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Spatial distribution and pollution assessment of heavy metals in urban soils from southwest China

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Abstract

To identify the concentrations and sources of heavy metals, and to assess soil environmental quality, 63 soil samples were collected in Yibin City, Sichuan Province, China. Mean concentrations of As, Pb, Zn, and Cu were 10.55, 61.23, 138.88 and 56.35 mg/kg, respectively. As concentrations were comparable to background values, while Pb, Zn, and Cu concentrations were higher than their corresponding background values. Industrial areas exhibited the highest concentrations of As, Pb, Zn, and Cu, while the lowest concentrations occurred in parks. Statistical analysis was performed and two cluster groups of metals were identified with Pb, Zn, and Cu in one group and As in the other. Spatial distribution maps indicated that Pb, Zn, and Cu were mainly controlled by anthropogenic activities, whereas As could be mainly accounted for by soil parent materials. Pollution index values of As, Pb, Zn, and Cu varied in the range of 0.24–1.93, 0.66–7.24, 0.42–4.19, and 0.62–5.25, with mean values of 0.86, 1.98, 1.61, and 1.78, respectively. The integrated pollution index (IPI) values of these metals varied from 0.82 to 3.54, with a mean of 1.6 and more than 90% of soil samples were moderately or highly contaminated with heavy metals. The spatial distribution of IPI showed that newer urban areas displayed relatively lower heavy metal contamination in comparison with older urban areas.

Key words: urban soils; heavy metals; pollution assessment; spatial distribution

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Introduction

In recent decades, considerable attention has been paid to the problem of urban soil contamination with heavy metals to prevent further environmental deterioration and to examine applicable methods of soil remediation. Heavy metals in soils have been considered to be powerful tracers for monitoring the impact of human activities (Kelly et al., 1996; Manta et al., 2002). Known for peculiar characteristics such as unpredictable layering, poor structure, and high concentrations of trace elements (Kabata-Pendias and Pendias, 1992; Tiller, 1992), urban soils can be served as recipients of large amounts of heavy metals from multiple sources including municipal wastes (Schuhmacher et al., 1997), vehicular emissions (Harrison et al., 1981), industrial wastes (Rashed, 2010), and coal and fuel combustion (Li et al., 2001). These activities lead to emission of heavy metals into the air and their subsequent deposition into urban soils (Chen et al., 2005).

Heavy metals in urban soils have been widely studied due to their ubiquity, toxicity and persistence. In urban areas, heavy metals can be readily transferred into the human body as a consequence of dermal contact absorption, inhalation, and ingestion (Ferreira-Baptista et al., 2005; Lim et al., 2008). Then the metal can typically accumulate in human body due to their non-biodegradable nature and long biological half-lives for elimination. It has been found that heavy metals in urban soils may have toxic effects on human health (Ahmed and Ishiga, 2006; De Miguel et al., 1998), especially on the children (Li et al., 2004; Ljung et al., 2006; Poggio et al., 2009). For example, low-level Pb exposure can be harmful to enzyme systems involved in blood production and high-level Pb exposure can even affect intelligence of human (Babula et al., 2008; De Miguel et al., 2007).

Studies on heavy metal contamination in urban soils assist in developing strategies to protect urban environments and human health against long-term accumulation of heavy metals. Numerous studies have been conducted in developed countries (Geagea et al., 2008; Imperato et al., 2003; Madrid et al., 2006; Pen-Mouratov et al., 2008; Zhang, 2006). However, differences among cities such as population density and industrial activities, as well as traffic density may have some influence on the results of individual studies. Moreover, little information is available

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from these cities with various heavy industries and rapid economic development.

Yibin City, a rapidly developing city in China has undergone rapid urbanization and industrialization in recent years. The local government has been striving to construct ecological cities in the upper reaches of the Yangtze River to protect the Three Gorges Reservoir. Previous studies of heavy metal concentrations in agricultural soils of rural areas have been conducted in Yibin City (Wang et al., 2008, 2009). However, concentrations and spatial distribution patterns of heavy metals in the urban soils of Yibin City remain unknown. Therefore, the main objectives of this study were to investigate the contents and spatial distribution of heavy metals in urban soils, and to assess the soil contamination levels in Yibin City.

1 Materials and methods

1.1 Study area

Yibin City (103°36′E–105°20′E, 27°50′N–29°16′N) is a major industrial city in Sichuan Province, south-west China. It covers an area of 1123 km^2 and has about 890,000 inhabitants. It has a subtropical monsoon humid climate with a mean annual temperature of 17.9°C and a mean annual rainfall of 1168 mm. The city consists of three administrative districts, Jiangbei District (including Shangjiangbei Area and Xiajiangbei Area), Cuiping District, and Nan'an District. The Jiangbei and Cuiping District are relatively older districts in comparison with the Nan'an District.

