

Article

The Identification of Soil Heavy Metal Sources and Environmental Risks in Industrial City Peri-Urban Areas: A Case Study from a Typical Peri-Urban Area in Western Laizhou, Shandong, China

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Abstract: During the past several decades, soil heavy metal contamination has emerged as an environmental affliction and subject of study. Soil heavy metal contamination in peri-urban areas is more severe and intricate. The western region of Laizhou City, as a typical industrial city exhibiting vigorous factory, agricultural, and extraction activities, possesses substantial research merit and can offer a noteworthy example for the analysis of heavy metal contamination in the peri-urban areas of industrial cities. We procured 271 surface soil samples (0–20 cm) from the western peri-urban areas of Laizhou City. Through statistical scrutiny, it was discerned that the concentrations of As, Cd, Hg, and Pb surpassed the local baseline concentrations, with the ranking of the coefficient of variation being $Hg > 1 > Cd > 0.5 > Pb > Cu > As > Ni > Zn > Cr$. Subsequently, we investigated the potential origins of heavy metals through correlation analysis, principal component analysis, and geostatistical analysis and elucidated the primary origins of heavy metals based on the existing land-use scenario: PC1 (As, Cd, Cu, Pb, and Zn) predominantly originated from agricultural pursuits, transportation, and industrial production; PC2 (Cr and Ni) was correlated with soil parent materials; and PC3 (Hg) was attributed to industrial production and open-pit mining of minerals. According to the analysis of the Nemero comprehensive index and potential ecological risk index, the soil environmental risk in the study area was within a controllable range. However, the continuous enrichment of heavy metals in the soil should receive sufficient attention, and continuous monitoring of the site is recommended. This study attempts to use a combination of existing land-use scenarios and statistical analysis methods to analyze the heavy metal pollution conditions in peri-urban industrial cities. Although this study has shortcomings, it provides valuable information for the study of heavy metal sources and environmental risks in typical industrial city suburbs.

Keywords: industrial city peri-urban areas; soil heavy metals; data analysis; pollution source identification; environmental risk



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1. Introduction

With the acceleration of industrialization and agriculture, anthropic activities have contributed to the increase in heavy metal contents in soil [1–3]. Soil heavy metal pollution is more serious than expected in China [4]. Additionally, high concentrations of heavy metals in soil have been recorded in Russia [5], India [6], Iran [7], Nepal [8], North America [9], Canada [10], Spain [11], and Germany [12]. It has been widely considered by the public and the government. The Action Plan for Soil Pollution Prevention and Control was issued by China in 2016 and aimed to effectively prevent and control soil pollution. Heavy metals can

damage plant growth and threaten human health through dermal contact, breathing, and the ingestion of soil particles [13,14] due to their ubiquity, hypertoxicity, persistence, and bioaccumulation in the food chain. For example, cadmium (Cd) can cause renal dysfunction, kidney damage, anemia, and cancer [15]; mercury (Hg) can cause serious damage to the body's central nervous system and digestive system [16]; and chromium (Cr) affects the formation and development of respiratory organ tumors [17]. Lead (Pb) toxicity can cause cognitive behavioral problems, even at low concentrations [18].

Generally, heavy metals in soil are divided into natural sources and artificial sources. The composition of geological parent material determines the natural contents of heavy metals in soil [19,20]. However, heavy metals are continuously input into the soil as a result of human activities, including fossil fuel combustion, the application of pesticides and fertilizers, sewage irrigation, and mining [21–26], resulting in a sharp deterioration in soil quality. Therefore, accurate identification of the sources of heavy metals is the key to preventing or reducing soil pollution by heavy metals [26,27]. Multivariate statistical analysis is an effective means to identify the characteristics of soil heavy metal pollution and distinguish natural sources and human inputs [28–30]. In addition, soil properties have strong spatial variability [31,32]. Thus, it is necessary to be able to describe the spatial distribution on a regional scale and conduct a reasonable assessment of environmental risks [33]. At present, geostatistical analysis has been used extensively in the study of spatial variations and risk assessments of soil heavy metals [34]. Jie et al. [35] used the kriging method to investigate the contamination conditions and spatial distributions of soil heavy metals. Fei et al. [36] further visually obtained the spatial distribution characteristics and risk probability maps of heavy metal soil pollution in specific areas using geostatistics.

Urban areas are frequently considered more contaminated than other regions, and a great deal of research on soil pollution caused by heavy metals has been conducted in urban areas [37–42]. However, the soils in suburban areas not only contain exogenous heavy metal pollutants that migrate from urban areas to input sites but also overlap with the increasingly strong heavy metal emissions generated by human activities and industrial production in urban fringe areas, leading to the exacerbation of soil heavy metal pollution in urban fringe areas [43]. Therefore, soil heavy metal pollution in suburban areas is more serious and complex than that in urban areas [44]. These areas are characterized by highly variable agricultural and industrial activities, which result in simultaneous exposure to multiple sources of emissions. Researching and evaluating heavy metal pollution in soil is essential for implementing strategies to protect ecological health and grain safety. There is relatively little research on heavy metals in the environment of typical industrial city suburbs, and most related studies have focused on certain types of urban suburbs, such as mining cities [44] and developed cities [45].

The western part of Laizhou is a typical industrial city suburb. With the advancement of industrialization and urbanization, such as the continuous exploration of various mineral deposits, there may be some environmental issues with the soil, including heavy metal contamination. On the other hand, the research region is one of the main grain-producing areas in Shandong Province, and excessive use of fertilizers and unreasonable spraying of pesticides are also major reasons for the accumulation of heavy metals in soil [46]. To date, there is little public information about soil heavy metal pollution in Laizhou City, and there is a lack of systematic analysis and research, which has led to certain obstacles in the study of heavy metal distributions in the region and has affected the assessment and treatment of heavy metal pollution.

