ORIGINAL ARTICLE

Spatial pattern of heavy metal concentration in the soil of rapid urbanization area: a case of Ehu Town, Wuxi City, Eastern China

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Abstract Urbanization has an important effect on the soil ecosystem. Anthropogenic activities during urban sprawl are the sources and methods of migration, transition, and concentration of heavy metals in the soil. In this paper, the Yangtze River delta, the fastest urbanization area in China, was selected to study the influence of urbanization on heavy metal concentration in the soil. The analysis was performed on 218 samples collected from a typical industrial town in the delta. The content of six heavy metals (Hg, Cd, As, Pb, Cu, and Zn) was measured, which showed that the soil was seriously polluted by heavy metals, particularly Hg. In the study area, human activities were the major factors for the heavy metal concentration in the soil. The average content of Hg in the 0–500 m zone from the urban core was 1.4 mg/kg, about five times higher than the grade II threshold of the Chinese Environmental Quality Standard for Soils. The content of Hg, Cd, Pb, and Cu decreased gradually with increasing distance from the core of the built-up area, especially Hg. The influence of the anthropogenic activities was found to be anisotropic, and effects in the direction of 315° and 225° were most significant. Hg pollution in the 225° direction of the study area was more serious. These results suggested that the urbanization process affects not only the content but also the spatial pattern of heavy metal concentration in the soil.

Keywords Urbanization · Soil · Heavy metal concentration · Spatial pattern

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Introduction

Urbanization can be viewed as a typical element of economic growth and industrial advancement. Due to urban expansion and industrial development, the impacts of human land-use activities on the soil ecosystem are of great concern (Hu et al. 2004; Li et al. 2001). Urban expansion caused by urbanization makes a transition from massive cultivated lands to industrial lands. This trend can lead to the collapse of agro ecosystem and a serious degradation of land quality (Li et al. 2003; Chen et al. 2001). Human activities such as industrial production, transportation construction, and urban expansion have negative influences on the soil environment, especially in terms of heavy metal concentration (Zhang et al. 2012; Zhong et al. 2007). Heavy metals enter soils through different pathways, including atmospheric fallout (Simonson 1995), automotive exhaust emission (Harrison et al. 1981), urban household sewage discharge (Al-Khashman and Shawabkeh 2006), solid waste stacking (Zheng et al. 2005), and the production and use of agricultural means of production (Lu et al. 1992). Moreover, the circulation and enrichment of heavy metals in soil threatened public health and food security (Järup 2003). Heavy metal contamination in soil can be considered the "cancer" of soil, as there is no quick, cost-effective way to amend the functions of the contaminated soil; therefore, a great deal of economic, human, and material resources would be required

in restoring the contaminated soil. In addition, heavy metal pollution events have been of common occurrence in recent years (Liu et al. 2012).

There are numerous studies on the relationship between urbanization and heavy metal soil contamination, which have made great achievements (Hu et al. 2004; Tan et al. 2006; Feng et al. 2004; Han et al. 2006; Zhang et al. 2009; Krishna and Govil 2005). However, the number of soil sampling sites in several studies was lower than the minimum quantity required for geostatistical analysis (>80) (Tang and Yang 2007). On the other hand, the soil sampling sites were mostly located in the core of built-up areas, and focused on the influence of anthropogenic activities on heavy metal concentration in their direct interface—soil (Tan et al. 2006; Feng et al. 2004; Han et al. 2006; Zhang et al. 2009; Krishna and Govil 2005). A few of these studies accounted for the impact of human activities on the whole region, including urban cores, suburbs, and rural areas, and improved the existence of such affect in different zones (Amini et al. 2005). In addition, this impact may occur in a gradual manner in which the heavy metal content in the soil would change with the increase of distance from the core of the built-up area (Sun et al. 2010). Urbanization has an impact on not only the content but also the spatial pattern of heavy metal concentration in the soil, such as the spatial variation and orientation of heavy metals in the soil (Chamannejadian et al. 2013). A total of 218 available high-density soil samples were collected at different distances from the core of the built-up area in this study, to (1) evaluate the influence of anthropogenic activities on the heavy metal content in the soil surrounding a rapid urbanization area, (2) determine the existence of a gradual effect, and (3) assess the impact of urbanization on spatial patterns of heavy metal concentrations in the soil.

