ORIGINAL ARTICLE

# Spatial distribution and contamination assessment of heavy metals in urban topsoil from inside the Xi'an second ringroad, NW China

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**Abstract** The paper reports the spatial distribution and contamination level of heavy metals (Co, Cr, Cu, Mn, Ni, Pb, Zn and V) in urban topsoil from the interior area of the second ringroad of Xi'an city, China, based on X-ray fluorescence spectroscopy measurements. Geostatistical analysis shows that Co, Cu, and Pb have similar spatial distribution patterns. Heavy traffic density mainly contributed to the high concentrations of Co, Cu and Pb. The spatial distribution of Cr coincides with the industrial activity, whereas the spatial distribution of Zn differs from other heavy metals. The high concentrations of Zn coincide with heavy traffic and high population density. For Mn, Ni and V, natural factors are important in controlling their distribution. The calculated geoaccumulation indices indicate that urban topsoil inside the Xi'an second ringroad was uncontaminated by Cr, V, Mn and Ni, while Pb, Cu, Co and Zn are classified as uncontaminated to moderately contaminated with means of 0.64, 0.46, 0.26 and 0.21, respectively. The Nemero synthesis pollution index of these heavy metals revealed that the topsoil inside Xi'an second ringroad has been heavily contaminated due to anthropogenic activity.

Keywords Heavy metal  $\cdot$  Soil  $\cdot$  Spatial distribution  $\cdot$  Contamination assessment  $\cdot$  GIS  $\cdot$  Xi'an

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#### Introduction

Soil, a crucial component of the urban ecosystem, is not only a transmitter of many pollutants to the atmosphere, groundwater and plants, but also a medium for plants to grow or a pool to dispose of undesirable materials (Chen et al. 1997). Nowadays, soil is one of the most important, but also endangered, parts of the environment, because many heavy metals can accumulate in topsoil from the atmosphere, depositing by sedimentation, impaction and interception (Sezgin et al. 2003), and posing a potential threat to human health and ecological systems (Chen et al. 2008). Several studies have indicated that accumulation of toxic metals in fatty tissue can affect the function of organs and disrupt the nervous system or the endocrine system (Batjargal et al. 2010; Shi et al. 2008). Some heavy metals can also interact directly with DNA to cause mutations, which can induce carcinogenesis if not treated immediately (Batjargal et al. 2010; Rico et al. 2009).

Heavy metals in urban soil have attracted a great deal of attention from governments, regulatory bodies and researchers concerned with reducing the risk of environmental pollution. According to the numerous studies, anthropogenic activities, such as motor vehicles (Chen et al. 2005; Lee et al. 2006; Madrid et al. 2002; Manta et al. 2002; Wei et al. 2009; Yang et al. 2011; Zhu et al. 2001), industrialization (Li et al. 2004; Manta et al. 2002; Morton-Bermea et al. 2009) and urbanization (Gallego et al. 2002; Raghunath et al. 1999; Shi et al. 2008), make a significant contribution to the accumulation of hazardous metal in urban soil (Batjargal et al. 2010; Morton-Bermea et al. 2009). To effectively decrease the risk of heavy metal pollution in urban areas and establish reliable protection approaches, it is necessary to understand spatial distribution patterns and contamination levels of heavy metals.

**Fig. 1** Sketch indicating sampling sites in Xi'an city



Studies of spatial distribution of heavy metals in urban soil contribute to identification of hot-spot areas and assessment of their potential sources (Imperato et al. 2003; Lee et al. 2006; Li et al. 2004; Morton-Bermea et al. 2009). Some research has been conducted in many areas to the study of the spatial distributions of heavy metals in urban soil based on GIS methods (Facchinelli et al. 2001; Li et al. 2004; Mielke et al. 2000; Norra et al. 2001; Romic and Romic 2003; Tao 1995). Contamination assessment of heavy metals in urban area is often established by comparing the heavy metal concentration with background values or by quantification in terms of an accumulation factor (for example pollution index) (Jung 2001).

