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ORIGINAL ARTICLE

Spatial distribution of heavy metals in soils: a case study of Changxing, China

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Abstract Six hundred and sixty-five soil samples were taken from Changxing County in Zhejiang Province, China, to characterize the spatial variability of Hg Cd, Pb, Cu, As and Cr. The geostatistics and geographic information system (GIS) techniques were applied, and the ordinary kriging and lognormal kriging were used to map the spatial patterns of the six heavy metals. Hg, Pb, Cu and As were fitted to the spherical model with a range of 85.75, 82.32, 86.10, and 23.17 km, respectively. Cr was fitted to the exponential model with a range of 6.27 km, and Cd was fitted to the linear model with a range of 37.66 km. Both Pb and Cu had strong spatial dependence due to the effects of natural factors including parent material, topography and soil type. Hg, Cd, Cr and As had, however, moderate spatial dependence, indicating an involvement of human factors. Meanwhile, based on the comparison between the original data and the guide values of the six metals, the disjunctive kriging technique was used to quantify their pollution risks. The results showed that only Cd and Hg exhibited pollution risks in the study area. The pollution source evaluated was closely corresponded with the real discharge of industrial production and the application of organomercury pesticides. The results of this study provide insight into risk assessment of environmental pollution and decision making for agricultural production and industrial adjustment of building materials.

Keywords Geostatistics · Heavy metals · Spatial variability · Geographic information system

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Introduction

Heterogeneity is an inherent quality of soils. In a natural landscape, it represents a wide variety of soil attributes, both spatial and volumetric, as a result of interactions of the processes that rule soil formation. When the soil is cropped, additional sources of heterogeneity are caused by agricultural practices. Although basic principles of classical statistics consider that soil variability occurs entirely at random (Júnior et al. 2006), many studies indicate strong spatial dependence of soil attributes (Journel and Huijbregts 1991). Therefore, geostatistical methods should be adopted in order to better understand spatial variation of soil attributes (Zhao et al. 2005).

Geostatistics, based on the theory of regionalized variables, enables the interpretation of results based on the structure of their natural variability, taking into consideration spatial dependence within the sample space. The analysis of dependence is based on the structure of the semivariogram, which demonstrates the existence of spatial dependence and the category of soil attributes (Goovaerts 1997, 1999). An important contribution of geostatistics is the assessment of uncertainty about unsampled values. The uncertainty assessment usually takes the form of a probability map of exceeding critical values, such as regulatory thresholds in soil pollution or criteria for soil quality. It also needs expert knowledge for decision making such as delineation of contaminated areas where remedial measures should be taken or areas of high soil quality where specific management plans can be developed. Geostatistics is thus increasingly used in the characteristics of spatial variability and risk assessment of soil pollution (Steiger et al. 1996; White et al. 1997; Lin et al. 2001; Romic and Romic 2003; McGragh et al. 2004).

Heavy metals in soils have been shown to be very useful indicators of environmental pollution worldwide and been the subject of much attention because of their peculiar pollutant characteristics. In the past decades, the natural input of heavy metals to soils due to pedogenesis has been exceeded by the anthropogenic input, even on a regional scale (Bacon et al. 1992; Kelly et al. 1996; Manta et al. 2002; Zhang and Ke 2004). This urgent pollution status is also the case in China. The high environmental pollution risk of heavy metals in main agricultural areas of Zhejiang Province has been reported by Liu et al. (2006). They discussed a real risk of Cu, Zn or Cd in the Hangzhou–Jiaxing–Huzhou Plain, and concluded the pollution genesis to the irrational farming practices. As another dominating area for agricultural production of Zhejiang Province, Changxing County also had similar farming practices as those of Hangzhou–Jiaxing–Huzhou Plain. It seems likely that the potential risk of heavy metals exists likely in Changxing County.

The aim of the present work was thus to elucidate the spatial distribution of heavy metals in soils of Changxing County. Our specific objectives were (1) to examine the spatial dependency and the variation mechanism of heavy metals in soils, (2) to map the spatial distribution and risk assessment of soil heavy metals, and (3) to provide basic knowledge for environmental management and agricultural policy making.

Material and methods

Study area

The research focused on the cultivated land of Changxing County in northern Zhejiang Province around Taihu Lake. The county covers 1,430 km^2, of which 30.5% is cultivated land. Sixteen towns were included in this area: Zhicheng, Baixian, Meishan, Shuikou, Jiapu, Huaikan, Xiaopu, Erjieling, Sian, Lincheng, Hongxingqiao, Lvshan, Lijiaxiang, Hongqiao, Wushan and Heping (Fig. 1). As a typical agricultural and most developed region of the South Yangtze River, the study area is densely dotted with drainage ditches that form a network of waterways and is the primary food production zone in Zhejiang Province. Soils are mostly paddy soils within the plain area, and rice is the dominant crop.

