RESEARCH ARTICLE



Source identification and risk assessment of heavy metal contaminations in urban soils of Changsha, a mine-impacted city in Southern China

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Abstract The urban soils suffered seriously from heavy metal pollutions with rapid industrialization and urbanization in China. In this study, 54 urban soil samples were collected from Changsha, a mine-impacted city located in Southern China. The concentrations of heavy metals (As, Cd, Co, Cu, Mn, Ni, Pb, and Zn) were determined by ICP-MS. The pollution sources of heavy metals were discriminated and identified by the combination of multivariate statistical and geostatistical methods. Four main sources were identified according to the results of hierarchical cluster analysis (HCA), principal component analysis (PCA), and spatial distribution patterns. Co and Mn were primarily derived from soil parent material. Cu, Pb, and Zn with significant positive relationships were associated with mining activities and traffic emissions. Cd and Ni might be affected by commercial activities and industrial discharges. As isolated into a single group was considered to have correlation with coal combustion and waste incineration. Risk assessment of heavy metals in urban soils indicated an overall moderate potential ecological risk in the urban region of Changsha.

Keywords Heavy metal · Urban soil · Spatial distribution · Multivariate analysis · Source identification · Potential ecological risk

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Introduction

Urban soil is an integral part of the urban ecosystem closely related to human health and life quality of city residents. Serving as sinks, urban soils may become enriched in various contaminants from human activities and atmospheric depositions. The contaminants will transmit from urban soils into the atmosphere, surface water, and plants, which could directly or indirectly threat human health through inhalation, dermal contact, and ingestion. Among various types of the contaminants, heavy metal pollutions have attracted considerable attention due to their biotoxicity and irreversibility (Chen et al. 2008, Ding and Hu 2014). Once transferred into the human body, the heavy metals can be accumulated because of their long residence time and non-biodegradable nature (Cui et al. 2005). The toxicity of heavy metals in urban soils has been widely investigated to evaluate its adverse impacts on local inhabitants. Long-term exposure to heavy metals may cause damage to certain organs and induce cancers (Jarup 2003). In order to assess the potential risk of heavy metals in urban soils to the ecosystem, evaluation models, such as potential ecological risk index and Nemerow comprehensive index, have been employed to quantify the pollution levels (Ma et al. 2015, Zhou et al. 2014).

The contaminations of heavy metals in many urban soils primarily originated from anthropogenic sources by urban and industrial expansion, such as mining and smelting activities, municipal and industrial wastes disposal, motor vehicle emissions. and fossil fuel combustion (Chen et al. 2014, Gallagher et al. 2008). Identification and discrimination of heavy metal sources have become an important issue for pollution control and soil remediation (Hu and Cheng 2013, Luo et al. 2015). Since the origins of heavy metals were normally contributed by several factors, such as anthropogenic activities and natural existence, sources of heavy metals in urban soils were

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sometimes indistinguishable (Burt et al. 2014, Nanos and Rodríguez Martín 2012). Numerous surveys have been carried out to investigate the concentrations, distributions, and sources of heavy metals in urban and periurban soils. Due to the complex behavior and heterogeneity of urban soils, the concentrations and distribution of heavy metals varied over space (Luo et al. 2007). The multivariate statistical methods in conjunction with the geographic information system (GIS) were previously used as powerful analysis tools to distinguish and identify different sources of heavy metals in soils (Mihailović et al. 2015). Normally, the heavy metal sources of covariation were inferred indirectly through multivariate statistical analysis (for instance, correlation analysis, hierarchical cluster analysis, and principal component analysis) (Li et al. 2015, Wang et al. 2012). In addition, the spatial distributions of heavy metals were mapped by GIS to infer their sources in spatial scale. Factorial kriging analysis is one of the most common interpolation methods applied in environmental research (Guo et al. 2012, Mihailović et al. 2015).

