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ORIGINAL ARTICLE

Spatial pattern and distribution regularity of soil environmental quality in East China

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Abstract Random numbers of heavy metal (Cd, As, and Pb) concentrations were created by Monte Carlo simulation of 16 representative sites in five soil zones from published papers for subsequent research. Ordinary kriging and significant analyses were used in a dataset of the 16 areas. Concentration of heavy metals in topsoil was found to be mainly influenced by zonality, non-zonality, and anthropogenic inputs. Nevertheless, human-induced correlations among heavy metals could not be detected on a regional spatial scale. Soil zone and landform were found to be representations of zonality and non-zonality, respectively. It was found that in east China, heavy metals accumulated in the south, especially the southwest, and were not uniform in each soil zone, indicating that zonality was not the unique influencing factor of soil environment quality. Both soil zone and landform may be responsible for heavy metal concentrations. An analysis of variance, coefficients of divergence analysis, was applied to assess the effects of soil zone and landforms on the variation of heavy metal concentrations in the study region. The influence of soil zone and landform was relatively homogeneous in areas of the north; while in the south, landform played a dominating role. Therefore, soil environment quality formed mainly under the effect of landform. This was mainly due to the fact that landform in fluences other natural environment elements including climate, further impacting the division of soil zone.

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1 Introduction

Soil is a natural component of the Earth, serving a variety of vital functions, including food production. Soil environmental quality can provide some scientific guidance for proper land use, control of soil contamination, and eco-environmental layout. A comprehensive understanding of heavy metal accumulation can be achieved by mapping distribution, tracing potential sources, and assessing risks, and can inform strategies to minimize pollution or exposure.

Heavy metals can have adverse effects on human health in high concentrations due in part to their persistence in the environment (e.g., Eze et al. 2010; Karim et al. 2014; Karimi Nezhad et al. 2014; Xu et al. 2013). Concentration of heavy metals in topsoil depends on bedrock elemental composition as well as several physical and chemical properties that control pedogenesis (e.g., Wilcke et al. 1999). Human-induced enrichment of heavy metals in soil is attributed to many activities. Several productive activities such as mining, smelting, industry, and power production have been identified as sources of metals in the environment (e.g., Huang et al. 2007; Cai et al. 2012; Lu et al. 2012; Sun et al. 2013). Anthropogenic inputs often contribute to an increase in the concentration of toxic heavy metals only locally and are not detected on a large scale (Nanos and Rodríguez Martín 2012). The natural concentration of heavy metals in these soils polluted by human-input tends to remain low owing to the composition of geological parent material (Shan et al. 2013).Meanwhile, significant geogenic enrichment has also been reported recently (e.g., Kelepertzis et al. 2013).

On the macroscopic scale, the main factors influencing concentrations of heavy metals are soil type and landform (Jing 1962; Taghipour et al. 2011). Soil types commonly characterized by high concentrations of heavy metals have been the subject of much research (Zhang and Guan 1994; Navas and Machín 2002). Meanwhile, a few scholars have shown that each geomorphic domain was an independently operating system with its own geochemical character, and soils varied from domain to domain (Graf et al. 1991; Ansari et al. 2000). According to Jing (1962) and Xi and Zhou (1982), soil type and landform are representations of zonality and non-zonality regulation, respectively. Zonality, which is really latitudinal zonality, means geographic crust components and their complexes roughly extend along latitude lines and generally vary with latitude, as opposed to variations associated with geological landform (Kelepertzis et al. 2013). China steps over several latitude zones, and forms multiple soil zones under the influence of zonal factors (Xi and Zhou 1982).