1.2 Soil sampling

A total of 63 topsoil samples (0–5 cm) were collected from different functional areas in Yibin City (Fig. 1) including 13 samples collected from parks, 12 from commercial areas, 14 from main roadside areas, 8 from residential areas, and 16 from industrial areas. Soil samples were obtained by mixing 5–10 subsamples from each site. About 1 kg of each soil sample was collected using a stainless steel spade and stored in self-sealing plastic bags. The spade was washed with deionized water and wiped dry with paper towels between each use. Geographical coordinates of sampling locations were recorded at each sampling point with a GPS.

1.3 Analytical procedures

All soil samples were air-dried, gently ground, and sieved through a 2 mm polyethylene sieve to remove stones, coarse materials and other debris. A portion of each sample was then further ground and homogenized with an agate mortar to pass through a 0.15 mm polyethylene sieve. All handling procedures were carried out without contacting any metals to avoid potential cross-contamination of the samples.

Soil samples were digested with nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) using Method 3050B of the US EPA (1996). Concentrations of Cu, Pb, and Zn in the digestion solution were determined by flame atomic absorption

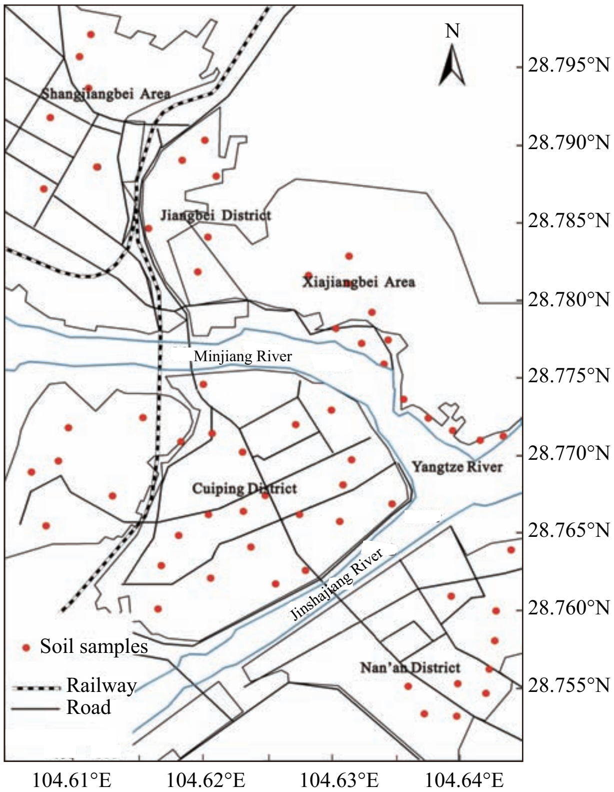

Fig. 1 Location of the study area and distribution of sampling sites in Yibin City.

spectrometry (GF-AAS, Vario 6, Jena Co. Ltd., Germany), whereas for determination of As concentrations, hydride generation atomic fluorescence spectroscopy (HG-AFS, AFS-2202, Haiguang Instrumental Co., China) was used.

Standard reference materials obtained from the Center of National Standard Reference Materials of China, as well as blank samples, were included in each batch of analyses for quality assurance and quality control (QA/QC) procedures. Results were considered satisfactory when within a range of ±10% from certified values. All samples were analyzed in duplicate and results were accepted when the relative standard deviation was within 5%.

1.4 Methods of heavy metal pollution assessment

To assess contamination level of the heavy metals, a pollution index (PI) for each metal and an integrated pollution index (IPI) for these four metals were calculated. The PI of each metal was defined as the ratio of its concentration to the background value of the corresponding metal using the following equation (Chen et al., 2005; Wei and Yang, 2010):

$$PI = C/S$$

where, PI is the pollution index corresponding to each sample, C (mg/kg) is the measured concentration of each heavy metal, and S (mg/kg) is the background value. In this study, the soil background values of As, Pb, Zn, and Cu in Sichuan Province used were 10.4, 30.9, 86.5, and 31.1 mg/kg, respectively (CNEMC, 1990). The PI of each metal was classified as either low (PI ⩽ 1), moderate (1 < PI ⩽ 3) or high contamination (PI > 3).

The integrated pollution index (IPI) of these four heavy metals was defined as the mean PI value for these four metals, and was then classified as low (IPI ⩽ 1), moderate (1 < IPI ⩽ 2), or high contamination (IPI > 2) (Chen et al., 2005; Wei and Yang, 2010).

