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Short Communication

Assessment of Heavy Metal Pollution in Chinese Suburban Farmland

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Abstract

A total of 650 arable soil samples were collected from the suburbs of Liaocheng, Shandong Province, China to identify the concentrations and the sources of heavy metals and to assess the environmental quality of soil. Subsequently, the contents of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in the samples were analyzed. The investigation revealed that the mean concentrations of As, Cd, Hg, Ni, Hg, and Cd exceeded their corresponding background values. The maximum concentrations of As, Cd, Hg, and Cu are all higher than the allowable values. Each soil sample had a low or slight pollution index (*PI*) of As, Cr, Cu, Ni, Pb, and Zn, whereas the *PI* values of Hg and Cd were high; more than 39.7% and 18.6% of the samples were classified as being moderately or heavily contaminated by Hg and Cd, respectively. Using multivariate statistical approaches (cluster analysis and principal component analysis), four factors controlling the heavy metal variability were identified; these accounted for more than 80% of the total variance. The As, Zn, Ni, Cu, and Cr levels were controlled by the parent material in the soils. Cd and Hg levels were controlled mainly by anthropic sources, such as industrial activities, manure, and the burning of coal; however, the parent materials of the soils may partially control the concentrations of Cd and Hg. The abnormally high levels of heavy metals mostly coincided with industrial locations.

Keywords: heavy metals, contamination, spatial distribution, suburban farmland

Introduction

Due to their environmental toxicity and persistence, soil heavy metals are toxic to plants, animals and humans through water and food chain transport. Rapid urbanization, industrialization, and increases in the release of agrochemicals into the environment has led to increasing public concern regarding the potential accumulation of heavy metals in agricultural soils [1-3]. In China, heavy metal pollution in agricultural soils has become serious with the rapid industrialization and urbanization during the last two decades. Studies indicate that the background soil levels of heavy metals are low in China, while water, plants, soil and air have been found to be polluted by heavy metals due to anthropogenic activities in recent decades in some studied cases; this can affect human health via the food chain or through direct intake [3, 4]. During the past decade, numerous studies associated with heavy metal contamination in agricultural soils have been carried out focusing on the “sources” and “sinks” of soil heavy metals, the evaluation of the environmental quality of soil, the spatial distribution characteristics, food safety and the pollution evaluation method [2, 5-8]. However, less developed and medium-sized cities remain less studied.

As a spatial transition zone between urban and rural areas, suburban farmland plays an important role in providing regional food security and protecting the environment and the ecosystem. Soil heavy metals in suburban farmland may have been deposited and may accumulate more quickly than in rural areas due to rapid urban industrial development.

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Some researchers have indicated the need for a better understanding of suburban agricultural soil pollution [6, 9], and, indeed, increasing research has focused on heavy metals in suburban farmland soils. Heavy metals in suburban soils may come from mining, smelting, waste disposal, vehicle exhausts, urban effluent, sewage sludge, fertilizer application, pesticides, and other sources [9-12]. Understanding the spatial distribution of suburban topsoil heavy metals is important for environmental management and agricultural production. The spatial variability of soil heavy metals is a significant part of ecosystem evaluation and environmental supervision. Geostatistics has successfully been applied to investigate and map soil heavy metals [2, 9, 13]. Yang [9] suggested that spatial principal component analysis was helpful in identifying anthropogenic sources of soil heavy metals, which could reveal both the spatial structures and the relationships among multiple variables at a given spatial scale.

In the North China Plain, one of China's most important agricultural regions, soil heavy metal contamination caused by intensive anthropogenic activities has become an urgent problem. In the past two decades, research on heavy metal concentrations in agricultural soils, forest soils, urban soils, and urban parks has been conducted in Beijing, which is located in the northern portion of the North China Plain [14]. However, the spatial variability in heavy metal deposition and the subsequent soil pollution levels remain unknown in the relatively underdeveloped cities in this area. Based on the observed data, the spatial variability of heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) in suburban areas of Liaocheng is investigated using techniques such as geostatistics, statistics, and geographical information systems (GIS) for the purpose of evaluating heavy metal spatial distribution and the scale of variability. This provides efficient estimates for soil environmental monitoring and valuable information for regional soil quality management.