Some research based on data from the survey of the soil environment of the Ministry of Land and Resources, People's Republic of China (Cheng et al. 2014a; Li et al. 2014) has focused on the distribution of heavy metals across China. A survey of background values of soil elements in China started in the mid-1970s (China National Environmental Monitoring Centre 1990). The survey included analysis of background values in all provinces and soil types. From April 2005 to December 2013, the national soil pollution survey was carried out for the first time. Preliminary results announced by the Ministry of Environmental Protection of the People's Republic of China & Ministry of Land and Resources of the People's Republic of China (2014) presented the condition of soil pollution in China in different land use types and in several typical land parcels. However, the non-zonality/zonality of heavy metals in soil, and the factors influencing soil environment quality were little discussed at the national scale in these studies. For this reason, this study seeks to (i) investigate the spatial pattern and distribution regulation of heavy metals in the soil of China, (ii) determine the crucial factors controlling soil environmental quality.

2 Data sources and methods

2.1 Study area and data sources

The whole territory of China was divided into four great soil regions based on soil features and natural environment. In these four great soil regions, different soil zones were further divided mainly according to the zonal soil types and their related bioclimatic conditions (Xi and Zhou 1982). Because of the obvious difficulties of covering such a vast area, data in east China were collected from published papers. In order to reduce error, 16 sites distributed across five soil zones were chosen for being representative in terms of landform and relatively unaffected by anthropogenic activities (Fig. 1). The summary statistics of soil heavy metals are listed in Table 1.

2.2 Analytical methods

2.2.1 *Uncertainty analysis*

Uncertainty is an error between the true value and the measured or estimated value, and it originates from inaccurate measurements, lack of data, and model assumptions. In this paper, the uncertainty analysis was conducted using Monte Carlo simulation to create random numbers in each region based on statistics and distribution (Rajabi and Ataie-Ashtiani 2014) with Oracle Crystal Ball software version 11.1.2.2.000. Parameters of the Monte Carlo simulation of 16 representative sites in five soil zones, including minimum value (Min), maximum value (Max), mean value (Mean), standard deviation (Stdev), and the distribution assumption are listed in Table 1.

2.2.2 *Spatial analysis*

ArcGIS 10.0 was used to integrate physiographic and soil information. The spatial distribution of heavy metals in soils of east China was interpolated using ordinary kriging interpolation. Significant analysis was carried out by One-

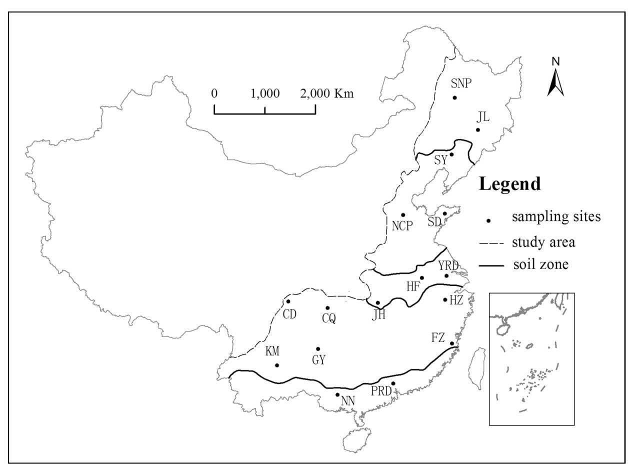

Fig. 1 Main sites of the field studies and sampling. (1) *NN* Nanning city; *PRD* Pearl river delta; *FZ* Fuzhou city; *KM* Kunming city; *CQ* Chongqing city; *GY* Guiyang city; *CD* Chengdu plain; *HZ* Hangzhou city; *JH* Jianghan plain; *YRD* Yangtze river delta; *HF* Hefei city; *NCP* North China plain; *SD* Shandong province east; *SY* Shenyang city; *JL* Jilin city; *SNP* Songnen plain. (2) I Lateritic *red* earth zone; II *Red* earth and *yellow* earth zone; III *Yellow brown* earth and *yellow* cinnamon *soil zone*; IV Brown earth, cinnamon *soil* and *dark loessal soil zone*; V *Dark brown* earth, phaiozem and chernozem zone

