



Overview of trace metals in the urban soil of 31 metropolises in China



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ABSTRACT

This overview provides an up-to-date assessment of the trace metal contamination (As, Cd, Cr, Cu, Hg, Ni, Pb, Sb, Se, and Zn) in urban soils of 31 metropolises in China. This systematic soil geochemical survey summarizes the characteristics of trace metals in Chinese urban soils, including concentration, accumulation, spatial distribution, and major sources. Mercury was ranked first followed by Cd and Se in geo-accumulation among all of the contaminant metals in urban soils in China; this finding is likely due to the Hg and Se emissions from fossil fuels. However, the lack of studies on Se contamination in urban soils, not only in China but also in the rest of the world, implies that Se contamination may have been unobserved for a long time. Shanghai, Kunming, Shenyang, and Changsha may be some of the most heavily contaminated Chinese cities based on the concentrations, spatial dimensions, and associations among the contaminant metals. Numerous hotspots with high concentrations of metals were found in Changsha, Shanghai, and Shenyang, clearly indicating a significant contribution from both the metallurgical industry and smelt mining to the contamination of urban soils. Conversely, the levels of Sb, Cu, and Cd in Kunming originated from their naturally high geochemical background in soils. Heavy Se contamination was found in Guiyang and Taiyuan. The natural source of Se may be important in defining the pattern of pollution in Guiyang, whereas anthropogenic sources are likely more accurate than is the natural background in Taiyuan city. We review the existing limits and types of pollutants in the current soil guidelines and find that an international agreement on the range of the limits and the types of pollutants contained in the soil guidelines is urgently needed.

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1. Introduction

The scale and pace of urbanization and industrialization have rapidly increased since China underwent an overall reform 34 years ago. Statistics show that the number of cities in China grew from 122 in 1978 to 655 in 2011, whereas the urban population unpredictably rose from 17.9% of the total population in 1978 to 51.3% in 2011 (National Bureau of Statistics of China, 2011, 2012). Consequently, a steady rise in the emission of trace metal contamination in urban areas has been observed. The pattern of contamination is evident not only in local, highly concentrated sites in urban areas, but also in areas of low contamination that are widely dispersed throughout soil, atmosphere,

water, and plants in the Earth's critical zones. The contamination has resulted in the deterioration of the air, soil, and water quality in China (Chan and Yao, 2008; Cheng, 2003; Fu et al., 2012; Luo et al., 2012; Rose and Shea, 2007; Shao et al., 2006; Wong et al., 2006).

Urban areas are the most densely populated regions of the world because of their strong industrial and economic activities. Urban soil, which is strongly influenced by anthropogenic activities, differs greatly from natural soils (Bullock and Gregory, 2009) and receives a major proportion of trace metal emissions from industrial, commercial, and domestic activities. Urban soils continuously accumulate organic and inorganic pollutants from either localized or diffuse sources (Luo et al., 2012). The typical diffuse pattern of these contaminations and the proximity of urban soils to humans increase the risk of human exposure through inhalation, ingestion, or dermal contact (Abrahams, 2002; Ajmone-Marsan and Biasioli, 2010). Therefore, recognizing the spatial distribution and concentrations of toxic and potentially toxic elements in urban soils in China is vitally important.

Approximately 32 urban soil contamination and geochemical surveys or projects have been initiated in China since the 1990s, covering cities in areas such as the Yangtze River Delta and the Pearl River Delta in Eastern and Southern China, the Bohai Rim in the north, the

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Old Industrial Zone in the northeast, the Chengdu–Chongqing Plain in the west, and some cities in the northwest (Table 1). The methodological approaches employed in the majority of the studies show that the sampling schemes and analytical procedures used in these urban soil geochemical surveys vary (Luo et al., 2012). The sampling strategies can be divided into the following two categories based on their sampling patterns:

- (i) A random sampling pattern reflects different land use purposes or functional zones such as residential, commercial, and industrial areas. The soil samples are randomly collected from urban parks, green lands, and city roadsides (Cai et al., 2013; Chen et al., 2010a, 2010b; Guo et al., 2012; Liang et al., 2009; Lu and Bai, 2010; Lu et al., 2007; Sun et al., 2010; Wang and Qin, 2007; Zhang and Ke, 2004; Zhang et al., 2009; Zheng et al., 2008; Zhou et al., 2008). In most cases, the sampling points in this pattern are heterogeneously distributed throughout areas with serious contamination and do not cover the entire city area. However, trace metals usually accumulate in the urban topsoil and appear to be more concentrated in the central or older areas of cities because of their longer development history compared to the suburbs. The concentration of metals in urban soils obtained for this sampling pattern may not give an accurate overview of the concentration and distribution of trace metals in an entire city area.
- (ii) The systematic sampling pattern is also known as “grid or cell sampling” in China. The sampling points in studied areas are randomly distributed based on a regular grid of $n \times n$ km (usually 1×1 km), with each grid having at least one sampling point (Cheng et al., 2013; Lee et al., 2006; Li and Huang, 2007; Li et al., 2010, 2013; Yang et al., 2011). This sampling strategy is used to reduce the possibility of a sampling bias that may

result from collecting an unrepresentative average sample because of the high portion of subsamples from the same region. Several subsamples are taken and mixed thoroughly to obtain a bulk sample for the sampling site. The profile of the top 0 to 10 cm or 0 to 20 cm of soils at each sampling point is taken using sampling tools from the two sampling patterns (Table 1).

The trace elements determined in these studies are highly diverse in both type and number and vary from city to city and from project to project in the same city. Except for those shown by Fuzhou, Fujian Province, less than eight heavy metal elements (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) have been reported in all of the 32 existing urban soil geochemical surveys (Table 1). The elements most commonly studied in the 32 projects are Cu and Pb (29), Zn (26), Cd (22), and Cr (20); As (9), Hg (10), and Ni (15) received less attention.

These differences in the variability of the sampling schemes, analytical procedures, and areas covered hinder meaningful comparisons among these studies. An international agreement on the range of elements and standardized methodologies to produce comparable datasets in different locations is urgently needed (Ajmone-Marsan and Biasioli, 2010; Luo et al., 2012). Nevertheless, the distribution, dispersion, and geochemical characteristics of some trace metals in the urban soils of Chinese cities and many other cities around the world have been widely compared and discussed in recent reviews (Ajmone-Marsan and Biasioli, 2010; Luo et al., 2012; Wei and Yang, 2010).

Systematic geochemical mapping is the best method available for assessing and monitoring changes in the levels of chemical elements on the Earth's surface. As a result, in 1999 China launched a new soil geochemical mapping project called the National Multi-Purpose Regional Geochemical Survey (NMPRGS) (Li et al., 2014).

Table 1
Sampling methods and heavy elements found in the 32 existing urban soil geochemical surveys.

	No of samples	Sampling pattern	Sampling depth/cm	Determined elements							Reference	
				As	Cd	Cr	Cu	Hg	Ni	Pb		Zn
Baoji	10	Systematic	10									Li and Huang (2007)
Beijing	773	Random	20									Zheng et al. (2008)
	261	Systematic	20									Cheng et al. (2013)
	285	Systematic	20									Li et al. (2010)
	127	Random	20									Chen et al. (2010a), Xia et al. (2011)
Changchun	352	Systematic	20									Yang et al. (2011)
Changsha	112	Random	20									Zhou et al. (2008)
Chongqing	48	Random	20									Li et al. (2006)
Fuyang	286	Random	20									Zhang et al. (2009)
Fuzhou	179	Systematic	20									Chen (2008)
Guangzhou	40	Random	10									Lu et al. (2007)
	78	Random	20									Cai et al. (2013)
Guiyang	50	Random	5									Li et al. (2012)
Haerbin	23	Random	10									Meng et al. (2009)
Hangzhou	82	Random	10									Zhang and Ke (2004)
	182	Random	10									Lu and Bai (2010)
Hefei	169	Systematic	20									Yuan et al. (2010)
Hong Kong	48	Systematic	15									Li et al. (2004)
	594	Random	10									Li et al. (2001)
Lanzhou	88	Random	10									Zhao et al. (2010)
Nanjing	138	Random	20									Lu et al. (2003)
Shanghai	273	Random	10									Shi et al. (2008)
Shenyang	93	Systematic	15									Li et al. (2013)
	36	Random	5									Sun et al. (2010)
Shijiazhuang	220	Systematic	20									Cui et al. (2011)
Ürümqi	428	Systematic	20									Tan et al. (2012)
Xi'an	53	Random	15									Yin and Zhao (2006)
	78	Systematic	20									Chen et al. (2012)
Xiamen	20	Random										Liang et al. (2009)
Xuzhou	21	Random	10									Wang and Qin (2007)
Yibin	63	Random	5									Guo et al. (2012)
Zhengzhou	30	Random	20									Gu et al. (2009)
				9	22	20	29	10	15	29	26	

The sampling year of each city is listed in Table 2. The topsoils from 0 to 20 cm in depth were collected at a density of one sample/1 km² and at two samples/1 km² in urban areas. Deep soils at 150 to 200 cm in depth were collected at a density of one sample/4 km² in the NMPRGS to study any anthropogenic influences on the surface layers. A total of 52 elements (Ag, As, Au, B, Ba, Be, Bi, Br, Cd, Ce, Cl, Co, Cr, Cu, F, Ga, Ge, Hg, I, La, Li, Mn, Mo, N, Nb, Ni, P, Pb, Rb, S, Sb, Sc, Se, Sn, Sr, Th, Ti, Tl, U, V, W, Y, Zn, Zr, Si, Al, Fe, Mg, Ca, Na, K, and total C), organic carbon, and pH were measured for each soil sample using a multi-element analytical scheme (Zhang, 2005). A special analytical quality control method was designed and followed during the analysis (Ye, 2005). Up to 2012, an area of 1.65 million km² has been covered by the NMPRGS in Eastern China, which contains both urban and developed agricultural regions. More than 510,000 soil samples were taken, resulting in approximately 27.8 million analytical data points. The samples contained the 52 elements listed above and organic carbon and had various pH levels.

Several different types of Chinese cities exist, including provincial capital cities or the municipalities directly controlled by the central government, prefecture-level cities, and county-level cities. The provincial capital cities are the most-densely populated and have strong industrial and economic activities. The study areas include 31 provincial capital cities (Fig. 1). The 31 provincial capital cities had a total area of 15,196 km² and an urban population of 134.63 million, approximately 10% of the total national population, by the end of 2011 (National Bureau of Statistics of China, 2011). The urban population of each of Chongqing, Shanghai, and Beijing is more than 10 million people; Chengdu, Guangzhou, Nanjing, Shenyang, Tianjin, Wuhan, Xi'an, and Zhengzhou have populations ranging from 5 million to 10 million; Changchun, Changsha, Fuzhou, Guiyang, Haikou, Hangzhou, Harbin, Hefei, Huhhot, Jinan, Kunming, Lanzhou, Nanchang, Nanning, Shijiazhuang, Taiyuan, Ürümqi, and Xining have populations ranging from 1 million to 5 million; and Lhasa and Yinchuan have populations less than 1 million.

In this paper, the concentration, spatial distribution, and contamination levels of 10 trace elements (As, Cd, Cr, Cu, Hg, Ni, Pb, Se, Sb, and Zn) selected from the NMPRGS are compared to provide a consistent national geochemical overview of the contamination of trace metals in urban soils of 31 provincial capital cities in China.

2. Data processing

2.1. Concentration of metals

The average, median, maximum, and minimum values of the 10 trace metals in urban topsoils (0 cm to 20 cm) and deep soils (150 cm to 180 cm) were calculated for each city individually and all together. The national background soil values (NBSVs) (Chen et al., 1991) and the target and intervention values recommended by the Dutch Ministry of Housing, Spatial Planning, and Environment (VROM, 2000) are listed in Tables 2 and 3 to facilitate the assessment and comparison of concentration levels of these elements.

2.2. The geo-accumulation index and contamination classes

Identifying a single metal contaminant or the co-contamination of multiple metals in urban areas is often performed by comparing the metal concentration with established guidelines or by quantifying an accumulation index based on background values. The concentration of trace metals in urban soils at a certain depth is generally composed of the natural geologic signature of the substrate plus the metal concentration from natural signature, and exogenous variables from various sources of contamination. The concentration of a trace metal in urban deep soil may represent the metal's background value in all soils.

The geo-accumulation index (I_{geo}) values introduced by Muller (1969) were employed for different metals to obtain an overview of the trace metal contamination levels in the topsoil of each city.