1.5 Statistical analysis

All statistical analyses in this study were performed using SPSS 11.5 software. Principal component analysis (PCA) and cluster analysis (CA) were used to distinguish the different groups of heavy metals. PCA with varimax rotation was performed on log-transformed data. CA was performed to classify heavy metals from different sources on the basis of similarities in their chemical properties. Geochemical maps of heavy metals were obtained using the extension of geostatistical analyst of geography information system (GIS) software (Arc GIS, version 93).

2 Results and discussion

2.1 Heavy metal concentrations

Concentrations of As, Pb, Zn and Cu in the urban soils of Yibin City, together with soil background values of heavy metals in Sichuan Province, are presented in Table 1. The concentration ranges of As, Pb, Zn, and Cu were 5.95–15.10, 20.29–223.85, 36.16–362.15 and 19.15–163.32 mg/kg, with mean values of 10.55, 61.23, 138.88, and 56.35 mg/kg, respectively. Mean concentrations of the heavy metals in the urban soils decreased in the order of Zn > Pb > Cu > As. Concentrations of As were comparable to the background values, while Pb, Zn, and Cu concentrations were, respectively, 1.98-, 1.61-, and 1.78-fold higher than their corresponding background values. The concentrations of Pb, Zn, and Cu varied greatly, while As concentrations were quite homogeneous across the city. Based on the coefficients of variation (CV), these analyzed heavy metals can be classified into two groups: As, with CV values lower than 0.3; and Pb, Zn, and Cu, with CV values higher than 0.5. It has been reported that CV values of heavy metals dominated by natural sources are relatively low, while CV values of heavy metals affected by anthropogenic sources are quite high (Han et al., 2006). Accordingly, Pb, Cu, and Zn concentrations in urban soils tend to be affected by anthropogenic activities, while As may more often be associated with natural sources.

The concentrations of As, Pb, Zn, and Cu in different functional areas are shown in Fig. 2. Concentrations of Pb, Cu, and Zn in each functional areas were higher than their corresponding background values. As concentrations in soils from parks and main roadside areas were lower than the background values, while concentrations in commercial, residential, and industrial areas slightly exceeded these values. The highest mean concentrations of As, Pb, Zn and Cu were found in industrial areas with mean values of 11.84, 96.3, 189.22, 79.96 mg/kg, respectively. Industrial activities such as coal combustion, metallurgy, and metal manufacturing processes were the dominant sources of heavy metals in these industrial areas. The lowest mean concentrations of As, Pb, Zn and Cu were observed in parks with mean values of 9.3, 37.7, 86.4 and 37.25 mg/kg, respectively. Compared with commercial and residential areas, the mean concentrations of Pb, Cu, and Zn in main roadside soils were higher and their mean concentrations were 78.50, 61.99 and 181.09 mg/kg, respectively. In contrast, mean concentrations of As were lower in main roadside soils in comparison with commercial and residential areas.

Table 1 Heavy metal concentrations and coefficients of variations in urban soils in the Yibin City and background values in Sichuan Province

Heavy metal	Concentrations (mg/kg)				SD	CV	BV (mg/kg)
	Min	Max	Median	Mean			
As	5.95	15.10	10.13	10.55	2.72	0.26	10.4
Pb	20.29	223.85	46.83	61.23	38.73	0.63	30.9
Zn	36.16	362.15	111.78	138.88	71.98	0.52	86.5
Cu	19.15	163.33	51.63	56.35	28.74	0.51	31.1

SD: standard deviation; CV: coefficients of variation; BV: background values.

Pb, Zn, and Cu have been most studied in the urban soils worldwide (Table 2). As shown in Table 2, concentrations of the metals in the urban soils of Yibin City were much lower than those reported from many large and/or industrialized cities (i.e., Nanjing, Torino, Stockholm, Hamburg, London, Madrid, Naples, and Palermo). Concentrations of Zn and Cu in the analyzed soils were higher than those in Hong Kong and Seville, whereas the concentrations of Pb were lower. However, Pb, Zn, and Cu concentrations were similar to those measured in cities such as Bangkok, Damascus, Tallinn and Oslo.

2.2 Correlation coefficient analysis

The relationships between heavy metals can provide important information on heavy metal sources and pathways (Manta et al., 2002). Pearson correlation coefficients and their significance levels in the urban soils of Yibin were summarized in Table 3. Significant correlations were found among Zn and Cu, suggesting that these heavy metals may originate from a common pollution source. As showed only weak positive correlations with the other heavy metals, suggesting a different source than for Pb, Zn, and Cu.

2.3 Multivariate analysis results

PCA and CA were applied to assist in identification of pollutant sources. The CA shows that the heavy metals can be classified into two clusters using a criteria value of rescaled distance between 5 and 10 (Fig. 3). Cluster I contained Pb, Cu, and Zn. These three heavy metals were well-known pollutants in urban soils and may originate from a common anthropogenic source. Cluster II contained As which may originate from the soil parent materials. A natural source was suggested by its separate clustering from the other heavy metals.