Table 1 Descriptive statistics of heavy metals in different regions

Region	n	Heavy metal	Min (mg kg^{-1})	Max (mg kg^{-1})	Mean (mg kg^{-1})	Stdev (mg kg^{-1})	Assumption	landform	Soil zone	Reference
PRD	212	Cd	0.072	1.116	0.366	0.282	Lognormal	Plain	I	Chai et al. (2004)
		As	0.88	57.97	13.88	9.66	Lognormal			
		Pb	1.92	216.70	58.01	22.66	Lognormal			
NN	179	Cd	0.28	1.11	0.69	0.248	Lognormal	Low-middle mountain	I	Liu (2004)
		As	8.74	69.96	23.20	23.20	Lognormal			
		Pb	14.68	414.96	65.56	68.69	Lognormal			
FZ	46	Cd	0.04	0.80	0.30	0.12	Lognormal	Hilly basin	I	Chen (2008)
		As	1.39	42.19	8.28	3.97	Lognormal			
		Pb	22.80	1071.70	89.83	94.00	Lognormal			
CD	86	Cd	0.021	2.12	0.323	0.155	Lognormal	Basin	II	Wang (2005)
		As	3.00	20.57	10.41	3.85	Lognormal			
		Pb	15.51	108.72	56.73	27.77	Lognormal			
GY	416	Cd	0.025	4.52	0.392	0.41	Normal	Middle mountain	II	Deng et al. (2006)
		As	1.08	98.00	23.55	24.63	Lognormal			
		Pb	9.40	229.80	50.94	15.04	Lognormal			
KM	45	Cd	0.285	0.968	0.548	1.39	Lognormal	Middle-high basin	II	Chen et al. (2004)
		As	3.098	41.011	14.549	36.95	Lognormal			
		Pb	23.666	264.794	56.984	69.50	Lognormal			
HZ	528	Cd	0.003	0.84	0.11	0.09	Lognormal	Plain	II	Xu et al. (2012)
		As	0.004	35.20	7.80	6.38	Lognormal			
		Pb	1.4	69.5	23.6	52.12	Lognormal			
CQ	455	Cd	0.04	19.105	0.5359	1.29	Lognormal	Low Mountain	II	Yang (2005)
		As	0.232	272.513	11.168	26.90	Lognormal			
		Pb	7.425	309.175	56.403	50.40	Lognormal			
JH	42	Cd	0.103	0.629	0.303	0.18	Lognormal	Plain/low mountain	III	Yu et al. (2008)
		As	3.7	20.24	10.977	6.50	Lognormal			
		Pb	25.49	46.09	35.855	14.33	Lognormal			
YRD	18,773	Cd	0.103	9.09	0.1809	0.72	Lognormal	Plain/hill	III	Yang (2010)
		As	–	–	–	–	Lognormal			
		Pb	11.4	2521.0	31.8179	111.89	Lognormal			
HF	180	Cd	0.069	0.577	0.131	0.10	Lognormal	Plain	III	Dong (2007)
		As	5.65	14.69	9.17	7.15	Lognormal			
		Pb	19.68	34.68	25.41	11.98	Lognormal			
NCP	19,084	Cd	0.109	0.184	0.15	0.02	Lognormal	North China plain	IV	Liu (2012)
		As	6.65	12.5	9.57	1.24	Lognormal			
		Pb	18.1	27.2	22.63	10.0	Lognormal			
SD	13,674	Cd	0.01	10.54	0.12	0.16	Lognormal	Hill	IV	Dai et al. (2012)
		As	0.8	162.3	6.62	8.83	Lognormal			
		Pb	8.9	934.9	27.66	5.41	Lognormal			
SY	2256	Cd	0.15	1.179	0.113	0.20	Normal	Songliao plain	IV	Hou et al. (2011)
		As	0.971	11.807	6.84	12.26	Lognormal			
		Pb	5.518	124.018	24.2	55.0	Normal			
SNP	20,701	Cd	0.01	4.2	0.095	0.099	Lognormal	Plain	V	Zhang et al. (2011)
		As	1.4	41	8.68	9.05	Lognormal			
		Pb	1.9	119.5	22.0	0.79	Lognormal			
JL	1490	Cd	0.04	1.59	0.13	0.066	Lognormal	Middle-low mountain	V	Dong (2012)
		As	3.6	45.9	10.43	5.32	Lognormal			
		Pb	20.3	92.5	27.44	12.35	Lognormal			

