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C. R. Biologies 332 (2009) 558–566

COMPTES RENDUS BIOLOGIES

Ecology / Écologie

The spatial variability of heavy metal distribution in the suburban farmland of Taihang Piedmont Plain, China

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Received 29 November 2008; accepted after revision 14 January 2009

Available online 28 February 2009

Presented by Philippe Morat

Abstract

The understanding of the spatial variability of soil heavy metals is an important precondition for suitably monitoring and evaluating eco-environment quality in a primary agricultural production zone. 100 topsoils were sampled from the Zhengding County of the urban-rural transition zone in Taihang Piedmont Plain, China. The contents of eight heavy metals Cu, Zn, Cr, Ni, Pb, Cd, Hg and As were tested for each soil sample, and their spatial patterns were analyzed by using the semivariogram approach of geostatistics, with which the kriging method was used to estimate the unobserved points. Then GIS technology was employed to produce spatial distribution maps of the 8 elements. The results showed that the concentration of Cd exceeded its background level. The local pollution from Cd was attributed to the anthropogenic influence. The concentrations of the eight heavy metals are relatively lower than the critical values of the national soil quality standard. The correlation distance of soil heavy metals ranged from 3.28 to 11.63 km, with the eight heavy metals having moderate spatial dependence. Cu, Cr, Ni, Pb and As were associated with and controlled by parent material. The spherical model was fitted to the semivariograms of Cu, Cr, Cd, Hg, Pb and As, and the Zn and Ni were fitted with the Gaussian model and the linear model, respectively. The results are helpful for improving agricultural and forest ecosystem in the region. ***To cite this article: P.G. Yang et al., C. R. Biologies 332 (2009).***

Keywords: Suburban farmland; Heavy metal-contamination; Spatial variability; Eco-environment; China

1. Introduction

The evaluation of soil environmental quality can provide some scientific warranty for proper landuse, soil contamination control and eco-environmental layout. Its

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1631-0691/$ – see front matter
doi:10.1016/j.crvi.2009.01.004

precondition depends on considering the spatial distribution of soil heavy metal contents. The problems associated with the characterization of heavy metals in the majority of sites are often due to multiple sources of pollution. Soil heavy metal information has undoubtedly implications for agriculture product and food safety [1–3].

Soil heavy metals have been a very useful indicator of environmental quality worldwide and been the subject of much attention because of their peculiar characteristics. Suburban farmland is a spatial transition zone between urban and rural, which plays an important role in providing regional food security to urban and local residents and protecting the environment or ecosystem; it has experienced a more rapid growth of industrialization than in rural areas in China. Soil heavy metals in suburban farmland may have deposited and accumulated as a result of the urban rapid industrial development. Heavy metal contents are influenced by natural and anthropogenic factors including parent materials, land use, application of pesticides and fertilizers etc. It can accumulate in crops and may lead to the damage and alteration of animal or human physiological functions by food chain [2–4]. Understanding the spatial distribution of suburb topsoil heavy metals is critical for environmental management and agricultural production.

Geostatistics has successfully been applied in investigating and mapping soil heavy metals [5–12]. A main contribution of semivariogram is to reveal the spatial change properties of sampled values that belong to the regional variables. The kriging interpolating can provide spatial distribution, which assumes that the distance or direction between sample points reflecting the spatial correlation can be used to explain the variation on a regional scale. Although there have been some papers on the spatial distribution of soil heavy metals, some conclusions were not always agreed on for different sampling intervals [13–17].

The spatial variability of soil heavy metals is an important part of environmental supervision and ecosystem evaluation. In the Taihang Piedmont Plain, a dominating area for agricultural production of Hebei Province, little attention has been paid to the spatial variability of soil heavy metals. Zhengding County is the typical suburban farmland in the Taihang Piedmont Plain. The spatial variability of heavy metals (Cu, Zn, Cr, Ni, Pb, Cd, Hg and As) was investigated using statistics, geostatistics and geographical information system (GIS) techniques, in order to find out heavy metal scale variability and spatial distribution maps and provide satisfactory and efficient estimates for soil environmental monitoring, and provide valuable information for the regional soil quality management [18–20].