To better study heavy metals in the soil of the western suburbs of Laizhou, we adopted a combination of multivariate statistical analysis, geostatistical analysis, the Nemero comprehensive index, and potential ecological risk index methods. The main purpose of this study was to (1) clarify the concentration of heavy metals in the soil, (2) characterize their spatial distribution, (3) explore and analyze the possible sources of soil heavy metal pollution, and (4) reveal environmental risks [47].

2. Materials and Methods

2.1. Study Area

Laizhou (119°33′–120°18′ E, 36°59′–37°28′ N) is west of Yantai, Shandong, China, and has a total area of 1928 km² and a population of nearly 0.84 million people. The region of Laizhou has a typical temperate continental monsoon climate and warm and wet conditions in summer. The annual average temperature is 12.5 °C, and there are strong seasonal temperature variations. Rainfall events are strong and seasonal, with an annual average value of 809 mm (mostly in June, July, and August). There are 16 major rivers, such as the Wang River, and the terrain of Laizhou is high in the southeast and low in the northwest. The mineral resources include iron, gold, copper, talc, magnesite, and granite, and industries include chemical, construction, machinery, textile, etc. Additionally, Laizhou has a long history of rapid development in recent decades. Zhao et al. evaluated the potential risk of heavy metals in Laizhou City [47]. Some researchers have studied the current situation of heavy metal pollution in the topsoil on the south coast of Laizhou Bay [48,49].

2.2. Soil Sampling and Analysis

In accordance with the principles of accessibility, typical representativeness and elimination of human factors, and comprehensive consideration of landforms, soil types, and land utilization types, ArcGIS 10.5 was used to arrange the sampling points in Figure 1 on a digital base map according to a regular grid. The sampling points were divided into 3–5 multipoint combinations according to the shape of plum blossoms, discarding animal and plant residues, discarding gravel, etc., during collection. A total of 271 topsoil samples (0–20 cm) were collected and placed into polyethylene sealed bags. Then, the samples were air-dried at room temperature (20–25 °C), passed through 2 mm nylon sieves, and stored in plastic bottles [50].

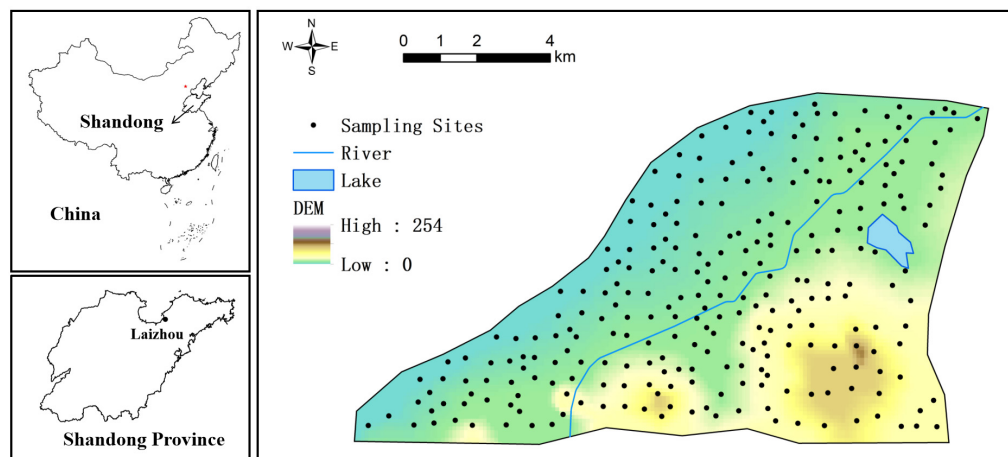


Figure 1. Area of study and sampling sites.

Soil sample experiments were carried out in the Analysis and Testing Laboratory, Yantai Coastal Geological Survey Center, China Geological Survey. This laboratory has a national CMA testing qualification. The heavy metal content of the soil was determined via the rock mineral analysis (DZG20.01-2011) procedure [51]. Heavy metal testing was performed as follows: 0.5 g soil samples were weighed and put into polyvinyl chloride (PVC) digestion containers. The samples were digested with a 10 mL mixture of HCL, HF, HNO₃, and HCLO₄, and the digested solutions were diluted with 2% HNO₃ to a final volume of 50 mL. The concentrations of As and Hg in the samples were measured via atomic fluorescence spectrometry (AFS) [52]. The concentrations of Cu, Cr, Mn, Ni, Pb, and Zn were determined via inductively coupled plasma–mass spectrometry (ICP–MS) [53]. Environmental soil standard reference material and repetitive and blank samples were

used to control the precision and accuracy. During the testing process, 10% parallel sample analyses and two sample blanks were used for each batch of samples. The qualification rate of parallel samples was 100%, and the sample blanks were less than the detection limit of the method. The national certified materials and parallel samples were used to check the quality of the analysis results.

2.3. Statistical Analysis

2.3.1. Descriptive and Multivariate Statistical Analysis

Descriptive statistical methods were used to preliminarily analyze heavy metals in the soil samples and determine the overall trends of the statistical indicators, including minimum, maximum, median, average, standard deviation, variation coefficient, skewness, and kurtosis. Correlation analysis, cluster analysis, and principal component analysis were carried out to reveal whether the elements were of the same origin or had similar geochemical behaviors [54]. All the statistical analyses were performed using SPSS 19.0.

2.3.2. Geostatistical Analysis

Soils are highly heterogeneous and spatially continuous [55]; even within the same soil texture area, the values of soil properties at different points are not the same. The major roles of geostatistics in environmental edaphology are the estimation, prediction, and mapping of soil properties in unsampled areas [56]. Prior to geostatistical analysis, the heavy metal data need to be tested for a normal distribution. The semivariance function was used to describe the structural and random characteristics of the regional variables, including the linear model, spherical model, exponential model, and Gaussian model. The parameters included the nugget (C_0), sill (C_0+C), range (range), determination coefficient (R^2), residual error (RSS), and nug/sill ratio $C_0/(C_0+C)$. GS+9 was used to fit the model, and the optimal variogram model was selected based on the principle of maximizing R^2 and minimizing the RSS.