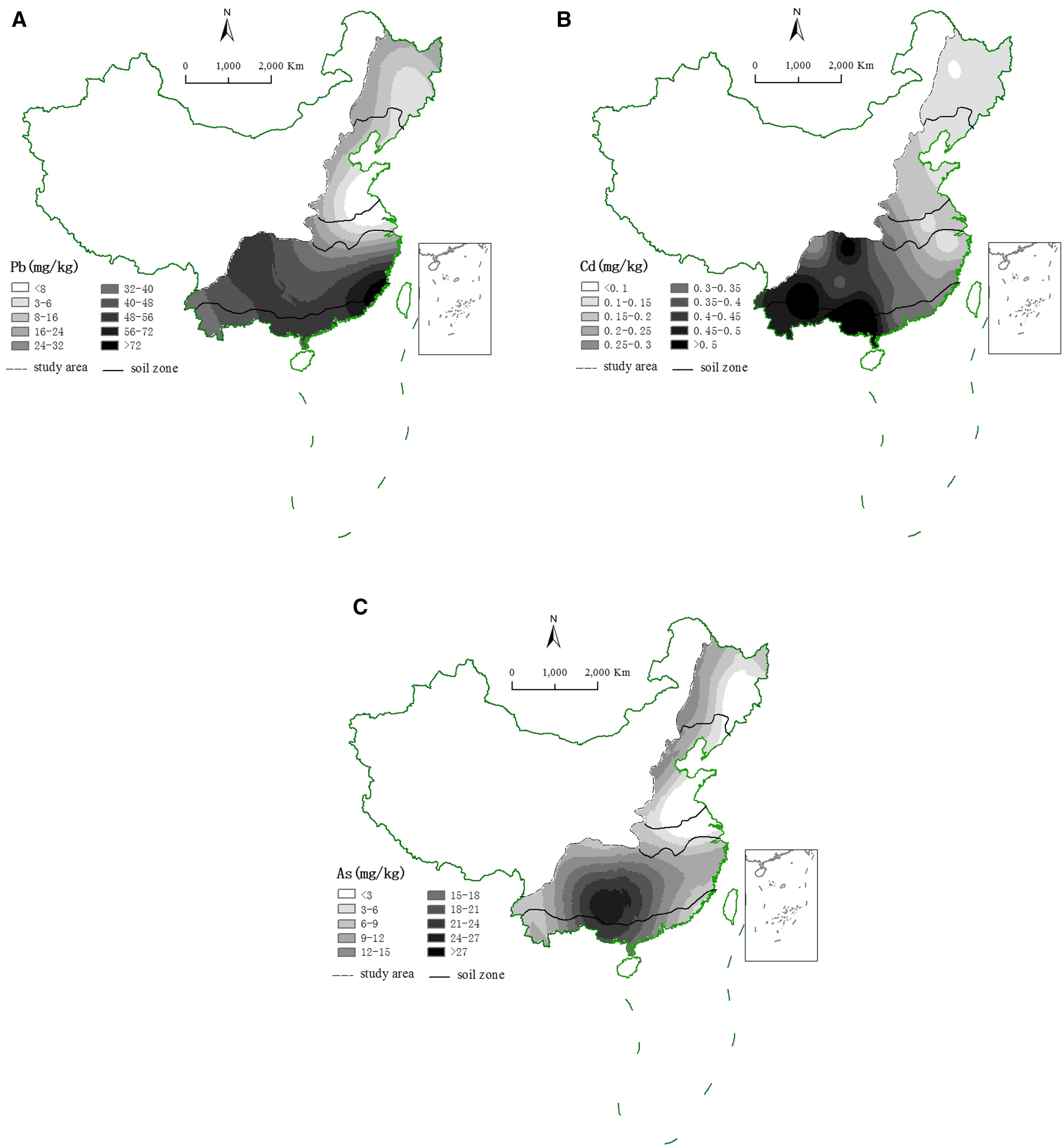

Fig. 2 Distribution of Cd, Pb and As concentrations in east China

way ANOVA with SPSS 17.0 to test whether soil environment quality was consistent in each soil zone.