Differences caused by soil heterogeneity and anthropogenic sources result in spatial variation of heavy metals concentrations, making it necessary to conduct studies in different areas. China, the largest developing country in the world, has undergone rapid economic development and urbanization during recent decades, resulting in serious environmental pollution. Several studies on heavy metal contamination in urban soil have been conducted in China (Chen et al. 1997; Duzgoren-Aydin et al. 2006; Shi et al. 2008; Wang and Qin 2006; Wang et al. 2006; Yang et al. 2011). Xi'an, the biggest city in northwest China, is experiencing rapid urbanization and industrialization. With the rapid increase in urban population in Xi'an city, pressures on the urban environment from human activities are more serious. Research on its urban soil environment is very necessary for sustainable development of Xi'an city. However, there is very little information available regarding heavy metal contamination of urban soil in Xi'an. Li and Feng (2010) reported heavy metals contamination in urban soil from the industrial district of Xi'an. To obtain the more information on the spatial distribution and contamination level of heavy metals in Xi'an, the objectives of this study were: (1) to determine the concentrations of heavy metals in urban topsoil collected from inside the Xi'an second ringroad; (2) to assess the contamination levels of heavy metals in the urban topsoil based on the geoaccumulation index ( $I_{geo}$ ) and the Nemero synthesis pollution index (NPI); and (3) to characterize the spatial distribution of heavy metals in urban topsoil from inside the Xi'an second ringroad.

### Materials and methods

### Overview of study area

Xi'an city, the capital of Shaanxi province, is located in the middle of Wei River valley (107°40'-109°49'E and  $33^{\circ}39'-34^{\circ}45'N$ ) (Fig. 1). The city, over 800 m above sea level, is bounded by Qinling Mountain on its south and east and by Loess Plateau at the north. The annual average temperature is 13–15 °C, and the average annual rainfall is 500-700 mm. The prevailing wind direction is northeast in winter and autumn, but southwest in summer and spring (XAMBS 2010). The main soil type is cinnamon (Song 1988). Xi'an, the largest city and the important center of economy, education, culture, manufacturing and high-tech industries in northwestern China, has an area of 3,580 km<sup>2</sup> and an urban population in excess of 6,470,000 in 2009. The growth rate of gross domestic product (GDP) was 14.5 % in 2010. There were more than 1.12 million motor vehicles in Xi'an in 2010, and this number has been increasing by 26.1 % per year.

### Soil sampling and experimental analysis

The soil samples were collected from inside Xi'an second ringroad with 3S technology. Soil sampling sites in the study area were chosen with a  $1,120 \times 1,120$  m grid using GIS software. Then, combining with Google Earth remote sensing images, coordinates of all sampling sites were defined. During field sampling, the concrete sampling sites were adjusted under actual field conditions, and the actual latitude and longitude coordinates of sampling sites were recorded simultaneously by GPS. Finally, the samples diagram was generated in ArcGIS software.

A total of 78 topsoil (0–20 cm) samples were collected from the interior of the Xi'an second ringroad (Fig. 1) during July to August 2010. At every sampling site, four sub-topsoil (0–20 cm) samples were collected from the four corners of a  $2 \times 2$  m grid using a stainless steel shovel and mixed to a composite sample of ~1.5 kg by a quartile method. The collected soil samples were stored in polyethylene bags for transport and storage. All soil samples were air-dried naturally in the laboratory at room temperature.

The air-dried soil samples were then sieved through a 0.9 mm nylon mesh to remove large debris, stones, and pebbles. Then, 100 g of each sieved soil sample was ground with a vibration mill until fine particles (<0.075 mm) were obtained. The concentrations of Co, Cr, Cu, Mn, Ni, Pb, Zn and V in these soil samples were measured by XRF (X-ray fluorescence spectrometry) (Lu et al. 2010). For quality assurance and control (QA/QC), duplicate samples and standard reference materials (GSD-12 and GSS1) (Lu et al. 2010), obtained from the Center of National Standard Reference Material of China, were prepared and analyzed using the same procedures. The analyzed precision, measured as relative standard deviation, was routinely 3–5 %. The quality control gave good precision (SD < 5 %).