The geo-accumulation indexes were calculated as in Eq. (1):

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

where C_n is the measured concentration of n -th element in the topsoil, and B_n is the geochemical background for the n -th element. In this study, the median concentrations of the different metals in all 1011 urban deep soil samples were computed to be the geochemical background values for calculating the I_{geo} values. The deep soil samples for each city were considered at a national scale or at a municipal administration scale. The constant 1.5 was introduced to compensate for possible variations in the background values caused by lithogenic variations. The different I_{geo} values for each trace metal are classed as follows: $I_{geo} \leq 0$, uncontaminated; $0 < I_{geo} \leq 1$, uncontaminated to moderately contaminated; $1 < I_{geo} \leq 2$, moderately contaminated; $2 < I_{geo} \leq 3$, moderately to heavily contaminated; $3 < I_{geo} \leq 4$, heavily contaminated; $4 < I_{geo} \leq 5$ heavily to extremely contaminated; and $I_{geo} \geq 5$, extremely contaminated. The results for the calculation of I_{geo} are summarized in Table 4.

2.3. Pollution indexes (PIs) and the integrated pollution index (IPI)

A PI for each metal and the Nemerow IPI (IPI_N) of metals were calculated using Eqs. (2) (Lee et al., 2006) and (3) (Luo et al., 2012) to assess the degree of metal pollution in a given city.

$$PI_i = C_i/T_i^i \quad (2)$$

$$IPI_N = \left[(IPI_{avg}^2 + IPI_{max}^2) / 2 \right]^{1/2} \quad (3)$$

where C_i is the concentration of a given metal in topsoil samples and T_i is the corresponding target concentration from the circular on target values and the intervention values for the established soil remediation (VROM, 2000). The PIs and IPI_N of trace metals in the urban soil of each city in China are summarized in Table 4.

The IPI_N is divided into five levels from none to heavy pollution to indicate the pollution degree. The relationships between IPI_N and the metal pollution level are as follows: $IPI_N \leq 0.7$, safe; $0.7 < IPI_N \leq 1.0$, precaution; $1.0 < IPI_N \leq 2.0$, slight pollution; $2.0 < IPI_N \leq 3.0$, moderate pollution; and $IPI_N \geq 3.0$, heavy pollution.

2.4. Concentration contour values chosen and their implications

Evaluating the degree of contamination in urban areas is often established by comparing the metal concentration with the guidelines (Jung, 2001). Studying the spatial distribution of heavy metals in urban soils is necessary for identifying hotspots and assessing the potential sources of pollutants (Imperato et al., 2003; Lee et al., 2006; Li et al., 2004; Morton-Bermea et al., 2009; Reimann and de Caritat, 2005).

The overall objective of contamination evaluation is the sustainable use of soil by maintaining a level of functionality consistent with current and intended use based on soil quality guidelines. However, minimal current legislation relates directly to urban soil quality and soil protection in China despite the limitations of agricultural soil quality as given in the Environmental Quality Standards for Soils [Chinese Environmental Protection Administration (CEPA, 1995)].

However, the Dutch Target and Intervention Values (VROM, 2000), the Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health [Canadian Council of Ministers of the Environment (CCME, 1999)], the Soil Guideline Values (SGVs) (Environment Agency, 2009c) and Regional Screening Levels (RSLs) for chemical

Table 2
Statistical summaries of trace metal concentrations in urban deep soil (150–180 cm) from Chinese cities ($\text{mg}\cdot\text{kg}^{-1}$).

	Sampling year	n	As				Cd				Cr				Cu			
			Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max
Beijing	2003	61	8.2	8.3	3.4	14.8	0.089	0.090	0.050	0.130	57	58	40	83	19.6	20.1	10.9	28.7
Changchun	2000	16	11.2	11.2	4.0	15.7	0.082	0.084	0.060	0.100	64	65	45	76	21.6	22.3	13.9	28.0
Changsha	2002	49	14.5	13.4	6.9	31.7	0.116	0.078	0.042	0.868	94	91	66	138	26.0	25.1	14.9	41.3
Chengdu	1999	25	12.7	13.3	3.2	19.4	0.133	0.120	0.099	0.220	81	81	60	102	29.6	30.7	18.1	40.5
Chongqing	2005	31	6.1	5.3	2.6	16.1	0.122	0.114	0.055	0.228	80	79	53	112	26.8	25.8	15.1	51.4
Fuzhou	2002	20	6.4	7.0	4.0	9.0	0.238	0.245	0.090	0.380	53	54	34	75	22.8	23.0	15.0	34.0
Guangzhou	2005	24	19.0	15.5	2.0	62.0	0.212	0.197	0.029	0.673	44	44	15	76	21.9	23.5	6.0	42.0
Guiyang	2007	20	28.2	22.9	14.9	64.5	0.427	0.275	0.057	1.970	109	103	68	177	60.7	48.4	24.5	118.0
Harbin	2005	16	7.9	8.9	2.7	13.7	0.075	0.080	0.048	0.118	49	54	23	65	17.1	20.5	4.0	25.1
Haikou	2004	11	4.6	5.1	2.0	7.0	0.064	0.039	0.021	0.196	70	70	26	108	16.8	19.9	7.7	25.9
Hangzhou	2004	66	5.8	4.7	2.8	20.0	0.120	0.111	0.055	0.374	66	57	40	127	19.5	16.0	7.1	84.3
Hefei	2002	9	11.9	12.9	7.9	13.8	0.063	0.065	0.041	0.083	80	83	65	86	26.9	27.6	21.3	29.2
Huhhot	2004	20	9.0	7.8	2.7	18.6	0.103	0.100	0.064	0.186	57	61	36	77	19.4	21.0	10.2	30.0
Jinan	2002	15	12.2	11.3	10.2	18.4	0.125	0.110	0.080	0.280	77	73	59	139	26.3	25.0	20.0	44.1
Kunming	2006	23	8.7	8.6	3.3	14.3	0.457	0.293	0.042	2.947	97	95	75	119	77.8	77.6	22.9	177.3
Lanzhou	2006	24	16.2	13.0	7.1	61.3	0.133	0.134	0.087	0.175	62	64	44	80	21.7	22.1	14.3	28.0
Lhasa	2007	9	20.5	19.2	10.6	34.4	0.123	0.120	0.100	0.150	42	42	34	56	21.6	20.6	17.2	28.8
Nanchang	2002	40	8.2	8.7	3.5	13.7	0.093	0.070	0.030	0.200	69	73	26	163	23.5	23.0	10.3	54.0
Nanjing	2001	72	10.7	11.0	4.0	28.9	0.162	0.140	0.036	0.470	83	83	39	128	34.4	30.7	16.1	208.0
Nanning	2006	48	18.0	13.7	3.9	60.6	0.114	0.080	0.024	0.740	73	75	40	97	24.1	22.8	9.4	41.6
Shanghai	2004	42	7.5	7.3	4.9	12.8	0.155	0.120	0.075	0.800	89	88	71	158	28.9	27.5	15.5	68.5
Shenyang	2004	35	12.0	9.0	4.3	136.0	0.184	0.110	0.059	1.440	74	74	48	100	39.8	27.2	16.7	430.0
Shijiazhuang	2004	16	10.2	10.8	3.0	14.1	0.086	0.090	0.050	0.110	63	63	38	74	20.1	20.6	8.9	24.3
Taiyuan	2002	32	10.2	10.5	5.5	13.1	0.124	0.110	0.080	0.360	68	68	53	80	22.5	20.8	17.2	45.4
Tianjin	2005	42	12.7	11.9	6.8	30.5	0.157	0.150	0.100	0.340	82	82	64	94	29.5	28.7	18.0	40.2
Ürümqi	2007	15	13.3	11.9	9.2	22.1	0.136	0.140	0.112	0.160	58	59	47	65	30.1	30.1	23.3	41.1
Wuhan	1999	56	12.3	12.4	5.2	20.1	0.144	0.095	0.030	0.590	90	89	62	117	34.1	32.3	24.9	53.8
Xi'an	2005	63	11.5	12.2	2.7	16.2	0.130	0.128	0.064	0.417	71	75	29	85	26.7	26.8	7.6	60.9
Xining	2007	9	11.6	11.6	10.6	12.5	0.151	0.140	0.130	0.210	74	74	65	86	24.5	23.4	19.5	37.1
Yinchuan	2006	48	10.8	11.0	4.3	16.1	0.115	0.115	0.054	0.179	57	61	32	70	19.2	19.9	7.5	28.5
Zhengzhou	2004	54	7.5	7.9	3.7	11.3	0.080	0.079	0.051	0.113	64	64	41	86	13.7	14.3	6.2	21.4
Mean of all cities		1011	11.1	10.0	2.0	136.0	0.141	0.110	0.021	2.947	73	73	15	177	26.9	24.3	4.0	430.0
NBSV ^a			11.2				0.097				61			23.0				

^a The national soil background (Chen et al., 1991).

contaminants (USEPA, 2012) have emphasized soil management legislation in the Netherlands, Canada, the UK and the USA, respectively. The Dutch soil guidelines consider different land-use types and are based on extensive studies of both the human and ecotoxicological effects of soil contaminants. The concentrations of metals in soils that are below the target values indicate sustainable soil quality, whereas concentrations above the target values indicate contamination. The intervention values generally indicate that the effect of contaminants on the soil environment is serious and requires soil remediation. The Dutch soil guidelines are used as the basis for the concentration contour values of all metals in this study, except Se, facilitating an evaluation of the degree of contamination and delineating the spatial distribution of metal contamination.

The concentration contour values of Se were selected from the Canadian Soil Quality Guidelines because Se is not listed in the Dutch Guidelines. Some metal concentration contour values were also taken from the SGVs based on an integrated analysis of their concentration in urban soils. At least three representative concentration contour values for each metal and their selection criteria are presented in Table 5.

3. Trace metal concentrations

3.1. Mercury

Hg is a persistent, toxic, and bio-accumulative heavy metal. Soil is enriched with Hg when the concentration is $\geq 0.100 \text{ mg}\cdot\text{kg}^{-1}$ (Connor and Schaklette, 1975; Gustin and Lindberg, 2000). The legislative limit for clean soils in the Dutch Guidelines considers $0.3 \text{ mg}\cdot\text{kg}^{-1}$

to be the optimum level for Hg (VROM, 2000). The SGV of Hg for residential areas in the UK is $1 \text{ mg}\cdot\text{kg}^{-1}$ (Environment Agency, 2009b), the same as the Environmental Quality Standard (GB 15618-1995) for agricultural soils given by the CEPA (1995). The Hg concentrations in urban soils are categorized into four groups. The first group has low levels of Hg in soils ($\text{Hg} \text{ mg}\cdot\text{kg}^{-1} < 0.1000 \text{ mg}\cdot\text{kg}^{-1}$), indicating sustainable soil quality. The second group has moderate levels of Hg ($0.100 \text{ mg}\cdot\text{kg}^{-1} \leq \text{Hg} < 0.300 \text{ mg}\cdot\text{kg}^{-1}$), indicating an enriched-Hg soil. The third group has high Hg concentrations ($0.300 \text{ mg}\cdot\text{kg}^{-1} \leq \text{Hg} < 1.00 \text{ mg}\cdot\text{kg}^{-1}$), indicating that the urban soils should be examined. The fourth group has Hg concentrations equal to or greater than $1.000 \text{ mg}\cdot\text{kg}^{-1}$, indicating that the soils may be a potential health risk.

The Hg concentrations in urban topsoils and deep soils in all of the investigated Chinese cities are presented in Tables 2 and 3. The average deep soil Hg concentrations in 14 of the 31 cities are significantly elevated compared to the NBSVs, whereas 17 cities have values lower than $0.065 \text{ mg}\cdot\text{kg}^{-1}$. The mean Hg concentration ranges from $0.018 \text{ mg}\cdot\text{kg}^{-1}$ to $0.210 \text{ mg}\cdot\text{kg}^{-1}$ in urban deep soils.

The urban topsoils in most cities, including Changchun, Changsha, Chongqing, Harbin, Haikou, Hefei, Jinan, Lhasa, Nanchang, Nanjing, Nanning, Shanghai, Shenyang, Shijiazhuang, Taijing, Taiyuan, and Wuhan, have moderate Hg levels ($0.100 \text{ mg}\cdot\text{kg}^{-1} \leq \text{Hg} < 0.300 \text{ mg}\cdot\text{kg}^{-1}$). Low concentrations of Hg ($< 0.100 \text{ mg}\cdot\text{kg}^{-1}$) in urban topsoil are present in Huhhot, Lanzhou, Ürümqi, Yinchuan, and Zhengzhou. High average Hg concentrations ($\geq 0.300 \text{ mg}\cdot\text{kg}^{-1}$) are found in Beijing, Fuzhou, Guangzhou, Guiyang, Hangzhou, Kunming, Shanghai, and Xi'an.