Materials and methods

Study area

This study was conducted in Ehu Town of Yangtze River Delta, Wuxi City, Eastern China, which extended from $120°28'24''$ to $120°35'59.49''$E and $31°28'12.02''$ to $31°35'33.67''$N. Ehu Town is located in the northern subtropical zone, and has a subtropical monsoon humid climate. The climate is characterized by well-marked seasons, temperate climate, much rainfall, sunshine, and long frost-free days, with an average temperature of 2.8 °C in January and 28 °C in July. The annual rainfall is approximately 1048 mm. Yellow–brown soil, paddy soil, and moist soil are the main soil types in the study area. The GDP of Ehu Town's was RMB 0.93 billion in 2003, and reached RMB 2.9 billion in 2010 with an average annual growth rate of 26.4 %.

Sampling and chemical analyses

Due to the fact that the study area is surrounded by water in two directions (east, south), the research was carried out in the northwest and southwest parts of the study area. Taking urban built-up areas as centers, 10 buffer zones were established (buffer radius = 500 m) (Fig. 1), covering an area of 97.95 km². The study area was then divided into 500 × 500 m plots, besides Ehu Town. Four to eight soil samples were collected within a distance of 20 m around the center of each plot using a polyvinylchloride shovel and were mixed to make one composite sample. The sampling depth was 20 cm for all samples. If there were settlements, roads or other built-up lands in the center of plots, a soil sample could be obtained nearby. Each composite sample was packed into a polyethylene bag and then taken back to laboratory. A total of 218 soil samples were collected across the whole area in September 1–15, 2009. The Global Position System (GPS) was applied to obtain the coordinates of each sampling point.

Contents of six heavy metals, including Mercury (Hg), Cadmium (Cd), Arsenic (As), Lead (Pb), Copper (Cu) and Zinc (Zn), were examined in this study. All soil samples were air-dried timely with a polyvinylchloride salver at room temperature and ground in an agate mortar to pass through a 0.149 mm sieve for chemical analysis. For each sample, nearly 0.2 g of dried soil was digested by a $HCl–HClO_4–HNO_3–HF$ mixture. Then, the concentration of Cd was determined using inductively coupled plasma mass spectrometry (ICP-MS). The concentration of As and Hg in the samples were measured by inductively coupled plasma atomic fluorescent spectroscopy (ICP-AFS). Finally, the content of Cu, Pb, and Zn were measured using inductively coupled plasma atomic emission spectroscopy (ICP-AES).

Methods

Semivariogram analysis

The semivariogram analysis is an effective spatial statistical method that explains the spatial change of objects based on the regionalization variable theory. It is a technology that can quantitatively research regional variables that have random and structural characters on a temporal-spatial domain. This analysis can be computed using function (1) (Liu et al. 2006):