Geostatistic methods and mapping of element concentration

Geostatistics, a tool for studying and predicting the spatial structure of geo-referenced variables, focus on spatial objects and spatial correlation (Chen et al. 2008). Developed originally to predict the probability distributions of ore grades for mining operations, geostatistics is now widely applied in many disciplines including soil science (Krasilnikov et al. 2008). It can be used to build the agrochemical distribution and to map the physicochemical properties of soil globally. To improve the significant level and avoid the distortion of the analysis results, a normal distribution of variables is often desirable in geostatistics analysis (Chen et al. 2008; Gallego et al. 2002; McGrath et al. 2004).

Kriging is based on the assumption that the parameter being interpolated can be treated as a regionalized variable (Xie et al. 2011). It is regarded as the best spatial covariance interpolation method, providing optimal interpolation (Chen et al. 2008). The kriging estimator is a linear combination of the observed values, suitably weighted (Xie et al. 2011). There are several types of kriging, including simple kriging, ordinary kriging, universal kriging, etc. Ordinary kriging is sufficiently robust for estimating the monitor values of an un-sampled area (Krasilnikov et al. 2008). It has been widely used in groundwater modeling, soil mapping and other fields. The weights of ordinary kriging are derived from the kriging equations using a semivariance function (Chen et al. 2008; Xie et al. 2011). The spatial distribution maps of all studied heavy metals in topsoil from inside Xi'an second ringroad were generated by the ordinary kriging interpolation method with ArcGIS software.

Methods of heavy metal pollution assessment

Contamination levels of all studied heavy metals in topsoil collected from inside Xi'an second ringroad were characterized by the geoaccumulation index ( $I_{geo}$ ) and Nemero synthesis pollution index (NPI). The  $I_{geo}$ , defined by Müller (1969), has been widely used to evaluate the intensity of historical heavy metal pollution or the contamination degree of soil heavy metals (Ji et al. 2008; Krishna and Govil 2008; Wei et al. 2009). It is expressed as

$$I_{\text{geo}} = \log_2(C_n/1.5B_n) \tag{1}$$

where  $C_n$  is the measured concentration of heavy metal *n* in the soil sample and  $B_n$  is the corresponding background value of heavy metal *n* in Shaanxi soil (CNEMC 1990). The following classification applies (Krishna and Govil 2008): uncontaminated ( $I_{\text{geo}} \le 0$ ), uncontaminated to moderately contaminated ( $0 < I_{\text{geo}} < 1$ ), moderately contaminated ( $1 < I_{\text{geo}} < 2$ ), moderately to heavily contaminated ( $2 < I_{\text{geo}} < 3$ ), heavily contaminated ( $3 < I_{\text{geo}} < 4$ ), heavily to extremely contaminated ( $4 < I_{\text{geo}} < 5$ ), extremely contaminated ( $5 < I_{\text{geo}}$ ).

The Nemero synthesis pollution index (NPI) was calculated (Cai et al. 2010; Chen et al. 2008; Li and Feng 2010; Zhang et al. 2009) by

$$NPI = \sqrt{\frac{\left(\frac{C_i}{S_i}\right)_{\max}^2 + \left(1/n\sum_{i=1}^n \frac{C_i}{S_i}\right)^2}{2}}$$
(2)

where  $C_i$  is the measured concentration of heavy metal *i* in the soil sample and  $S_i$  is background value of heavy metal *i*. In this study,  $S_i$  is the background content of element *i* in Shaanxi soil (CNEMC 1990). NPI  $\leq 0.7$  means that the soil is safe,  $0.7 < \text{NPI} \leq 1$  indicates that the soil is guard level,  $1 < \text{NPI} \leq 2$  corresponds to slightly polluted soil,  $2 < \text{NPI} \leq 3$  means that the soil is noderately polluted, and 3 < NPI signifies that the soil is heavily polluted by heavy metals (Chen et al. 2008; Li and Feng 2010).