Soil sampling and analysis

In accordance with soil types and the uniformity of soil sampling distribution in the study area, 665 soil samples were collected from different locations of the plain and valley of the study area in 2003 at an interval of 4 km. Distribution of sampling points is presented in Fig. 2. All soil samples were taken at a depth of 0–15 cm and air-dried. Gravels and coarse organic matter or plant root residues were removed. Samples were thoroughly mixed and ground to pass a 0.149-mm sieve, then stored in plastic bags prior to chemical analysis.

Soil pH (soil:H_2O ratio = 1:2) was measured using a pH meter with a glass electrode. It ranged from 4.26 to 8.00 with a mean of 5.58 (Fig. 3). Organic matter (OM) was determined by potassium dichromate wet combustion procedure (Agricultural Chemistry Committee of China 1983). Cation exchange capacity (CEC) was measured according to the procedure of Hendershot and Duquette (1986). Available phosphorus (AP) was extracted using NH_4F–HCl or $NaHCO_3$ (base on the

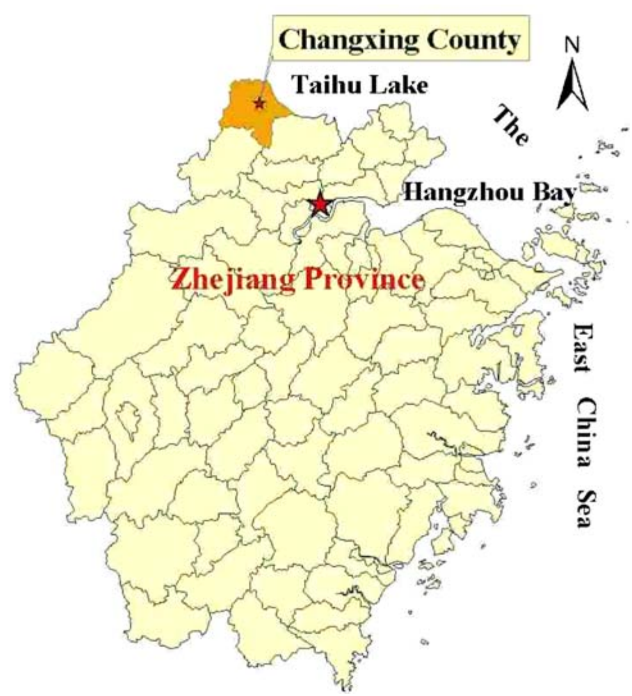

Fig. 1 Study area

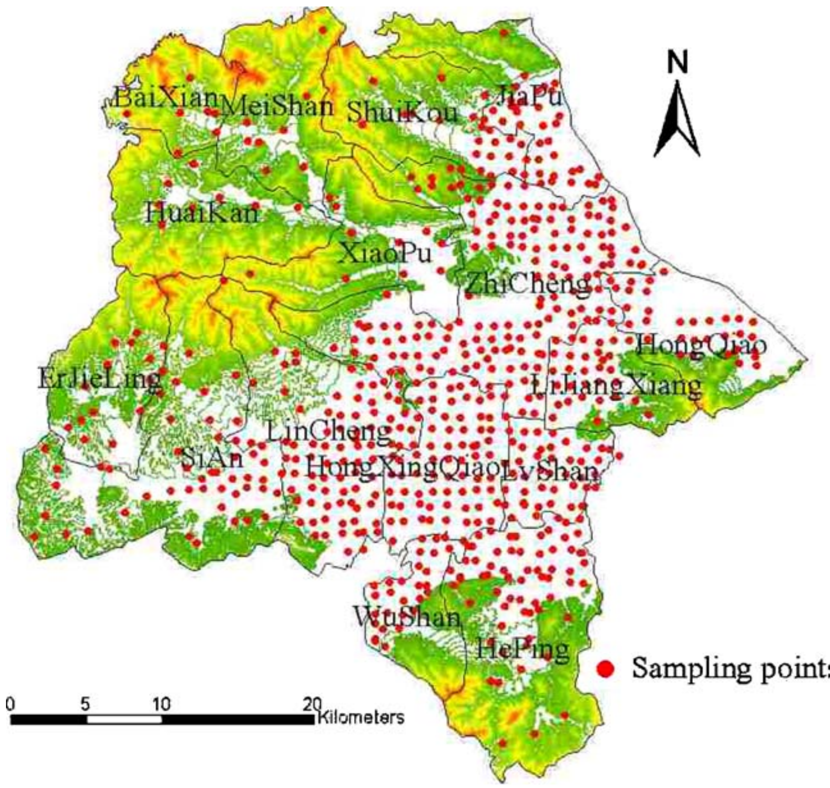

Fig. 2 Distribution of sampling locations

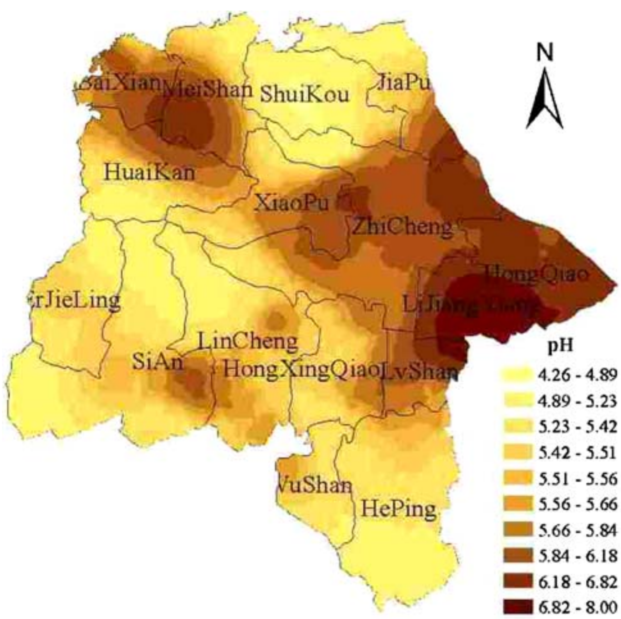

Fig. 3 Filled contour maps of soil pH by ordinary kriging

pH values), and analyzed using the molybdenum-blue method (Agricultural Chemistry Committee of China 1983). Available potassium (AK) was extracted using NH_4OAc and then measured by flame emission spectrometry (Agricultural Chemistry Committee of China 1983).

Total contents of heavy metals, were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) for Cu and Cd, by absorption fluorescence spectrometry (AFS) for As and Hg, and by X-ray fluorescence spectrometry (XRFS) for Cr and Pb after soil samples were digested with a mixture of nitric acid (HNO_3) and perchloric acid ($HCLO_4$).

Geostatistical methods

Geostatistics is based on the theory of a regionalized variable (Matheron 1963), which is distributed in space (with spatial coordinates) and shows spatial auto correlation such that samples close together in space are more alike than those that are further apart. Geostatistics uses the technique of variogram (or semivariogram) to measure the spatial variability of a regionalized variable, and provides the input parameters for the spatial interpolation of kriging (Krige 1951; Webster and Oliver 2001). Variogram was used in the study to analyze discrete soil samples. The function is expressed as:

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

where $Z(x_i)$ is the value of the variable Z at location of x_i, and $N(h)$ is the number of pairs of sample points separated by the lag distance h.