Material and Methods

Study Area

Liaocheng (35º47′ to 37º02′N, 115º16′ to 116º32′E) is an important industrial city in Shandong Province in eastern China (Fig. 1). It covers an area of 8,715 km^2 and has approximately 6,350,000 inhabitants. It has a typical continental monsoon climate, including warm temperatures, semi-humid conditions, well-defined seasons, and is rich in water resources. The mean annual temperature is 14ºC, and the average mean precipitation is 570 mm. The soil of Liaocheng is typically moist; the soil textures mainly include sandy soil, loam soil, and clay soil. It is an important cotton grain base both in the Shandong province and in the whole country.

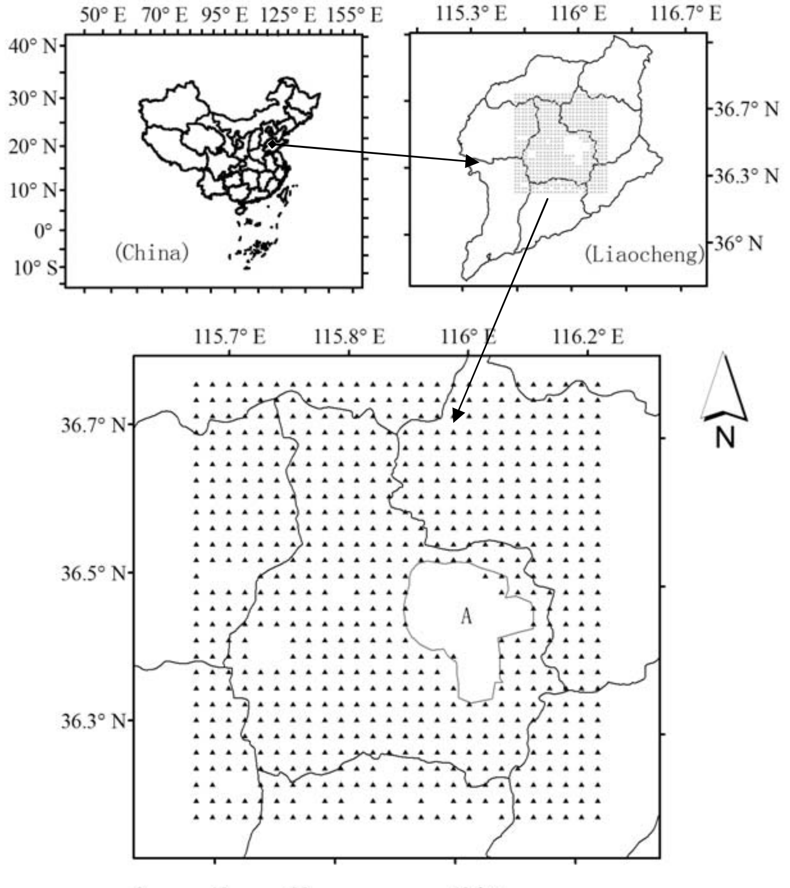


Fig. 1. Map of the sampling sites in Liaocheng. A: Main urban area of Liaocheng.

Table 1. Summary statistics of heavy metal concentrations in agricultural soils of Liaocheng, and background values of Shandong Province. All concentrations are in mg/kg dry weight.

	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Mean	10.74	0.14	62.59	21.75	0.04	26.35	21.34	63.06
SD	1.75	0.03	5.57	3.10	0.02	3.11	2.30	7.38
BC	9.30	0.08	66.00	24.00	0.02	25.80	25.80	63.50
Guideline value	15	0.20	90	35	0.15	40	35	100
C.V. (%)	16.3	20.4	8.9	14.2	54.7	11.8	10.8	11.7
Geom. M.	10.70	0.14	62.56	21.74	0.04	26.27	21.42	63.10
Geom. S.D.	2.17	0.03	5.60	3.32	0.06	3.20	2.58	7.91
Min.	1.90	0.08	39.40	13.50	0.01	16.40	14.70	42.50
Max.	17.60	0.34	83.70	41.50	0.26	37.70	30.60	93.70
Skewness	0.25	1.35	0.00	0.88	3.34	0.19	0.18	0.12
Kurtosis	1.14	3.89	1.03	2.72	9.12	0.57	0.09	0.80
Range	15.70	0.26	44.30	28.00	0.25	21.30	15.90	51.20
Distribution	Lognormal	Lognormal	Lognormal	Lognormal	Lognormal	Normal	Normal	Normal

SD – standard deviation, BC – background concentrations in the soils of Shandong [17], Guideline value – soil environmental quality standard GB 15618–1995, C.V. – coefficients of variation, Geom.M – geometric mean, Geom.S.D – geometric standard deviation, Min. – minimum value, Max. – maximum value.