In recent decades, the urban ecosystem suffered serious from heavy metal pollutions with rapid industrialization and urbanization in many cities of China (Wei et al. 2009). Elevated heavy metal levels in urban soils may increase exposure to harmful materials and pose potential risk to ecosystem and humans. Changsha City, located in Southern China, was chosen as the study region due to the serious soil contamination (Wang et al. 2010) and few studies concerning the heavy metal pollution in soils from downtown area. The objectives of this study were (1) to investigate the concentration and distribution of heavy metals, (2) to identify the possible sources, and (3) to assess the potential ecological risk.

Materials and methods

Site description

Changsha City (111° 54'–114° 15' E, 27° 51'–28° 40' N) is the capital of Hunan Province located in Southern China. The total area of this city is 11,819.5 km² and the population is approximately 7.31 million. The climate of Changsha was characterized as humid subtropical monsoon climate with average annual precipitation of 1360 mm. Changsha is a rapid developing city where nonferrous mining and metallurgy are the economic mainstays. Mining and smelting activities were reported to cause soil contamination in a relatively large scale (Liao et al. 2005, Liu et al. 2005). As a mine-impacted city, the soils have been significantly polluted by heavy metals in Changsha (Wang et al. 2010). The research region was focused on the downtown area, within the second ring road of Changsha. This region with relative high density of people and heavy traffic pollution is separated into two parts by the Xiangjiang River, main tributary of the Yangzi River. The residential area and campus are mainly located in the western part, while the business district, hospitals, and railway stations are mostly located in the eastern part.

Sample collection and preparation

A total of 54 topsoil samples (0–10 cm) were collected in the downtown area of Changsha City in November 2015. The sample sites are illustrated in Fig. 1. Three to five subsamples were taken using a stainless steel scoop around each of the sampling site. The subsamples were mixed to get one composite sample and then sealed in a polyethylene bag. After being air dried, the samples were grinded and sieved through a 0.15-mm nylon mesh. The homogenized fractions were stored in polyethylene bags until analysis.

Heavy metal analysis

The fine soil samples were digested according to Method 3051A (USEPA 2007) with minor modification. The mixture of HNO₃ and HCl (3:1, v/v) was employed to extract heavy metals from soil samples using microwave digestion (MDS-6G, Sineo Microwave Chemistry Technology Co. Ltd, Shanghai, China). The digestion solutions were filtered through 0.45-µm cellulose acetate membrane filters (Xinya Purify Device Company, Shanghai, China) and then made up to 50 mL by ultrapure deionized water (18.2 M Ω cm⁻¹, Direct-Q 3, Millipore SAS, France). The concentrations of As, Cd, Co, Cu, Mn, Ni, Pb, and Zn were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7700x, Tokyo, Japan). The method detection limits of the ICP-MS method were 195, 7, 10, 37, 52, 74, 56, and 108 µg/kg for As, Cd, Co, Cu, Mn, Ni, Pb, and Zn, respectively. A method blank was performed throughout the entire pretreatment procedure for each batch of sample preparation. The digestion efficiency was verified using certified reference material (CRM) soil GBW07444 (urban dust) obtained from CRM/ RM Information Center (Beijing, China). The recoveries for all the heavy metals were in the range of 91–105 %.

Statistical analysis

Pearson's correlation analysis, hierarchical cluster analysis (HCA), and principal component analysis (PCA) were selected to investigate and identify the possible sources of heavy metals in the urban soils. The HCA approach was based on the Ward's method and the squared Euclidean distance. The principal components (PCs) were transferred from the original variables using eigenanalysis (Lee et al. 2006). Varimax rotation with Kaiser normalization was applied to extract PCs according to the eigenvalue with the variance >10 % and cumulative variances >70 % (Ma et al. 2015). The validity

Fig. 1 Distribution of sampling sites in Changsha City



of the PCA was confirmed by Kaiser-Meyer-Olkin (KMO) measure and Bartlett's test. Multivariate analysis techniques in this study were performed by SPSS v.13.0 for Windows. The spatial distributions of heavy metals were plotted by the geostatistical method. The concentrations of heavy metals at unsampled or unmeasured locations were predicted using the ordinary kriging interpolation technique (Guo et al. 2012). This approach was performed by the geography information system (GIS) software.