2.2.3 Coefficients of divergence (COD)

An analysis of variance was applied to assess the effects of soil type and landforms on the variation of heavy metal concentrations in the study region. In this context, soil zones may have strong linear association with each other in total heavy metal concentrations, yet landform may have substantial influence. For this analysis, COD was defined as:

$$COD_{jk} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}}\right)^2}$$

where ‘n’ is the number of heavy metal components (i.e., three); ‘x_{ij}’ is the average concentration of component ‘i’ at location ‘j;’ and ‘j’ and ‘k’ represent two monitoring locations of the same soil zone(Sarnat et al. 2010). The leading influencing factor was assessed using COD.

3 Results and discussion

3.1 Spatial distribution of heavy metals

Cd, As, and Pb concentrations exhibit a large spatial variability in their distribution (Fig. 2). The summary statistics of soil heavy metals are listed in Table 1.

Average concentrations of Cd ranged from 0.09 to 0.69 mg kg^{-1}. The high concentration (above 0.3 mg kg^{-1} (Environmental Protection Department of the People’s Republic of China 2008)) occurred in the south, especially the southwest, including NN, KM, CQ, GY, CD, and JH. SNP, SY, SD, HF, and JL had concentrations of Cd were lower than 0.15 mg kg^{-1} and were mostly located in the north.

Average Pb concentrations ranged from 22.63 to 89.83 mg kg^{-1}. The soil in Fuzhou had the highest average Pb concentration at 89.83 mg kg^{-1}, followed by CD, NN, KM, PRD, CQ, and GY all above 50 mg kg^{-1}. These hotspots of Pb were mainly distributed in the south and southeast coast of China. Remarkably low Pb concentrations (<27 mg kg^{-1}) were observed in north and central China at NCP, SNP, HZ, SY, and HF (Table 1).

Average As concentrations ranged from 6.62 to 23.55 mg kg^{-1}. The highest average As concentration of 23.55 mg kg^{-1} in topsoil was found in GY, and the lowest mean, 6.62 mg kg^{-1}, was found in SD. The largest region with high As concentration was the Yunnan-Guizhou Plateau.

Though the distribution patterns of Cd, Pb, and As differ greatly in the region of east China, there is some similarity in that the three metals are all relatively concentrated in the south. Also, this variability is in accordance with the landform distribution in China. The Qinling Mountains are a geographical boundary as well as a climate barrier between the north and the south of China. South of this boundary, there is a drastic decrease in the percentage of plains and an increase in mountainous conditions. Under the influence of geologic processes, heavy metals are taken to the surface from parent material (Wang et al. 2006). Therefore, trace element concentration in surface soil has consistency and inheritance with deep soil, which may lead to spatial differences and could explain the basic pattern of soil environmental quality (Chen and Zhou 2012).