Sampling and Analysis

Based on an electronic map of Liaocheng (scaled 1:250,000), uniform grids of 2×2 km were obtained for the whole study area; each grid center is a sampling point (Fig. 1). Topsoil samples (0-20 cm) were collected from 650 different grids in the whole suburban Liaocheng city in 2008 (Fig. 1). Certain sampling point locations varied within small limits according to the specific topography, land use, and soil type provided by topographic maps with a scale of 1:50000, soil maps with a scale of 1:100,000, and aerial photos with a scale of 1:10,000. Soil samples were obtained by mixing five subsamples from each site within a 20×20 m area and recorded for the central point position using a GPS device. Approximately 1 kg soil samples were collected at each location using a stainless steel spade and stored in self-sealing plastic bags.

Chemical Analysis

All soil samples were air-dried, ground, and sieved through a 2 mm nylon sieve to remove coarse materials and other debris. Then a portion of each sample was further ground with a pestle and mortar until all particles passed through a 0.15 mm nylon sieve. For the analysis of total heavy metal concentrations, one gram of each dry soil sample was digested in Teflon tubes with a mixture of perchloric acid ($HClO_4$), nitric acid (HNO_3), and hydrogen fluoride (HF). The solution of each digested sample was analyzed by inductively coupled plasma atomic absorption spectrometry (ICP/AES) for the following heavy metals: As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn [15]. Standard reference materials obtained from the Center of National Standard Reference Materials of China and blank samples were inserted with each batch of samples (1 blank and 1 standard for each 10 samples) for quality assurance and quality control. The analytical precision, measured as relative standard deviations, were all less than 10%. All samples were analyzed in duplicate and results were accepted when the relative standard deviation was within 5%. The results met the accuracy demand of the Technical Specification for Soil Environmental Monitoring HJ/T 166-2004 [16].

In this study, principal component analysis (PCA) and cluster analysis (CA) were employed. Because the soil heavy metal concentrations varied greatly, PCA and CA were performed on the standardized datasets to minimize the effects of differences in measurement units and variance [5]. Additionally, statistical calculations were performed within each cluster to allow for further comparison between clusters [14]. Other statistical analyses included the mean, the standard error, the maximum, the minimum and the coefficient of variation.

To assess the environmental quality of soil, a pollution index (*PI*) was introduced for each metal. The *PI* of each heavy metal is defined as the ratio of its concentration to the background value of the corresponding metal of Shandong using the following equation [14]:

$$PI = C_i / S^i \qquad (1)$$

...where *PI* is the pollution index corresponding to each sample, C_i is the measured value of each metal (mg/kg), and S_i is the background value (mg/kg). In this study, the soil background values of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were taken from measurements in the Shandong Province and the values were 9.30, 0.08, 66.00, 24.00, 0.02, 25.80, 25.80, and 63.50 mg/kg, respectively [17]. The PI of each metal was calculated and classified as either low ($PI \leq 1$), slight ($1 < PI \leq 2$), moderate ($2 < PI \leq 3$), or high ($PI > 3$) [14].

All of the above statistical calculations were carried out with SPSS 13.0 and Origin Pro 8.0 software packages. The environmental soil quality maps of eight heavy metals spatial distributing pattern were produced using a Kriging interpolation technique in ArcGIS 9.3.

Results

Descriptive Statistical Parameters

The statistical results of the geometric mean and the geometric standard deviation are shown in Table 1. The mean value was used when the concentrations of the soil heavy metal had a normal distribution, and the geometric mean value was used when the concentration of the heavy metal had a lognormal distribution [14].