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

where $Z(x_i)$ is the regionalization random variable, h is the separation distance between two sample sites, and $Z(x_i)$ and
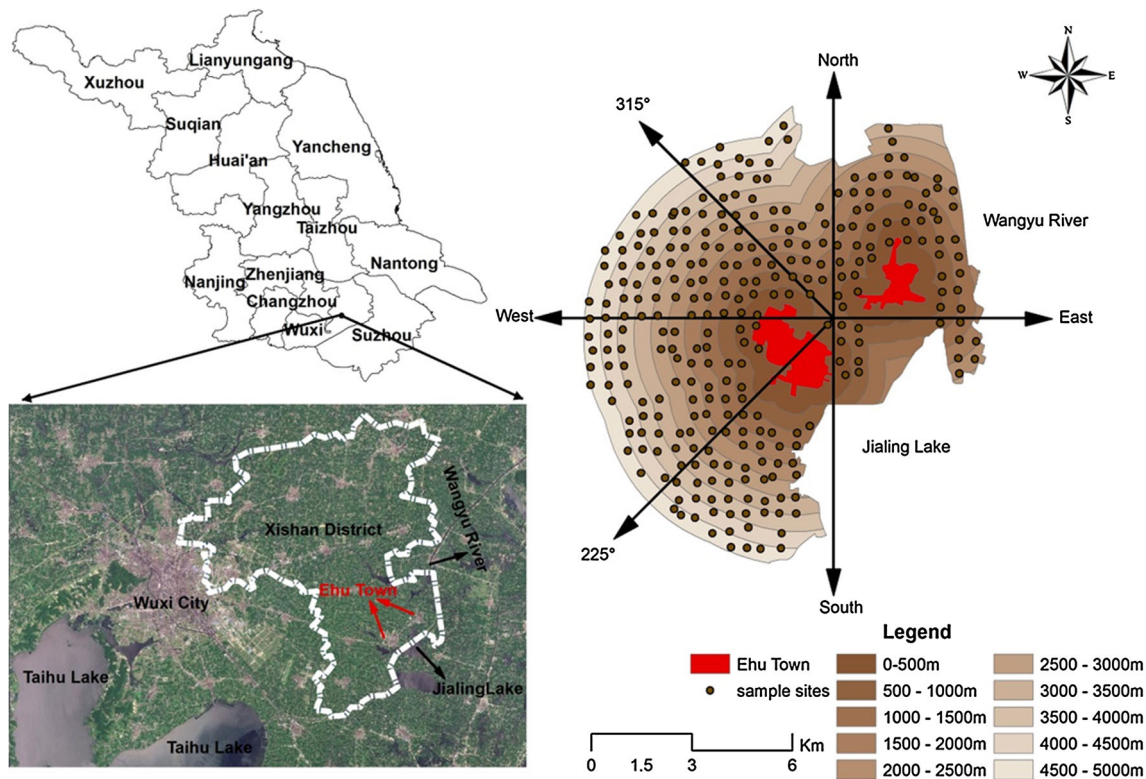
Fig. 1 Study area and the distribution of samples

$Z(x_i + h)$ are the values of $Z(x)$ at location x_i and location $x_i + h$.

Four important parameters can be obtained through the calculation of the variogram, namely nugget (C_0), range, sill ($C_0 + C$), and the mean variance. Nugget (C_0) and sill ($C_0 + C$) reflect the random and structural aspects of the spatial objects, respectively. In general, the $C_0/C_0 + C$ ratio (NSR) reflects the spatial correlation characteristics. If the ratio of $C_0/C_0 + C$ (NSR) is less than 0.25, then the variable has strong spatial dependence, i.e., the objects have the same attribute in space. Between 0.25 and 0.75, the spatial object has moderate spatial dependence, and for greater than 0.75, the spatial object only has weak spatial dependence (i.e., the feature is very complex in its spatial distribution pattern). The mean variance is related to the spatial variation of the object; the more complex the variation of the spatial object, the greater the mean variance.

Spatial autocorrelation analysis

Spatial autocorrelation analysis is an important method to explore the comparability of the spatial objects. This method takes the spatial position and the spatial attribute of the object into account to study the interaction between them. Moran's I is adopted in this study, and the formula is as follows:

$$I = \frac{n}{\sum_{i=1}^{n} \sum_{j=1}^{n} W_{ij}} * \frac{\sum_{i=1}^{n} \sum_{j=1}^{n} W_{ij}(x_i - \bar{x})(x_j - \bar{x})}{\sum_{i=1}^{n} (x_i - \bar{x})^2} \quad (2)$$

where I is Moran's I, and W_{ij} is the position weight matrix (PWM).