The geostatistical methods proposed by Kriger in the 1970s have overcome the shortcomings and limitations of studying the spatial variation or spatial distribution of soil properties [57]. Kriging is a regression algorithm based on a covariance function for spatial modeling and prediction of random fields and includes ordinary kriging, universal kriging, cokriging, and disjunctive kriging. Kriging is a typical and famous geostatistical estimation technique that has been successfully applied to soil heavy metal mapping [58–60]. Among the various interpolation methods, ordinary kriging, in particular, is one of the most popular and robust geostatistical methods widely used [61].

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

where $Z(xi)$ represents the value of the variable Z at position xi , h is the log, and $N(h)$ represents the data logarithm of interval h . Interpolation and mapping by kriging were performed using ArcGIS 10.5.

2.4. Assessment of Heavy Metal Pollution

2.4.1. Nemero Synthesis Index Method

The Nemero synthesis index is a multifactorial, integrated assessment approach in which a single-factor pollution index is combined in a certain way [62]. It is only necessary to compare these results with the corresponding classification standards to determine the integrated environmental quality of environmental factors. The calculation formula of the comprehensive index is as follows:

$$Pi = \frac{Ci}{Si} \quad (2)$$

$$P = \sqrt{\frac{P_{imax}^2 + P_{ave}^2}{2}} \quad (3)$$

where P_i represents the single-element pollution index, i represents different trace elements, C_i is the measured concentration, S_i represents the threshold value, P_{imax} stands for the maximum pollution index, and P_{ave} represents the average value of all the elements.

2.4.2. Potential Ecological Risk Index Method

The potential ecological risk index method was developed by the Swedish scientist Hakanson in 1980 to evaluate heavy metal pollution and its ecological hazards [63]. This method considers not only the content of heavy metals in soil but also the synergistic effect of multiple elements and comprehensively considers the ecological, environmental, and toxic effects of heavy metals. Therefore, it is widely used in ecological risk assessment. The calculation formula is as follows:

$$E_r^i = T_r^i \times \frac{C_i}{C_n^i} \quad (4)$$

$$RI = \sum_{i=1}^n E_r^i \quad (5)$$

In the above equation, E_r^i is the potential ecological risk index of the i th heavy metal element, and RI is the comprehensive potential risk index of multiple heavy metal elements. Similarly, the toxicity response coefficient of the i th heavy metal is T_r^i (Hg = 40, Cd = 30, As = 10, Pb = Ni = Cu = 5, Cr = 2, and Zn = 1) [63]. C_i is the measured value of the i th heavy metal and the evaluation value of the i th heavy metal element (using local soil geochemical background values, [64]).

The evaluation criteria for the potential ecological risk index method are shown in Table 1.

Table 1. Evaluation standards for the potential ecological risk index [63].

E_r^i	Potential Ecological Risk Level	RI	Potential Ecological Risk Level
$E_r^i < 40$	Slight	$RI < 150$	Slight
$40 \leq E_r^i < 80$	Medium	$150 \leq RI < 300$	Medium
$80 \leq E_r^i < 160$	Strong	$300 \leq RI < 600$	Strong
$160 \leq E_r^i < 320$	Very strong	$600 \leq RI < 1200$	Very strong
$E_r^i \geq 320$	Extremely strong	$RI \geq 1200$	Extremely strong

3. Results

3.1. Spatial Distribution of Heavy Metals

The distributions of eight heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) in the study area are shown in Figure 2. Overall, the distribution of heavy metals showed a polarized trend, with some heavy metals having high concentrations in the northern part of the study area and low concentrations in the southern part (As, Cd, Hg, and Pb), while others had the opposite trend (Cr and Ni). High values of Zn and Cu were scattered throughout the area. The distribution characteristics of the heavy metals Cr and Ni in the study area were similar, with a symmetrical boundary line in the middle, a higher south and a lower north. As, Cd, Cu, Pb, and Zn all had high distributions in the northwestern region. All the elements exhibited low-concentration bands in the coastal areas in the western part of the study area.