Information generated through variogram was used to calculate sample weighing factors for spatial interpolation by a kriging procedure, using the nearest 16 sampling points and a maximum searching distance equal to the range distance of the variable (Isaaks and Srivastava 1989; Lark and Ferguson 2004). Variogram plots were acquired by calculating variogram at different lags. Linear, exponential and spherical models were selected in order to acquire information about the spatial structure as well as the input parameters for kriging interpolation.

The linear function is:

$$\gamma(h) = C_0 = 0$$

$$\gamma(h) = AH \quad h > 0$$

The exponential function is:

$$\gamma(h) = C_0 + C\left(1 - \mathrm{e}^{-(h/a)}\right) \quad h = 0$$

The spherical function is:

$$\gamma(h) = C_0 + C\left[\frac{3}{2}\frac{h}{a} - \frac{1}{2}\left(\frac{h}{a}\right)^3\right] \quad 0 < h \leq a$$

$$\gamma(h) = C_0 + C \quad h > a$$

where C_0 is the nugget variance ($h = 0$), represents the experimental error and field variation within the minimum sampling spacing. Typically, the semivariance increases with increasing lag distance to approach or attain a maximum value or sill ($C_0 + C$) equivalent to the population variance. C is the structural variance and a is the spatial range across which the data exhibit spatial correlation. A is the slope.

For the assessment of pollution risks, the disjunctive kriging technique was used to estimate the probability that the true values of soil heavy metals at unsampled points exceed the specified thresholds. It is assumed that the content of a heavy metal is a realization of a random variable $Z(x)$, where x denotes the spatial coordinates in two dimensions.

Data treatment

Occurrence of exceptional values can lead to the data discontinuity and this would violate the geostatistics theory. In this study, we considered the data out of the extent of ($A \pm 3s$) as exceptional values (Liu et al. 2004, 2006), where A denotes the average value for each heavy metal and s is the standard deviation. Data exceeding the values ($A \pm 3s$) were replaced with the maximum or minimum values within ($A \pm 3s$) in the raw data set.

As in conventional statistics, a normal distribution for a variable under study is desirable in linear geostatistics (McGraph et al. 2004). Serious violation of normality, such as too high skewness and outliers, can impair the variogram structure and the kriging results. It is often observed that environmental variables are lognormal (Krige 1951, 1960; Sichel 1952) or positively skewed (Zhang et al. 1995; Zhang and Selinus 1998), and data transformation is necessary to normalize such data sets. Outliers in a data set can make the variogram exhibit erratic behavior, whereas data transformation can dampen the difference between extreme values (Gringarten and Deutsch 2001). Thus the data that were not normally distributed were logarithmically transformed in order to normalize positively skewed data sets in this study.