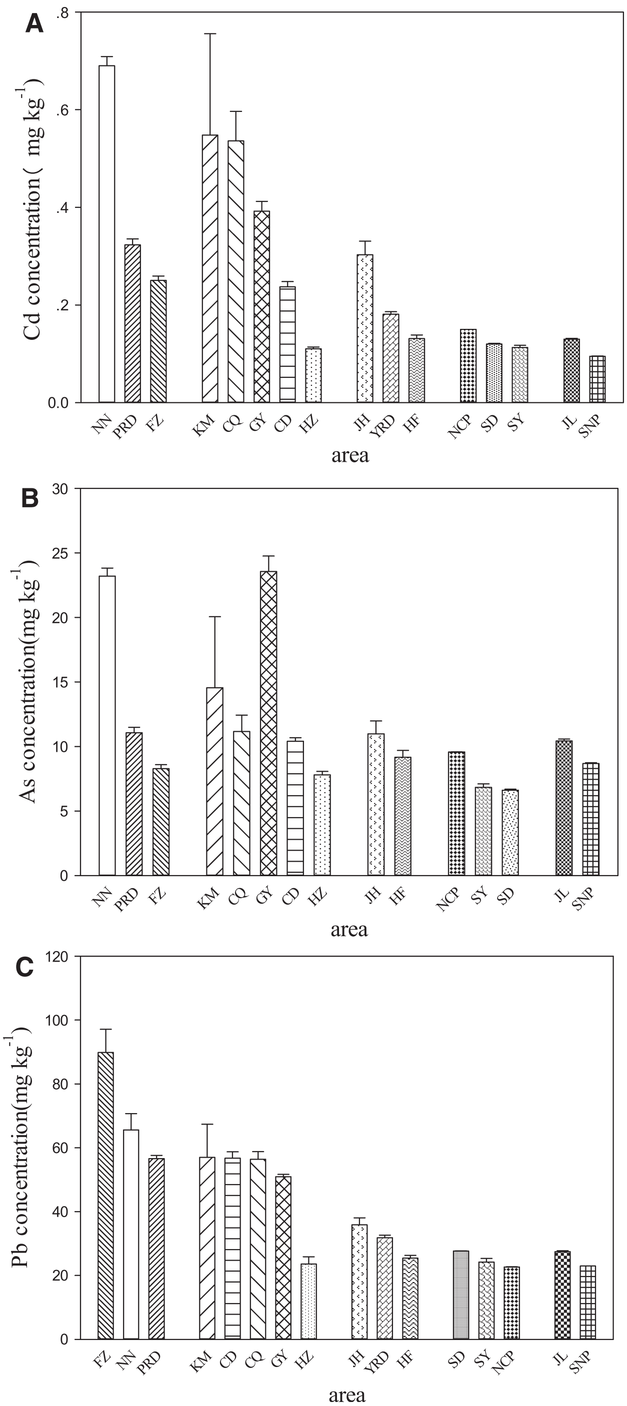

Fig. 3 Concentration of Cd, Pb and As in different *soil zones*

3.2 Heterogeneity of heavy metals in soil zones

Heavy metals differed greatly ($p < 0.05$) in diverse areas of the same soil zone (Fig. 3). For zone I, NN, PRD, and FZ had different heavy metal concentrations ($p < 0.05$), and their landforms were hilly basin, plain, and low-middle mountains, respectively. In zone II, concentrations of heavy metal in KM, CQ, and GY were relatively homogeneous, mainly due to high background levels of the Yunnan-Guizhou Plateau (National Environmental Protection Agency of the People's Republic of China 1993). In contrast, HZ had completely different concentrations of heavy metal ($p < 0.05$) with the landform of plain. In zone III, heavy metals were significant different among JH, PRD, and HF ($p < 0.05$), the landform of which were plain/low mountain, plain/hills, and plain. In zone IV, heavy metal concentrations were different among NCP, SD, and SY ($p < 0.05$), which belonged to east China low plain, hilly basin, and Songliao plain, respectively. In zone V, JL and SNP, with the landform of middle/low mountain and plain, respectively, also had different heavy metal concentrations ($p < 0.05$).

Since concentrations of heavy metals varied within soil zones, zonality was not the only factor that influenced soil environment quality. In each soil zone, concentrations of heavy metals varied with the change of landform. This diversity of heavy metal concentrations and landforms were almost synchronous. According to other research (Wang et al. 2006; Cheng et al. 2014b), the main source of heavy metals in soils were parent materials. Under the effect of geologic agent, parent materials were diverse in different landforms. This result mainly agreed with previous research. Nanos and Rodríguez Martín (2012) found that in northeast Spain, the spatial distribution of heavy metal concentrations showed a large variability with the highest concentrations in the mountain ranges and the lowest in the plains. Chen and Zhou (National Environmental Protection Agency of the People's Republic of China 1993) found that heavy metals of moisture soil in the Yangtze River basin were significantly different from the Huaihe River basin due to different sediment sources.