Hg				Ni				Pb				Sb				Se				Zn			
Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max
0.070	0.031	0.009	0.563	24.9	24.8	15.8	39.8	19.2	19.2	14.4	26.0	0.71	0.71	0.31	0.99	0.09	0.08	0.05	0.18	58	59	41	75
0.019	0.018	0.012	0.036	27.6	27.4	16.9	42.6	26.9	26.6	23.1	29.7	0.69	0.68	0.43	0.99	0.11	0.10	0.09	0.16	63	63	46	71
0.092	0.068	0.034	1.058	26.9	26.4	14.0	39.9	28.1	25.5	18.2	55.1	1.50	1.10	0.69	7.47	0.30	0.30	0.11	0.54	74	68	36	121
0.074	0.051	0.008	0.550	36.8	37.0	23.5	48.9	22.9	21.9	17.6	31.2	0.84	0.84	0.25	1.48	0.08	0.05	0.02	0.30	76	75	42	112
0.060	0.056	0.019	0.118	31.6	31.4	20.2	50.0	26.1	26.1	18.3	42.1	0.65	0.63	0.24	1.17	0.11	0.10	0.06	0.24	82	80	57	145
0.134	0.105	0.050	0.530	23.4	24.0	14.0	34.0	66.0	68.5	49.0	85.0	0.36	0.33	0.23	0.64	0.28	0.28	0.13	0.46	122	126	81	194
0.210	0.155	0.028	0.670	15.7	14.0	5.0	30.0	50.8	47.5	25.0	126.0	1.40	1.27	0.15	4.59	0.43	0.41	0.25	0.74	71	72	22	143
0.202	0.175	0.044	0.430	54.2	49.0	27.5	120.0	36.7	36.1	18.8	50.1	1.59	1.42	0.87	2.92	0.46	0.43	0.12	0.83	110	105	59	182
0.034	0.021	0.010	0.125	22.1	24.5	8.7	30.5	21.7	22.2	16.4	28.5	0.56	0.66	0.20	0.80	0.09	0.08	0.04	0.16	57	63	31	75
0.049	0.057	0.011	0.070	25.5	29.0	9.0	45.4	21.0	20.4	13.9	32.3	0.43	0.43	0.20	0.65	0.33	0.38	0.12	0.59	37	33	21	67
0.152	0.058	0.021	4.091	27.6	24.5	13.7	52.8	25.4	20.5	12.7	201.4	0.51	0.42	0.23	2.90	0.17	0.12	0.03	1.64	71	64	33	134
0.048	0.022	0.013	0.144	33.1	34.3	23.2	38.1	24.8	25.6	19.9	26.4	0.89	0.90	0.68	1.05	0.09	0.08	0.05	0.15	58	60	44	63
0.018	0.019	0.010	0.025	24.4	24.6	14.0	37.3	19.9	19.0	15.0	27.2	0.78	0.70	0.27	1.40	0.09	0.09	0.05	0.16	54	58	29	81
0.024	0.020	0.013	0.050	32.5	31.2	27.1	42.8	23.1	21.3	17.5	45.3	1.13	0.99	0.81	2.67	0.12	0.10	0.07	0.37	66	65	52	105
0.193	0.119	0.032	1.235	44.1	42.5	23.3	66.6	43.6	42.7	25.1	94.9	1.36	1.10	0.34	4.01	0.17	0.16	0.12	0.23	90	87	42	133
0.035	0.018	0.010	0.249	28.3	30.1	18.1	33.1	20.3	19.8	16.4	25.9	0.93	0.96	0.65	1.14	0.15	0.15	0.10	0.26	61	64	37	79
0.092	0.075	0.016	0.250	21.0	19.3	17.6	26.7	31.0	30.4	28.5	34.2	1.24	1.10	0.77	2.15	0.09	0.09	0.06	0.14	65	65	57	73
0.086	0.042	0.011	1.613	24.5	23.8	9.6	37.9	29.5	27.5	17.9	90.7	0.73	0.78	0.28	1.31	0.21	0.19	0.09	0.43	72	68	43	149
0.052	0.028	0.014	0.440	40.0	39.6	17.5	60.8	24.9	23.3	2.9	52.6	1.03	1.05	0.31	1.80	0.14	0.09	0.04	0.55	82	78	47	154
0.118	0.120	0.054	0.269	23.1	21.9	6.9	40.5	25.3	24.3	12.7	50.0	5.88	2.51	0.77	23.75	0.50	0.49	0.19	0.94	67	65	22	124
0.102	0.075	0.039	0.420	36.7	37.1	26.4	46.9	27.7	27.3	18.1	51.8	0.69	0.56	0.31	4.73	0.15	0.14	0.06	0.50	108	107	72	177
0.046	0.040	0.016	0.110	31.1	31.2	21.7	42.8	28.1	25.7	12.7	56.1	0.80	0.78	0.35	1.64	0.19	0.19	0.09	0.32	76	70	44	244
0.022	0.020	0.007	0.045	28.9	29.7	16.3	33.8	19.6	20.4	12.5	22.6	0.84	0.89	0.25	1.08	0.08	0.08	0.05	0.13	59	60	34	66
0.041	0.034	0.017	0.120	27.4	27.7	21.2	36.4	20.2	19.8	14.7	29.0	0.81	0.87	0.35	1.04	0.20	0.15	0.07	0.60	63	59	52	95
0.048	0.030	0.016	0.422	35.3	35.4	24.2	45.3	24.9	24.7	19.6	30.3	0.94	0.90	0.51	1.76	0.17	0.14	0.07	0.84	83	83	55	119
0.041	0.023	0.014	0.161	27.5	27.0	21.6	32.3	18.2	18.1	14.6	21.6	0.91	0.90	0.75	1.08	0.24	0.20	0.12	0.70	72	72	67	79
0.051	0.046	0.014	0.184	39.5	39.0	28.0	56.8	24.8	24.6	16.7	32.6	1.00	1.01	0.43	1.66	0.20	0.16	0.06	0.41	80	78	59	118
0.171	0.059	0.010	2.225	31.1	31.6	10.4	64.2	23.9	23.4	19.1	48.3	0.83	0.85	0.34	1.25	0.09	0.09	0.03	0.14	66	70	26	83
0.077	0.027	0.015	0.478	26.1	26.1	23.0	29.3	24.1	22.1	19.0	37.2	0.22	0.23	0.13	0.32	1.02	0.98	0.89	1.17	64	60	54	97
0.023	0.017	0.009	0.077	24.7	25.5	10.4	34.5	19.4	19.6	11.7	25.5	0.90	0.90	0.52	1.34	0.10	0.11	0.05	0.15	52	56	22	76
0.025	0.023	0.013	0.087	20.4	21.3	9.3	27.9	18.1	17.9	15.5	20.5	0.68	0.71	0.32	0.93	0.07	0.07	0.03	0.12	42	45	19	59
0.082	0.042	0.007	4.091	29.9	29.2	5.0	120.0	26.0	23.4	2.9	201.4	1.11	0.84	0.13	23.75	0.19	0.13	0.02	1.64	72	68	19	244
0.065			27.0					27.0				1.20				0.29				74			

On a national scale, the first groups that have a low Hg concentration in urban topsoils are mainly distributed in Northern and Northwestern China, as shown in Fig. 2. The second and third groups are located in Eastern and Southeastern China, where the more developed regions or ancient cities, such as Beijing and Xi'an, are located. These results imply that an anthropogenic impact is associated with the rapid development of industrial and urban regions in China.

3.2. Cadmium

Cadmium (Cd) is an extremely toxic metal and an environmental hazard. Tables 2 shows that the average Cd concentrations in deep soils of 23 cities range from 0.103 mg·kg⁻¹ to 0.457 mg·kg⁻¹, higher than the NBSV (0.097 mg·kg⁻¹) (Chen et al., 1991). Eight cities have Cd concentrations lower than 0.097 mg·kg⁻¹.

The guideline value for the Environmental Quality Standard for Soils (II) in China is 0.300 mg·kg⁻¹ (CEPA, 1995). The average Cd concentrations in the topsoils of Kunming, Shenyang, Changsha, Xi'an, Guiyang, Tianjin, Shanghai, Guangzhou, and Wuhan are over the limit (Table 3), and among those cities, two extremely high Cd concentrations are found in Shanghai (90.9 mg·kg⁻¹) and Kunming (69.9 mg·kg⁻¹).

The average Cd concentrations in topsoils in Kunming, Shenyang, Shanghai, and Changsha are 2.087, 1.161, 1.091, and 0.876 mg·kg⁻¹, respectively, which are each above 0.800 mg·kg⁻¹, the target value for soil remediation established by VR0M (2000). The lowest concentration of Cd in topsoils is found in Haikou and Huhhot, which both average below 0.100 mg·kg⁻¹, likely because the developmental stage of the

two cities is different from the other cities. Haikou and Huhhot were not developed until the 1990s, resulting in a relatively short metal accumulation process.

3.3. Lead

Pb is a major source of pollution in urban soils. The average Pb concentration in deep soils of the investigated cities is equal to the NBSV (27 mg·kg⁻¹) (Chen et al., 1991). The deep soils in Fuzhou and Guangzhou have an average Pb content of 66 and 50.8 mg·kg⁻¹, respectively, which may imply that these two cities have a high geochemical background of Pb in the soil-forming parent materials. Remarkably low Pb concentrations (<20 mg·kg⁻¹) were observed in Beijing, Huhhot, Ürümqi, Yinchuan, and Zhengzhou in this survey (Table 2).

The urban soils of all cities have an average Pb content of 45.3 mg·kg⁻¹ in topsoils (at 0 cm to 20 cm) (Table 3). Unexpectedly, 29 of the 31 cities have an average soil Pb content of <100 mg·kg⁻¹, which is remarkable compared to the 102 mg·kg⁻¹ average found in 34 European cities (Luo et al., 2012), the 231 mg·kg⁻¹ in Baltimore (Yesilonis et al., 2008), the 395 mg·kg⁻¹ in Chicago (Cannon and Horton, 2009) in the USA, the 140 mg·kg⁻¹ in Mexico (Morton-Bermea et al., 2009), the 208 mg·kg⁻¹ in Islamabad (Ali and Malik, 2011), the 122 mg·kg⁻¹ in Sialkot (Malik et al., 2010), Pakistan, the 262 mg·kg⁻¹ in Naples (Imperato et al., 2003), and the 253 mg·kg⁻¹ in Palermo (Manta et al., 2002). These observations imply that leaded gasoline was banned before the peak number of cars was reached, although vehicle emissions are considered to be the principal source of



Fig. 1. Schematic map of China.

Pb in soils in urban environments (Christoforidis and Stamatis, 2009; Feng et al., 2012; Li et al., 2004; Yang et al., 2011).

High average topsoil Pb concentrations of $115.8 \text{ mg}\cdot\text{kg}^{-1}$, ranging from $3.4 \text{ mg}\cdot\text{kg}^{-1}$ to $2055.0 \text{ mg}\cdot\text{kg}^{-1}$, in Shenyang and $103.6 \text{ mg}\cdot\text{kg}^{-1}$, ranging from $25.3 \text{ mg}\cdot\text{kg}^{-1}$ to $2521.0 \text{ mg}\cdot\text{kg}^{-1}$, in Shanghai exist and are significantly higher than the corresponding Pb content in deep soil (Table 2). The topsoil values exceed the target limit of $85 \text{ mg}\cdot\text{kg}^{-1}$ established for soil remediation (VROM, 2000). Shenyang was once a heavy industry base, and Shanghai has long been a center of industry in China. Therefore, Pb in the topsoil at Shenyang and Shanghai was affected by anthropogenic contributions.