Data sets were analyzed with different software packages. The descriptive statistical parameters were calculated with Microsoft EXCEL® and SPSS®. Maps were produced with GIS software ArcGIS® and its extension of Spatial Analyst®. The geostatistical analyses and the probability calculation were carried out with GS+®.

Results

Descriptive parameters and probability distribution of the raw data set

The representative statistical summary of the available data sets for six metals was given in Table 1. It was noted that the kurtosis (–0.22 to 2.03) and skewness (0.14–0.97) values for Cu, Cr and As were low, suggesting a normal distribution of the raw data and thus no need for data transformation before geostatistics analysis. Kurtosis (3.30–4.92) and skewness (1.12–1.79) values for Hg, Cd and Pb were high and the raw data sets were logarithmically transformed before performing geostatistical analysis. Logarithmic transformation resulted in reduced skewness (–0.10 to 0.12) and kurtosis (0.58–2.64) values of Hg, Cd and Pb, and the transformed data sets passed the lognormal tests.

Meanwhile, the coefficients of variation of Hg and Cd were 54.85 and 36.32%, respectively, higher than those of Pb, Cr, Cu and As, suggesting that Hg and Cd had greater variation among the soils samples and thus

Table 1 Statistical summary of heavy metal contents in the topsoil collected from the study area

Soil attributes	Min	Max	Mean	SD	Kurtosis	Skewness	CV (%)	Guide value	Number
pH	4.26	8.00	5.75	0.75	1.31	1.14	12.97	–	–
Hg (mg/kg)	0.03	0.49	0.15	0.08	3.61	1.69	54.85	0.30	44
Logarithm of Hg	–1.56	–0.31	–0.87	0.22	0.58	–0.10	–	–	–
Cd (mg/kg)	0.02	0.48	0.19	0.07	4.92	1.79	36.32	0.30	36
Logarithm of Cd	–1.61	–0.32	–0.76	0.14	2.64	0.01	–	–	–
Pb (mg/kg)	16.10	63.40	32.94	7.00	3.30	1.12	21.24	250	0
Logarithm of Pb	1.21	1.80	1.51	0.09	0.99	0.12	–	–	–
Cr (mg/kg)	31.00	111.00	66.97	15.30	–0.22	0.31	22.85	250	0
Cu (mg/kg)	10.70	39.00	23.81	5.40	–0.15	0.14	22.70	50	0
As (mg/kg)	2.12	16.20	8.13	2.41	2.03	0.97	29.60	30	0

Min minimum, *Max* maximum, *SD* standard deviation, *CV* coefficient of variation, *Number* number exceeding guide value

would be possibly influenced by the extrinsic factors such as human activities.

Geostatistical analysis

The ranges of semivariograms for Hg, Pb and Cu were similar and about 85 km (Table 2), and were much greater than those for Cd, Cr and As. In addition, the smallest range of semivariogram, presented by Cr, was 6.27 km. This confirmed the rational of the sampling density, which was 4 km interval for the precise environmental survey of the six heavy metals tested in this study. The sampling interval could thus be increased for the elements with larger ranges such as Hg, Pb and Cu.

Capitalizing on the spatial correlation between the available data, ordinary and lognormal kriging techniques were used here to predict attribute values at unsampled points of cultivated land. The experimental semivariograms of soil heavy metals with the fitted models are presented in Fig. 4. The results showed that soil Cd data were fitted with the linear model; Cr fitted with the exponential model; and the other four heavy metals were all best fitted with the spherical model. The attributes of the semivariograms for each soil heavy metal were also summarized in Table 2. All of the Nug/Sill ratios for the six metals were less than 0.72.

Spatial distributions and risk assessment

Mapping metal contents is often a preliminary step towards decision making, such as delineation of polluted areas or identification of zones that are suitable for crop growth. For soil pollution, a straightforward approach is to delineate all contaminated locations where the estimated pollutant content exceeds the regulatory threshold.

Figure 5 presents the spatial patterns of the six heavy metals in soils of Changxing County generated from their semivariograms. All the metals had distinct geographical distribution. The spatial distribution maps showed similar geographical trends, especially for Hg, Pb, Cr and Cu, with high contents both in the northeast and southwest areas.