Because there appeared to be outliers near pollution sources, the normal distribution of each soil heavy metal was tested. Soil heavy metals did not have normal distributions, except for Ni, Pb, and Zn; most of the distributions showed positive skewness and kurtosis values greater than zero ($p<0.05$), which affects the results of the chemometric analysis. However, after log-transformation [18], all of the transformed variables, including As, Cd, Cr, Cu, and Hg, nearly fit a normal distribution (Table 1). The distribution of the data was tested with the Shapiro-Wilk method ($p<0.05$).

The maximum measured values of all of the eight heavy metals were more than double the minimum values, with large spatial ranges. The mean concentrations plus the standard deviations of Hg and Cd are 0.04±0.02 mg·kg^{-1} and 0.14±0.03 mg·kg^{-1}, respectively. These values are significantly higher than the background concentration, indicating possible pollution at some points in the study area. The Pb (mean±S.D.) exhibited a lower concentration than the background value; this was most likely caused by weathering and lithogenic fluxes of rich parent materials. The observed maximum concentrations of As, Cd, Hg, and Cu are all higher than the guideline values [19], particularly for the heavy metals Hg and Cd.

The coefficient of variation (C.V.) values of eight soil heavy metals ranged from 8.9% to 54.7%, indicating that they had moderate variations. Hg has the highest C.V. value (54.7%), indicating that Hg would have the highest possibility of being affected by the extrinsic factors such as anthropogenic activities. Cr has the lowest C.V. value from the eight heavy metals (8.9%), suggesting that Cr has a weak variation and may more often be associated with natural sources (Table 1). The sequence of the other C.V. values was Cd>As>Cu>Ni>Zn>Pb.

Correlation Coefficient Analysis

The Pearson correlation coefficients of the heavy metals in the agricultural soils of Liaocheng are summarized in Table 2. Generally, the relationships between heavy metals can provide important information on the sources and pathways of heavy metals [20]. Significant correlations were found between As, Cd, Cr, Cu, Ni, Pb, and Zn; among them, the correlation coefficients of Zn-Ni, Zn-Cu, and Ni-Cu are all higher than 0.7, suggesting that these heavy metals may originate from a common source, such as agricultural activities. However, Hg showed only weak positive correlations with the other heavy metals, suggesting that it has a different source than the other heavy metals.

Multivariate Analysis Results (PCA)

Cluster analysis (CA) was applied to assist in the identification of pollutant sources. The results are shown in Fig. 2. The distance axis represents the degree of association between groups of variables. The eight heavy metals can be classified into six clusters using the criteria of a rescaled distance between 0 and 5. Cluster I contained Ni, Zn, and Cu. There are significant correlations between these three heavy metals, indicating that they have naturally associated relationships. Clusters II to VI each contained only one heavy metal.

PCA was used to further analyze the relationships between the soil heavy metals. The total variance explained by the PCA and the component matrices for the eight heavy metal concentrations in agricultural soils from Liaocheng are listed in Table 3. The results of the PCA suggest that the eight heavy metals could be reduced to four components, which accounted for more than 80% of the total variance of the data (Table 3). The communalities of the variables were 2% for Hg, 33% for Cd, 56% for As, 84% for Ni, 94% for Cu and Pb, and 96% for Zn and Cr.

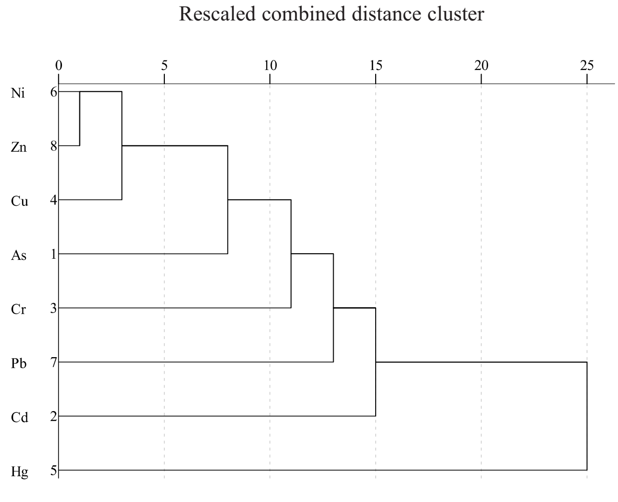


Fig. 2. Dendrogram of the hierarchical cluster analysis of the heavy metal concentrations in the agricultural soils of Liaocheng. The distance axis represents the degree of association between groups of variables.