3.4. Zinc

The average Zn concentrations in urban topsoils and deep soils are 112 and $72 \text{ mg}\cdot\text{kg}^{-1}$, respectively, indicating an elevated concentration in the topsoils. An extreme variability in the Zn concentrations in urban topsoils was found at $13 \text{ mg}\cdot\text{kg}^{-1}$ to $2180 \text{ mg}\cdot\text{kg}^{-1}$ (Table 3), whereas the Zn concentration ranges from $59 \text{ mg}\cdot\text{kg}^{-1}$ to $244 \text{ mg}\cdot\text{kg}^{-1}$ in deep soil (Table 2). Exceedingly high concentrations of Zn were observed in the 10 cities, each of which at least one sample has a concentration above $720 \text{ mg}\cdot\text{kg}^{-1}$, which is the intervention value proposed (VROM, 2000). Zn concentrations of $2180 \text{ mg}\cdot\text{kg}^{-1}$ in Wuhan, $1353 \text{ mg}\cdot\text{kg}^{-1}$ in Kunming, $1268 \text{ mg}\cdot\text{kg}^{-1}$ in Shenyang, and $1094 \text{ mg}\cdot\text{kg}^{-1}$ in Shanghai were observed, all of which require immediate remediation. The VROM proposed a Zn concentration of $140 \text{ mg}\cdot\text{kg}^{-1}$ as the limit of sustainable soil quality, but average Zn concentrations are higher than the Dutch limit in Shanghai ($244 \text{ mg}\cdot\text{kg}^{-1}$), Kunming ($205 \text{ mg}\cdot\text{kg}^{-1}$), Shenyang ($167 \text{ mg}\cdot\text{kg}^{-1}$), Guangzhou ($166 \text{ mg}\cdot\text{kg}^{-1}$), Tianjin ($155 \text{ mg}\cdot\text{kg}^{-1}$), Chengdu ($141 \text{ mg}\cdot\text{kg}^{-1}$), and Wuhan ($140 \text{ mg}\cdot\text{kg}^{-1}$), indicating that Zn contamination exists in these cities.

3.5. Copper

All cities in this study have an average Cu content of $38.2 \text{ mg}\cdot\text{kg}^{-1}$ in the topsoils (at 0 cm to 20 cm), which is remarkably lower than the $46 \text{ mg}\cdot\text{kg}^{-1}$ average found in 34 European cities (Luo et al., 2012), the $45 \text{ mg}\cdot\text{kg}^{-1}$ in Baltimore (Yesilonis et al., 2008), the $150 \text{ mg}\cdot\text{kg}^{-1}$ in Chicago (Cannon and Horton, 2009) in the USA, the $101 \text{ mg}\cdot\text{kg}^{-1}$ in Mexico (Morton-Bermea et al., 2009), the $74 \text{ mg}\cdot\text{kg}^{-1}$ in Naples (Imperato et al., 2003), and the $63 \text{ mg}\cdot\text{kg}^{-1}$ in Palermo (Manta et al., 2002).

The average Cu concentrations in the urban topsoils in Kunming ($126.7 \text{ mg}\cdot\text{kg}^{-1}$) and Shanghai ($63.8 \text{ mg}\cdot\text{kg}^{-1}$) are higher than the soil quality guideline value of $63 \text{ mg}\cdot\text{kg}^{-1}$ for residential areas or parklands (CCME, 1999). The Cu input from anthropogenic sources to the urban topsoils in Shanghai may be higher than that in Kunming because of low geochemical Cu background value of $28.9 \text{ mg}\cdot\text{kg}^{-1}$ in Shanghai compared to $77.8 \text{ mg}\cdot\text{kg}^{-1}$ in Kunming (Table 2).

In Wuhan, Xi'an, Changsha, Chengdu, Shanghai, Guiyang, Kunming, Guangzhou, Hefei, Nanjing, Shenyang, and Tianjin, each topsoil has an average Cu content that is greater than the soil target value of $36 \text{ mg}\cdot\text{kg}^{-1}$ established by VROM (2000). The other 19 cities have low Cu concentrations in urban topsoils (Table 3).

3.6. Selenium

Se, despite being an essential element in human and animal health, is toxic above certain levels. China has been considered a Se deficient country with a low NBSV ($0.29 \text{ mg}\cdot\text{kg}^{-1}$) for Se (Chen et al., 1991). Keshan disease, an endemic cardiomyopathy, occurs in a broad geographic belt with low Se concentrations, stretching from the Heilongjiang Province in the northeast to the Yunnan Province in the southwest (Tan, 1989; Tan et al., 1999). The mean Se concentration in the world soils is $0.40 \text{ mg}\cdot\text{kg}^{-1}$. However, no information is available regarding Se contamination in urban soils in Chinese cities,

implying that Se contamination in urban soils may have been overlooked for a long period of time.

Tables 2 and 3 show that the Se contents of the deep soils in cities have a range of $0.02 \text{ mg}\cdot\text{kg}^{-1}$ to $1.64 \text{ mg}\cdot\text{kg}^{-1}$, with a mean value of averages $0.19 \text{ mg}\cdot\text{kg}^{-1}$. The average is lower than the NBSV of $0.29 \text{ mg}\cdot\text{kg}^{-1}$ and the world mean Se value of $0.40 \text{ mg}\cdot\text{kg}^{-1}$. Twenty cities, including Beijing, Changchun, Chengdu, Chongqing, Hangzhou, Harbin, Hefei, Huhhot, Jinan, Kunming, Lanzhou, Lhasa, Nanjing, Shanghai, Shenyang, Shijiazhuang, Tianjin, Xi'an, Yinchuan, and Zhengzhou, have deep soil Se concentrations that are less than the average Se value of $0.19 \text{ mg}\cdot\text{kg}^{-1}$; whereas the mean Se content in the topsoil is $0.40 \text{ mg}\cdot\text{kg}^{-1}$, indicating an enrichment of Se in urban topsoil.

Two extremely high Se concentrations in topsoils were found in Wuhan ($10.80 \text{ mg}\cdot\text{kg}^{-1}$) and Taiyuan ($10.40 \text{ mg}\cdot\text{kg}^{-1}$). Fourteen cities, including Changsha, Chengdu, Guangzhou, Guiyang, Hefei, Jinan, Nanning, Shanghai, Shenyang, Shijiazhuang, Taiyuan, Tianjin, Wuhan, and Xining, have Se concentrations higher than the average value in topsoils ($0.4 \text{ mg}\cdot\text{kg}^{-1}$), confirming that China is a Se deficient country despite the enrichment of Se in the topsoils.

Among all elements, Se has one of the narrowest intake ranges between dietary deficiency ($<40 \mu\text{g}\cdot\text{day}^{-1}$) and toxic levels ($>400 \mu\text{g}\cdot\text{day}^{-1}$) (Fordyce, 2005). No recommended regulatory values for Se in urban soils in China exist, and therefore, the data were compared with the guideline value of $1 \text{ mg}\cdot\text{kg}^{-1}$ for residential areas and parkland provided by the Canadian Council of Ministers of the Environment (CCME, 1999).

The average Se value in all of the cities does not exceed the regulatory limits despite the evident enrichment of Se in the urban topsoil. However, the maximum Se value in the topsoil in Beijing, Changsha, Guiyang, Hangzhou, Hefei, Jinan, Kunming, Nanchang, Nanjing, Nanning, Shanghai, Shenyang, Shijiazhuang, Taiyuan, Tianjin, Wuhan, and Xining is more than the limit of $1 \text{ mg}\cdot\text{kg}^{-1}$, implying that some Se-contaminated areas exist in Chinese cities.

3.7. Antimony

Sb and its compounds are hazardous to human health and may be carcinogenic (Fowler and Goering, 1991; Gebel, 1997; Hammel et al., 2000). The United States Environmental Protection Agency (USEPA, 1979), the European Union (Filella et al., 2002), and many other countries (Ettler et al., 2007; Flynn et al., 2003; He and Yang, 1999; He et al., 2012; Maher, 2009; Wilson et al., 2010) also consider Sb to be a pollutant with priority interest. China plays an important role in global anthropogenic Sb emissions because the country is the highest producer of Sb (He et al., 2012) and the total atmospheric Sb emissions from coal combustion (Tian et al., 2011). The Sb concentration in urban soils in China is scarce except in Shanghai (Dai and Li, 2010), Dalian in the Liaoning Province, Xiamen in the Fujian Province, and Xuzhou in the Jiangsu Province (Wang and Qin, 2005). Average Sb contents in other areas of China were reported in a review paper by He et al. (2012), implying that Sb contamination in urban soils is of minimal concern.

The average Sb content in urban deep soils is $1.11 \text{ mg}\cdot\text{kg}^{-1}$, which is almost equivalent to the NBSV ($1.20 \text{ mg}\cdot\text{kg}^{-1}$). The highest average concentration of $5.88 \text{ mg}\cdot\text{kg}^{-1}$ and the maximum value of $23.75 \text{ mg}\cdot\text{kg}^{-1}$ of Sb in deep soils are found in Nanjing, Guangxi Province (Table 2), where very large, large, and medium Sb deposits account for 34.4% of the total Sb deposits in China, indicating areas with high Sb geochemical background. The lowest average Sb concentration of $0.22 \text{ mg}\cdot\text{kg}^{-1}$ was detected in Xining City.

The Sb levels in urban topsoils range from $0.05 \text{ mg}\cdot\text{kg}^{-1}$ to $73.20 \text{ mg}\cdot\text{kg}^{-1}$ (mean = $1.72 \text{ mg}\cdot\text{kg}^{-1}$), indicating a wide range of the Sb concentrations. The target and intervention values of Sb proposed by the Netherlands are 3.0 and $15.0 \text{ mg}\cdot\text{kg}^{-1}$ (VROM, 2000), respectively. The Sb guideline for agricultural areas, residential areas, and parklands in Canada is $20 \text{ mg}\cdot\text{kg}^{-1}$ (CCME, 1999). The average Sb

concentrations in the topsoils exceed the target value in Nanning, Shanghai, Changsha, Shenyang, and Kunming, whereas the maximum value is higher than the intervention value and the guidelines in Shenyang, Shanghai, Nanning, Guangzhou, and Kunming, showing the urban soils that are contaminated with Sb.

3.8. Nickel

The average Ni content in deep soils and topsoils is 29.9 and $29.2 \text{ mg}\cdot\text{kg}^{-1}$, respectively, both of which are slightly higher than the NBSV ($27 \text{ mg}\cdot\text{kg}^{-1}$). The range of the mean values in deep soils is $15.7 \text{ mg}\cdot\text{kg}^{-1}$ to $54.2 \text{ mg}\cdot\text{kg}^{-1}$ and is almost equivalent to the range of $15.9 \text{ mg}\cdot\text{kg}^{-1}$ to $50.8 \text{ mg}\cdot\text{kg}^{-1}$ in topsoils, indicating that the Ni content likely originated in natural parent materials.

The target and intervention values for Ni established by the Dutch are 35 and $210 \text{ mg}\cdot\text{kg}^{-1}$, respectively (VROM, 2000). Kunming, Guiyang, Chengdu, Wuhan, Nanjing, Shanghai, and Tianjin have an average Ni concentration that is higher than the target value, indicating that the soils should be cleaned. The remaining 24 cities have soil Ni levels at a sustainable level.

The maximum Ni concentrations of $348.9 \text{ mg}\cdot\text{kg}^{-1}$ and $157.6 \text{ mg}\cdot\text{kg}^{-1}$ were detected in the topsoil of Xi'an and Haikou, respectively. Both of these values exceed the SVG of $130 \text{ mg}\cdot\text{kg}^{-1}$ for residential soils given by the UK (Environment Agency, 2009c), indicating that these isolated sites have unsuitable levels of Ni for residential soils. Kunming is another city with an average Ni concentration that is higher than the guideline of $50 \text{ mg}\cdot\text{kg}^{-1}$ for agricultural, residential, park, commercial, and industrial lands as established by CCME (1999), indicating Ni contamination in the topsoils in Kunming.

3.9. Chromium

The Cr concentration in the topsoils was $76 \text{ mg}\cdot\text{kg}^{-1}$ on average with a range of $37 \text{ mg}\cdot\text{kg}^{-1}$ to $114 \text{ mg}\cdot\text{kg}^{-1}$, whereas an average of $73 \text{ mg}\cdot\text{kg}^{-1}$ was found in the deep soil with a range of $42 \text{ mg}\cdot\text{kg}^{-1}$ to $109 \text{ mg}\cdot\text{kg}^{-1}$, both of which are comparable with the corresponding background value of $61 \text{ mg}\cdot\text{kg}^{-1}$. The range of average concentrations in the deep soils is $42 \text{ mg}\cdot\text{kg}^{-1}$ to $109 \text{ mg}\cdot\text{kg}^{-1}$, whereas the range in the topsoils is $37 \text{ mg}\cdot\text{kg}^{-1}$ to $114 \text{ mg}\cdot\text{kg}^{-1}$, indicating an evident natural contribution.

For Cr, the target value for clean soil and the intervention value for soil remediation as established by VROM are $100 \text{ mg}\cdot\text{kg}^{-1}$ and $380 \text{ mg}\cdot\text{kg}^{-1}$ (VROM, 2000), respectively. An average Cr concentration that is higher than the target value was found in Kunming, Guiyang, Changsha, Jinan, and Shanghai, indicating that the soils should be cleaned. The remaining 26 cities have sustainable Cr soil quality levels. Extremely high Cr concentrations in urban topsoils were also reported in Shanghai, implying that the studied area needs to be promptly cleaned because of Cr contamination.