Compared with the guide values in Table 1, the overproof probability of each metal was estimated. The results showed that only Cd and Hg exhibited the pollution risk. The risk patches resulted from disjunctive kriging were given in Fig. 6. For soil Hg, southeast Zhicheng and west Lijiaxiang were the areas with high pollution risk, where the estimated probability (Ω[Hg $\geq$ 0.3 mg/kg]) reached 0.40–0.67 (Fig. 6). For soil Cd, the overproof probability (Ω[Cd $\geq$ 0.3 mg/kg]) exhibited in general the same risk patches but more serious than that of Hg. Besides the area of Lijiaxiang, the highest risk for Cd also distributed in the northwest

Table 2 Best-fitted semivariogram models of heavy metals and their parameters

Soil attributes	Model	C_0	$C + C_0$	$C_0/C + C_0$	Range (km)	R^2
Hg	Spherical	0.028	0.10	0.27	85.75	0.95
Cd	Linear	0.018	0.024	0.72	37.66	0.90
Pb	Spherical	0.0041	0.017	0.24	82.32	0.94
Cr	Exponential	61.20	230.30	0.27	6.27	0.32
Cu	Spherical	14.60	66.62	0.22	86.10	0.97
As	Spherical	2.84	6.66	0.43	23.17	0.93

C_0 nugget variance, C structural variance, $C + C_0$ sill variance

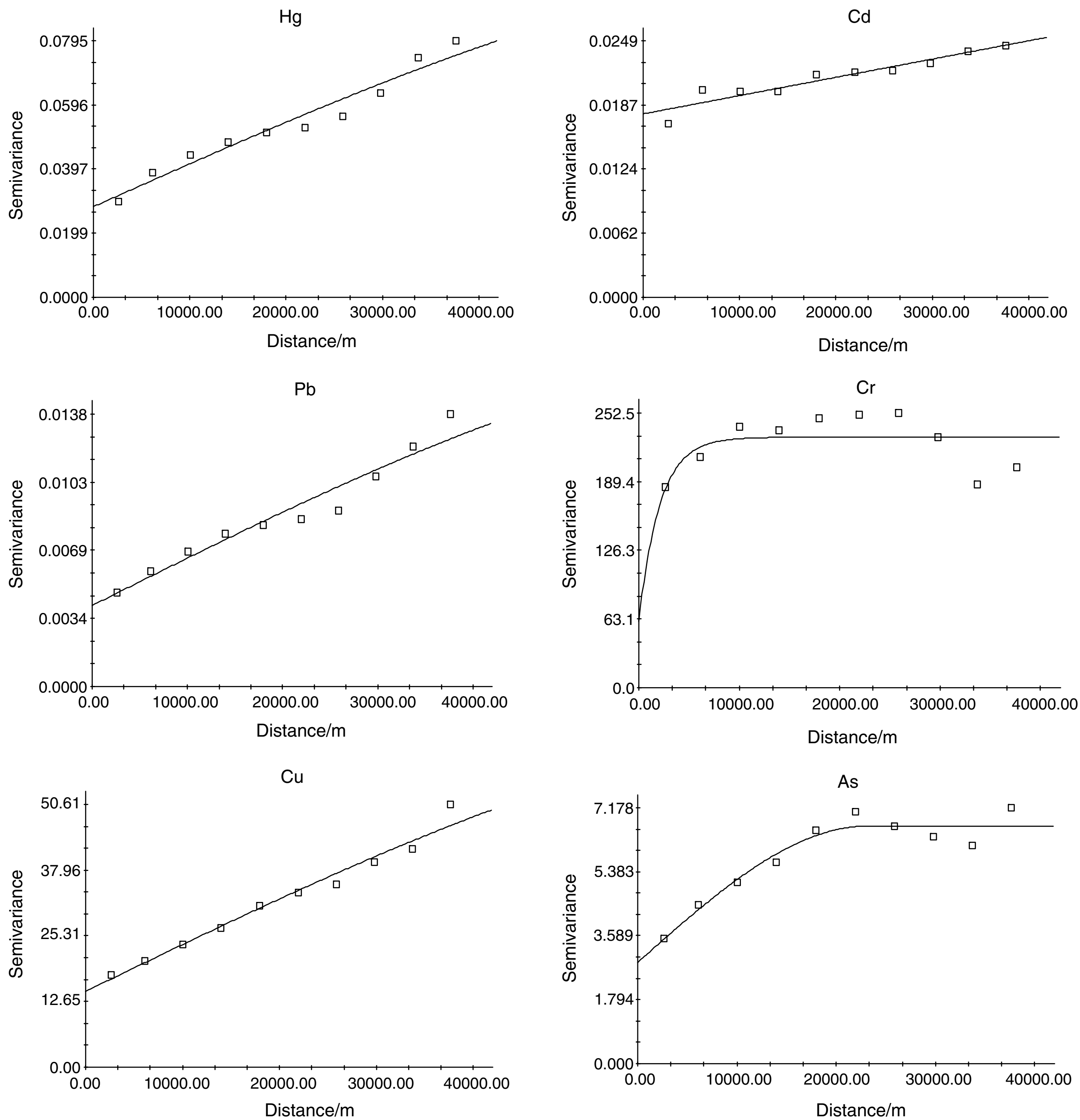

Fig. 4 Experimental semivariograms of soil heavy metals with fitted models

of the studied region—Meishan town. Thus, these areas with overproof probability in high level would contribute to the environmental pollution and ultimately threaten the health of human and other living organisms. Elsewhere of the region with low estimated probability less than the guide thresholds (0.3 mg/kg for both Hg and Cd), however, can be regarded as safe for crop growth.