Risk assessment

To evaluate the degree of heavy metal pollution in urban soils from Changsha City, the Nemerow comprehensive index (P_s) was introduced in this study. The P_s is calculated according to the following equations (Wu et al. 2015):

$$C_f^i = C_s^i / C_n^i \tag{1}$$

$$P_s = \sqrt{\frac{\left(C_{f ave}^i\right)^2 + \left(C_{fmax}^i\right)^2}{2}} \tag{2}$$

where C_s^i (mg/kg) represents the measured content of heavy metal *i* in the collected urban soil, C_n^i (mg/kg) represents the geochemical background value heavy metals *i* in the local soil, C_f^i represents the single pollution index of heavy metal *i*, C_{fave}^i is the average value of C_f^i , and C_{fmax}^i is the maximum value of C_f^i of all the heavy metals. The P_s is classified into five groups (Ma et al. 2015): $P_s < 1$: uncontaminated; $1 \le P_s < 2$: slightly contaminated; $2 \le P_s < 3$: moderately contaminated; $3 \le P_s < 5$: strongly contaminated; and $P_s \ge 5$: seriously contaminated.

To assess the harmful effect of soil heavy metal contamination on the urban ecosystem, the potential ecological risk index (RI) proposed by Hakanson (Hakanson 1980) was employed here. The RI is defined according to the following equations:

$$E_r^i = T_r^i \times C_f^i \tag{3}$$

$$RI = \sum_{i=1}^{n} E_r^i \tag{4}$$

where T_r^i is the toxic response factor of heavy metal *i*. Based on the results of Hakanson (Hakanson 1980) and Madiseh (Madiseh et al. 2009), the T_r^i values for As, Cd, Co, Cu, Mn, Ni, Pb, and Zn are 10, 30, 1, 5, 1, 2, 5, and 1, respectively. E_r^i represents the potential ecological risk coefficient of heavy metal *i*. The degree of E_r^i is classified into five groups: $E_r^i < 40$, low risk; $40 \le E_r^i < 80$, moderate risk; $80 \le E_r^i < 160$, considerable risk; $160 \le E_r^i < 320$, high risk; and $E_r^i \ge 320$, very high risk. The potential ecological risk index can be categorized into four classes as reported in a previous study (Ma et al. 2015): *RI*<100: low risk; $100 \le RI < 200$: moderate risk; $200 \le RI < 400$: considerable risk; *RI* ≥ 400 : very high risk.

Results and discussion

Heavy metal concentrations

The descriptive statistical results of heavy metal concentrations, such as maximum, minimum, mean, standard deviation, and coefficient of variation, in the urban soils collected from Changsha City are summarized in Table 1. Compared with the natural background values of soils from Hunan Province, the mean values of the investigated heavy metals in this study were higher than the reference values (CNEMC 1990), except Co and Ni. The maximum values for As (32.36 mg/kg), Cd (2.74 mg/kg), Co (28.42 mg/kg), Cu (138.46 mg/kg), Mn (943.89 mg/kg), Ni (98.25 mg/kg), Pb (176.49 mg/kg), and Zn (416.84 mg/kg) were 2.1, 21.7, 1.9, 5.0, 2.1, 3.1, 5.9, and 4.4 times higher than the corresponding reference values, indicating that heavy metal pollutions existed in the study region. A large coefficient of variation (>70 %) was found in Cd and Zn, which represents the concentrations of these two heavy metals varied widely in the study region.

The probability distributions of heavy metal concentrations were examined by the One-Sample Kolmogorov-Smirnov (K-S) normal test. The results illustrated that the concentrations of As, Cd, Co, Mn, Pb, and Zn were normally distributed (p > 0.05) in the collected urban soils (Wang et al. 2012), while Cu and Ni concentrations could not pass the test (p < 0.05) indicating discharges of Cu and Ni in some subareas (Zhou et al. 2014). The positive skewness value revealed that all the measured heavy metals skewed toward lower concentrations. Also, the positive kurtosis value of most heavy metals represented steeper distribution than normal (Chen et al. 2012), except Mn.