3.10. Arsenic

The negative effects of As on the environment and on public health have caused considerable global attention. The main source of As in soils is its parent material, but As accumulation in urban environments is most often attributed to fossil fuel combustion, particularly in coal, metal-processing industries, and mining activities (Ajmone-Marsan and Biasoli, 2010). Tian et al. (2010) estimated that in China, As emissions from coal combustion reached 635.57 t in 1998 and 2205.50 t in 2007 and have resulted in serious environmental pollution in the country. However, comprehensive and detailed studies on As in urban soils in Chinese cities are still limited. Of the 33 existing literature, only nine focused on cities in China (Chen, 2008; Cui et al., 2011; Guo et al., 2012; Li et al., 2006, 2013; Tan et al., 2012; Yang et al., 2011; Yin and Zhao, 2006; Zhou et al., 2008).

Table 3
Statistical summaries of trace metal concentrations in urban topsoil (0–20 cm) from Chinese cities ($\text{mg}\cdot\text{kg}^{-1}$).

	n	As				Cd				Cr				Cu			
		Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max
Beijing	261	7.7	7.6	4.4	17.3	0.193	0.168	0.083	0.902	58	57	46	129	31.4	27.8	12.8	164.5
Changchun	63	12.8	12.4	6.1	28.4	0.207	0.162	0.066	1.760	62	61	48	79	29.7	27.0	17.7	104.9
Changsha	196	19.8	16.3	8.5	84.1	0.876	0.509	0.160	6.663	108	100	69	250	38.5	35.3	17.3	116.5
Chengdu	100	13.4	12.1	6.1	27.4	0.231	0.190	0.110	0.710	80	80	59	143	52.2	45.6	31.5	221.0
Chongqing	120	5.8	4.9	1.1	18.0	0.278	0.284	0.063	0.650	75	71	47	137	29.9	28.6	14.5	92.4
Fuzhou	90	6.6	6.2	1.3	32.5	0.258	0.250	0.060	1.060	37	37	8	71	31.2	28.5	4.0	98.0
Guangzhou	86	18.8	18.0	2.3	37.4	0.344	0.307	0.045	0.928	55	52	17	95	48.0	38.5	9.0	378.0
Guiyang	80	26.2	22.5	7.2	65.1	0.495	0.420	0.140	2.010	112	113	62	155	56.0	53.7	19.7	128.0
Harbin	64	8.3	9.0	3.6	15.2	0.159	0.137	0.041	0.554	56	60	28	73	24.5	24.5	7.3	64.2
Haikou	44	3.7	3.5	1.3	9.8	0.088	0.069	0.023	0.259	73	46	17	309	20.2	15.3	3.5	64.9
Hangzhou	226	7.0	6.7	3.8	17.2	0.188	0.169	0.057	1.321	62	60	43	98	30.5	29.1	8.4	133.4
Hefei	36	11.1	10.9	8.4	18.5	0.188	0.168	0.069	0.540	72	71	61	112	37.4	34.0	25.5	85.7
Huhot	78	8.4	6.3	4.2	16.2	0.099	0.086	0.035	0.224	48	43	29	74	15.1	12.2	7.1	45.7
Jinan	64	11.2	11.4	4.4	16.3	0.191	0.175	0.060	0.720	108	80	56	740	31.2	28.5	15.9	66.7
Kunming	86	15.6	12.1	3.9	90.0	2.087	0.672	0.073	69.87	110	106	79	197	126.7	103.1	27.9	462.2
Lanzhou	80	11.5	12.3	3.8	15.4	0.263	0.258	0.089	0.478	64	66	24	91	28.3	27.1	8.8	76.9
Lhasa	36	20.7	18.6	12.5	36.5	0.128	0.120	0.079	0.210	42	39	28	59	22.7	22.1	14.9	34.0
Nanchang	160	9.5	9.0	3.4	26.1	0.186	0.150	0.059	0.750	69	71	21	117	28.5	26.5	9.9	146.0
Nanjing	287	10.8	10.2	5.3	42.9	0.253	0.210	0.080	2.220	82	79	29	188	42.2	38.4	9.6	278.0
Nanning	157	15.0	12.0	3.3	65.2	0.248	0.189	0.045	1.540	56	55	29	119	23.7	21.5	8.3	64.5
Shanghai	167	9.5	8.7	6.0	21.0	1.091	0.370	0.120	90.90	114	96	72	1341	63.8	47.9	21.2	420.0
Shenyang	140	12.8	10.4	4.8	160.0	1.161	0.560	0.110	30.40	80	78	63	129	58.2	45.6	21.3	586.0
Shijiazhuang	42	9.6	9.5	7.2	16.2	0.287	0.215	0.140	1.080	72	69	57	147	27.8	27.2	19.2	45.5
Taiyuan	128	9.9	10.3	4.9	20.6	0.181	0.170	0.092	0.640	74	70	56	158	33.1	25.5	17.7	750.0
Tianjin	132	11.2	11.1	6.7	21.3	0.413	0.281	0.105	5.392	93	84	65	346	50.1	42.6	21.5	253.8
Ürümqi	56	14.2	13.0	10.3	23.2	0.208	0.200	0.140	0.360	59	60	48	67	35.2	34.8	25.3	43.9
Wuhan	224	13.7	13.0	6.4	39.6	0.338	0.280	0.080	4.980	91	89	46	271	50.0	42.4	18.8	1440
Xi'an	192	11.2	11.5	5.8	18.9	0.501	0.281	0.149	3.900	77	77	55	174	37.6	34.1	14.1	96.1
Xining	28	12.0	12.2	9.1	14.8	0.241	0.175	0.140	0.700	59	58	46	75	28.6	25.5	18.2	100.2
Yinchuan	154	11.1	11.7	3.7	17.0	0.172	0.173	0.036	0.403	57	60	19	81	21.7	23.5	5.8	43.8
Zhengzhou	221	8.1	8.2	3.9	14.8	0.151	0.145	0.069	0.826	70	70	34	103	17.4	17.9	5.6	46.3
Mean of all cities ^a	3799	11.4	10.2	1.1	160.0	0.394	0.210	0.023	90.90	76	72	8	1341	38.2	31.1	3.5	1440
NBSV ^b		11.2				0.097				61				23.0			
Target value ^c		29				0.80				100				36			
Intervention value ^c		55				12				380				190			

^a Mean of all cities.

^b NBSV is the national background soil value in China (Chen et al., 1991).

^c Target and intervention values recommended by the Dutch Ministry of Housing, Spatial Planning and Environment (VROM, 2000).

^d The guideline value for agricultural, residential and parkland established by the Canadian Council of Ministers of the Environment (CCME, 1999).

Tables 2 and 3 show that similar to Cr and Ni, the mean As concentrations in urban topsoils and deep soils are 11.4 and 11.1 $\text{mg}\cdot\text{kg}^{-1}$ (Chen et al., 1991), respectively, both of which match the background value of 11.2 $\text{mg}\cdot\text{kg}^{-1}$. The highest average As content of 26.2 $\text{mg}\cdot\text{kg}^{-1}$ in topsoil is found in Guiyang, Guizhou Province, and the lowest mean, 3.7 $\text{mg}\cdot\text{kg}^{-1}$, is found in Haikou City.

The limits for the guideline values are set at 12 $\text{mg}\cdot\text{kg}^{-1}$ in Canada, and 32 $\text{mg}\cdot\text{kg}^{-1}$ for residential areas in the UK. However, 29 and 55 $\text{mg}\cdot\text{kg}^{-1}$ are the threshold and intervention values, respectively, for clean soils in the Netherlands.

The maximum value of 160.0 $\text{mg}\cdot\text{kg}^{-1}$ As in topsoils was found in Shenyang, but the mean is only 12.8 $\text{mg}\cdot\text{kg}^{-1}$, implying that at least one sampling point needs prompt cleaning of As contamination.

4. Distribution patterns and source identification

4.1. Mercury

The distribution of metal concentrations in urban soils as obtained using geochemical mapping facilitates a reliable estimation of the influence of contamination on the measured element concentrations (Reimann and de Caritat, 2005) and identifies hotspots with high metal concentration.

Figs. 3a, 4a and 5a show the distribution pattern of Hg in urban topsoils in Chengdu, Guangzhou and Xi'an, respectively. The downtown

areas of the cities are marked by extensive anomalies of Hg in topsoils and a decreasing trend from the center of the city to its suburbs is often observed.

The links between Hg contamination in urban topsoils and the geochemical background in deep soils are documented in Figs. 3 to 5. The maps demonstrate that the spatial distribution of Hg in deep soils deteriorates with the lower background levels of Hg in Chengdu (Fig. 3a vs. b). A similar distribution pattern is also present for Beijing, Fuzhou and Shanghai, indicating a remarkably increased Hg input to the topsoils from various anthropogenic sources. The distribution pattern of Hg in urban deep soils has a higher background level of Hg in Guangzhou (Fig. 4a vs. b), Guiyang, Kunming and Xi'an (Fig. 5a vs. b) than that in natural parent materials. This finding implies that the Hg contaminations in the urban topsoils of cities such as Guangzhou, Guiyang, Kunming and Xi'an may be originated from a high geochemical background and various anthropogenic sources.

Unexpectedly, the Hg in urban soils in Chinese cities, which has the largest I_{geo} mean value, can be classified as moderately contaminated (Table 4). Furthermore, the I_{geo} values of Hg in Beijing, Changchun, Chengdu, Harbin, Hefei, Jinan, Nanjing, Shenyang, Tianjin, and Xi'an range from 2 to 3, indicating a moderate to heavy Hg contamination in these cities. Urban soils in Changsha, Fuzhou, Guangzhou, Hangzhou, Kunming, Lanzhou, Nanchang, Shanghai, Shijiazhuang, Taiyuan, Wuhan, Ürümqi, Xining, Yinchuan, and Zhengzhou are classified as moderately contaminated with Hg. Chongqing, Guiyang, Haikou, and