Table 1	Summary	statistics	of heavy	/ metal	concentrations	in top	soil of Xi	'an and	reference	value	(mg	kg <sup>-1</sup>	)
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Element	Minimum	5 %	25 %	Median	Mean	75 %	90 %	95 %	Maximum	Reference value <sup>a</sup>
Co	14.5	15.6	16.8	18.5	19.3	20.9	23.1	24.1	38.9	10.6
Cr	71.0	72.8	75.4	77.9	81.1	82.2	94.2	99.2	130.9	62.5
Cu	27.2	29.7	34.5	39.4	54.3	47.2	62.4	84.3	792.0	21.4
Mn	518.8	619.7	655.0	671.7	671.5	688.2	712.5	720.2	780.3	557
Ni	23.7	30.8	33.5	34.8	34.5	35.6	37.1	37.4	39.1	28.8
Pb	26.0	28.5	38.9	45.4	59.7	60.4	80.2	123.1	506.5	21.4
Zn	68.6	72.8	84.3	104.3	186.2	141.7	196.0	250.1	4964.8	69.4
V	63.8	78.1	83.3	85.9	85.2	87.6	89.4	90.1	97.5	66.9

<sup>a</sup> CNEMC (1990)

# **Results and discussion**

### Descriptive statistics

The descriptive statistics for heavy metal concentrations in urban topsoil of the studied area, as well as background values of Shaanxi soil (CNEMC 1990), are shown in Table 1. The mean concentrations of Co, Cr, Cu, Mn, Ni, Pb, Zn and V in soil inside the Xi'an second ringroad are seen to be 19.3, 81.1, 54.3, 671.5, 34.5, 59.7, 186.2 and 85.2 mg kg<sup>-1</sup>, respectively, all of which are higher than the corresponding background values of Shaanxi soil. Cu, Pb and Zn in the soil samples are significantly higher than their background values in Shaanxi soil. The 75 % percentile values of Pb, Zn and Cu are 2.8, 2.0, 2.2 times the background values of Shaanxi soil, respectively, and the maximum concentrations in soil inside Xi'an's second ringroad are 37.0, 23.7 and 71.5 times the background values of Shaanxi soil, respectively. The large difference between the minimum and maximum Co, Cr, Cu, Pb and Zn concentrations, especially for Cu, Pb and Zn, shows large variations of these heavy metals concentrations in the study area, indicating that the main sources of these heavy metals are human activities.

Some elements in urban topsoil are essential for plant growth. However, the metals create toxicity to plants once their concentrations in topsoil exceed specific thresholds. In the study area, the concentrations of Pb, Zn and Cu in some sampling sites exceeded the grade III limit value of Chinese Soil Environmental Quality Standard (GB15618-1995) (SEPAC 1995), meaning that the normal growth of plants will be threatened if the concentrations of these heavy metals continue to increase. Therefore, it is necessary to carry out essential measures to prevent further enrichment of these heavy metals. Spatial distribution of heavy metals

To determine the spatial distribution of heavy metals, kriging interpolation was used in the study. Prior to geostatistical analyses, data transformation was carried out. The skewness, kurtosis and the significant level of Kolmogrov-Smirnov (K–S) test for normality are provided in Table 2. The kurtosis values of all heavy metals contents  $\gg 0$ , especially for Cu, Pb and Zn, indicating that the studied heavy metals inside the Xi'an second ringroad are strongly positively peaked, with some extremely high values in the data sets. Skewnesses values of Cu, Pb, Zn, Co and Cr > 1, show that these elements are positively skewed towards lower concentrations, as confirmed by the fact that their median concentrations are lower than the mean concentrations. Logarithmic transformation of the heavy metal concentration data reduced the skewness and kurtosis. However, the most log-transformed data did not pass the normality test (Table 2). Compared with logarithmic transformation, all the Box-Cox transformed data passed the normality test. Therefore, Box-Cox transformation was applied to the concentration of Co, Cr, Cu, Mn, Ni, Pb, Zn and V before interpolation. In the study, ordinary kriging was used to create spatial distribution maps of Co, Cr, Cu, Mn, Ni, Pb, Zn and V.