Correlation analysis

The relationships among measured heavy metals were investigated by Pearson's correlation analysis. As listed in Table 2, Cd. Cu. Mn. Pb. and Zn were found significantly positively correlated (p < 0.01) with each other. Besides, As was significantly correlated (p < 0.01) with Cd (r = 0.434), Pb (r=0.420), and Zn (r=0.391); Co was significantly correlated (p < 0.01) with Cu (r = 0.560), Mn (r = 0.632), Pb (r = 0.500), and Zn (r=0.467); and Ni was significantly correlated (p < 0.01) with Cd (r = 0.618), Mn (r = 0.485), Pb (r = 0.472), and Zn (r=0.543). Although the significant positive relationship could not always be concluded to a common source, the correlation interelements could still provide interesting information on the source and pathway of heavy metals (Lu et al. 2010). For example, Cd, Cu, Pb, and Zn with significant positive relationships could be derived from anthropogenic sources (mining and smelting activities). Also, As, Co, and Ni might have originated from different sources compared to the other elements.

Multivariate analysis

The hierarchical dendrogram was generated from HCA. As illustrated in Fig. 2, three distinct clusters were identified according to the degree of association between heavy metals. Cluster 1 (C1) including Cd, Cu, Ni, Pb, and Zn could be associated with anthropogenic inputs, such as traffic and industrial pollution (Chen et al. 2014). Cluster 2 (C2) consisting of Co and Mn might be mainly derived from a natural origin. Different from other elements, As classified into Cluster 3 (C3) was originated from anthropogenic pollution, such as fossil fuel combustion through atmospheric deposition.

The PCA approach was performed to further identify the sources of measured heavy metals in this study. Before the PCA, the sampling adequacy and sphericity were examined by KMO measure and Bartlett's test. For the KMO measure, the original variables were factorized efficiently and the KMO index was calculated through comparison of the values of correlations between variables and those of the partial correlations. The Bartlett's test checked if the observed correlation

 Table 1
 Descriptive data of heavy metal concentrations in urban soils from Changsha City

	Min (mg/ kg)	Max (mg/ kg)	Mean (mg/ kg)	Std. deviation (mg/kg)	Coefficient of variation (%)	K-S (Normal) <i>p</i>	Skewness (mg/kg)	Kurtosis (mg/kg)	Background values ^a (mg/ kg)
As	8.10	32.36	20.61	5.09	24.7	0.951	0.157	0.142	15.7
Cd	0.05	2.74	0.74	0.60	81.0	0.184	1.272	1.551	0.126
Со	3.21	28.42	11.65	4.83	41.5	0.568	1.055	1.870	14.6
Cu	13.39	138.46	37.09	19.93	53.7	0.040	2.906	12.142	27.8
Mn	84.15	943.89	467.10	213.65	45.7	0.304	0.511	-0.477	459
Ni	8.03	98.25	20.23	13.58	67.1	0.011	4.073	21.093	31.9
Pb	14.65	176.49	50.14	29.52	58.9	0.085	2.095	5.915	29.7
Zn	29.86	416.84	111.28	81.36	73.1	0.063	1.909	3.990	94.4

^a Soil background values for heavy metals in Hunan Province of China

 Table 2
 Pearson's correlation matrix for heavy metal concentrations

	As	Cd	Co	Cu	Mn	Ni	Pb	Zn
As		0.001	0.664	0.024	0.027	0.063	0.002	0.003
Cd	0.434^{b}		0.011	0.000	0.000	0.000	0.000	0.000
Со	0.060	0.344 ^a		0.000	0.000	0.041	0.000	0.000
Cu	0.307 ^a	0.521^{b}	0.560^{b}		0.001	0.010	0.000	0.000
Mn	0.301 ^a	0.601^{b}	0.632^{b}	0.425 ^b		0.000	0.000	0.000
Ni	0.255	0.618 ^b	0.279 ^a	0.347^{a}	0.485 ^b		0.000	0.000
Pb	0.420 ^b	0.722^{b}	0.500^{b}	0.804 ^b	0.547 ^b	0.472^{b}		0.000
Zn	0.391 ^b	0.830 ^b	0.467 ^b	0.780 ^b	0.544 ^b	0.543 ^b	0.883 ^b	

The right upper part is significant level; the left lower part is correlation coefficient

^a Correlation is significant at the 0.05 level (two-tailed)