Among diverse landforms, parent materials generate different element concentrations (Nechaev and Isphording 1993; National Environmental Protection Agency of the People's Republic of China 1994). Natural concentrations of heavy metals depend on their concentrations in parent materials. Some research (Ren and Yu 2012) has shown soil developed from carbonatite rock to be rich in certain elements due to a high degree of weathering compared with soils developed from neutral alluvium. As the red earth and yellow earth zone was the most prevalent in south China, this study examined HZ and GY to analyze the influence of parent materials on the concentrations of heavy metals. According to Wang and Zuo (2009), the parent material of HZ and GY were neutral alluvium and carbonatite rock, respectively. Although both HZ and GY were in the red earth and yellow earth zone, the concentrations of heavy metals differed significantly ($p < 0.05$). In the long-term soil-forming process, heavy metals in parent material were reassigned.

3.3 Dominating factor of soil environment quality

Krudysz et al. (2008) defined CODs for pollutant distributions greater than 0.20 as being "relatively heterogeneous" or dissimilar using results from numerous sites. In this study, COD was used to measure whether the influence of soil zone and landform was equivalent on concentrations of heavy metals. The pairwise COD value of zone IV was set as a reference value COD_0. Since the zone's landform, soil type, and other soil-forming factors were relatively homogeneous, its soil environment quality was considered to be influenced by both zonality and non-zonality equally. CODs of other soil zones larger than COD_0 indicated that zonality was the main influencing factor on heavy metal concentration. Otherwise, the soil environmental quality formed under the interaction of zonality and non zonality. For the current analysis, the absolute concentrations of Cd, As, and Pb from each soil zone were assessed using CODs. CODs ranged from 0.12 to 1.03 in soil zones (Fig. 4).

The CODs were larger than COD_0 in zones I, II, and III, indicating that landform played a dominant role in heavy metal concentrations. COD in zone V was below COD_0, suggesting that the influences of zonality and non-zonality were relatively homogeneous.

Zone IV and V are in north China, while zone I, II, and III are in south China. In the north, plain is the general landform. Therefore, zonality was the single influencing variable at the macroscopic scale. Nevertheless, landform and parent materials varied greatly in the south, especially for zone II, leading to the a decrease in the influence of zonality on heavy metal concentrations. Landform restricts other natural environment elements, such as climate, vegetation, soils, hydrology, etc. (Li et al. 2013). Meanwhile, soil zones were divided mainly according to the zonal soil types and their related bioclimatic conditions (Nanos and Rodríguez Martín 2012). Therefore, the division of soil zones was influenced by landform to some

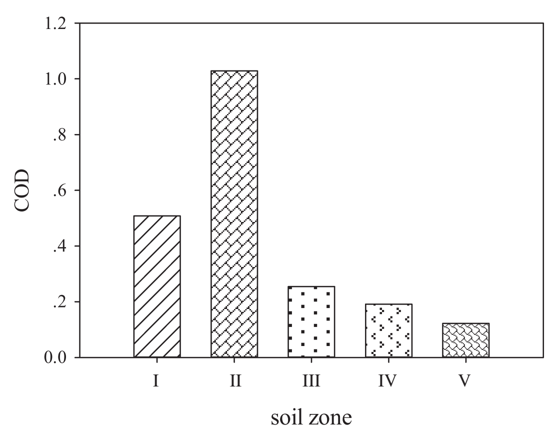

Fig. 4 CODs of different zones

extent. For east China, it was the landform that dominated the heavy metal concentrations in soil.

4 Conclusions

Concentrations of heavy metals varied greatly not only with latitude, but also with soil zone for east China. Under the mountainous conditions in the south, especially in areas with high background levels, concentrations of heavy metals in soil were relatively higher than the north, due to the fact that geologic processes took heavy metals to the surface from parent material. Although soil type had some influence on soil environment quality locally, this influence occurred when landforms were relatively uniform. Soil zone and landform, which are examples of zonality and non-zonality regulation, were alien to each other in the geochemistry process. Soil zone was also influenced by landform, which was the basic cause of observed spatial patterns as well as an independent factor of soil environment quality in east China. Further research, such as regional soil management and soil environment function, could be carried out according to landform regionalization.

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