2. Materials and methods

2.1. Regional status

The study area, Zhengding County near the Shijiazhuang City of the Capital of Hebei Province, lies in 114°22′–114°45′E and latitude 38°4′–38°22′N, and occupies a 486 km^2 of total area. The landform is flat with an average elevation of 75 m. The Hutuo River runs through the southern border of the County. Soils develop on alluvial sediment material, and there are soil types including drab soil, Chao soil and paddy soil. It belongs to the continental monsoon climate area, and the average annual temperature and rainfall are 12.2 °C and 530 mm, respectively. Because locating at the transitional zone from urban to rural areas, the land was affected intensely by human activities for agricultural production. 62.6% of the land was used for agricultural farming, 16.8% of the land was occupied by residential settlements, factories and roads (including railways, highways), and the rest was occupied by forests, riverways and an airport. The samples came from agricultural areas where winter wheat and summer maize are the dominant crops. This county is also a major production base for grain, vegetables, fruits and oil plants.

2.2. Sample collection

Considering on the complexity of the land use, a total of 100 soil samples were collected in the simple random sampling method, which whole sampling points are an irregular grid that can represent cultivated areas and allow geostatistical treatment. Fig. 1 showed the distribution of sampling points. Actually the density of sampling varies within small limits due to difficult sampling conditions in the study area. Each sample was a mixture of 5 topsoil cores (0–20 cm in depth, approximately 200 g total weight) which were taken from within a 20 × 20 m area; the central point position was recorded with a GPS device.

2.3. Laboratory analysis

All soil samples were air-dried at room temperature and ground in an agate mortar to pass through a 100-mesh plastic sieve. The soil pH was measured by a glass electrode in a 1:5 soil/water suspension. Cu, Zn, Cr, Ni, and Pb were extracted by aqua regia digestion (HNO_3, HCl and H_2O_2) of the soil fraction in

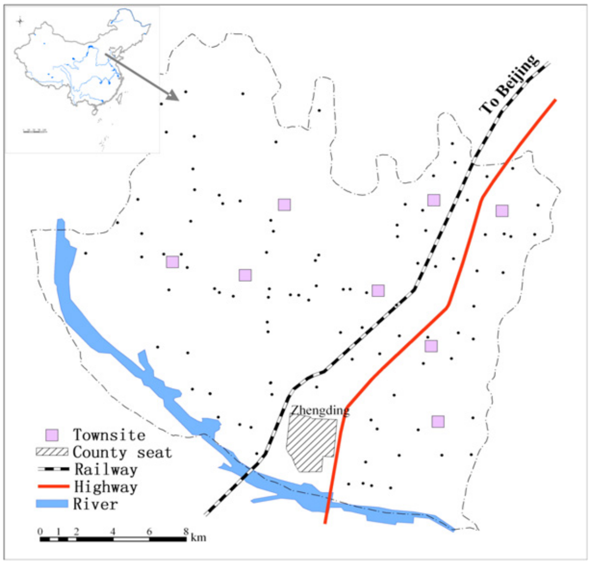

Fig. 1. The sketch map of geographical location and sampling points.

a microwave (Milestone Ethos 900 plus Mod. 44062) in accordance with the ISO 11466 procedure (International Organization for Standardization, 1995), then the total concentrations of Cu, Zn, Ni, Cr were measured by flame atomic absorption spectrometry (AAS), and Cd and Pb by graphite furnace AAS, as well as As and Hg was determined by atomic fluorometry (Agricultural Chemistry Committee of China 1983) [1].

2.4. Spatial structure analysis

Common descriptive statistics and histograms do not incorporate the spatial locations of data into their defining computations. The four main operations of linear geostatistics (variances of estimation and dispersion, regularization and kriging) involve only the structural function of the random function (covariance or variogram). Thus, every geostatistical study begins with the construction of a model designed to characterize the spatial structure of the regionalized variable studied.

Soil heavy metals are typical regionalized variables. The presence of a spatial structure where observations close to each other are more similar than those that are far apart (spatial autocorrelation) is a prerequisite to the application of geostatistics [20]. The experimental semivariogram measures the average degree of dissimilarity between sampled values and a nearby data value [18–20], and thus can depict autocorrelation at various distances.