To further investigate the relationships among heavy metals, PCA was performed. This indicates that As, Pb, Zn, and Cu concentrations can be grouped into two components which accounted for 84.46% of the total variance (Table 4). The heavy metal classification from PCA was

Table 2 Heavy metal concentrations in urban soils worldwide

City	Sample number	Pb (mg/kg)	Zn (mg/kg)	Cu (mg/kg)	Reference
Nanjing	138	107.3	162.6	66.1	Lu et al., 2003
Hong Kong	58	94.6	125	23.3	Li et al., 2004
Hong Kong	236	88.1	103	16.2	Lee et al., 2006
Torino	70	149	183	90	Biasioli et al., 2006
Stockholm	7	101	171	71	Linde et al., 2001
Seville	12	92.1	91.4	38.4	Ruiz-Cortés et al. 2005
Hamburg	30	218.2	516	146.6	Wilcke et al., 1998
London	53	294	183	49	Culbard et al.,1988
Damascus	22	17	84	30	Möller et al., 2005
Oslo	300	34	130	24	Tijhuis et al., 2002
Tallinn	532	63	121	–	Bityukova et al., 2000
Madrid	55	161	210	72	De Miguel et al., 1998
Naples	173	251	262	74	Imperato et al., 2003
Palermo	70	252	151	77	Manta et al., 2002
Bangkok	30	48	118	42	Wilcke et al., 1998
Yibin	63	61.23	138.88	56.35	This study