Table 2. Pearson correlation matrix for heavy metals in the agricultural soils of Liaocheng.

Element	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
As	1.00							
Cd	0.43*	1.00						
Cr	0.53*	0.38*	1.00					
Cu	0.62*	0.36*	0.50*	1.00				
Hg	0.07	0.05	0.02	0.08*	1.00			
Ni	0.60*	0.45*	0.55*	0.74*	0.02	1.00		
Pb	0.46*	0.32*	0.32*	0.48*	0.06	0.46*	1.00	
Zn	0.66*	0.52*	0.57*	0.77*	0.09**	0.83*	0.52*	1.00

*Significant correlation at the 0.01 level (2-tailed).
**Significant correlation at the 0.05 level (2-tailed).

Table 3. The total variance explained and the component matrices for the heavy metals.

Total Variance Explained									
Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	4.277	53.46	53.46	4.277	53.46	53.46	3.294	41.18	41.18
2	0.992	12.41	65.87	0.992	12.41	65.87	1.220	15.25	56.43
3	0.711	8.89	74.76	0.711	8.89	74.76	1.089	13.61	70.04
4	0.661	8.27	83.02	0.661	8.27	83.02	1.039	12.98	83.02
5	0.536	6.70	89.73						
6	0.417	5.21	94.94						
7	0.251	3.13	98.07						
8	0.155	1.93	100.00						

Component Matrix								
Element	Component Matrix				Rotated Component Matrix			
	1	2	3	4	1	2	3	4
Zn	0.913	-0.031	-0.034	-0.045	0.808	0.329	0.255	0.102
Ni	0.865	-0.182	-0.098	-0.076	0.823	0.294	0.177	-0.049
Cu	0.846	-0.007	-0.243	-0.125	0.813	0.339	0.021	0.117
As	0.801	0.036	-0.017	-0.077	0.712	0.262	0.219	0.159
Cr	0.700	-0.196	0.126	-0.375	0.792	-0.110	0.212	-0.008
Pb	0.643	0.134	-0.327	0.609	0.283	0.897	0.138	0.071
Hg	0.238	0.949	0.067	-0.170	0.075	0.064	0.045	0.990
Cd	0.615	-0.022	0.717	0.302	0.289	0.142	0.937	0.051

Extraction Method: Principal Component Analysis. Eight components extracted.

According to Table 3, all the elements were well represented by four components. The initial component matrix indicated that Zn, Ni, Cu, As, and Cr were associated, displaying high values in the first component (F1). Hg showed a high value in the second component (F2). However, Pb showed a greater value in F1 and was also partially represented in the fourth component (F4). Additionally, Cd showed a high value in the third component (F3) and was also partially represented in F1. Nevertheless, the rotation of the matrix eliminated ambiguities. As shown in Table 3, F1 includes Zn, Ni, Cu, As, and Cr; F2 includes Pb; F3 includes Cd; and F4 includes Hg.

Table 4. Pollution index of heavy metals in the agricultural soils of Liaocheng.

	PI			Number of samples			
	Min	Max	Mean	Low	Slight	Middle	High
As	0.20	1.89	1.15	102	548	0	0
Cd	1.00	4.25	1.75	0	529	114	7
Cr	0.60	1.27	0.95	495	155	0	0
Cu	0.56	1.73	0.91	546	104	0	0
Hg	0.50	13.00	2.00	19	373	180	78
Ni	0.64	1.46	1.02	277	373	0	0
Pb	0.57	1.19	0.83	635	15	0	0
Zn	0.67	1.48	0.99	350	300	0	0

Because the contents of Zn, Ni, Cu, and Cr are close to the background value, the source for these heavy metals is likely the parent soil materials. Cd and Hg are well known pollutants in agricultural soils and may originate from a common anthropogenic source, such as industrial activities, manure, and the burning of coal; the parent materials of the soils may partially control their concentrations.