Geostatistics uses the technique of semivariogram to measure the spatial variability of a regionalized variable, and provides the input parameters for the spatial interpolation of kriging [21]. The value of the experimental semivariogram for a separation distance of h (referred to as the lag) is half the average squared difference between the value at $Z(x_i)$ and at $Z(x_{i+h})$,

$$\gamma(h) = \frac{1}{2} Var\big[Z(x+h) - Z(x)\big] \tag{1}$$

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

where: $N(h)$ is total number of pairs of sample points separated by the lag distance h. For irregular sampling, it is rare for the distance between the sample pairs to be exactly equal to h, therefore, the lag distance h is often represented by a distance band. $Z(x_i)$ is the measured sample value at point i, $Z(x_i + h)$ is measured sample value at point $i + h$. The variogram model is chosen from a set of mathematical functions that describe spatial relationships. The appropriate model is chosen by matching the shape of the curve of the experimental variogram to the shape of the curve of the mathematical function.

The fitted model provides information about the spatial structure as well as the input parameters such as nugget, sill and range for kriging interpolation. By fitting the appropriate variogram model, the distance-dependent coefficients can be estimated and graphically interpreted. In this study, the fitted spherical model and Gaussian model were selected.

Raw data were analyzed with different software packages. The descriptive statistical parameters were calculated with SPSS 11.0. The geostatistic analyses and the probability calculation were carried out with VarioWin 2.2. The maps of 8 elements spatial distributing pattern were produced using ArcGIS 8.3 software by kriging interpolation.

3. Results and discussion

3.1. Statistic descriptive parameters and normality test

To evaluate the raw data, the descriptive statistical parameters of soil heavy metals are presented in Table 1. The results showed that Cu, Zn, Ni, As had passed the Kolmogorov–Smirnov normality test (K-S $p < 0.05$), but other variables such as Pb, Hg, Cr and Cd had not passed. Since further geostatistic analysis need data to follow a normal distribution. Data transformation was carried out prior to the next analysis. Finally, all 8 heavy metals followed a normal distribution or a lognormal distribution.

Table 1
Descriptive statistics parameters and test for normality of soil heavy metals.

Item	Cu	Zn	Ni	Pb	Cr	Hg	As	Cd
Mean	21.22	69.96	25.04	18.80	57.77	0.08	6.16	0.15
China mean	20.00	67.70	23.40	23.60	53.90	0.04	9.20	0.074
Background value	21.7	62.0	28.8	20.0	63.9	0.023	12.1	0.075
Guide value	35	100	40	35	90	0.15	15	0.20
Standard deviation	3.42	7.19	4.59	3.92	8.77	0.06	1.50	0.04
C.V.	0.16	0.10	0.18	0.21	0.15	0.75	0.24	0.27
Minimum	11.10	46.30	12.90	12.30	32.80	0.02	2.17	0.09
Maximum	33.20	88.00	36.50	40.90	88.60	0.37	9.91	0.29
Skewness	0.352	0.073	−0.185	0.790	0.762	0.692	−0.116	0.488
Kurtosis	1.403	0.430	−0.283	2.401	1.959	0.438	0.177	0.643
K-S p	0.05	0.12	0.14	0.01	0.00	0.00	0.20	0.00
Distribution	normal	normal	normal	lognormal	lognormal	lognormal	normal	lognormal

$p < 0.05$, $n = 100$.

Table 2
Pearson correlation coefficients of topsoil heavy metals.

	Cu	Zn	Ni	Pb	Cr	Hg	As	Cd
Cu	1							
Zn	0.654*	1						
Ni	0.358*	0.205*	1					
Pb	0.267*	0.068	0.324*	1				
Cr	0.363*	0.489*	0.435*	0.033	1			
Hg	0.100	0.156	−0.331*	0.094	−0.243*	1		
As	0.444*	0.183	0.037	0.161	0.197*	0.139	1	
Cd	0.338*	0.514*	0.067	−0.010	0.254*	−0.034	0.013	1

* $p < 0.05$, $n = 100$.