Hg				Ni				Pb				Sb				Se				Zn			
Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max
0.406	0.282	0.020	3.312	24.1	24.0	16.8	43.2	32.8	30.1	16.8	100.2	0.97	0.87	0.42	3.75	0.26	0.23	0.07	5.26	90	84	48	289
0.133	0.087	0.029	1.434	25.2	25.0	23.0	28.0	32.8	30.5	23.8	86.6	1.11	0.98	0.53	3.98	0.26	0.23	0.14	0.80	88	79	60	240
0.212	0.180	0.057	0.950	27.4	25.6	16.8	109.0	48.5	40.9	26.0	216.4	3.34	2.86	1.18	14.13	0.26	0.23	0.25	1.66	133	108	63	512
0.669	0.330	0.110	15.40	38.5	38.0	29.2	49.2	57.0	46.0	26.0	311.0	1.24	1.14	0.58	4.13	0.47	0.42	0.16	0.81	141	125	89	517
0.118	0.103	0.026	0.503	30.3	29.7	18.1	71.1	33.0	31.3	21.0	85.0	0.80	0.74	0.23	3.44	0.40	0.38	0.06	0.73	104	95	56	330
0.517	0.290	0.010	4.730	15.9	15.7	4.1	24.8	71.5	66.0	20.0	353.0	0.73	0.58	0.05	2.52	0.29	0.25	0.03	0.79	129	121	20	404
0.531	0.406	0.052	2.444	19.5	17.9	6.0	35.1	86.5	73.5	29.0	464.0	2.80	2.16	0.37	26.93	0.38	0.34	0.24	0.92	166	128	25	826
0.372	0.260	0.092	5.59	40.4	41.0	16.5	71.8	53.0	47.9	23.6	168.0	2.56	2.26	0.75	8.82	0.54	0.52	0.36	2.89	126	120	54	224
0.151	0.098	0.011	0.871	23.0	24.6	11.2	29.2	32.7	29.1	17.1	119.5	0.88	0.89	0.26	1.80	0.99	0.92	0.04	0.44	83	83	36	171
0.125	0.060	0.003	0.54	33.3	18.2	5.2	157.6	19.7	17.3	6.6	73.9	0.46	0.40	0.18	2.82	0.23	0.24	0.06	0.73	56	44	13	189
0.324	0.248	0.027	3.549	25.3	25.0	14.0	38.0	38.3	36.1	15.5	196.4	0.90	0.79	0.41	7.44	0.33	0.32	0.07	1.55	102	92	45	655
0.161	0.097	0.010	1.092	27.5	27.4	21.3	39.7	40.5	34.4	24.2	90.8	1.32	1.09	0.63	4.11	0.37	0.37	0.14	3.54	104	93	54	279
0.017	0.015	0.005	0.037	19.3	16.3	10.4	35.8	16.6	16.2	10.1	23.0	0.69	0.59	0.42	1.46	0.67	0.46	0.04	0.22	45	38	24	78
0.177	0.110	0.016	1.857	30.6	29.6	22.2	45.0	36.7	31.5	12.6	115.9	1.46	1.15	0.37	7.09	0.11	0.08	0.11	1.51	97	83	51	505
0.385	0.254	0.070	2.981	50.8	46.9	28.0	99.6	76.1	60.8	30.3	335.1	3.13	1.98	0.69	22.50	0.40	0.35	0.13	4.14	205	155	59	1353
0.089	0.061	0.012	0.418	28.1	29.3	9.2	36.2	32.3	26.4	16.8	179.9	1.26	1.07	0.43	8.25	0.39	0.30	0.08	0.69	80	81	21	132
0.157	0.075	0.011	0.615	20.8	19.6	13.4	28.6	34.6	31.0	24.1	173.8	1.34	1.24	0.79	2.60	0.23	0.22	0.05	0.16	72	66	43	125
0.150	0.092	0.023	0.906	23.3	24.1	8.4	33.9	36.5	33.6	20.6	134.1	0.99	0.96	0.33	5.93	0.10	0.09	0.11	1.11	84	77	31	321
0.230	0.110	0.028	8.090	36.9	35.8	10.7	58.8	37.6	33.5	20.1	185.0	1.10	1.00	0.35	3.83	0.31	0.31	0.12	1.02	108	95	44	823
0.132	0.100	0.043	0.515	16.5	15.0	4.0	50.8	30.7	25.8	13.4	142.2	6.21	4.30	0.79	29.25	0.32	0.29	0.21	4.25	66	52	21	249
0.358	0.240	0.064	3.350	36.2	35.6	28.0	53.9	103.6	53.8	25.3	252.1	4.55	1.80	0.54	55.70	0.68	0.58	0.15	2.27	244	182	84	1094
0.269	0.205	0.041	1.930	34.1	33.3	25.0	59.1	115.8	68.5	3.4	2055	3.22	1.59	0.48	73.20	0.52	0.39	0.18	2.51	167	119	61	1268
0.114	0.104	0.052	0.220	28.1	27.7	23.7	41.1	32.9	31.4	20.9	59.5	1.18	1.03	0.74	3.96	0.46	0.40	0.36	1.27	113	96	66	384
0.123	0.094	0.027	0.85	29.5	28.4	20.5	75.8	28.7	25.9	14.2	60.2	1.04	0.95	0.47	7.96	0.63	0.58	0.19	10.4	89	78	58	337
0.294	0.188	0.042	1.889	35.8	34.1	26.2	96.9	58.5	37.4	21.1	2073	1.25	1.12	0.65	3.81	0.86	0.66	0.17	1.32	155	132	66	749
0.082	0.060	0.016	0.279	29.6	28.9	24.4	43.9	26.5	22.7	17.8	97.0	1.43	1.14	0.77	14.28	0.52	0.48	0.17	0.68	97	93	70	181
0.191	0.114	0.028	2.618	37.5	37.5	16.9	59.4	48.4	39.1	20.6	461.0	1.23	1.10	0.43	5.82	0.27	0.24	0.13	10.8	140	107	50	2180
0.411	0.234	0.023	2.600	33.5	32.8	18.3	348.9	42.3	35.0	22.6	258.8	1.16	1.05	0.53	11.00	0.53	0.37	0.11	0.66	112	96	53	740
0.081	0.064	0.010	0.302	26.8	27.4	21.3	33.9	28.5	26.2	18.6	66.5	0.23	0.22	0.12	0.46	0.27	0.24	0.61	2.30	79	74	51	141
0.055	0.040	0.008	0.461	26.0	27.6	7.9	37.7	21.2	21.2	13.0	46.0	0.95	1.00	0.41	1.51	0.90	0.86	0.05	0.36	61	67	17	123
0.069	0.053	0.010	0.555	21.0	21.9	8.5	39.1	22.1	21.1	15.9	47.3	0.82	0.84	0.30	1.50	0.19	0.20	0.07	0.51	54	55	23	112
0.245	0.134	0.003	15.40	29.2	28.3	4.0	348.9	45.3	34.0	3.4	2521	1.72	1.05	0.05	73.20	0.23	0.23	0.03	10.8	112	92	13	2180
0.065				27.0				27.0				1.20				0.29				74			
0.3				35				85				3.0				1.0 ^d				140			
10				210				530				15.0								720			

Lhasa are classified as uncontaminated to moderately contaminated because their I_{geo} values range from 0 to 1. The remaining two cities, Huhhot and Nanning, appear uncontaminated according to the definition by Muller (1969). These findings suggest that Hg contamination in urban soils is related to other trace metals, suggesting an increase in the anthropogenic inputs caused by rapid economic development.

Several studies have reported that the fossil fuel Hg emission in China is the largest anthropogenic Hg source (Feng, 2005; Jiang et al., 2006), which makes China the largest Hg emitter in the world (Pacyna et al., 2006, 2010; Pirrone et al., 2010; Streets et al., 2005; Tian et al., 2010; Wu et al., 2006). These Hg emissions from coal combustion may be deposited and accumulate in topsoil and are passed into urban areas by atmospheric deposition.

4.2. Cadmium

Table 5 shows the three concentration contours that represent the urban soil distribution of Cd: 0.30 mg·kg⁻¹, the maximum permissible concentration of Cd for agricultural soils in China (CEPA, 1995); 0.80 mg·kg⁻¹, the target values for clean soils (VROM, 2000); and 1.00 mg·kg⁻¹, the SVG values for Cd in soil (Environment Agency, 2009a). The estimated maps of Cd in the urban topsoils in Kunming, Shenyang, Changsha, and Xi'an are presented in Fig. 6.

Hotspots of high Cd concentration were identified using geochemical maps of the western part of the Shenyang Tiexi Industrial District

(Fig. 6a), which is the oldest and largest industrial zone in northeast China (Li et al., 2009). One of the largest metallurgical industries in China, the notorious Shenyang Smelting Plant was located in this area for 64 years until it was closed down by the government because of heavy pollution in 2000 (Sun et al., 2010). These findings suggest that the large-scale smelting operations greatly contributed to the urban soil contamination in the region.

In Xi'an, Cd has a spatial distribution pattern similar to that of Zn (not shown); both have contamination hotspots located between the second and third ring roads in the western section of the downtown area (Fig. 6b). Here, strong associations were found between these two metals, indicating that they originated from the same sources. Previous studies showed that Zn was mainly derived from industrial sources, traffic, and garbage in this area of city (Chen et al., 2012).

In contrast to the patterns observed in Shenyang and Xi'an, a high Cd concentration was found across the whole city of Changsha along the Xiangjiang River (Fig. 6c), upstream of which existed large-scale mining of Pb, Zn, and Cd metals (soil Cd mean concentration: 10.34 mg·kg⁻¹, maximum value: 219.9 mg·kg⁻¹) (Chen et al., 1999; Wei et al., 2009b; Zhang et al., 2012). This observation suggests that excess soil Cd was mainly derived from mining activities. Another area with a high Cd concentration is located west of Kunming and stretches from the northernmost area to the Dianchi Lake in the southernmost region (Fig. 6d). The high Cd concentrations in Kunming are likely due to the high geochemical Cd background level, whereas the low geochemical

Table 4
Geo-accumulation indexes (I_{geo}) and pollution indexes (PIs) of trace metals in urban soils of different cities in China.

	I_{geo}										PI												
	As	Cd	Cr	Cu	Hg	Ni	Pb	Sb	Se	Zn	As	Cd	Cr	Cu	Hg	Ni	Pb	Sb	Se	Zn	PI-ave	PI-max	IPI
Beijing	-0.69	0.51^a	-0.59	0.06	3.13	-0.63	0.19	-0.13	1.12	0.01	0.27	0.24	0.58	0.87	1.35	0.69	0.39	0.32	0.26	0.64	0.56	1.35	1.04
Changchun	-0.39	0.72	-0.66	-0.17	2.30	-0.70	-0.28	0.13	0.72	-0.11	0.44	0.26	0.62	0.82	0.44	0.72	0.39	0.37	0.26	0.63	0.49	0.82	0.68
Changsha	-0.02	2.90	-0.33	0.03	1.06	-0.53	0.34	1.02	0.08	0.38	0.68	1.09	1.08	1.07	0.71	0.78	0.57	1.11	0.47	0.95	0.85	1.11	0.99
Chengdu	-0.57	0.36	-0.61	0.18	3.13	-0.53	0.79	-0.03	2.32	0.32	0.46	0.29	0.80	1.45	2.23	1.10	0.67	0.41	0.40	1.01	0.88	2.23	1.70
Chongqing	-0.45	0.70	-0.66	-0.37	0.49	-0.64	-0.24	-0.22	0.91	-0.20	0.20	0.35	0.75	0.83	0.39	0.87	0.39	0.27	0.29	0.74	0.51	0.87	0.71
Fuzhou	-0.68	-0.51	-1.12	-0.15	1.71	-1.18	-0.52	0.55	-0.15	-0.55	0.23	0.32	0.37	0.87	1.72	0.45	0.84	0.24	0.38	0.92	0.63	1.72	1.30
Guangzhou	-0.31	0.22	-0.26	0.45	1.19	-0.11	0.28	0.56	-0.19	0.63	0.65	0.43	0.55	1.33	1.77	0.56	1.02	0.93	0.54	1.18	0.90	1.77	1.40
Guiyang	-0.39	0.26	-0.46	-0.37	0.50	-0.86	-0.03	0.27	0.63	-0.32	0.90	0.62	1.12	1.56	1.24	1.15	0.62	0.85	0.99	0.90	1.00	1.56	1.31
Harbin	-0.68	0.41	-0.52	-0.33	2.26	-0.67	-0.02	-0.16	0.94	-0.18	0.29	0.20	0.56	0.68	0.50	0.66	0.39	0.29	0.23	0.59	0.44	0.68	0.57
Haikou	-1.05	0.60	-0.54	-0.56	0.55	-0.38	-0.64	-0.48	-0.80	0.17	0.13	0.11	0.73	0.56	0.42	0.95	0.23	0.15	0.33	0.40	0.40	0.95	0.73
Hangzhou	0.00	0.18	-0.46	0.35	1.90	-0.54	0.31	0.53	1.03	0.09	0.24	0.24	0.62	0.85	1.08	0.72	0.45	0.30	0.37	0.73	0.56	1.08	0.86
Hefei	-0.80	0.94	-0.78	-0.15	2.29	-0.90	0.08	-0.03	2.56	0.21	0.38	0.23	0.72	1.04	0.54	0.79	0.48	0.44	0.67	0.74	0.60	1.04	0.85
Huhhot	-0.47	-0.59	-0.93	-1.06	-0.75	-0.94	-0.78	-0.60	-0.34	-0.97	0.29	0.12	0.48	0.42	0.06	0.55	0.19	0.23	0.11	0.32	0.28	0.55	0.44
Jinan	-0.60	0.21	-0.02	-0.27	2.56	-0.61	0.20	-0.03	1.41	0.00	0.39	0.24	1.08	0.87	0.59	0.87	0.43	0.49	0.40	0.69	0.60	1.08	0.87
Kunming	0.27	2.25	-0.37	0.12	1.11	-0.33	0.25	0.93	0.72	0.65	0.54	2.61	1.10	3.52	1.28	1.45	0.90	1.04	0.39	1.46	1.43	3.52	2.69
Lanzhou	-0.76	0.39	-0.59	-0.23	1.72	-0.68	0.12	-0.19	0.03	-0.25	0.40	0.33	0.64	0.78	0.30	0.80	0.38	0.42	0.23	0.57	0.49	0.80	0.66
Lhasa	-0.48	-0.49	-0.59	-0.45	0.48	-0.48	-0.40	-0.30	-0.50	-0.43	0.71	0.16	0.42	0.63	0.52	0.59	0.41	0.45	0.10	0.51	0.45	0.71	0.60
Nanchang	-0.45	0.82	-0.66	-0.27	1.25	-0.62	-0.18	-0.24	0.12	-0.29	0.33	0.23	0.69	0.79	0.50	0.66	0.43	0.33	0.31	0.60	0.49	0.79	0.66
Nanjing	-0.60	0.27	-0.60	-0.13	2.45	-0.68	0.11	-0.52	1.18	-0.12	0.37	0.32	0.82	1.17	0.77	1.06	0.44	0.37	0.32	0.77	0.64	1.17	0.94
Nanning	-0.45	1.04	-1.00	-0.53	-0.45	-0.99	-0.24	0.72	-0.12	-0.56	0.52	0.31	0.56	0.66	0.44	0.47	0.36	2.07	0.68	0.47	0.65	2.07	1.54
Shanghai	-0.21	2.60	-0.21	0.63	1.67	-0.62	1.34	2.45	1.36	0.61	0.33	1.36	1.14	1.77	1.19	1.03	1.22	1.52	0.52	1.74	1.18	1.77	1.51
Shenyang	-0.08	2.81	-0.47	0.51	2.16	-0.46	1.59	1.46	0.70	0.68	0.44	1.45	0.80	1.62	0.90	0.97	1.36	1.07	0.46	1.20	1.03	1.62	1.36
Shijiazhuang	-0.75	1.09	-0.40	-0.15	1.93	-0.66	0.11	-0.17	2.49	0.31	0.33	0.36	0.72	0.77	0.38	0.80	0.39	0.39	0.63	0.80	0.56	0.80	0.69
Taiyuan	-0.66	0.13	-0.46	0.09	1.27	-0.50	-0.05	-0.32	1.99	0.00	0.34	0.23	0.74	0.92	0.41	0.84	0.34	0.35	0.86	0.64	0.57	0.92	0.76
Tianjin	-0.67	0.88	-0.41	0.22	2.71	-0.57	0.66	-0.11	1.31	0.32	0.38	0.52	0.93	1.39	0.98	1.02	0.69	0.42	0.52	1.11	0.80	1.39	1.13
Ürümqi	-0.33	-0.01	-0.58	-0.36	1.25	-0.45	-0.04	0.08	-0.17	-0.15	0.49	0.26	0.59	0.98	0.27	0.84	0.31	0.48	0.27	0.69	0.52	0.98	0.78
Wuhan	-0.44	1.25	-0.56	0.04	1.47	-0.64	0.39	-0.30	1.14	0.26	0.47	0.42	0.91	1.39	0.64	1.07	0.57	0.41	0.53	1.00	0.74	1.39	1.11
Xi'an	-0.70	1.38	-0.55	-0.10	2.22	-0.50	0.27	-0.13	1.03	0.11	0.39	0.63	0.77	1.04	1.37	0.96	0.50	0.39	0.27	0.80	0.71	1.37	1.09
Xining	-0.53	0.20	-0.92	-0.30	1.00	-0.54	-0.22	-0.58	-0.71	-0.19	0.41	0.30	0.59	0.79	0.27	0.77	0.34	0.08	0.90	0.56	0.50	0.90	0.73
Yinchuan	-0.57	0.00	-0.68	-0.46	1.11	-0.56	-0.47	-0.51	0.31	-0.46	0.38	0.22	0.57	0.60	0.18	0.74	0.25	0.32	0.19	0.43	0.39	0.74	0.59
Zhengzhou	-0.56	0.35	-0.47	-0.30	1.00	-0.60	-0.28	-0.37	1.19	-0.30	0.28	0.19	0.70	0.48	0.23	0.60	0.26	0.27	0.23	0.39	0.36	0.70	0.55
Mean	-0.39	1.26	-0.53	0.07	1.96	-0.59	0.37	0.45	1.05	0.13	0.39	0.49	0.76	1.06	0.82	0.83	0.53	0.57	0.40	0.80	0.67	1.06	0.89
Max	0.27	2.90	-0.02	0.63	3.13	-0.11	1.59	2.45	2.56	0.68													