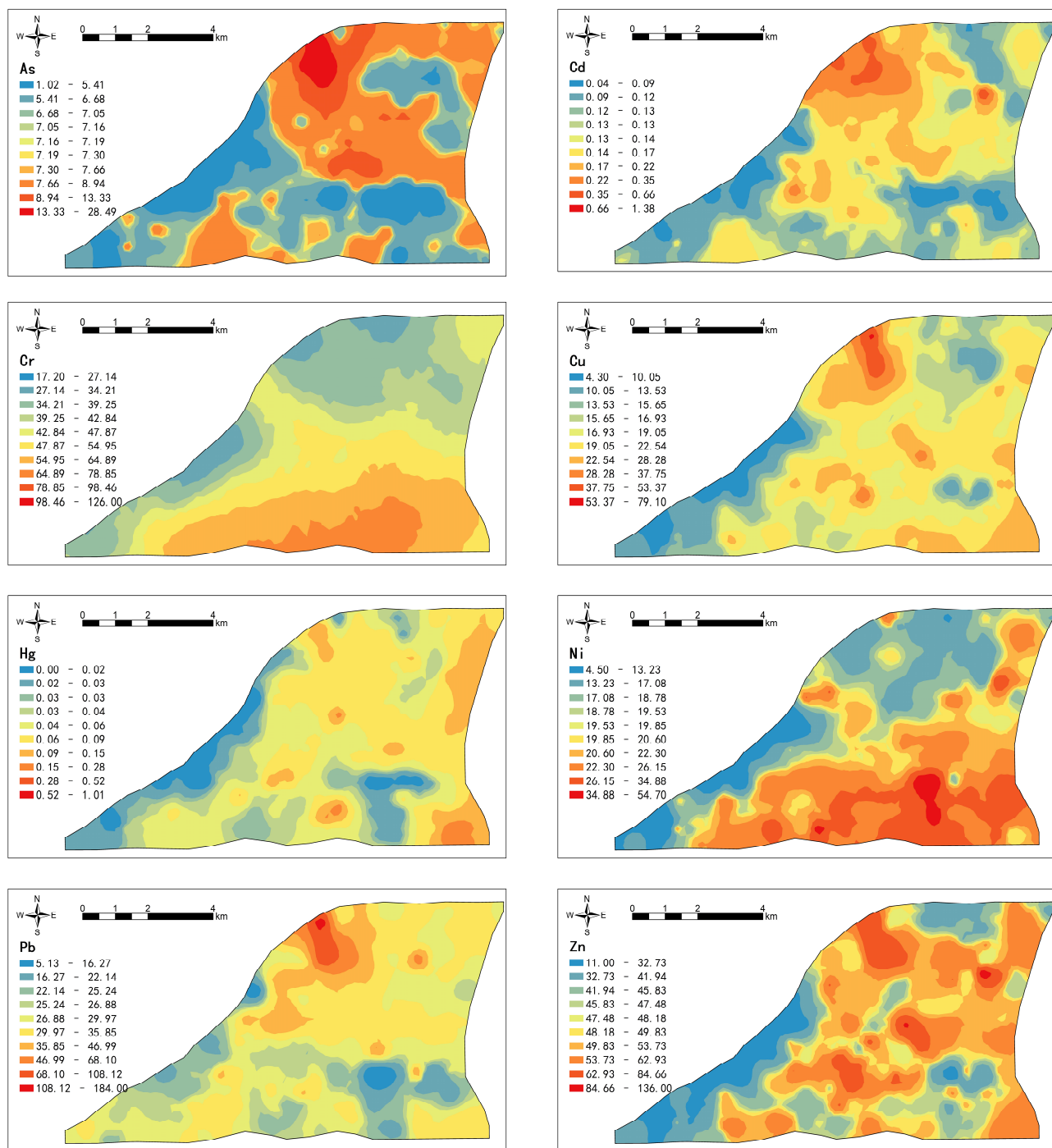


Figure 2. Distribution of 8 heavy metals in the study area ($\text{mg}\cdot\text{kg}^{-1}$).

3.2. Descriptive Statistics

The descriptive statistics of the eight heavy metals listed in Table 2 showed that the average contents of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were $7.13 \text{ mg}\cdot\text{kg}^{-1}$, $0.15 \text{ mg}\cdot\text{kg}^{-1}$, $48.40 \text{ mg}\cdot\text{kg}^{-1}$, $18.65 \text{ mg}\cdot\text{kg}^{-1}$, $0.06 \text{ mg}\cdot\text{kg}^{-1}$, $20.60 \text{ mg}\cdot\text{kg}^{-1}$, $30.99 \text{ mg}\cdot\text{kg}^{-1}$, and $48.89 \text{ mg}\cdot\text{kg}^{-1}$, respectively. The average values of Cr, Cu, Ni, and Zn were lower than the soil background values of heavy metals in Shandong Province [64]; however, the maximum values of eight elements were all higher than the background values in Shandong Province, which showed that they were all affected by human activities. The average values of As, Cd, Hg, and Pb were 1.12, 1.25, 2.00, and 1.14 times greater, respectively, than the corresponding soil background values of heavy metals.

Table 2. Statistics of heavy metal contents.

Elements	Minimum (mg·kg ^{−1})	Maximum (mg·kg ^{−1})	Median (mg·kg ^{−1})	Average (mg·kg ^{−1})	Standard Deviation	Variation Coefficient (%)	Background Values (mg·kg ^{−1})	Skewness	Kurtosis
As	1.02	28.50	7.00	7.13	2.67	37.45	6.40	2.83	19.29
Cd	0.04	1.38	0.13	0.15	0.11	73.33	0.12	6.81	65.24
Cr	17.20	126.00	46.50	48.40	15.34	31.69	57.00	1.26	3.85
Cu	4.30	79.10	17.50	18.65	8.18	43.86	26.00	2.90	15.60
Hg	0.002	1.01	0.04	0.06	0.08	133.33	0.03	7.91	84.18
Ni	4.50	54.70	20.00	20.60	7.00	33.98	24.60	1.64	6.52
Pb	5.13	184.00	28.50	30.99	14.20	45.82	27.20	6.30	57.39
Zn	11.00	136.00	47.10	48.89	16.11	32.95	60.40	1.42	5.61

The variation coefficient is the ratio of the standard deviation to the average and is used to compare the degree of data dispersion. According to Wilding's variation classification [65], the coefficients of variation for As, Cd, Cr, Cu, Ni, Pb, and Zn were 37.45%, 73.33%, 31.69%, 43.86%, 33.98%, 45.82%, and 32.95%, respectively, and exhibited moderate variation ($10\% < CV < 100\%$). A coefficient of variation (CV) greater than 50% usually indicates greater spatial variability [66]. In this case, complex external inputs and geological evolution may have been the major reasons for this phenomenon [67]. That of Hg was 133.33%, which was a strong variation ($CV > 100\%$). Compared with that of other heavy metals, the variation coefficient of Hg, in particular, was greater, indicating that the distribution of Hg was uneven and might have been affected by human factors. In terms of skewness, the values of the eight elements were ordered as $Hg > 1 > Cd > 0.5 > Pb > Cu > As > Ni > Zn > Cr$. These results were consistent with the classification of variation.

4. Discussion

4.1. Source Analysis of Heavy Metals

4.1.1. Correlation Analysis

In this study, correlation analysis, principal component analysis, and cluster analysis methods were used to analyze the data for Hg, Cd, Pb, Cu, As, Ni, Zn, and Cr, explore their potential sources, and determine the spatial distribution characteristics of the heavy metals to analyze their sources.