Discussions

The spatial variability of soil attributes can be affected by both soil pedogenic factors (such as parent materials) and human activities (such as industrial and agricultural production). In theory, the Nug/Sill ratio in the geostatistics can be regarded as a criterion to classify the spatial dependence of soil attributes. The ratio of

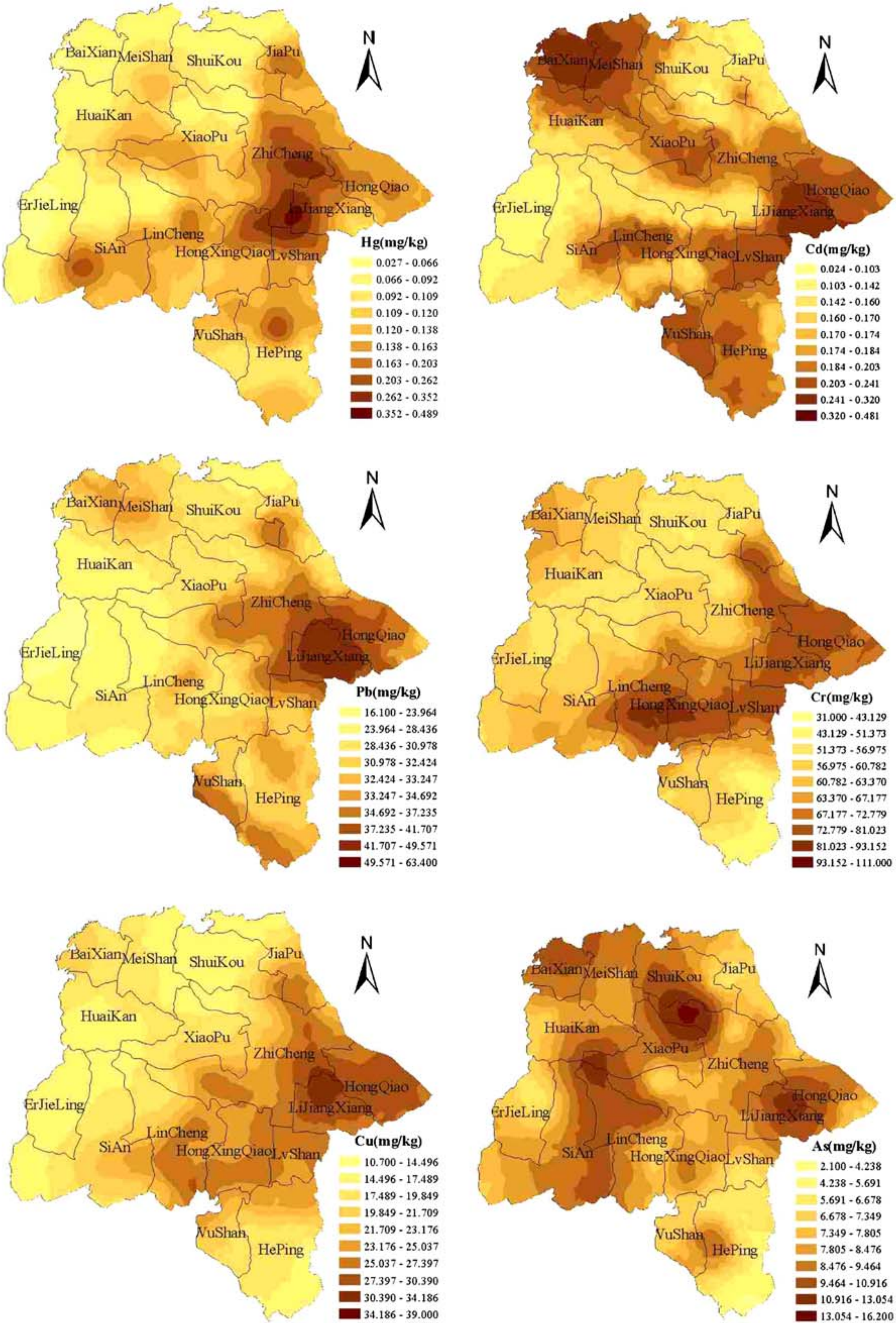

Fig. 5 Filled contour maps of soil Hg, Cd, Pb, Cr, Cu and As producted by ordinary Kriging

0.25 and 0.75 are two thresholds for the relative strength index of spatial correlations. The variable with a ratio of less than 0.25 is strongly spatial dependent; the variable with the ration between 0.25 and 0.75 is moderately spatial dependent; whereas the variable with the ratio greater than 0.75 is only weakly spatial dependent (Chien et al. 1997; Chang et al. 1998; Cambardella et al. 1994). As shown in Table 2, the Nug/Sill ratios of both Pb and Cu were less than 0.25, showing strong spatial dependence due to the effects of natural factors such as parent material, topography and soil types. The ratios of Hg, Cd, Cr and As were, however, between 0.25 and 0.75, which belonged to the scope of moderate spatial dependence, revealing that the anthropogenic factors such as industrial production, fertilization and other soil management practices changed their spatial correlation after a long process of utilization.