The maximum observation values of every heavy metal except for Zn were more than double the minimum values, which exhibited large spatial ranges. The average content plus standard deviation of Cd is $0.15 \pm 0.04\ \mathrm{mg\,kg^{-1}}$. The value is much higher than the background values that can be used to assess metal contamination in soil, indicating possible pollution at a few locations of the study area, which are probably caused by anthropogenic activity such as fertilizers and pesticides, vehicle exhausts and industrial fumes, burning of coal. The Zn, Hg, Cu, Cr, Ni, Pb and As (mean ± S.d.) exhibited lower contents than the background values that probably caused by weathering and lithogenic of rich parent materials.

According to the Standard of Chinese Environmental Quality for Soils (GB 15618–1995) (State Environmental Protection Administration of China, 1995) [19], the study area soils are feebly alkaline with an average pH value of 8.05. All heavy metal concentrations in the surface soil are lower than the guidance values established for cultivated areas, indicating that soil environmental quality in the study area was of little threat in terms of environment and human health.

The coefficient variation (C.V.) values of eight heavy metals in the study ranges from 0.10 to 0.75 indicating that they had moderate variations. The C.V. of Hg was 0.75, which is the highest of the 8 heavy metals, suggesting that Hg has the greatest variation among the soil samples and thus would have the highest possibility of being influenced by the extrinsic factors such as human activities. The lowest C.V. of the 8 heavy metals was Zn with a score of 0.1, suggests that Zn has a weak variation and its content was almost constant across the county (Table 1).

3.2. Correlation between soil heavy metals

Correlation measures the linear relationship between random variables. The Pearson correlation coefficients and their significance levels ($p < 0.05$) between all the variables are presented (shown in Table 2).

Strong positive correlations were observed between Cu and Zn, Ni, Pb, Cr, As, Cd except for Hg, indicating that Cu and Zn, Ni, Pb, Cr, As, Cd are closely related to each other, while Hg is poorly correlated with any other metal. A significant correlation was also observed