The correlation coefficient between two variables was calculated to determine whether there was a correlation [53,68]. Commonly, Pearson and Spearman correlation analyses are used to establish correlations; however, Spearman correlation analysis does not require that variables obey a normal distribution [69]. The Spearman correlation coefficients in Table 3 show that the Cd-Cu, Cd-Pb, Cd-Zn, Cr-Ni, Cu-Ni, and Cu-Zn concentrations were 0.56, 0.51, 0.59, 0.89, 0.51, and 0.77, respectively ($p < 0.01$). Strong correlations were found between Cd, Cu, Pb, and Zn, suggesting that they might come from the same sources; additionally, Cr and Ni might have the same origin. The correlation coefficients between Pb, As, Cr, and Ni were all less than zero, indicating that there was a weak correlation. Principal component analysis and cluster analysis were subsequently used to further explore the relationships between the heavy metals and determine their potential sources.

Table 3. Correlation matrix among the heavy metals.

Elements	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
As	1							
Cd	0.440 **	1						
Cr	0.043	0.075	1					
Cu	0.432 **	0.561 **	0.352 **	1				
Hg	0.377 **	0.444 **	0.069	0.394 **	1			
Ni	0.166 **	0.093	0.893 **	0.512 **	0.104	1		
Pb	−0.430 **	0.513 **	−0.070	0.328 **	0.493 **	−0.125 **	1	
Zn	0.333 **	0.592 **	0.356 **	0.773 **	0.423 **	0.458 **	0.369 **	1

** $p < 0.01$. Bold indicates correlation greater than 0.5.

4.1.2. Principal Component and Cluster Analysis

The Kaiser–Meyer–Olkin value for the soil samples was 0.733, and that of Bartlett’s test ($p < 0.001$) was low. The results showed that principal component analysis had good adaptability to the dataset [70]. Three principal components (PCs) were identified, as shown in Table 4, and cumulatively contributed 79% of the total variance. These PCs can be used to comprehensively characterize heavy metals [71]. A graphic of the components (PC1, PC2, and PC3) is displayed in Figure 3, showing the relevance of the heavy metals. Additionally, a cluster analysis of the heavy metals shown in Figure 4 revealed that they could be divided into three categories: (1) As, Cd, Cu, Pb, and Zn; (2) Cr and Ni; and (3) Hg, further verifying the results of the principal component analysis.

Table 4. Factor matrix of heavy metals.

Elements	Principal Component		
	PC1	PC2	PC3
As	0.81	−0.09	0.09
Cd	0.75	−0.12	−0.09
Cr	−0.05	0.94	0.05
Cu	0.81	0.29	0.19
Hg	0.14	0.05	0.97
Ni	0.03	0.96	0.01
Pb	0.81	−0.22	0.19
Zn	0.80	0.32	0.08
Percentage of variance (%)	40%	26%	13%
Percentage of cumulative variance (%)	40%	66%	79%

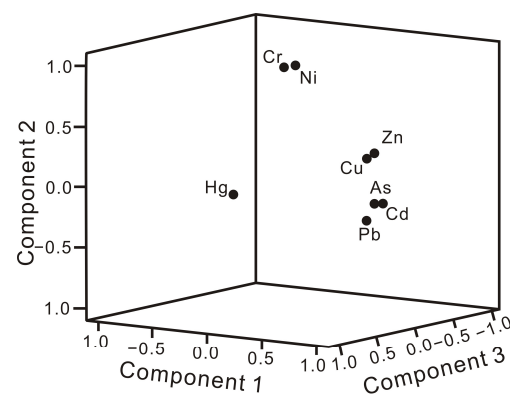


Figure 3. Loading of the three principal components.

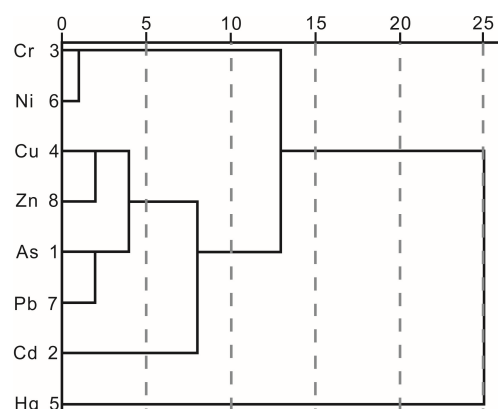


Figure 4. Clustering tree of heavy metals.

PC1 explained 40% of the variance, and As, Cd, Cu, Pb, and Zn had large loads of 0.81, 0.75, 0.81, 0.81, and 0.80, respectively. Zhuo et al. [72] and Huang et al. [44] demonstrated that the accumulation of Pb is related to coal burning in general. A large amount of Pb and Zn is released from automobile tire wear and corrosion of galvanized automobile parts [73,74]. In addition, Zn and its compounds have been extensively applied in agricultural chemical applications [75]. Cd and Cu are also common elements found in pesticides and fertilizers. Long-term application in agriculture is the main reason for the enrichment and increase in these heavy metals in soil [76]. Hence, As, Cd, Cu, Pb, and Zn in soil could be related mainly to agricultural practices or traffic activities.

PC2 accounted for 26% of the variance, and Cr and Ni had loads of 0.94 and 0.96, respectively. Previous studies have suggested that Ni and Cr in soil are mainly affected by the background geological materials, and there are strong correlations between Cr and Ni [50,77,78]. Additionally, Cr and Ni seem to be the least polluting elements in all Chinese cities [79]. The process of soil formation and the parent material are the primary factors influencing the contents and distributions of Cr and Ni in Shandong Province [80]. Clearly, soil Cr and Ni primarily originate from geological parent materials.

In PC3, only Hg had a large load of 0.97, and the mean value was twice as high as the background value in Shandong Province. The variation coefficient of Hg was the largest (133.33%), illustrating that human activities were a significant cause of element enrichment [81]. The Hg content in soil was correlated with the emission of industrial wastewater and flue gas [82]. Shandong Province is rich in mineral resources, as mentioned above, and the soil could be indirectly polluted by quarries through atmospheric sedimentation [83]. Therefore, the high content of Hg in soil has been closely related to the development of industry and mining in recent years.