Moran's I ranges from -1 to 1; when it is greater than 0, there is a positive correlation between the spatial properties of the object. In contrast, there is a negative correlation when Moran's I is less than 0. The spatial objects tend to combine with the increase of Moran's I, and the objects have good spatial pattern integrity. On the contrary, fragmental distribution occurs when Moran's I decreases. When Moran's I is 0, the spatial pattern of the objects is random.

Covariance analysis

Covariance is a parameter that measures the 'co-variation' between two variables, showing the degree of interaction of the two variables. The greater the covariance is, the more significant the interaction of the two variables. The formula is as follows:

$$\text{COV}(x, y) = E\{X - E(X)][Y - E(Y)]\} \quad (3)$$

where $\text{Cov}(x, y)$ is the covariance, x is the observed value a, y is the observed value b, $E(X)$ is the expectation of x, and $E(Y)$ is the expectation of y.

In this study, the greater the covariance is, the greater the influence that distance has on heavy metal content; on the other hand, smaller covariance indicates that distance has a lesser influence.

Results and analysis

The impact of urbanization on heavy metal concentrations in soil

The heavy metal content in soil at different distances from the core of the built-up area was obtained using ArcGIS 9.3 (zonal statistics as the table tool), as shown in Fig. 2. The summary statistics of the concentrations of the six heavy metals are provided in Table 1. The mean concentrations of As, Cd, Cu, Hg, Pb and Zn are 10.88, 0.19, 40.28, 0.93, 50.13 and 91.63 mg/kg, respectively. The results showed that the mean values of the heavy metals were all significantly higher than the corresponding background values; simultaneously, the pollution of four (Hg, Pb, Cu and As) heavy metals was the most serious. The rates of these four heavy metals were higher than their corresponding background values by 365, 46.57, 36.54 and 20.88 %, respectively. Furthermore, the Hg content in the soil exceeded the threshold of grade II in the Chinese Environmental Quality Standard for Soils. Specifically, its value was approximately three times higher than the threshold of the soil quality standard.

Figure 2 showed that the content of three main heavy metals (Cd, Hg and Pb) in the soil tended to decrease with increasing distance from the core of the built-up area, revealing that urbanization had a significant impact on heavy metal concentrations in the region surrounding the city. The trend of Hg, Pb, and Cd content could be well simulated ($R^2 = 0.885$, $R^2 = 0.714$, $R^2 = 0.934$, respectively), from which we could conclude that the gradient change of heavy metal content in soil was associated with anthropogenic activities. This also showed that heavy metal pollution in urban soil caused by anthropogenic activities had a wide range of influence. Although the soil Hg content in the farthest zone from the built-up area (4,500–5,000 m zone) declined down to 0.6 mg/kg, it was still two times higher than the grade II threshold of the Chinese Environmental Quality Standard for Soils.

Among the different heavy metals, Hg changed mostly with the variation of distance to build-up area. The average Hg content was 1.4 mg/kg in the 0–500 m zone (cf. the grade II threshold of the Chinese Environmental Quality Standard for Soils, 0.3 mg/kg), and declined by 54.42 % to 0.6 mg/kg in 4,500–5,000 m zone. The results revealed that Hg was mainly concentrated in the core of the built-up area and its suburbs. It also suggested that point source pollution might be an important factor affecting the distribution of Hg pollution. Field survey showed that there were some high-polluting enterprises in the study area, such as a PVC chemical industry and a coal-based power station. People introduced high-polluting enterprises blindly, and such enterprises were sometimes lack of management. A great deal of sewage laced with Hg and Pb entered rivers, accelerating the heavy metal concentration in the soil. The content of Pb and Cd showed a descending trend with increasing distance from the core of the built-up area. The change of As, however, was relatively complicated, showing an undulating tendency with a slight increasing gradient, with the highest value (11.3 mg/kg) located in the 2,500–3,000 m zone from the built-up area. The trends of Cu and Zn content showed a decrease followed by an increment with increasing distance. The highest value of Cu content appeared in the 500–1,000 m zone (42.5 mg/kg), and the lowest value in the 2,000–2,500 m zone (37.2 mg/kg). The highest value of Zn was in the 4,500–5,000 m zone (95.5 mg/kg), while the lowest was found in the 3,000–3,500 m zone (88 mg/kg).