To further discriminate the natural and anthropogenic contributions, the correlation between six heavy metals and soil properties (OM, CEC, AP, AK and pH) was analyzed (Table 3). There existed close relationships between Pb and Cu ($r = 0.684^{**}$), between Cu and Cr ($r = 0.596^{**}$), between Cd and Pb ($r = 0.513^{**}$) and between Hg and Pb ($r = 0.427^{**}$). This might account for the similarities of the geographical distribution tendency, a consistent increasing fact in the areas of the northeast and the southwest, of all the tested heavy metals in Fig. 5. It might, to some degree, indicate a potential risk of combined pollution and its same genesis of pollution. Moreover, a close correlation existed between soil properties and the six tested heavy metals. The strong positive correlations were observed between all six heavy metals and soil pH ($p < 0.01$). This could be clearly seen from Figs. 3 and 5 which showed spatial patterns of soil pH and the heavy metals. The correlation coefficients varied from 0.093 to 0.359 ($p < 0.01$). Significant positive correlation was also found between AP and Cd ($r = 0.196$, $p < 0.01$) and Pb ($r = 0.252$, $p < 0.01$). Considering the relationship between accumulation of heavy metal (such as Cd) and fertilization (such as application of P fertilizer) suggested by many earlier studies (Yang et al. 1995; Carnelo et al. 1997), our results suggest that the tested six heavy metals were likely to be affected by the anthropogenic factors.

The mean and maximum contents of Pb, Cr, Cu and As (Table 1) were considered low as compared with the corresponding guide values suggested by Chinese Environmental Quality Standard for soils (GB 15618-1995) (State Environmental Protection Administration of China 1995). Thus these four elements are unlikely to exhibit a risk for environmental pollution or threat to human health. However, there were 36 and 44 samples that had Cd and Hg contents exceeded the guide values, suggesting the pollution fact of some cultivated lands in Changxing County. This coincided with the results of risk assessment.

In this study, the tested soils mainly distributed in the plain and valley of Changxing County. The parent materials were alluvial–lacustrine or coastal–alluvial aggradations, with the characteristics of being easily enriched by heavy metals (Zhejiang Soil Survey Office 1994). The high background values of both Hg and Cd in Lijiaxiang had been found in the latest soil survey (Zhejiang Soil Survey Office 1994). This might be a