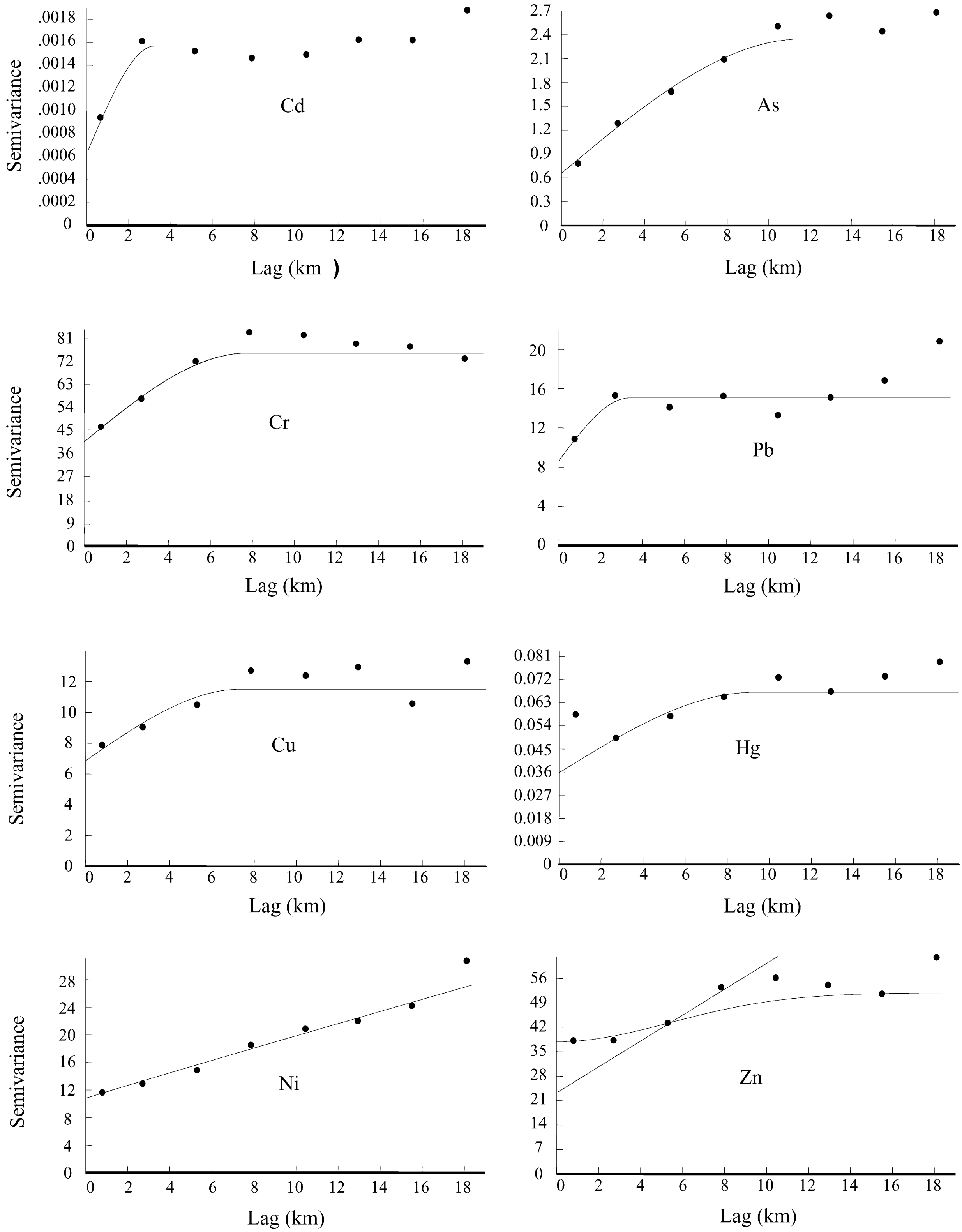

Fig. 2. Semivariograms model of topsoil heavy metals.

between Zn and Ni, Cr, Cd. Ni had strong positive correlation with Pb and Cr and negative correlation with Hg. A positive significant correlation was also observed between Cr and As, Cd and a negative significant correlation was found between Cr and Hg. It can be speculated that seven heavy metals came from the same soil parent material, climate and vegetation aside from Hg.

3.3. Spatial structure of soil heavy metals

Semivariograms were used to establish the degree of spatial continuity and the range of spatial dependence. The experimental semivariogram represents the variance of the sample value at various separation distances. Each of the experimental semivariograms can be described on three parameters: nugget, sill and range. Experimental semivariograms suggested that the theoretical spherical model is in reasonable agreement with the data. The Zn and Ni were fitted with the Gaussian model and the linear model (Fig. 2), respectively.

The largest nugget effect of Cr ($C_0 = 39.29$) indicated a strong random variance at short distance. The smallest nugget effect is Cd ($C_0 = 0.000696$), which showed relative variance and the sampling density is adequate to reveal the spatial structures. All soil heavy metals showed positive nugget, because of the sampling error, shorter distance variability, and random and inherent variability. More attention has been paid to the spatial correlation distance, as they play a more important role in kriging estimation.

The range of heavy metal contents varied from 3.28 to 11.63 km. The As had the largest range 11.63 km, which indicated that As was correlated and depended on the soil parent material. The Cd had a smallest range 3.28 km, which implies that the length of the spatial autocorrelation is much longer than the sampling interval of 1.5 km. Therefore, the current sampling design is appropriate for this study and it is expected that a good spatial structure will be shown on the interpolated map. These results are inconsistent with previous reports [8–10] that the correlation length is from 6.27 to 85.75 km. These results may indicate that the range of autocorrelation is influence by different study scales.

The $C_0/(C + C_0)$ ratio (Table 3) can be regarded as a criterion to classify the spatial dependence of soil properties. If the ratio is less than 25%, the variable has strong spatial dependence. If it is between 25% and 75%, the variable has moderate spatial dependence. With a ratio of greater than 75%, the variable shows only weak spatial dependence [13–15]. Usually, strong spatial dependence of soil properties can be attributed to intrinsic factors (soil formation), and weak spatial dependence can be attributed to extrinsic factors (soil management practices) [17–23]. The $C_0/(C + C_0)$ ratio of eight heavy metals between 25% and 75%, have moderate spatial dependence, indicating that the anthropogenic factors changed their spatial correlation through industrial production, fertilization and other soil management practices. The ratio of element As is near 28%, which is lowest one, suggesting that As had stronger spatial dependence due to the effects of natural factors such as the parent materials and topography. The ratio of element Zn is near 72%, which is the highest.