The spatial characteristics of heavy metals in soil

Semivariogram analysis, spatial autocorrelation analysis, and covariance analysis are the important methods to study the spatial pattern of variables. As shown in the previous section, Hg pollution was relatively serious; thus, we took Hg as an example to investigate the impact of urbanization on heavy metal concentrations in soil. The GS^+(v.7.0) software was used to compute the spatial pattern of Hg, with a total of 218 data samples. In view of the shape of the study area (Fig. 1), as well as the northwest (315°) and the southwest (225°) sections of the study area were mainly considered.

Semivariogram analysis

Soil Hg concentration in the study area had a significant spatial statistic characteristic (Table 2). The NSR for the whole region was 0.235, which showed that the distribution of Hg in soil was uniform relatively in space. The mean variance for the whole region was 0.393, and its distribution was well simulated by an exponential model. The range of the whole region was 5,530 m, indicating that the variability in Hg concentration tended to be stable in the scale of 5,530 m. The semivariogram function for the 315° section was very similar to that for the 225° section. The NSR and range for the 315° section of the study area were the same as the 225° section −0.14 and 6,610 m, respectively. Comparing with the whole study region, the NSR in the 315° and 225° sections was relatively low, which suggested that the effect of human activities on heavy
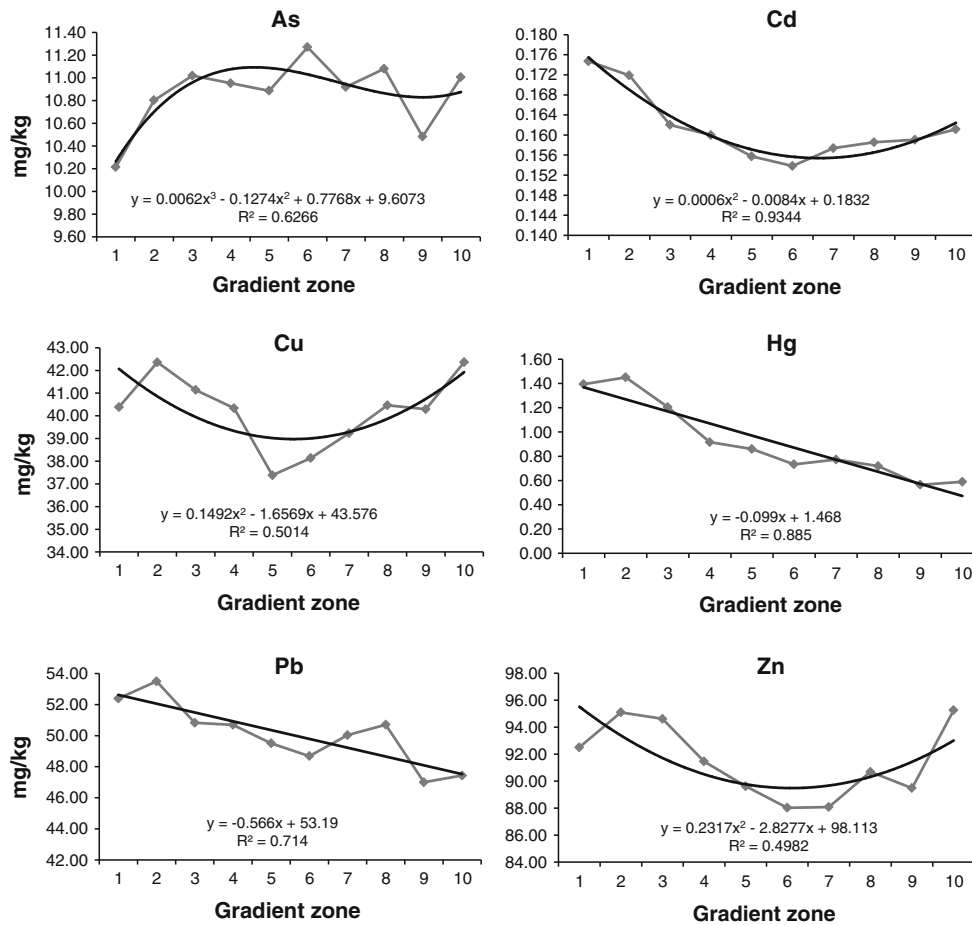
Fig. 2 The trend of six heavy metal soil concentrations according to distance from the built-up area. *Dashed lines* is trend line; 1–10 buffer zones represent 1–500, 500–1,000, 1,000–1,500, 1,500–2,000, 2,000–2,500, 2,500–3,000, 3,000–3,500, 3,500–4,000, 4,000–4,500, 4,500–5,000 m, respectively