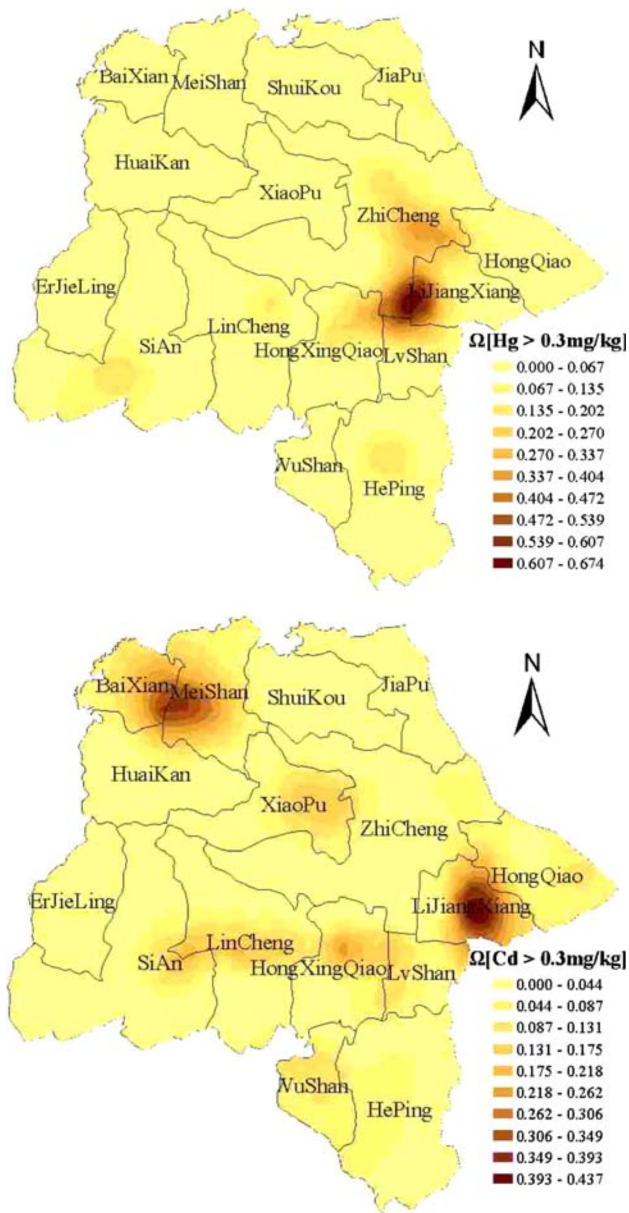

Fig. 6 The estimated probability maps of Hg and Cd

Table 3 The correlation coefficients between heavy metals and soil attributes

	OM	CEC	AP	AK	pH	Hg	Cd	Pb	Cr	Cu	As
OM	1										
CEC	0.402**	1									
AP	–0.020	–0.309**	1								
AK	0.202**	0.497**	0.019	1							
pH	–0.061	0.004	0.020	0.139*	1						
Hg	0.021	-0.128*	0.168*	-0.144*	0.202**	1					
Cd	0.091	0.002	0.196**	0.053	0.314**	0.146**	1				
Pb	0.169*	-0.085	0.252**	0.103	0.330**	0.427**	0.513**	1			
Cr	0.039	-0.035	0.057	0.110	0.225**	-0.004	0.119**	0.233**	1		
Cu	0.153*	-0.055	0.146*	0.095	0.359**	0.366**	0.361**	0.684**	0.596**	1	
As	0.136*	-0.006	0.085	0.127*	0.093**	-0.090*	0.041	0.169**	0.276**	0.110**	1

*$p < 0.05$,**$p < 0.01$, *OM* organic matter, *CEC* cation exchange capacity, *AP* available phosphorus, *AK* available potassium

likely reason accounting for the observed environmental risks of Hg and Cd.

It is note worthy that with the rapid development of industrialization in recent years, more and more township enterprises, e.g. the thermoelectricity, cement, mining, storage battery, and chemical industry enterprises, have emerged in Changxing County. Eighteen large-scale enterprises in the county were mainly located in northeast–southwest areas (Changxing County Bureau of Environment Protection 2004) (Fig. 7). These enterprises would inevitably present the potential risk of heavy metal pollution. Based on the similar geographical distribution between the Cd risk and the township enterprises, especially cement plants, it thus could be extrapolated that the pollution discharge from industrial production might partially contribute to the Cd pollution. These findings should be taken into account in decision making for industrial structure adjustment. As for the element Hg, main risk places generally happened in the southeast of the study area, where the topographical feature was relatively plain with an advanced agricultural cultivation history, especially rice cropping. The application of organo-mercury pesticide (2,250 mg/ha year) for pest control such as phenyl mercuric acetate and mercuric ethyl chloride in the middle 1960s of this area might be an alternative cause of the aggravation of the Hg pollution risk (State Environmental Protection Bureau 1990–1997; Hua and Shan 1996; Zhou and Wong 2000).

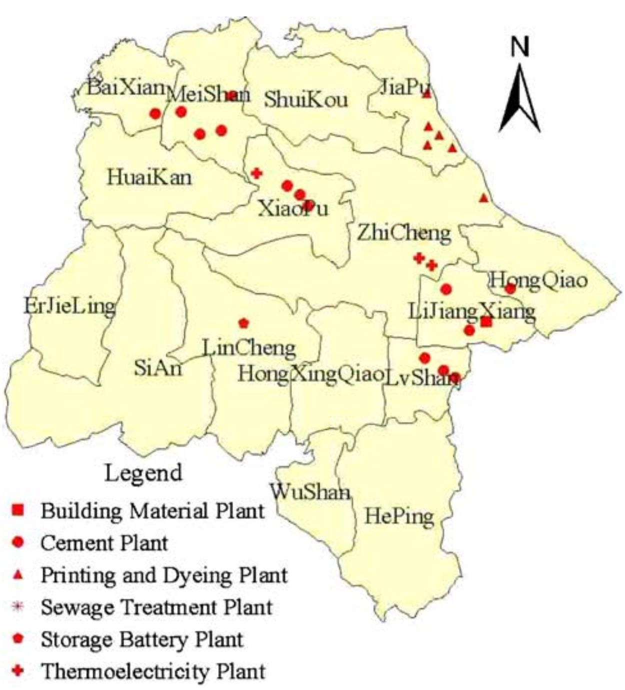

Fig. 7 Distribution of the main pollution source points

Conclusions

This study demonstrated that the spatial variability of six heavy metals in agricultural fields was apparent in Changxing County. Over the long history of land utilization, the spatial variability of investigated elements was influenced by both natural and anthropogenic factors. Among the six metals, Hg and Cd had high risks for environmental pollution and human health. Both natural (e.g., the high soil pH and background values) and anthropogenic factors (e.g., the discharge of industrial wastes and the application of special pesticides) had contributed to the genesis of pollution process. These results presented here could be used for decision making in structure adjustments of agriculture and industry in the polluted areas.

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