^a Number in bold represents a contamination metal in corresponding city.

Table 5

The concentration contour values of trace metals and the corresponding limits to legislation.

	Concentration contour value (mg·kg ⁻¹)		
	12 ^a	29 ^b	55 ^c
As	12 ^a	29 ^b	55 ^c
Cd	0.3 ^d	0.8 ^b	1.8 ^e
Cr	100 ^b	200 ^d	380 ^c
Cu	36 ^b	63 ^a	190 ^c
Hg	0.1	0.3 ^b	1.0 ^e
Ni	35 ^b	130 ^e	210 ^c
Pb	85 ^b	140 ^a	530 ^c
Sb	3.0 ^b	15 ^c	20 ^a
Se	0.4		1.0 ^a
Zn	140 ^b	360 ^a	720 ^c

^a Limit to residential/parkland or commercial land listed in the Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health (CCME, 1999).

^b Target value given by the Dutch soil guidelines (VROM, 2000).

^c Intervention values given by the Dutch soil guidelines Environmental Quality Standards for Soils (VROM, 2000).

^d Limits to agricultural soil given in its Environmental Quality Standards for Soils in China (CEPA, 1995).

^e Limit to the residential or allotment land listed in SVG (Environment Agency, 2009a, 2009b, 2009c).

Cd background values in Changsha, Xi'an, and Shenyang (Table 2) indicate that the urban soil contamination in these areas is more likely caused by other anthropogenic sources. This observation refines and confirms the results of the spatial pattern analysis.

The *I*_{geo} values of Cd in urban soils of the 31 cities vary from -0.59 to 2.90 (Table 4). Changsha, Shanghai, Shenyang, and Kunming show moderate to heavy Cd contamination. Nanning, Shijiazhuang, Wuhan, and Xi'an have *I*_{geo} values ranging from 1.04 to 1.38 and are classified as moderately contaminated. The *I*_{geo} values in Fuzhou, Huhhot, Lhasa, Ürümqi, and Yinchuan, however, are less than 0, indicating no Cd contamination.

Statistics show that urban soils of 260 km² (Table 6), approximately 1.7% of the total urban area, exceeded the limit for Cd soil allotment

(Environment Agency, 2009a). However, Shenyang, Changsha, Kunming, and Xi'an have a targeted value for urban soil area of 68, 56, 56, and 32 km², respectively, accounting for 81.5% of the total allotted area of soil. This observation indicates that the Cd contamination of urban soils in Chinese cities is primarily distributed in Shenyang, Changsha, Kunming, and Xi'an.

4.3. Lead

Before the 1980s, industrial factories, such as those for machinery, metal casting, smelting, textile processing, and the chemical industry, were located in the older and more central urban areas in China. These factories, which operated on the long-term scale of industrial production, contributed to the major pollution of the soils around them. However, with the continuous growth of the economy and rapid urbanization, many of these plants have been closed or moved to other locations because of serious pollution issues or changes in land use by the local government. The heavy metal pollution in the soils where these industrial factories and plants once stood remains an unresolved problem.

Fig. 7 shows the high Pb concentrations in the identified hotspots which were caused by an old smelting plant in Shenyang. In the central area of the old urban regions in Shanghai, Pb contamination in urban soils was caused by the long-term industrial activities. Therefore, these findings confirm that the primary source of Pb contamination is either old factory sites or the long-term industrial activities in the area.

The average *I*_{geo} value for Pb in urban topsoils is 0.37, which is remarkably less than those of Hg, Cd, and Se. Shanghai and Shenyang, with *I*_{geo} values >1, are classified as moderately contaminated. All of the other cities have an *I*_{geo} value <1 (Table 4), which indicates that the Pb contamination in urban soils may not be as serious as expected.

Overall, approximately 2.4% of total urban areas, equivalent to 364 km², exceeded the guideline value of 140 mg·kg⁻¹ for residential

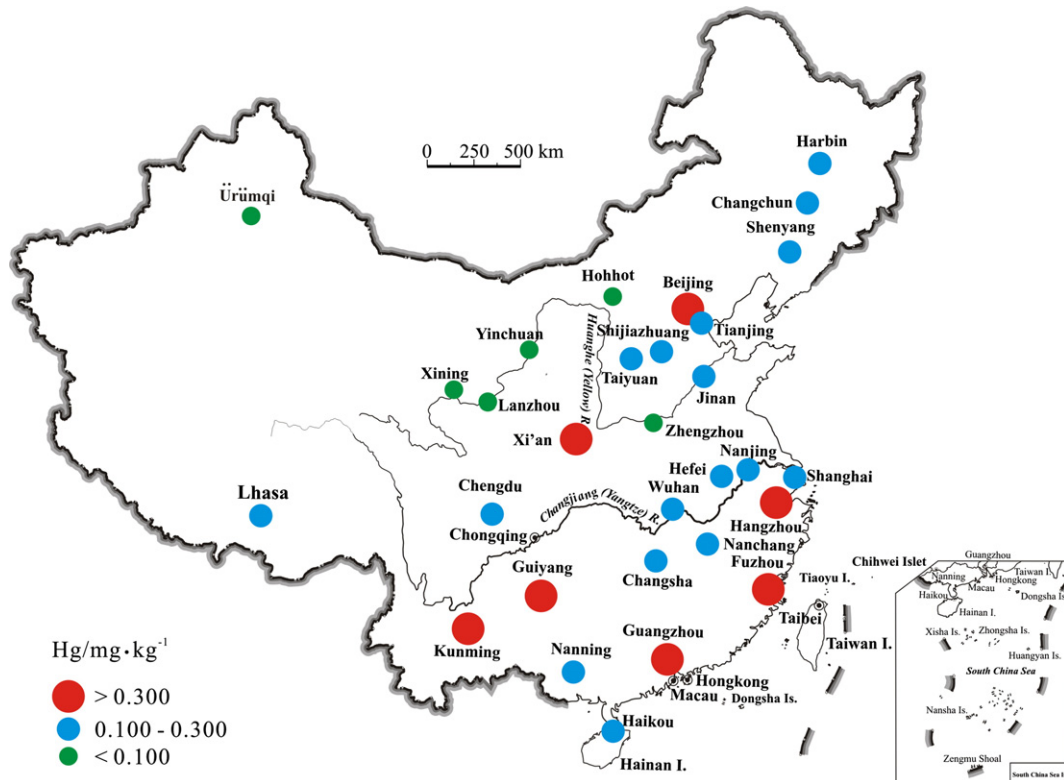


Fig. 2. Distribution of the average Hg concentration in the urban topsoils of different Chinese cities.

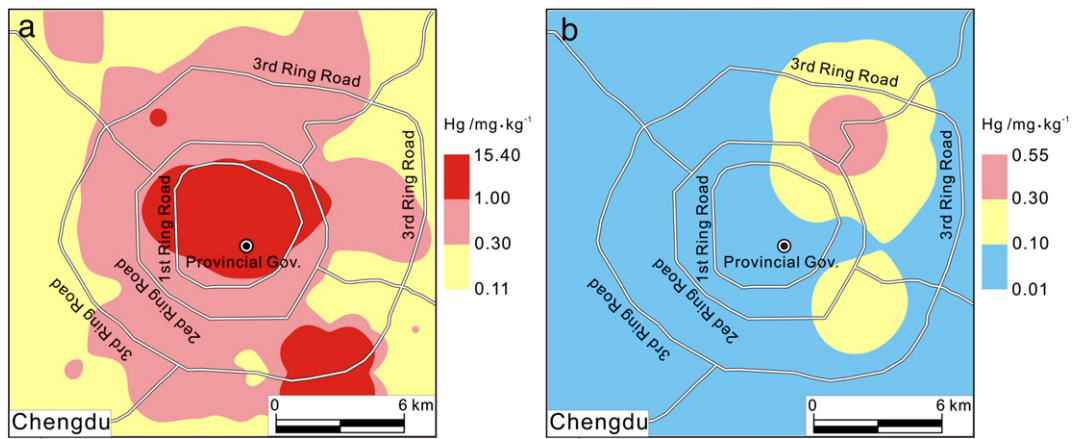


Fig. 3. Map of Hg distribution in the urban topsoil (a) and deep soil (b) in Chengdu.

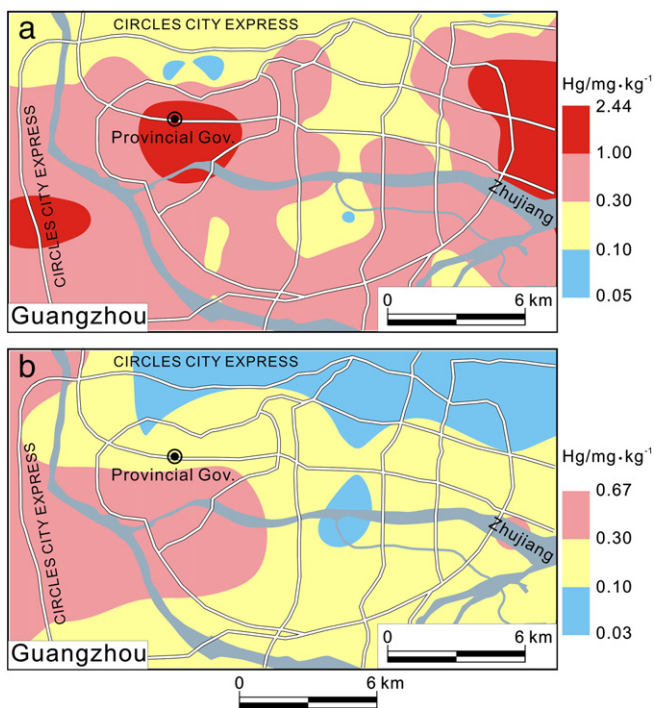


Fig. 4. Map of Hg distribution in the urban topsoil (a) and deep soil (b) in Guangzhou.