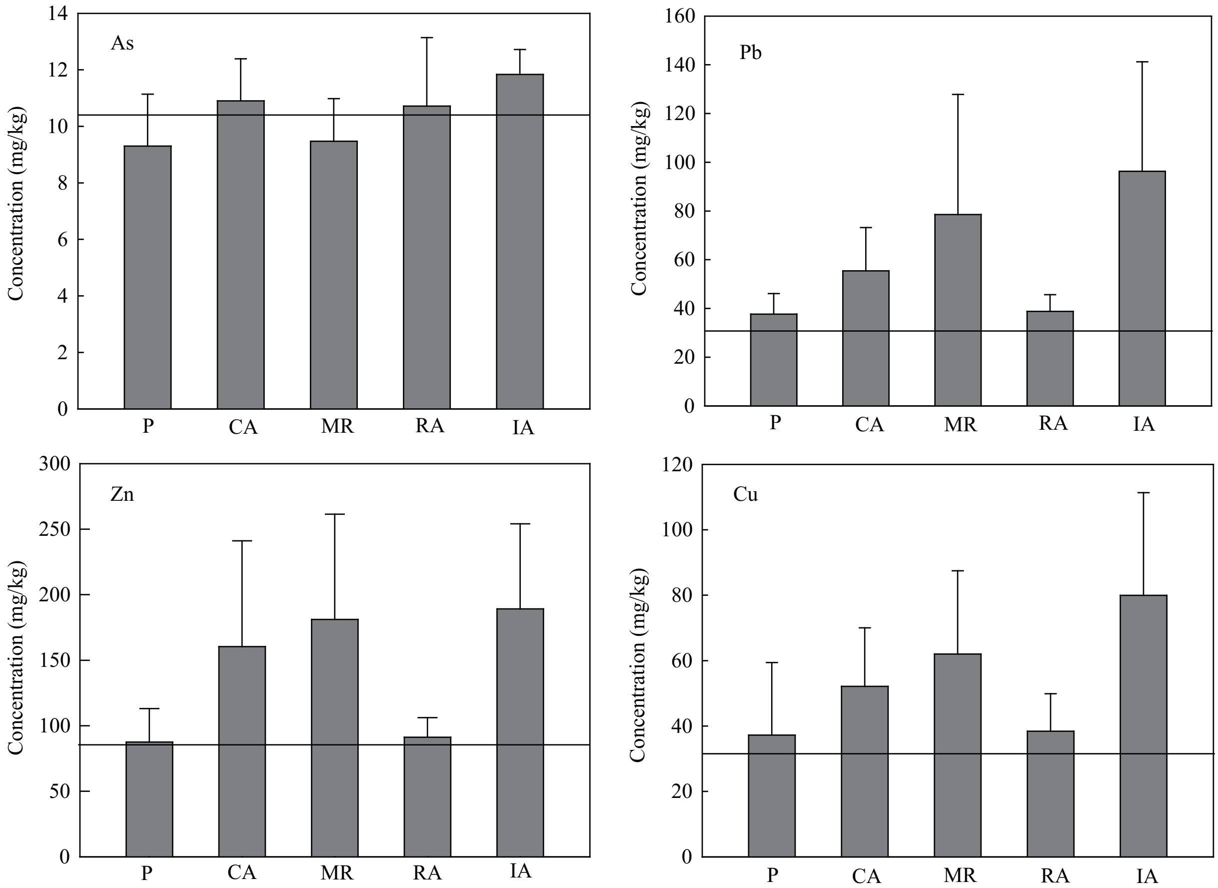

Fig. 2 Heavy metal concentrations in soils of different functional areas of Yibin City. P, CR, MR, RA, and IA refer to parks, commercial areas, main roadside areas, residential areas, and industrial areas, respectively. Horizontal lines represent the background values of As, Pb, Zn, and Cu in the soils of Sichuan Province.

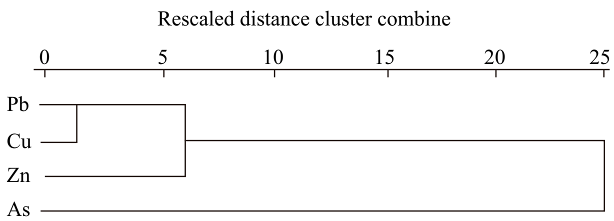

Fig. 3 Dendrogram of hierarchical cluster analysis of heavy metals in the urban soils of Yibin City.

consistent with the results from CA: PC1 with Cluster 1, and PC2 with Cluster 2. The first component (PC1) accounted for 58.63% of the total variance and showed the association of heavy metals such as Pb, Zn, and Cu dominated by anthropogenic inputs. As was unequivocally isolated in the second component (PC2) and accounted for 25.83% of the total variance. The communality of variables ranged from 0.78 for Cu to 0.99 for As. As showed a relatively weak association with the other heavy metals, suggesting a natural geochemical association with the soil parent materials.

Table 3 Pearson correlation matrix for heavy metals in the urban soils of Yibin City

Heavy metal	As	Pb	Zn	Cu
As	1	0.234	0.237	0.124
Pb	0.064	1	0.716*	0.663*
Zn	0.061	0.000	1	0.670*
Cu	0.331	0.000	0.000	1

The left lower part in significant level; the right upper part is correlation coefficients. * $p < 0.01$.

Table 4 Rotated component matrix for As, Pb, Zn, and Cu in urban soils of Yibin City

Heavy metal	Component		Communalities
	PC1	PC2	
As	0.11	0.99	0.99
Pb	0.87	0.16	0.79
Zn	0.88	0.16	0.80
Cu	0.89	–0.01	0.78
Rotation sum of squared loading			
Percentage of variance (%)	58.63	25.83	
Percentage of cumulative (%)	58.63	84.46	

2.4 Spatial distribution of heavy metals

GIS software can be used to produce spatial distribution maps and identify the potential sources of heavy metals in urban areas (Li et al., 2004). In the present study, the concentrations of As, Pb, Zn and Cu were interpolated using kriging.

Spatial distribution maps of As, Pb, Zn, and Cu in the urban areas of Yibin City are presented in Fig. 4. Pb, Cu, and Zn showed high values in the vicinity of industrial buildings and near road junctions and/or near major roads carrying large amounts of traffic. Heavy industries, including mechanical manufacturing plants, pharmaceutical plants, metallurgical plants, cement manufacturing plants, coal-fired power plants and chemical plants, mainly located in Jiangbei District of Yibin City. Cement-derived dust in cement plants contained high concentrations of heavy metals, especially Pb, Cu, and Zn (Han et al., 2006), which may be in turn accumulated in urban soils through atmospheric deposition. Atmospheric deposition of heavy