^b Correlation is significant at the 0.01 level (two-tailed)

matrix diverged significantly from the identity matrix. The large KMO value (0.822) and suitable significance of Bartlett's test (p < 0.001) obtained were demonstrating valid PCA approach (Fu et al. 2014). As presented in Table 3, a total of four PCs were extracted with eigenvalues higher than 1.0. The first component (PC1) explaining 33.0 % of the total variance showed positive and high loading (>0.5) for Cd, Cu, Pd, and Zn (whose coefficients were 0.510, 0.894, 0.829, and 0.808, respectively). The second component (PC2) explaining 22.2 % of the total variance showed high loading (>0.6) for Cd and Ni (whose coefficients were 0.676 and 0.897, respectively). The third component (PC3) explaining 20.1 % of the total variance exhibited strong loading (>0.75) for Co and Mn (whose coefficients were 0.869 and 0.782, respectively). The fourth component included only As with strong positive loading (>0.9) explaining 14.5 % of the total variance. The result of the PCA was in agreement with



Fig. 2 Dendrogram of HCA for heavy metal concentrations in urban soils of Changsha

 Table 3
 Rotated component matrix for heavy metal contents

Heavy metal	Component						
	1	2	3	4			
As	0.204	0.125	0.035	0.955			
Cd	0.510	0.676	0.179	0.294			
Co	0.377	0.033	0.869	-0.095			
Cu	0.894	0.072	0.282	0.087			
Mn	0.164	0.437	0.782	0.244			
Ni	0.182	0.897	0.156	0.043			
Pb	0.829	0.315	0.251	0.227			
Zn	0.808	0.451	0.196	0.185			
Eigenvalues	2.643	1.777	1.606	1.162			
% of Variance	33.0	22.2	20.1	14.5			
Cumulative %	33.0	55.2	75.3	89.8			

PCA loadings >0.5 are shown in bold. Extraction method: principal component analysis. Rotation method: varimax with Kaiser normalization. Rotation converged in seven iterations

the HCA for PC1 and PC2 corresponding to C1, PC3 corresponding to C2, and PC4 corresponding to C3.

Spatial distribution and source identification

In order to distinguish the anthropogenic activities from natural origin clearly, ordinary kriging interpolation was introduced to obtain the contour maps of the heavy metals analyzed in the present study. Spatial distribution patterns of As, Cd, Co, Cu, Mn, Ni, Pb, and Zn in the downtown area of Changsha City were presented in Fig. 3.

The elements of Co and Mn showed similar spatial distributions with high concentrations in the north region and the low concentrations in the south region as illustrated in Fig. 3c, e. Since the observed distribution trends for Co and Mn were gently without any hot-spot area and the mean concentrations of these two elements were close to or lower than the corresponding background values, Co and Mn in urban soils were defined uncontaminated and were primarily derived from soil parent material. This was consistent with the results of HCA and PCA, where Co and Mn were categorized into C2 and both exhibited strong positive loading on PC3.

The distribution maps of Cd, Cu, Ni, Pb, and Zn were similar with relatively high spatial variability as shown in Fig. 3b, d, f, g, h. This proved the results of the K-S normal test while the p values for Cd, Cu, Ni, Pb, and Zn were low. Impacted by lead and zinc smelters around Changsha City, the urban soils have been polluted by the heavy metals of Cd, Cu, Pb, and Zn. However, Cu, Pb, and Zn with strong positive loading (>0.8) on PC1 were not only associated with mining and smelting activities (a)



(b)

-28°17'

Fig. 3 Spatial distribution of heavy metal concentrations in urban soils of Changsha

but were also affected by traffic density and vehicular emission in urban soils. High concentrations of Pb might be attributed to the use of petrol with Pb additives. Although the leaded gasoline has been banned in China, the content of Pb in the urban soils may reflect long-term accumulation of historical Pb contamination from traffic emissions (Yang et al. 2011). Due to corrosion resistance and strength, copper alloy as a material is used in mechanical parts, such as baking systems and automotive radiators (Guo et al. 2012, Yang et al. 2011). The deterioration of the mechanical parts in vehicles contributed dramatically to the Cu content in urban soils. Zinc components are widely used in motor vehicles. Also, the tire tread contains 0.4 to 4.3 % of Zn (Chen et al. 2014). Corrosion of automobile parts and wear and tear of tire treads are the significant sources of Zn. In addition, the contaminations of Cd and Ni with high loading on PC2 might be derived from commercial

Fig. 3 continued.

activities and industrial discharges (Wei et al. 2009, Yang et al. 2011).