4.1.3. Spatial Distribution and Source Analysis of Heavy Metals

The previous section discussed the types and characteristics of heavy metals and their relationships. Below, we analyze the sources of heavy metals by studying their spatial distribution characteristics. First, the variation function model (Table 5) was used to explore the spatial variations in the two variables:

Table 5. Summary of the semivariogram models and corresponding parameters.

Soil Properties	Model	Nugget (C_0)	Sill (C_0+C)	Range (m)	R^2	RSS	$C_0/(C_0+C)$ (%)
PC1	Spherical	0.0443	0.0921	5480	0.949	1.7×10^{-4}	48.10
PC2	Spherical	0.0423	0.1286	8510	0.988	1.241×10^{-4}	32.89
PC3	Exponential	0.0254	0.0714	2040	0.966	6.6×10^{-5}	35.57
As	Exponential	0.0212	0.1584	810	0.730	3.223×10^{-3}	13.38
Cd	Spherical	0.0989	0.1988	4940	0.814	2.687×10^{-3}	49.75
Cr	Spherical	0.0332	0.1254	7170	0.984	1.854×10^{-4}	26.48
Cu	Exponential	0.0778	0.1976	4000	0.968	4.013×10^{-4}	39.37
Hg	Exponential	0.0254	0.0714	2040	0.966	6.6×10^{-5}	35.57
Ni	Spherical	0.0547	0.1534	8650	0.984	2.106×10^{-4}	35.66
Pb	Spherical	0.0444	0.1168	4190	0.670	3.066×10^{-3}	38.01
Zn	Spherical	0.0752	0.1514	13,600	0.933	2.814×10^{-4}	49.67

According to the results in Table 4, Cu, Hg, and PC3 conformed to the exponential model, and Cd, Cr, Ni, Pb, Zn, and PC1 and PC2 were in accordance with the spherical model. The effective variation range of each variable was 810~13,600 M. The R^2 values for As, Cd, Cr, Cu, Hg, Ni, Pb, Zn, PC1, PC2, and PC3 were 0.730, 0.814, 0.984, 0.968, 0.907, 0.984, 0.670, 0.933, 0.949, 0.988, and 0.966, respectively, indicating that the correlation between the model and the actual data was high, that the fitting effect was good, that the RSS was sufficiently small, and that the fitting error results met the requirements. Through the range change, we found that the spatial variation range of most heavy metal elements

was between 3000 and 10,000 m, indicating that they are widely distributed in the study area and that the sources may be relatively diverse; however, the range change was very small, at 810 m, indicating that the distribution range of these elements in the study area is very small and concentrated. Additionally, the sources may be single or small, such as a certain type of factory or a certain mine. In addition, the range of Zn was very large, at more than 10 km, which indicated that it is widely distributed in the study area and may have a wide range of sources, such as the soil matrix or farmland. The same may be the case for Ni because the range of Ni was more than 8 km.

According to the above optimal semivariogram theoretical models, the spatial distributions of the three principal components were described, as shown in Figure 5.