Table 3
Semivariogram models and parameters of topsoil heavy metals.

Heavy metal	Model	Nugget (C_0)	Sill ($C + C_0$)	$C_0/(C + C_0)$ (%)	Range (km)
Cd	Spherical	0.000696	0.0016	43.23	3.28
As	Spherical	0.657	2.35	27.96	11.63
Cr	Spherical	39.29	77.00	51.03	7.76
Cu	Spherical	6.86	11.51	59.60	7.37
Hg	Spherical	0.0021	0.0035	59.04	9.31
Ni	Linear	10.71	21.00	51.00	10.08
Pb	Spherical	8.48	15.41	55.03	3.42
Zn	Gaussian	37.14	51.76	71.75	8.92

3.4. Spatial distribution maps of soil heavy metals

ArcGIS is a valuable tool for interpreting spatial variability and environmental monitor. Information generated through semivariogram (nugget, sill, range) was used to calculate sample weighing factors for spatial interpolation by the simple Point Kriging procedure, using the nearest 8 sampling points and a maximum searching distance equal to the range distance of the variable. In particular, mapping the conditional probabilities of a soil property is of importance for management decisions, which are based on threshold values of this property, such as delineating safe or hazardous areas and identifying zones that are suitable for crop growth and those that must be treated.

Fig. 3 is the spatial distribution map generated based on the semivariograms of all heavy metals gathered from the study area. The maps showed similar geographical trends, especially for Cu, Zn, Ni and Cd, with both high contents at centre and northeast edge part of the County. At the other place, their concentrations are relatively low. Apart from the northwest and southeast directions, Zn values are generally high. It is concluded that the explanation for higher uncertainty is most likely attributed to land use. Similar to the grade in Fig. 3, it shows no risk of Pb in this study area. On the other hand, a high content of Ni distribution was found on the top of the study area. Hg is a dangerous metal, the higher soil