The spatial distribution of As with high concentrations in the south region and low concentrations in the north region (as shown in Fig. 3a) was distinctly different from the other elements. Less variability of As concentrations was found in the spatial distribution map. However, the mean and maximum concentrations of As were 1.3 and 2.1 times higher than the background value, respectively, implicating that the urban soils might be influenced by As contaminations. Commonly, As is considered to be an

indicator of coal combustion and waste incineration (Yang et al. 2011). The element of As isolated in C3 and PC4 was associated with industrial activities, such as fossil coal combustion, waste incineration, and metal refining and manufacturing.

Risk assessment

The Nemerow comprehensive index calculated through Eq. (1) and Eq. (2) was demonstrated in Fig. 4a. The P_s values of the sampling sites ranged from 0.63 to 4.87. Referring to

Fig. 4 Risk assessment for heavy metals in urban soils of Changsha: a Nemerow index; b ecological potential risk index

the P_s category mentioned before, the pollution degrees were between uncontaminated ($P_s < 1$) and strongly contaminated ($3 \le P_s < 5$). Thereinto, 31 sampling sites (57 % of total sites) with P_s value between 1 and 2 were slightly contaminated. Moreover, seven sites (13 % of total sites) with P_s values between 2 and 3 were grouped into moderately contaminated, and three sites (6 % of total sites) with $P_s > 3$ were considered as strongly contaminated. Only 13 sampling sites (24 % of total sites) with $P_s < 1$ were grouped to uncontaminated.

The average E_r^i values obtained by Eq. (3) were 13.1, 176.7, 0.8, 6.7, 1.0, 1.3, 8.4, and 1.2 for As, Cd, Co, Cu, Mn, Ni, Pb, and Zn, respectively, suggesting considerable risk for Cd in the urban soils. Due to relatively high T_r^i value and low geochemical background value, the E_r^i value of Cd was higher than the other heavy metals (Suresh et al. 2012). The potential ecological risk assessed according to Eq. (4) was illustrated in Fig. 4b. The *RI* ranged from 31.2 to 711.0 with arithmetic mean value of 151.7. According to the category of

RI, the heavy metals posed an overall moderate potential ecological risk in the urban region. However, 15 sampling sites with $200 \le RI \le 400$ were considerable risk, and 7 sampling sites with $RI \ge 400$ were grouped into very high risk. The results of potential ecological risk assessments indicated pollution control and soil remediation should be carried out in some locations.

Conclusions

The pollution degree and spatial distribution of heavy metals in urban soils collected from Changsha City have been investigated in this work. The concentrations of As, Cd, Co, Cu, Mn, Ni, Pb, and Zn were determined by ICP-MS method. The mean values of analyzed heavy metals were higher than the natural background values of soils from Hunan Province, except Co and Ni. Four distinct groups were classified according to the results of HCA and PCA. Furthermore, the pollution sources of heavy metals were identified by the combination of multivariate statistical and geostatistical methods. Mn and Co with similar distribution patterns and positive loading on PC3 were defined to uncontaminated and were primarily derived from soil parent material. Cu, Pb, and Zn with significant positive relationships and strong positive loading (>0.8) on PC1 were associated with mining activities and traffic emissions. Cd and Ni with positive loading on PC2 might be affected by commercial activities and industrial discharges. Also, Cd with moderate positive loading (0.510) on PC1 was originated from mining and smelting activities. As was isolated into C3 in HCA and PC4 in PCA, which was correlated with coal combustion and waste incineration. The pollution degrees categorized by the Nemerow index were between uncontaminated and strongly contaminated. The potential ecological risk assessment indicated an overall moderate risk in the urban region of Changsha.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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