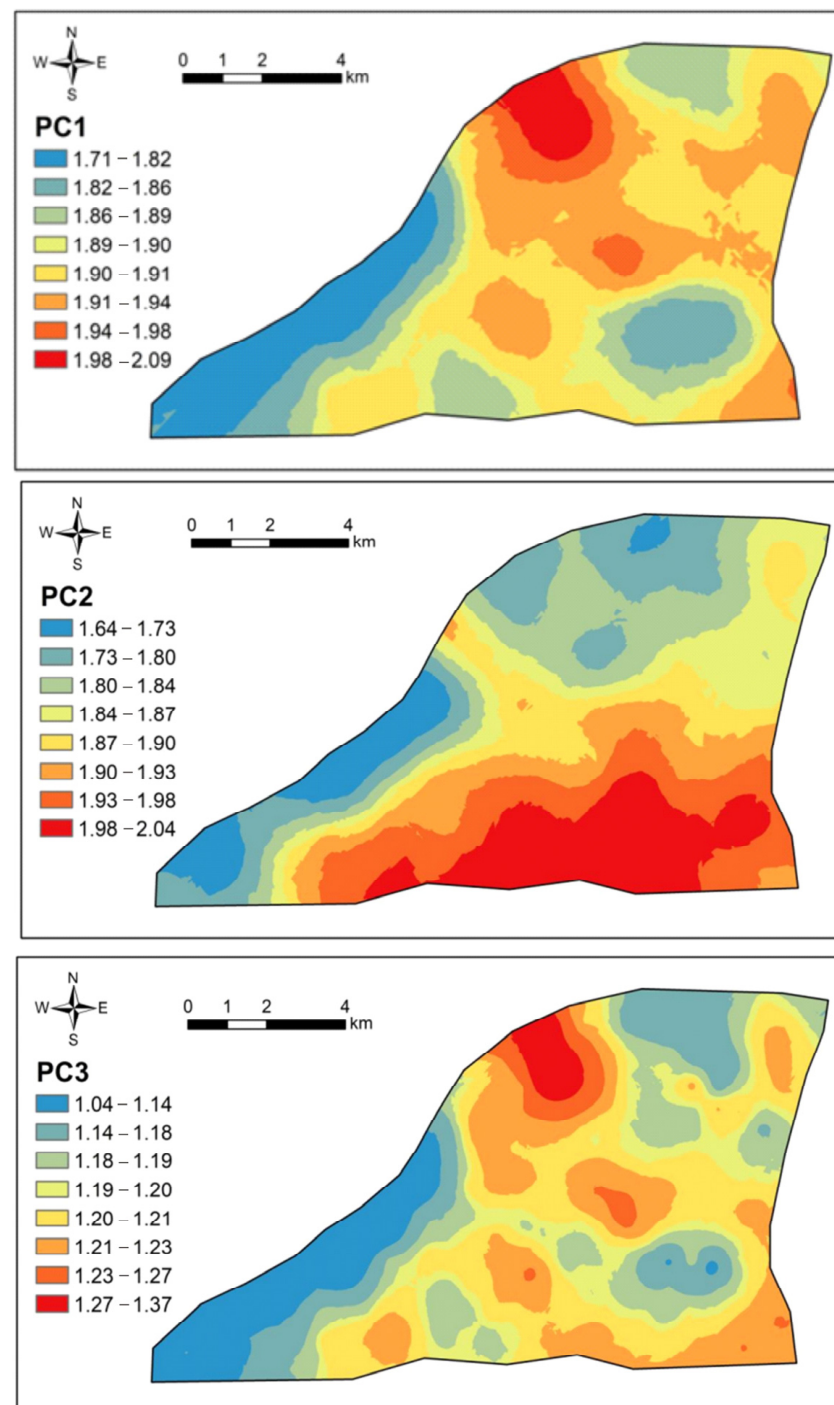


Figure 5. Spatial patterns of the principal components of heavy metals.

4.1.4. Source Analysis of Heavy Metals

The land-use type in the study area was determined through field surveys during the process of obtaining satellite images and soil sampling (Figure 6), and the source of heavy metals was more accurately inferred through the combination of these data with the data analysis results.

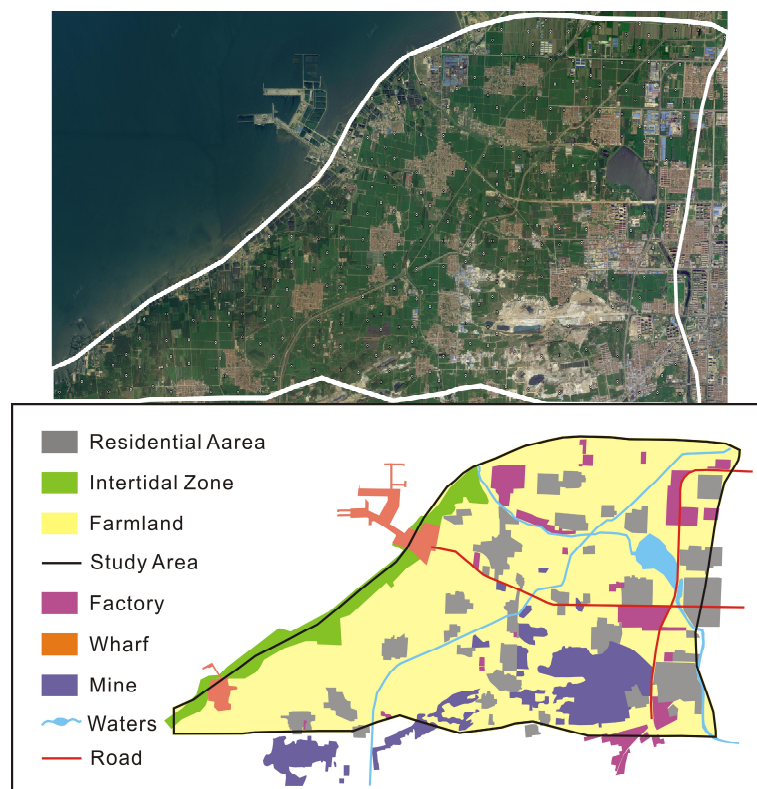


Figure 6. Land-use types in the study area.

PC1 mostly included Cd, Cu, Pb, and Zn, which were distributed in sheets, mostly in the middle, eastern and northern parts of the study area, and the highest was in the northwestern part. By comparing the distribution of factories and farmland in the study area, it was found that there was a large-scale paper mill in the northwestern hotspot. Paper pulping and wastewater removal caused by intensive industrial activities have been shown to be serious sources of heavy metals such as Cu, Zn, Cd, and Pb [84,85], which may indicate the presence of high heavy metal concentrations. There are also other types of factories in high-value areas. The area with a high PC1 is mainly covered by farmland, there is a winter wheat and corn rotation growing area in Laizhou, and compound fertilizers usually containing Zn are necessary for crop yield improvement [86]. As, Cd, and Cu are generally considered the landmark elements of agricultural activities. Therefore, during long-term cultivation, heavy metals continue to accumulate in the soil. Moreover, PC1 in the ore concentration area and residential area was in a low value state; therefore, it can be further speculated that the source of heavy metals in PC1 has little correlation with mining activities and daily life; therefore, it is speculated that the major sources of Cd, Cu, Pb, and Zn in PC1 in the study area can be divided into two categories: industrial production and agricultural planting.

The PC2 chart shows the distributions of Cr and Ni, revealing a decreasing trend from south to north, with the highest occurring in the southern region. Additionally, two bands extend northwestward and northeastward from the study area. A comparison of the land-use types (Figure 6) revealed that the high-value area in the southern region is located in a talc and magnesite mining area, and related processing and sorting plants occur in the northern region. Moreover, two strips trending northwest and northeast correspond

exactly to two highways. The northwest-trending highway leads to the dock, and the northeast-trending highway leads to the industrial park in Laizhou City. Frequent mining production and trade activities have led to significant transportation demands in the study area. Cr and Ni, two heavy metals, are present in the associated minerals and impurities of talc and magnesite. This is consistent with the previous speculation that Cr and Ni are influenced mainly by soil parent materials and that the southern soil parent materials are different from those in other regions. Therefore, comprehensive analysis suggested that Cr and Ni in the study area may mostly originate from the southern mining area and that As industrial activities may have spread throughout the research area.