metal concentration in the soil was distinct, while that of natural factors was low. The mean variance in the 315° section (0.476), however, was different than that of the 225° section (0.392), indicating that the spatial variation of soil Hg concentration in the 315° section was much more complex than that of the whole area.

Spatial autocorrelation analysis

Soil Hg concentration in the study area had a positive spatial correlation, and tended to converge in a spatial pattern (Moran's $I = 0.1356$). High Hg content was concentrated in the core of the built-up area and its surrounding region (up to 1,000 m away from the core), which suggested that Hg concentrations tended to gather together in space. Moran's I declined gradually with rising distance from 0.574 (at 389.38 m) to −0.068 (at 6,469.09 m), as shown in Fig. 3a. This revealed that the natural factors influencing the Hg concentration in soil increased with rising distance from the core of the built-up area; in

contrast, the structural factors (such as anthropogenic activities) decreased. This tendency also appeared in the 315° section, where the spatial variation is more abrupt (Fig. 3b). Here, the Moran's I value decreased from 0.518 at 323.88 m to −0.42 at 6,449.3 m, with the decline range being 0.94. This result suggested that the spatial variation of soil Hg concentration in the 315° section was much more complex than that of the whole area. The gradient in the 315° section was thus more obvious. The greatest value in the 225° section was 0.9 at 320.92 m, and the lowest was 0.017 at 4,664.06 m, which showed that the spatial pattern of Hg concentration was positively correlated (Fig. 3c); thus, the Hg pollution in the 225° section was more serious.

Covariance analysis

Variation of the Hg content weakened with increasing distance from the core of the built-up area (Fig. 4a), indicating a faded effect of human activities. This also revealed that the spatial variance of heavy metal concentration in

Table 1 The summary statistics values and its corresponding background values in study area (Unit: mg/kg)

|	As	Cd	Cu	Hg	Pb	Zn
Min	5.55	0.048	24.4	0.036	24.6	61.2
Max	17.9	11.6	108	4.38	110	204
Standard deviation (SD)	1.55	0.67	9.94	0.66	12.68	18.92
Mean	10.88	0.20	40.28	0.93	50.13	91.63
Corresponding background value	9	0.19	29.5	0.2	34.2	81.3
Grade II of The CEQSS[a]	30	0.3	50	0.3	250	200
EPA standard[b]	4	70	–	23	400	2,300
Trigger[c]	10	–	130	1	500	300

"–" represent no data

[a] The Chinese Environmental Quality Standard for Soils

[b] EPA, human health-screening criteria for residences (Sowana et al. 2011)

[c] England (Han et al. 2006)