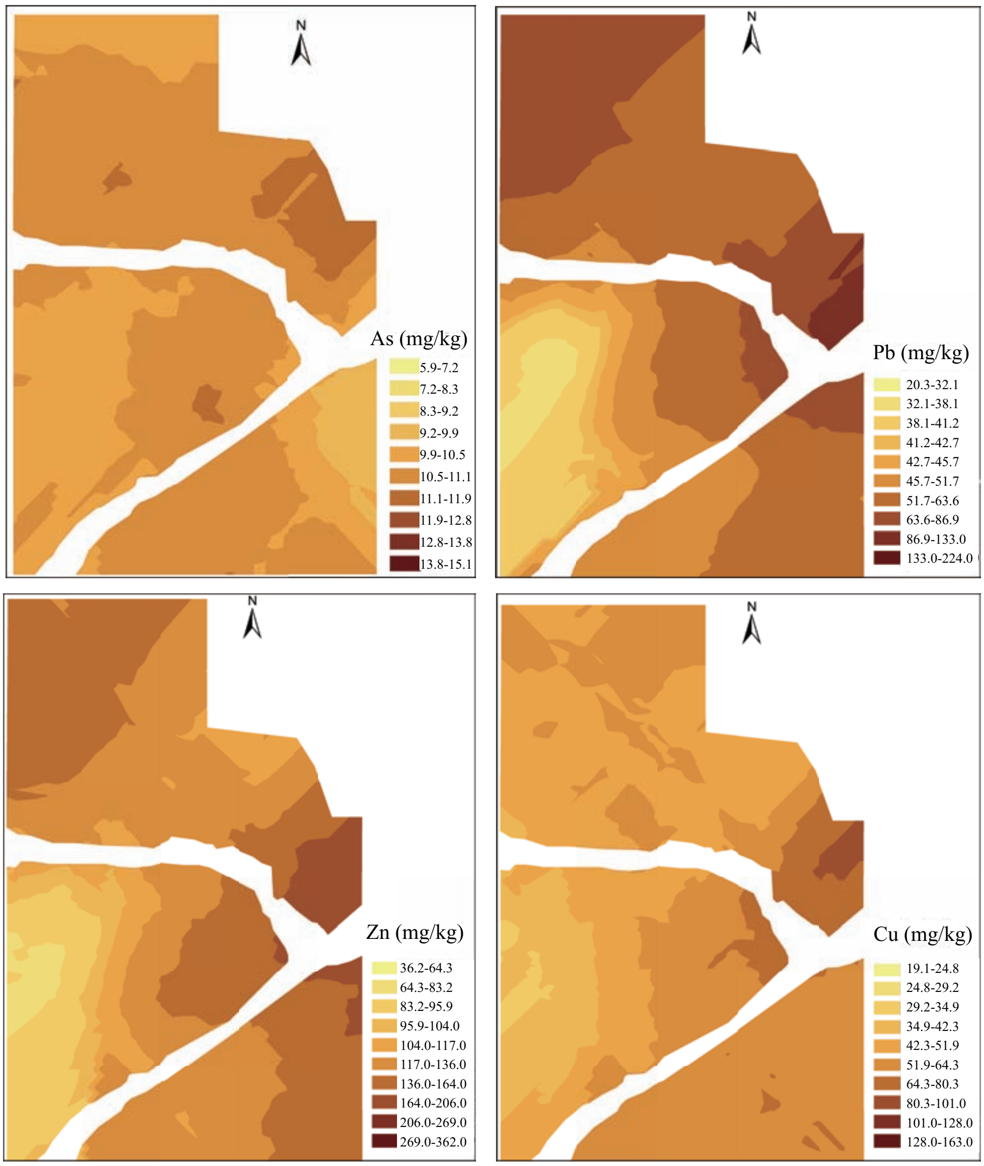

Fig. 4 Spatial distribution maps of As, Pb, Zn, and Cu in the urban soils of Yibin City.

metals was considered to be a significant factor in urban soil pollution (Lindström, 2001). It has been reported that high concentrations of Pb in urban soils were associated with vehicular exhausts arising from the use of leaded gasoline (De Miguel et al., 1997; Imperato et al., 2003; Saby et al., 2006). Although petrol with Pb additives has been banned in China, the high concentrations of Pb in the urban soils may reflect long-term accumulation of heavy metals from traffic emissions (Guo et al., 2008). In addition, Zn compounds have been employed extensively as antioxidants and as detergent/dispersant improvers for lubricating oils (De Miguel et al., 1999). Therefore, wear and tear of tyres contributed significantly to the Zn content in urban soils (Ellis and Revitt, 1982). Copper has been used in vehicular braking systems and in Cu-brass automotive radiators (Miner, 1993; Nimmo, 1998). The deterioration of mechanical parts in vehicles over time thus resulted in the accumulation of Cu and Zn in urban soils. Therefore, vehicular emission may play a part, in addition to industrial activities, in significant accumulation of heavy metal in the urban soils of Yibin City. Overall, the spatial patterns of heavy metals such as Pb, Cu, and Zn in the urban soils of Yibin City were associated with multiple factors including road density, location of major traffic roads, types of industries, and geomorphology of study area.

The spatial distribution of As was different from the other heavy metals. The spatial distribution pattern of As concentrations presented less variability and As concentrations in the urban soils were comparable with background values, suggesting that As concentrations in the soils of Yibin City have not been affected by anthropogenic activities.