PC3 (Hg) is distributed in blocks, mostly in the northwestern, central, and southern regions of the study area, without connecting into a large-scale sheet area. Almost every high-value area corresponds to a factory, especially the northwestern hotspot area, and the southeastern region also corresponds to a factory. The concentration in the ore concentration area is relatively low, which may be due to the long-term open-pit mining activities in the mining area, and the wastewater and gas discharged from mining production are usually accompanied by many heavy metals. Wang et al. [87] reported that contaminants released by mining activities led to the accumulation and enrichment of heavy metals. Wastewater irrigation, atmospheric deposition, and transportation in surrounding areas led to persistent outward movement of heavy metals [88]. However, in the large area of farmland with few factories, the concentration of Hg is very low, which indicates that the correlation between agricultural production activities and Hg is very small; therefore, it is speculated that there are two sources for PC3 (Hg): factories and mineral concentration areas, but mainly factories.

4.2. Environmental Risk Assessment of Heavy Metals

When the content of heavy metals in soil reaches a certain level, it may harm the surrounding environment or humans; thus, an environmental risk assessment of heavy metals is essential. Next, the Nemero synthesis index and potential ecological risk index were used to evaluate the ecological risk in the study area.

4.2.1. Nemero Synthesis Index

The heavy metal pollution levels in the topsoil of the study region are shown in Figure 7. The Chinese Soil Environmental Quality Assessment Standard (GB15618-2018) [89] was adopted for evaluation, with a mean soil pH of 6.45 [64]. Based on the Nemero synthesis index evaluation grade, a pollution level less than 0.7 is considered safe, a pollution grade ranging between 0.7 and 1 indicates that the environmental quality is in a critical state, and a pollution grade greater than 1 indicates that the environmental quality cannot meet the requirements of the evaluation standards. The pollution situation in the research region is not serious. A small part of the northwestern area is in a critical state, and people need to be vigilant there.

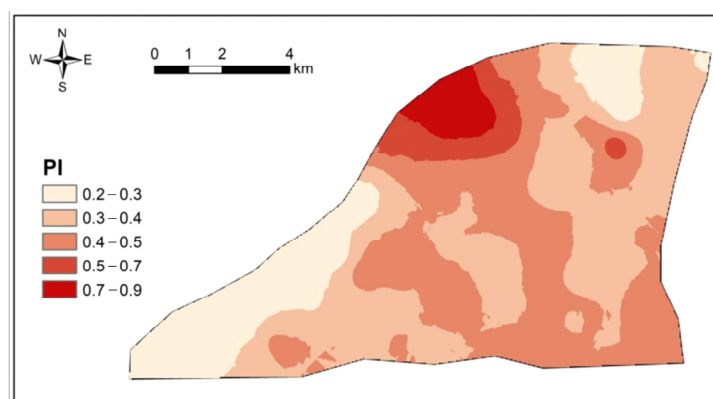


Figure 7. Environmental risk of heavy metals.

4.2.2. Potential Ecological Risk Index

According to the analysis of the potential ecological risk of single heavy metals, the potential ecological risk indices of Cr, Cu, Ni, Pb, and Zn in the soil of the study area were generally low (E_r^i , the maximum value was 33.82 and was less than 20 except for Pb), corresponding to the lowest degree of harm. When maximum value was 44.52, the potential ecological risk index was moderate. There were 60 moderate ecological risks, 7 strong ecological risks, 1 very strong ecological risk, and 1 extremely strong ecological risk for Cd (E_r , the maximum value was 353.85). There were 107 moderate ecological risks, 38 strong ecological risks, 12 very strong ecological risks, and 3 extremely strong ecological risks for Hg (E_r , the maximum value of high-value outliers was 118.24).

The comprehensive potential risk (RI) of eight heavy metal elements showed that only one sample reached a very strong ecological risk (Hg, high-value outlier, $RI = 1262.66$); there were six strong risks and two very strong risks, and the remaining samples had mild to moderate risk. Among them, 51 medium-risk samples were included. The risks mainly came from Hg and Cd, and the remaining samples had mild risk factors.

The potential ecological risks caused by heavy metals in the soil of the study area are notable, among which Hg has the highest contribution, Cd has a moderate risk, and the other elements basically have no risk. It is recommended that regular sampling and monitoring of the two elements be conducted and that the pollution sources be checked at the same time; for example, factory discharges should be checked for violations of regulations, such as the contents of heavy metals (such as pesticides) exceeding the standard.

5. Conclusions

By analyzing the correlation and spatial variation characteristics of eight heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) in the study area, we explored their potential sources and possible environmental impacts through environmental risk assessment, thus achieving the research objective of this article. Although the environmental risk of the soil in the study area is low, the average contents of As, Cd, Hg, and Pb are higher than the background values in Shandong Province, indicating that there is enrichment of heavy metals in the soil. The eight heavy metals can be divided into three major components. The contribution rates of As, Cd, Cu, Pb, and Zn in PC1 are 0.81, 0.75, 0.81, 0.81, and 0.80, respectively, which are mainly affected by agricultural activities, transportation, and industrial production in the study area. The contribution rates of Cr and Ni in PC2 are 0.94 and 0.96, respectively, which are mainly related to the local soil parent material, ore concentration areas, and major roads for mineral transportation. The contribution rate of Hg in PC3 is 0.97, which is mainly caused by the industrial production activities of plants, open-pit mining, and other factors. According to the two risk assessment methods, the environmental risk is generally relatively low. The main environmental risk comes from Hg and Cd, and their main source may be industrial production.

In this study, we evaluated only the sources and environmental risks of heavy metals in the suburbs of industrial cities based on existing data and land-use status. The major sources of heavy metals are 1. agricultural production activities, such as fertilizer and pesticides; 2. industrial production activities in factories, especially chemical plants; and 3. mining activities in mining areas. Due to the existence of factories and mining in the suburbs of industrial cities, heavy metal pollution risks are inevitable. In the suburbs of such typical industrial cities, controlling and preventing pollution is important. Long-term heavy metal testing is needed to determine whether and to what extent the soil has been polluted.

In a follow-up study, long-term monitoring will be carried out to conduct more in-depth research and provide additional information on the sources and environmental risks of heavy metals in the suburban areas of industrial cities.

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