Table 2 The results of Hg concentration in soil based on semivariogram analysis

Indictor	C_0	$C_0 + C$	Range (m)	$C_0/(C_0 + C)$	R^2	Mean variance	Model
Total	0.124	0.536	5,530	0.235	0.97	0.393	Exponential model
Northwest (315°)	0.126	0.9024	6,610	0.14	0.876	0.476	Exponential model
Southwest (225°)	0.126	0.9024	6,610	0.14	0.876	0.392	Exponential model
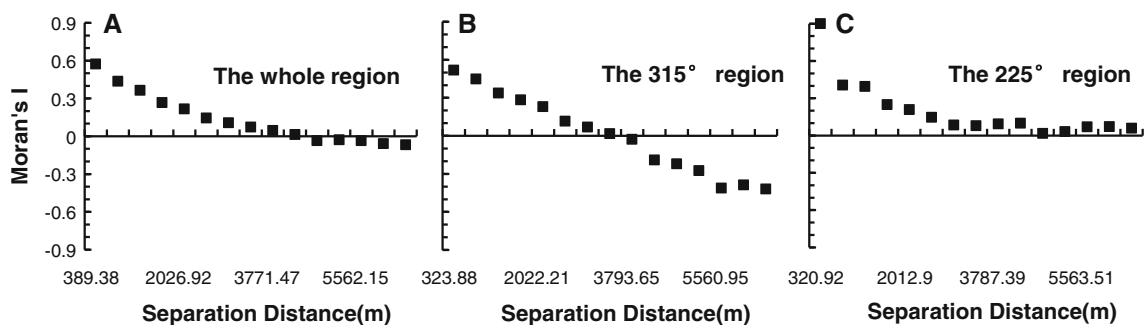
Fig. 3 The changes of Moran's *I* of Hg concentration
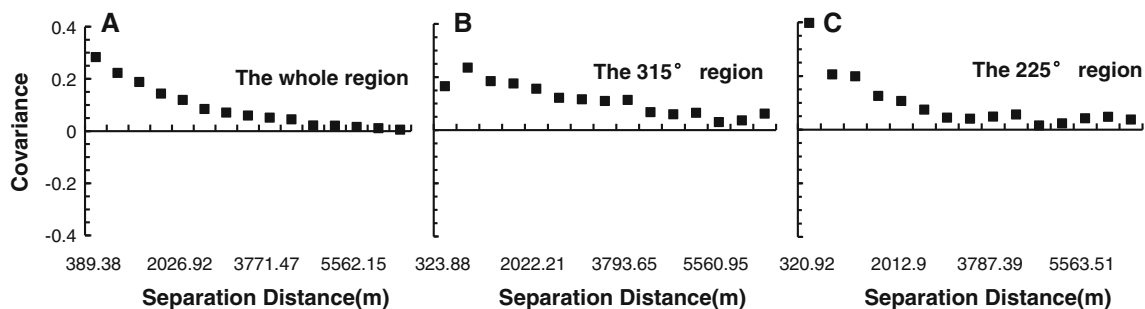
Fig. 4 The changes of covariance of Hg concentration

soil dropped with increasing distance from the core of the built-up area. The covariance was 0.28 at 389.38 m, and only 0.006 at 6,469.09 m, from which we could also determine that the tendency of the variation of heavy metal content became small with the decreased impacts of human activities. The decreasing rate of the covariance with

increasing distance from the core of the built-up area in the 315° section was lower than that for the whole study area (Fig. 4b), suggesting that soil Hg was mainly concentrated in the core of the built-up area and its suburbs (0–3,793.65 m). The covariance did not decrease obviously until the distance of 3,793.65 m from the core of the built-up area. The mean covariance was 0.09 for the whole area, and was 0.11 and 0.098, respectively, for the 315° and 225° sections. The change of the covariance was most abrupt for the 225° section, with its value decreasing from was 0.398 at 320.92 m to about 0.04 between 2,880 and 6,457 m (Fig. 4c). These results revealed a pattern of high pollution, high variance for Hg concentration showed in the small scale (built-up area), and high pollution, low variance in the large scale (the whole region).