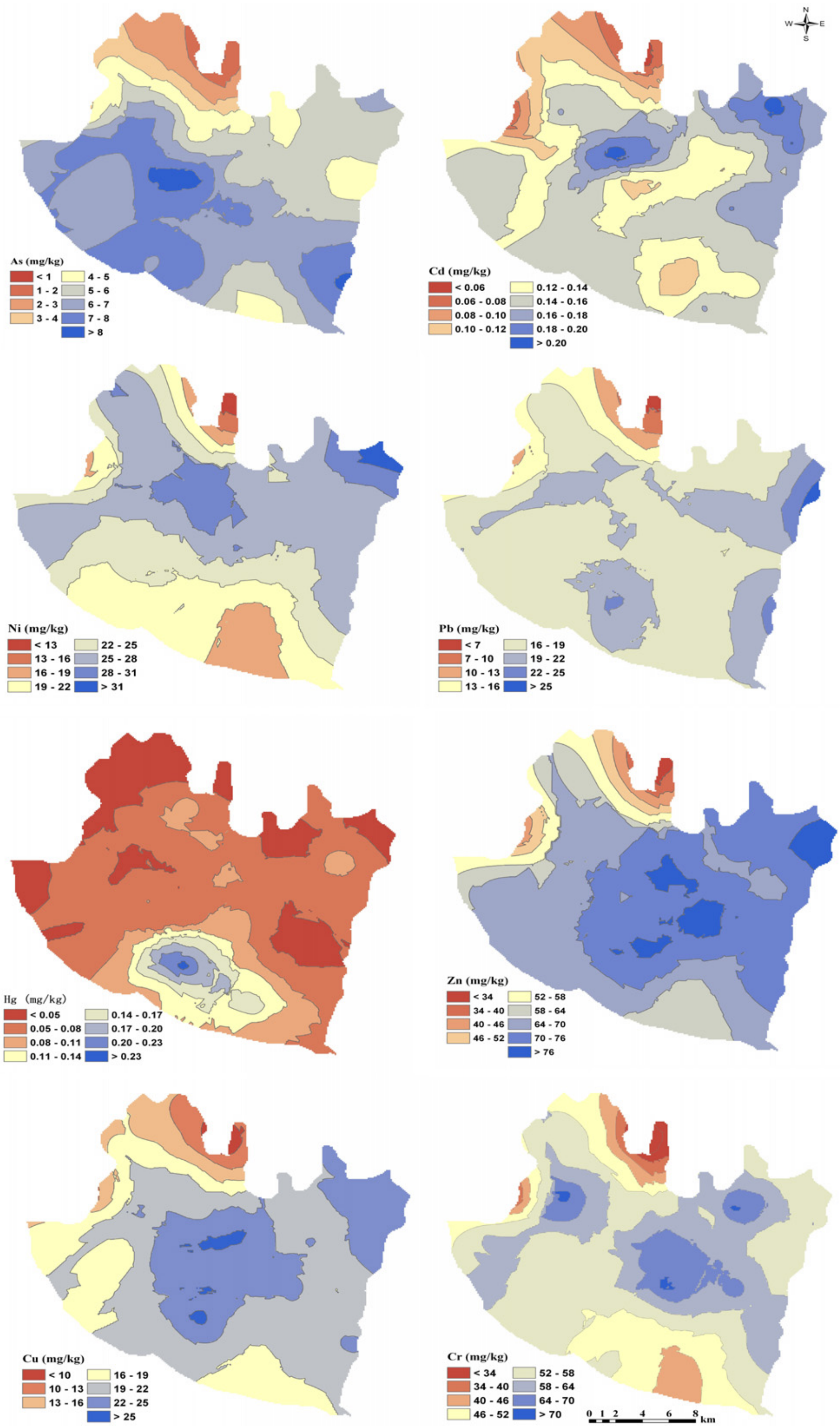

Fig. 3. The spatial distribution maps of 8 heavy metal contents.

Hg concentrations are found in the south edge of the bottom and the highest Hg area is the smallest. Ni and Hg exhibit apparent variance from north to south, while Zn and As display most variance from the northeast to southwest (Fig. 3).

Generally, anthropic inputs of Cr and Ni in fertilizers, limestone and manures are lower than the concentrations already present in the soil; Cu and Zn are common ingredients of some pesticides; Common sources of Pb in soils are manures, sewage sledges, vehicle exhausts and the burning of coal. Cd is a highly mobile and toxic element. In addition, using coal for heating during the winter period could have resulted in high Zn, Pb and Hg from air fallout [19–23]. Cu, Zn, Pb, Cd, Hg and As are mostly due to the different anthropic activities such as industrial, agricultural and transport. The abnormity with heavy metals mostly coincided with the industry locations. All 8 heavy metals that contain less than the threshold values can be regarded as safe for crop growth in the study areas.

4. Conclusions

Using the soil samplings of a relatively small scale in study area, an estimation of 8 heavy metal concentrations in suburban arable areas was tested. All data followed normal or lognormal distribution. A significant correlation was observed between Cu and Zn, Ni, Pb, Cr, As, Cd except for Hg, indicating that they were from the same origin as their soil parent materials and of the same soil formation factors. The local anomaly had been found with Cd, which is probably linked to anthropic and industrial activity.

The study results demonstrated that the spatial variability of eight heavy metals in suburban arable was apparent in the Zhengding County. Such studies could help validate procedures of spatial predictions that have limited measured data. This may be suitable for many problems in soil monitoring where heavy metal changes are relatively small and slow.

All heavy metal contents are less than the guidance values. Thus, even these eight elements are unlikely to exhibit a risk to the environment or a threat to human health at present, but the results are helpful for improving agricultural and forest ecosystem in the region [4–9, 14–23].

Acknowledgements

This work was supported by the National Basic Research Program of China (2005CB121106), the Major Project of National Natural Science Foundation of China (90502006), the Cooperative & Instructive Foundation of State Key Laboratory of Soil Erosion and Dryland Farming on the Loess Plateau (10501-HZ) and the Project of Monitoring Net for Soil Heavy Metal Pollution in Preponderance Agro-product Region of Hebei Province and One hundred-talent Plan of Chinese Academy of Sciences. The authors wish to thank to Prof. Yuanzhong Wang for his help in this research.

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