Heavy Metal Pollution Assessment

The PIs were calculated using the background value of heavy metals. As shown in Table 4, the *PI*s varied greatly across the different heavy metals. As, Cr, Cu, Ni, Pb, and Zn exhibited lower values, and all of the samples had low or small *PI*s, indicating that the concentrations of As, Cr, Cu, Ni, Pb, and Zn in the soil samples were comparable to the background values of the Shandong Province and there was not obvious pollution of the above six heavy metals in the agricultural soil samples. The *PI* value of Hg ranged from 0.50 to 13.00; more than 258 samples, approximately 39.7%, were classified as being moderately or heavily contaminated with Hg. Cd also exhibited higher PI values, ranging from 1.00 to 4.25; more than 121 samples, approximately 18.6%, were classified as being moderately or heavily contaminated with Cd. The maximum PIs for Hg and Cd were 13.00 and 4.25, respectively. Therefore, it is very likely that some soil samples in Liaocheng are highly polluted with Hg and Cd.

Spatial Distribution Characteristics of Soil Heavy Metals

ArcGIS can be used to produce spatial distribution maps of heavy metals and thus it can be used for interpreting spatial variability and environmental sources [3, 9]. A comparison of interpolation methods available in ArcGIS 9.3 showed that higher precise interpolations are obtained by ordinary Kriging; thus, the PIs of the eight heavy metals were interpolated using the Kriging interpolation method.

The spatial distribution maps of the PIs of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in the agricultural soils of Liaocheng are presented in Fig. 3. The maps show that the effects of heavy metals on soil quality are different. The effect of Hg and Cd are greater than the other metals with significant spatial heterogeneity.

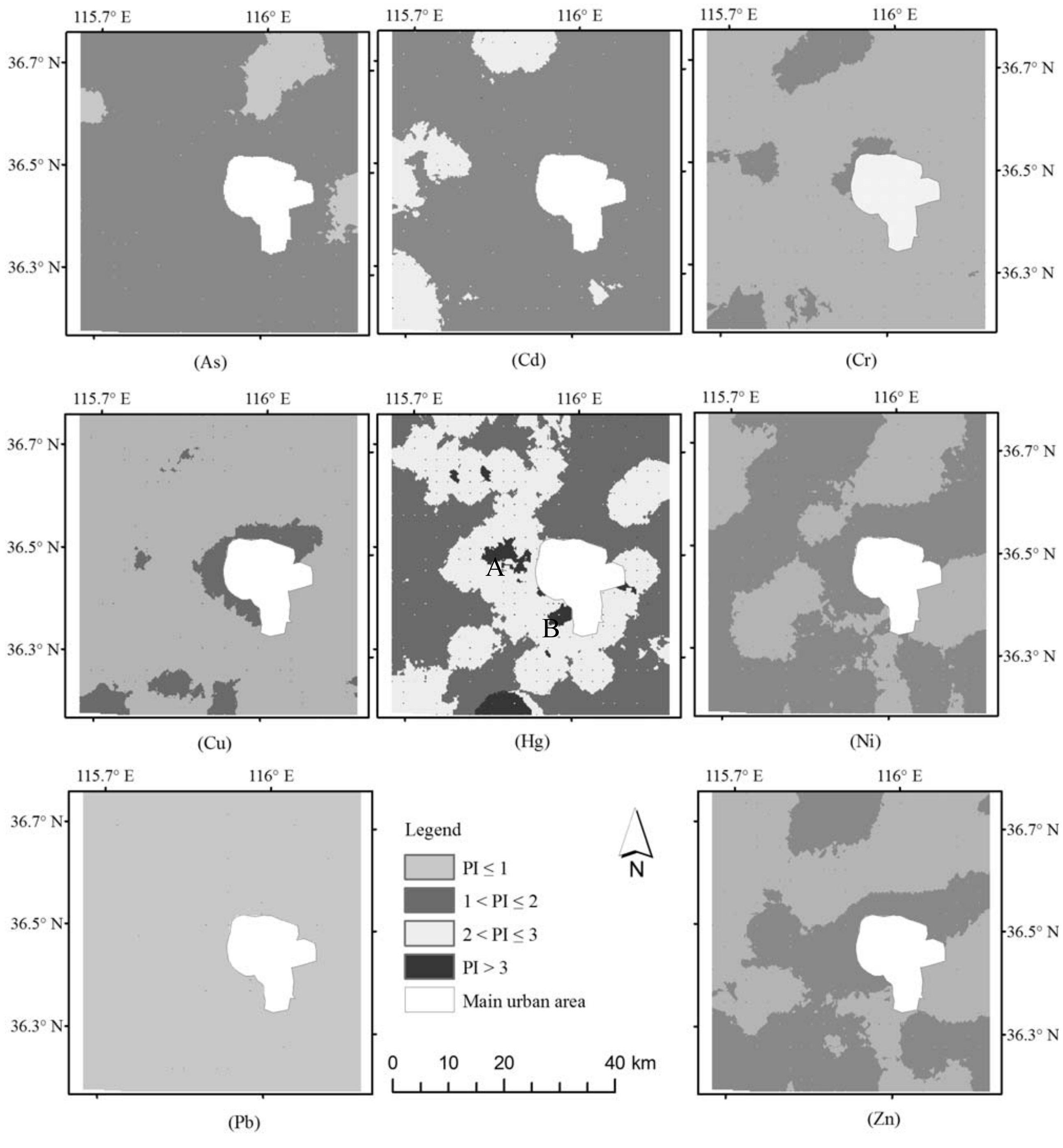
Hg is a dangerous heavy metal; the higher *PI* values of Hg concentrations are mainly located at the centre, northwestern, and southwestern parts of the study area. Elsewhere, the Hg concentrations are relatively low. For Hg, the proportions of moderately polluted ($2<PI\leq3$) and highly contaminated ($3<PI$) areas are 28% and 12%, respectively. However, higher *PI* values for Cd were found in the western and northern parts of the study area. For Cd, the proportions of moderately polluted ($2<PI\leq 3$) and highly contaminated ($3<PI$) areas are 17% and 1%, respectively. The values in Table 4 show no risk of heavy metal pollution from As, Cr, Cu, Ni, Pb, and Zn in this study area. However, the *PI*s of As, Ni, and Zn that are greater than one covered 91%, 63%, and 43% of the total area, respectively.