2.5 Heavy metal pollution assessment

The PI, calculated relative to the background values of heavy metals in the soils of Yibin City, varied greatly among the different heavy metals (Table 5). The PI value of As ranged from 0.24 to 1.93, with a mean value of 0.86. For parks, commercial areas, main roadside areas, residential areas and industrial areas, about 92.30%, 75.20%, 87.50%, 85.72% and 65.00%, respectively, of soil samples were classified as being at a low contamination level (Fig. 5), indicating no obvious As pollution in these urban soils. The PI value of Pb ranged from 0.66 to 7.24 and more than 95% of the soil samples were classed as being moderately or heavily contaminated with Pb. Furthermore, 8.33%, 14.29%, and 37.50% of soil samples in commercial areas, main roadside areas, and industrial areas, respectively, were heavily contaminated with Pb. The PI value of Zn varied from 0.42 to 4.19 with a mean value of 1.61 and about 80% of soil samples were classified as being moderately or

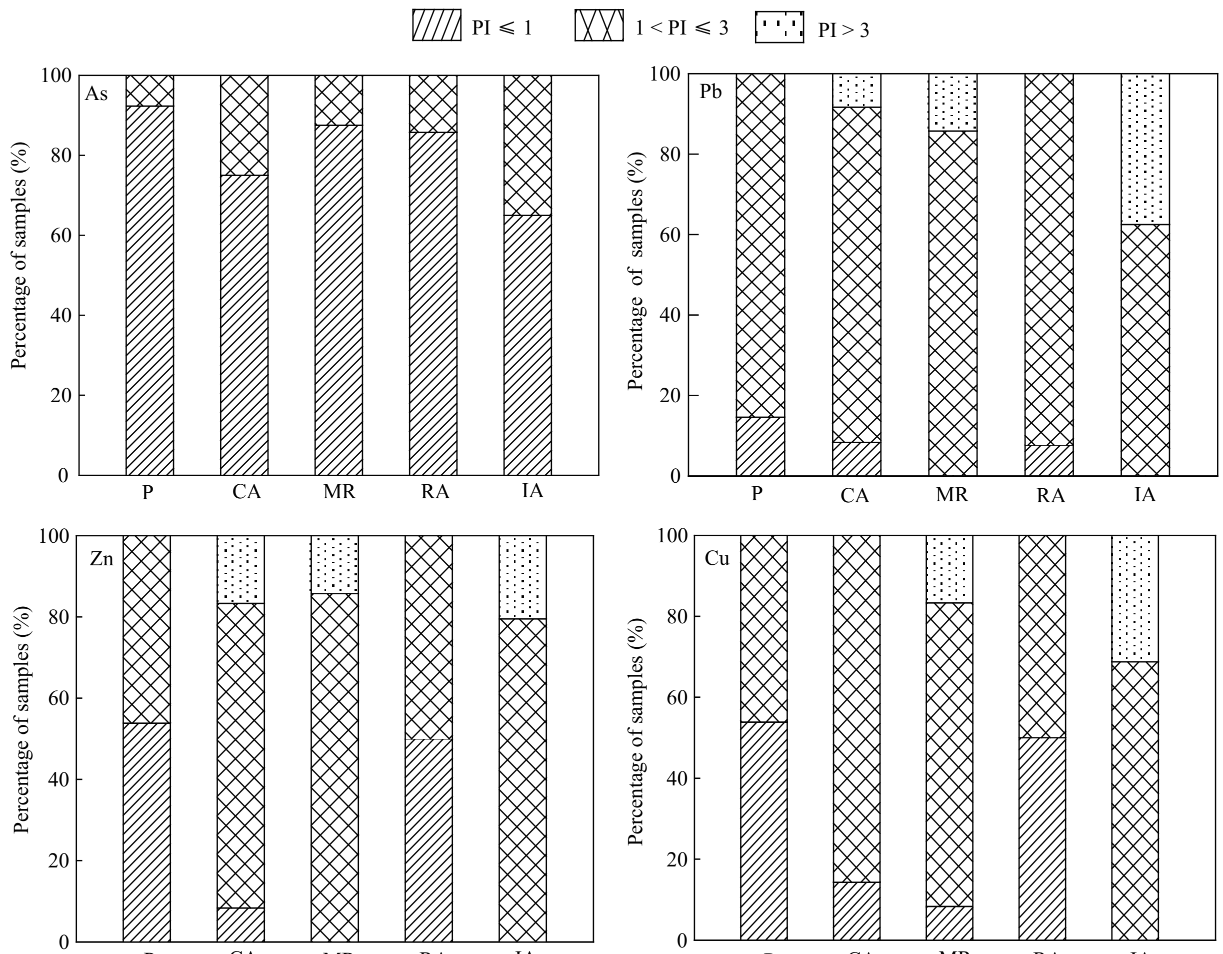

Fig. 5 Pollution characteristics of heavy metals in different functional areas of Yibin City. P, CA, MR, RA, IA refer to parks, commercial areas, main roadside areas, residential areas, and industrial areas, respectively. PI: pollution index.