The spatial distributions of Co, Cu, Mn, Ni, Pb, Zn and V in the topsoil inside of Xi'an's second ringroad are represented in Fig. 2. It can be seen that the spatial distribution characteristics of Co is similar to those of Cu and Pb, i.e., the high concentrations of Co, Cu and Pb are found in the south area, and low concentrations are found in the middle of the north area. The hot-spot areas of Co, Cu and Pb are mainly associated with heavy traffic density. In spite of the wide use of unleaded petroleum since 2000 in Xi'an, Pb is not liable to transfer, resulting in accumulation in urban soil due to pollution from previous decades. Copper is often used in car lubricants. It can be released to the urban environment as a

sets of soil neavy metals									
Data set	Parameter	Со	Cr	Cu	Mn	Ni	Pb	Zn	V
Raw	Skewness	2.48	2.49	8.26	-0.91	-1.49	5.79	8.67	-1.35
	Kurtosis	11.54	8.26	70.90	4.72	5.48	39.50	76.02	6.40
	K–S p	0.40	0.00	0.00	0.18	0.07	0.00	0.00	0.47
Log transformation	Skewness	1.28	1.97	3.87	-1.39	-2.10	2.11	3.92	-1.90
	Kurtosis	3.78	4.87	21.65	6.35	9.00	6.84	23.03	9.31
	K–S p	0.58	0.00	0.01	0.12	0.03	0.05	0.03	0.33
Box-Cox transformation	Skewness	-0.05	-0.59	-0.86	-0.04	-0.26	-0.89	-0.46	-0.04
	Kurtosis	-0.38	-0.33	0.99	3.38	1.13	1.47	-0.24	2.96
	K–S p	0.64	0.14	0.67	0.21	0.39	0.51	0.88	0.75
	Lambda	-1.95	-4 86	-144	3 24	4 20	-0.72	-0.99	4 03

 Table 2
 Skewness, kurtosis and significance level of Kolmogrov–Smirnov (K–S) test of raw, log-transformed, and Box–Cox transformed data sets of soil heavy metals

result of wear of automobile oil pumps or corrosion of metal parts, which come into contact with the oil. Traffic is the major source of Co, Pb, and Cu contamination. This finding is consistent with some related research findings (Chen et al. 2010; Li et al. 2004; Surthland et al. 2000; Wilcke et al. 1998; Yang et al. 2011). With regard to Zn, several hotspots were identified in its spatial distribution map. One Zn hotspot is located in the northwest region of Xi'an second ringroad, near the intersection of the railway and main road, while another hot-spot located in the middle-south is associated with high population density. For the spatial distribution of Cr, the areas with higher concentration  $(>80 \text{ mg kg}^{-1})$  are found in the west and northeast of the study area, the industrial districts of Xi'an city. The spatial diversities of Mn, Ni and V in the study area are comparatively small, differing from the other five elements. Their concentrations in urban topsoil samples collected from inside the Xi'an second ringroad are close to, or slightly higher than, the corresponding background Shaanxi soil values. We can therefore conclude that the spatial distributions of Mn, Ni and V in the study area are mainly controlled by natural factors. The contamination assessment in the next section confirms this finding.

#### Results of geo-accumulation assessment

The calculated  $I_{\text{geo}}$  results for heavy metals in topsoil collected from inside the Xi'an second ringroad are presented in Table 3. Geoaccumulation index ranges from -0.13 to 1.29, with an average of 0.26 for Co, -0.40 to 0.48 with an average of -0.22 for Cr, -0.24 to 4.62 with an average of 0.46 for Cu, -0.69 to -0.10 with an average of -0.32 for Mn, -0.87 to -0.14 with an average of -0.33 for Ni, -0.30 to 3.98 with an average of 0.64 for Pb, -0.60 to 5.58 with an average of -0.24 for V. Geoaccumulation index for Mn, Ni and V in all studied soil samples and  $I_{\text{geo}}$  for Cr

in 88.5 % of the topsoil samples are less than zero showing that Mn, Ni, V and Cr are uncontaminated in topsoil inside Xi'an's second ringroad. The mean values of  $I_{geo}$  for Co, Cu, Pb and Zn, and 89.7 %  $I_{geo}$  values for Co, 75.6 % for Cu, 61.5 % for Pb and 41.0 % for Zn between zero and one reveal uncontaminated to moderately contaminated soil. As shown in Table 3, 90.0 % of  $I_{geo}$  values for Co, 85.9 % for Cu, 83.3 % for Pb and 50.0 % for Zn exceed zero, while 1.3 % of  $I_{geo}$  values for Co, 10.3 % for Cu, 21.8 % for Pb and 9.0 % for Zn exceed one. This shows that most topsoil samples collected from inside the Xi'an second ringroad contained Co, Cu, Pb and Zn contamination at different levels, with large spatial variations in pollution levels, especially for Cu, Pb and Zn (Table 3) confirm this finding.