Discussion

The Yangtze River Delta is one of the main regions suffering from heavy metal pollution in China, and the main heavy metal pollutants found are Hg and Cd (Zhong et al. 2007). Anthropogenic activities, especially the three wastes (wastewater, waste gas, and waste solids) discharged from industrial factories, are the major cause of heavy metal soil pollution (Bai et al. 2011; Li et al. 2001). The anthropogenic activities (e.g. the three wastes and automotive exhaust emissions) concentrated in the built-up area, constantly aggravated the heavy metal soil pollution in the built-up areas (Chen et al. 1997). There is a social-economic-ecological complex system covering the city, suburbs, and countryside surrounding the city, which generates inherent economy connections, administration subordination relations, and ecological communications. The traffic links between the production and consumption areas, the administration relations between the management and employees, and the ecological relationship caused by the heat islands affect all and expand the scope of heavy metal soil pollution (Cheng 2003; Huamain et al. 1999; Steinnes et al. 1989).

Geostatistics is an effective tool to analyze the change and variance of spatial parameters (Liu et al. 2006). Different heavy metals exhibited different content and spatial pattern in the soil of study area. It was demonstrated that the gradient effect in soil heavy metal concentration due to urbanization and industrialization does exist. In this study, soil Hg and Pb were confirmed to be strongly influenced by anthropogenic activities and tended to decrease with increasing distance from the built-up area. However, soil Cu enriched by anthropogenic activities first show a decrease followed by an increase with increasing distance. This might be possibly explained by scattered township enterprises in the study area (Shi et al. 2007; Liu et al.

2006; Cai et al. 2010; Huamain et al. 1999). Cd content showed a gradient change; however, the variation was very tiny. Instead of anthropogenic activities, Cd might be contributed by parent materials of the soil. Intensity of human activity was at the peak in urban built-up area where a huge volume of wastes involving heavy metal were released. However, the emission of heavy metal was less in suburb and countryside (Razo et al. 2004). Heavy metal soil pollution in urban would be expanded to suburb and countryside by the traffic links, the administration relations and the ecological relationship. This was the main reason for the gradient effect of soil heavy metal enrichment (Sun et al. 2010).

Meanwhile, the spatial pattern of the heavy metal concentration in the soil was found to be anisotropic. Prevailing wind, pollution sources and parent materials of soil were predominately responsible for the anisotropy of heavy metal concentration (Razo et al. 2004; Zhang et al. 2006; Bai et al. 2011). The study area was located in East Asia where southeast monsoon was powerful. Therefore, the heavy metal content changed distinctly in 315° section of study area. On the contrary, serious heavy metal pollution occurring in 225° section may be related to the existence of broad lake there. Because heavy metal content in sediment was higher than that in water and other soils, moreover the distribution of heavy metal in sediment was relatively uniform (Zhou et al. 2008). In addition, the distinct gradient change in different sections also might be due to the difference of human activity intensities, production methods and pollution sources distribution.

Conclusion

The results showed that the urbanization process affects not only the heavy metal content, but also the spatial pattern of the heavy metal concentration. In the study area, soil Hg, Cu, Pb and As were significantly influenced by anthropogenic activities. It was found that contents of three heavy metals (Hg, Cd and Pb) in soil tended to decrease with increasing distance from the built-up area, and exhibited a significant variation with the gradient, especially for Hg. This indicated that the gradient effect of heavy metal concentration among urban, its suburb and countryside does exist. The soil Hg pollution was most serious in Ehu Town. Therefore, local government and the environmental protection agencies should pay more attention to the problem of Hg soil pollution, and take effective actions to manage the high-polluting enterprises to strengthen the collection and treatment of high Hg content sewage. Moreover, the spatial pattern of heavy metal concentration was different among the whole study area, the 315° section and the 225° section.

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