Table 5 Pollution index (PI) of heavy metals in the urban soils of Yibin City

Heavy metal	PI		
	Min	Max	Mean
As	0.24	1.93	0.86
Pb	0.66	7.24	1.98
Zn	0.42	4.19	1.61
Cu	0.62	5.25	1.78

highly contaminated with Zn. For commercial areas, main roadside areas, and industrial areas, about 14.29%, 12.5%, and 16.67%, respectively, of soil samples were classified as being highly contaminated with Zn. The PI of Cu was in the range of 0.62–5.25, with a mean value of 1.78, and approximately 87% of the samples were moderately or highly contaminated with Cu. For main roadside and industrial areas, about 16.67% and 31.25% of soil samples were heavily contaminated with Cu, respectively. Overall, Pb, Zn and Cu pollution in urban soils followed the decreasing order of industrial areas > main roadside areas > commercial areas > residential areas > parks, while As decreased in the order industrial areas > commercial areas > residential areas > main roadside areas > parks.

The IPI of all the analyzed samples varied from 0.82 to 3.54, with a mean of 1.61. The spatial distribution of IPI is presented in Fig. 6. Most of the urban soil samples from the older urban areas of Jiangbei and Cuiping Districts were identified as high or moderate contamination, which can be attributed to intensive anthropogenic activities and long-term accumulation of heavy metals. In contrast, relatively low heavy metal pollution levels were found in

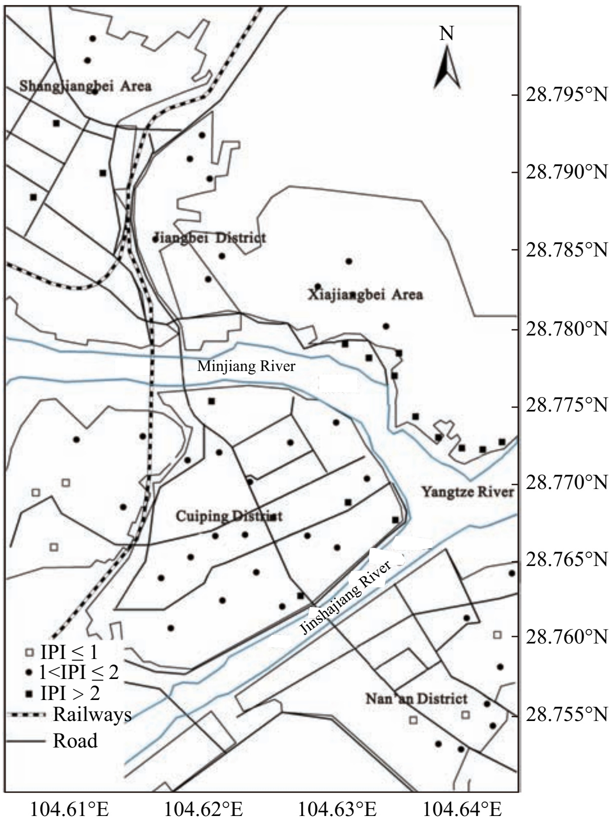

Fig. 6 Spatial distribution of integrated pollution index (IPI) of heavy metals in the urban soils of Yibin City.

the newer urban areas of Nan'an District. Therefore, the development history of urban areas may be an important factor determining the accumulation of heavy metals in urban soils.

About 22.22% of all samples were highly contaminated with heavy metals, with IPI values higher than 2. These highly contaminated samples were mainly located close to main roadsides and manufacturing plants such as power plants with coal-consumption, cement plants, and mechanical plants. Approximately 68.25% of soil samples showed moderate contamination with IPI values between 1 and 2. Only about 9.53% of all samples were classified as a low contamination level.

3 Conclusions

A total of 63 urban soil samples collected from five different functional areas in Yibin City were analyzed for As, Pb, Zn, and Cu. As concentrations in the soils were similar to background values, whereas Pb, Zn, and Cu concentration exceeded their corresponding background values. Results of combined multivariate statistical analyses and spatial distribution patterns of heavy metals indicated that industrial activities and vehicle emissions represented the most important sources of Pb, Zn, and Cu contamination, whereas As concentrations were dominated by soil parent materials.

Heavy metal contaminations in the urban soils were assessed using pollution and integrated pollution indexes, respectively. There was no obvious As contamination, but about 95%, 80%, and 87% of the soil samples were moderately or highly contaminated with Pb, Zn, and Cu, respectively. Soil samples in older urban areas exhibited moderate or high metal contamination due to intensive anthropogenic activities and long-term accumulation of heavy metals. In contrast, soil samples from newer urban areas displayed relatively lower pollution.

Acknowledgments

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