## Integrated pollution index

The Nemero synthesis pollution index (NPI) of heavy metals in all topsoil sampling points was calculated from Eq. (2). The results are provided in Fig. 3. The NPI ranges from 1.3 to 51.1, with an average of 3.2. The NPI values of all soil samples are bigger than one, suggesting that urban topsoil inside Xi'an's second ringroad experienced different extents of heavy metal contamination. The mean value of NPI and NPI for 11 sampling sites (14.1 %) exceeded three, indicating that the soil is heavily polluted with heavy metals, while the NPI in 43 sampling sites (55.1 %) were between one and two, and in 24 sampling sites (30.8 %) between two and three, indicating that the soil is lightly or moderately polluted with heavy metals. Spatial differences of NPI for heavy metals in urban topsoil inside the Xi'an second ringroad are shown in Fig. 3. It is seen that the midwest of the study area is heavily contaminated with heavy metals. About 45 % of the areas have heavy pollution, 47 % area moderate pollution, and only 8 % area slight pollution. The heavily polluted areas are mainly





located at the heavy traffic sites, industrial areas and the old residential area with dense population, whereas the slightly polluted areas correspond to the new urbanization region. Therefore, the pollution spatial distribution can be attributed to urbanization, industrialization and accumulation of heavy metals.

#### Fig. 2 continued





Table 3 The calculated results of  $I_{geo}$  and the samples percentage of different  $I_{geo}$  values

Elements	Со	Cr	Cu	Mn	Ni	Pb	Zn	V
I <sub>geo</sub>								
Minimum	-0.13	-0.40	-0.24	-0.69	-0.87	-0.30	-0.60	-0.65
Maximum	1.29	0.48	4.62	-0.10	-0.14	3.98	5.58	-0.04
Mean	0.26	-0.22	0.46	-0.32	-0.33	0.64	0.21	-0.24
SD	0.23	0.16	0.65	0.08	0.10	0.71	0.82	0.08
Samples (%)								
$I_{geo} \leq 0$	9.0	88.5	14.1	100	100	16.7	50.0	100
$0 < I_{\text{geo}} < 1$	89.7	11.5	75.6	0	0	61.5	41.0	0
$1 < I_{\text{geo}} < 2$	1.3	0	7.7	0	0	18.0	7.7	0
$2 < I_{\text{geo}} < 3$	0	0	1.3	0	0	2.5	0	0
$3 < I_{\text{geo}} < 4$	0	0	0	0	0	1.3	0	0
$4 < I_{\text{geo}} < 5$	0	0	1.3	0	0	0	0	0
$5 < I_{\rm geo}$	0	0	0	0	0	0	1.3	0

SD standard deviation

**Fig. 3** Spatial distribution of the NPI of heavy metals in the studied area



## Conclusions

The concentrations, spatial distribution and contamination by Co, Cr, Cu, Pb, Zn, Mn, Ni and V in urban topsoil from inside the Xi'an second ringroad in NW China have been studied. The concentrations of all studied heavy metals are higher than their background levels in Shaanxi soil, especially for Co, Cr, Cu, Pb, and Zn. Co, Cu and Pb have similar spatial distribution patterns. Heavy traffic mainly contributed to the high concentrations of Co, Cu and Pb. The spatial distribution of Cr concentrations coincides with industrial activity, while the spatial distribution of Zn differs from that of the other heavy metals. High concentrations of Zn coincide with areas of heavy traffic and high population density. Natural factors are important in the distributions of Mn, Ni, and V. The geoaccumulation index assessment shows that Mn, Ni, V and Cr in urban topsoil inside the Xi'an second ringroad are uncontaminated, while Pb, Cu, Zn and Co are contaminated to different levels. Comprehensive assessment results of heavy metals indicate that the urban topsoil collected inside the Xi'an second ringroad exhibited different levels of heavy metal contamination. Urbanization, industrialization and perennial accumulation of heavy metals are the key underlying factors governing heavy metal contamination in the urban topsoil inside the Xi'an second ringroad.

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