2015, 2019), Liang et al. (2017), Wang et al. (2019), and Jiang et al. (2021).

Conclusions

Fifty-eight (58) samples of surface soil from the Zhuxianzhuang mining region were collected. The Cr, Zn, Pb, Fe, Mn, and Mo contents of the samples were analyzed using descriptive statistics, multivariate statistics, geo-statistics, and geo-accumulation indexes. Mo, with an average of 26.39 times its background level and exerting risk of moderate and moderately heavy pollution, was found to be distributed mainly in sampling areas 2 and 4. Six heavy metals showed cold spots in sampling area 3. Cr was found to be derived mainly from parent material and mining activities, with contributions of 48.86% and 37.24%, respectively. Zn, Pb, and Mo were affected mainly by mining activities, with contributions of 73.21%, 80.32%, and 93.68%, respectively. Fe and Mn were found to be derived mainly from industrial and agricultural production, with contributions of 53.57% and 88.69%, respectively.

Local government departments need to carry out control measures, such as chemical and ecological restoration, depending on the spatial distributions, degrees of pollution, and pollution sources of heavy metals. Risk prevention in the Zhuxianzhuang mining region is suggested based on pollution evaluation and determination of the sources of heavy metals. Zn and Pb, which polluted to a mild degree the surface soil in the collapsed park of the Zhuxianzhuang mining region, were derived mainly from industrial and agricultural production and mining activities. Therefore, measures such as clean mining and smelting, mining restoration, and the treatment of industrial wastewater, waste gases, and residues, should be carried out to relieve pollution by Zn and Pb. Mo, which have

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heavily polluted the north and south of the study area, were found to come mainly from metal smelting and the application of molybdenum-based fertilizers. Therefore, strict supervision of Mo smelting and control of molybdenum fertilizer application should be strengthened in order to reduce the risk of pollution by Mo.

The spatial distributions, hot spots, and sources of heavy metals were studied and determined in this study; however, the risks associated with contamination by heavy metals were not considered. The impacts of pollution by heavy metals on animal, plant, and human health needs to be studied more thoroughly in the future. The pollution level of each heavy metal simulated by the geo-accumulation index method may be lower than the measured pollution degree of the heavy metal. The Nemerow composite pollution index should be used in the future to accurately determine the positions of heaviest pollution and the pollution levels by heavy metals. The geo-accumulation index should be combined with the Nemerow composite pollution index in follow-up studies.

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Conflict of Interests. The authors declare that they have no competing interests.

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