Previous studies have suggested that common sources of As and Hg in soils are sewage irrigation, manure application, and coal burning; Cd is a highly mobile element and often input through sewage irrigation. Generally, anthropogenic inputs of Cr and Ni from fertilizers and manures are lower than the concentrations already present in the soil. Zn and Cu are common ingredients of some pesticides and are also common in manure [3, 21, 22]. As, Cd, Cu, Hg, Pb, and Zn are mostly due derived from different anthropogenic activities such as agriculture, industry, and transportation. The abnormal heavy metal concentrations mostly coincided with industrial locations [9]. For example, there are large thermal power plants, chemical enterprises, and machinery manufacturing enterprises in zones “A” and “B” in Fig. 3.

Conclusions

A total of 650 agricultural soil samples collected from suburban areas of Liaocheng were analyzed for the following heavy metals: As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn. All data followed normal or lognormal distributions. Among the eight heavy metals, the mean concentrations of As, Cd, Hg, and Ni exceeded their corresponding background values. Additionally, the maximum concentrations of As, Cd, Hg, and Cu are all higher than the guideline values (GB 15618-1995), particularly the heavy metals Hg and Cd. All soil samples had low or slight *PI*s of As, Cr, Cu, Ni, Pb, and Zn. Hg and Cd exhibited high *PI* values (39.7% and 18.6%, respectively), and the soils were classified as being moderately or heavily contaminated with these heavy metals. Significant correlations were observed between As, Cd, Cr, Cu, Ni, Pb, and Zn, suggesting that they may originate from a common source. However, Hg showed only weak positive correlations with the other heavy metals, suggesting a different source. The abnormally high heavy metal concentrations mostly coincided with the industrial locations.

Results of the combined multivariate statistical analyses (cluster analysis and principal components analysis) and the spatial distribution patterns of heavy metals showed that anthropogenic sources, such as industrial activities, manure, and the burning of coal, represent important sources of Cd and Hg contamination, while the parent soil materials may partially control their concentrations. In contrast, Zn, Ni, Cu, As, and Cr are close to the background values, so the parent soil materials are likely to be the



(As) (Cd) (Cr) (Cu) (Hg) (Ni) (Pb) (Zn)

Fig. 3. Spatial distribution maps of the PIs of the heavy metals As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn in the agricultural soils of Liaocheng. A: thermal power plants (5.6 million mw unit) and B: industrial park.

sources for those heavy metals. Additionally, there is a strong spatial variability of soil pollution. There is a close relationship between urban areas and heavy metal pollution of soils.

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