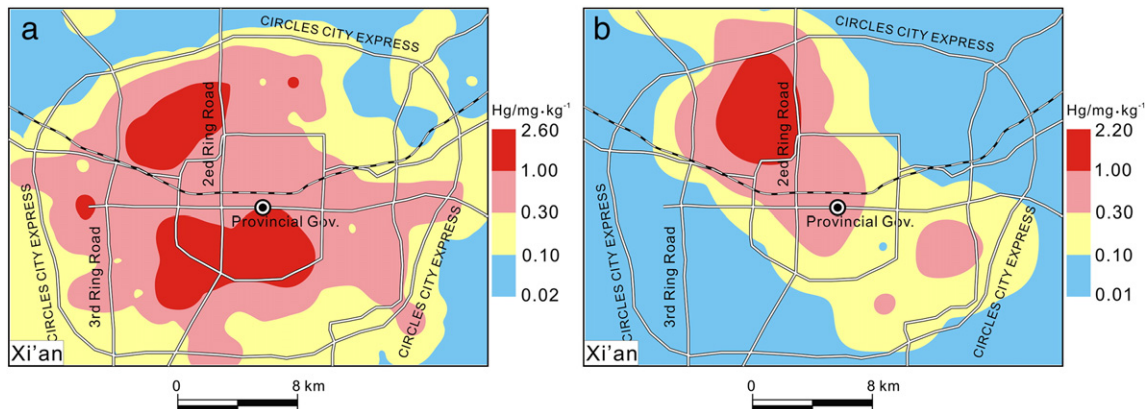


Fig. 5. Map of Hg distribution in the urban topsoil (a) and deep soil (b) in Xi'an.

and parkland soils (CCME, 2009). The Pb-contaminated areas are primarily located in Shanghai (108 km²), Shenyang (80 km²), Guangzhou (32 km²), Chengdu (24 km²), and Kunming (24 km²) (Table 6); these cities account for 73.6% of the total contaminated areas. This observation suggests that considerable attention should be given to Shanghai and Shenyang because of the severity of their contamination.

4.4. Zinc

The spatial patterns of Zn in Shanghai and Kunming indicate that high Zn concentrations (≥ 140 mg·kg⁻¹) cover almost the entire urban area (Fig. 8), indicating a significant Zn contamination. A high Zn concentration exists in the contamination hotspots in Shenyang (Fig. 9a) and along the Xiangjiang River in Changsha (Fig. 10) and the Zhujiang River in Guangzhou (Fig. 9b). No evident patterns of distribution of Zn were observed in deep soils, which strongly indicates a great contribution from anthropogenic sources, as highlighted in previous studies (Cai et al., 2013; Chen et al., 2007; Li et al., 2013; Shi et al., 2008; Sun et al., 2010; Wei et al., 2009b).

The limits of the target and intervention values for Zn listed by the Dutch are 140 and 720 mg·kg⁻¹ (VROM, 2000), respectively. The guideline values of 200 mg·kg⁻¹ for residential areas and parkland and 360 mg·kg⁻¹ for commercial and industrial regions were established by the Canadians (CCME, 1999). These values are useful for assessing the risks to human health and the environment that arise from exposure to contaminated soils. Nationally, approximately 12,536 km² of urban topsoils accounts for 82.5% of the total area that has a concentration of Zn lower than 140 mg·kg⁻¹, which implies

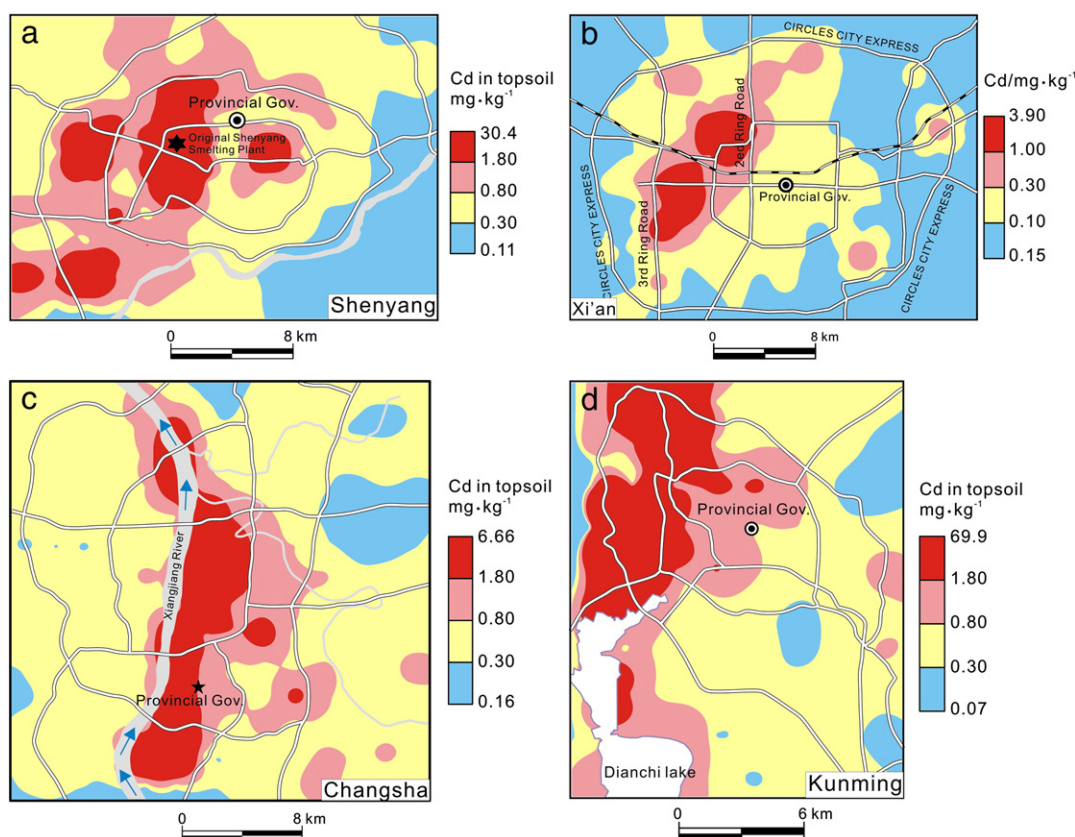


Fig. 6. Distribution of Cd in the urban soils of Shenyang (a), Xi'an (b), Changsha (c), and Kunming (d).

sustainable soil quality. However, an area of 1064 km², accounting for 7.0% of the total area, exceeds the guideline value of 200 mg·kg⁻¹ and is unfit for residential and parkland use. An area of 292 km² was found to be unsuitable for commercial and industrial land use, and an area of 72 km², distributed in Shanghai (16 km²), Shenyang (16 km²), Wuhan (16 km²), Guangzhou (12 km²), Xi'an (4 km²), Nanjing (4 km²), and Kunming (4 km²), requires immediate remediation because topsoil Zn concentrations exceed 720 mg·kg⁻¹ (Table 6).

4.5. Copper

The target and intervention values of Cu in the Netherlands are 36 and 190 mg·kg⁻¹ (VROM, 2000), respectively. The guideline values for residential areas and parkland and commercial and industrial lands in Canada are 63 and 91 mg·kg⁻¹ (CCME, 1999), respectively. The high Cu content in topsoil and deep soil is widely distributed in Kunming, Yunnan Province and is observed more often in topsoils than in deep soils (Fig. 11). A highly similar distribution pattern between the topsoils and the deep soils clearly shows the impact of the native parent material on the spatial distribution of Cu in Kunming. High Cu concentrations are found in two locations where the Cu geochemical background is low, at the old smelting plant in Shenyang and in the central area of the old urban regions in Shanghai (Fig. 12), indicating that Cu contaminations are derived from anthropogenic sources.

Cu in urban soils has a low average I_{geo} value of 0.07, which implies a lower level of Cu contamination compared to Pb, Zn, and Cd. A total area of 112 km² exceeded the intervention value of 190 mg·kg⁻¹ proposed by VROM (2000). Kunming has the largest area (44 km²) of Cu contamination and the highest average Cu content (126.7 mg·kg⁻¹) compared to other cities. In addition to Kunming, Shanghai (28 km²) and Guangzhou,

Shenyang, and Tianjin (8 km²) are cities with the most widespread Cu contamination in Chinese cities (Table 6).

4.6. Selenium

The atmospheric emissions of Se (and Hg) from coal combustion are the primary sources of anthropogenic discharge and pollution. Discriminating between anthropogenic and natural sources is important in defining the extent of pollution in areas where environmental legislation has not yet established intervention limits for soils. In China's geological complexity and the rapid development of its economy, elevated concentrations of Se in urban soils are likely strongly influenced by anthropogenic contributions. In fact, recent studies have showed that the Se emissions from coal combustion in China have increased from 639.69 t in 1980 to 2352.97 t in 2007 (Tian et al., 2010), a rapidly increasing growth rate of 4.9%. The increasing Se emissions have elevated the atmospheric Se concentration in urban areas, resulting in significant atmospheric pollution. The wide dispersion and deposition of dry and wet Se over soil result in negative effects on the environment and public health.

Fig. 13 presents the distribution of Se in the urban topsoils of Guiyang in the Guizhou Province and Taiyuan in the Shanxi Province, which are two of the largest coal-producing and coal-combusting provinces. The samples with a high Se concentration ≥ 1.00 mg·kg⁻¹ accounted for approximately 43.8% and 28.9% of the total samples from each city, respectively, demonstrating the negative effects of Se on the environment and public health according to the guideline value (1 mg·kg⁻¹) suggested by the CCME (2009).

However, naturally occurring Se may be important in defining the pattern of pollution in Guiyang urban areas where high Se geochemical background is observed (mean Se content: 0.46 mg·kg⁻¹). Although Se emission from coal combustion in the Guizhou Province was 93.51 t in

Table 6
Summaries of the area and extent of metal contamination and the percentage of total area of each city in China.

	Total area ^a	As ≥ 55 ^b		Cd ≥ 1.80		Cr ≥ 380		Cu ≥ 190		Hg ≥ 1.000		Ni > 130		Pb ≥ 140		Sb ≥ 20		Se ≥ 1.000		Zn ≥ 720		Total		
		Area ^c	% ^d	Area	%	Area	%	Area	%	Area	%	Area	%	Area	%	Area	%	Area	%	Area	%	Area ^e	% ^f	
Beijing	1044									60	13.6							4	0.7			64	6.1	
Changchun	256									4	0.9											4	1.6	
Changsha	784	20	31.3	68	26.2									8	2.2			16	2.8			112	14.3	
Chengdu	400							4	3.6	40	9.1			24	6.6							68	17.0	
Chongqing	480																							
Fuzhou	360									36	8.2			12	3.3							48	13.3	
Guangzhou	344							8	7.1	44	10.0			32	8.8	4	6.7				12	16.7	100	29.1
Guiyang	320	16	25.0	4	1.5					8	1.8			8	2.2			140	24.6			176	55.0	
Harbin	256																							
Haikou	176											8	66.7									8	4.5	
Hangzhou	904									32	7.3			4	1.1					4	0.7		40	4.4
Hefei	144									4	0.9									20	3.5		24	16.7
Huhhot	312										0.0													
Jinan	256					4	33.3			4	0.9									8	1.4		16	6.3
Kunming	344	12	18.8	56	21.5			44	39.3	20	4.5			24	6.6	4	6.7	12	2.1	4	5.6	176	51.2	
Lanzhou	320													8	2.2							8	2.5	
Lhasa	144													4	1.1							4	2.8	
Nanchang	640																			4	0.7		4	0.6
Nanjing	1148			12	4.6			4	3.6	24	5.5			16	4.4					4	0.7	4	5.6	
Nanning	628	4	6.3											4	1.1	16	26.7	44	7.7			68	10.8	
Shanghai	668			12	4.6	4	33.3	28	25.0	36	8.2			108	29.7	24	40.0	80	14.1	16	22.2	308	46.1	
Shenyang	560	12	18.8	56	21.5			8	7.1	16	3.6			80	22.0	12	20.0	16	2.8	16	22.2	216	38.6	
Shijiazhuang	168										0.0									8	1.4		8	4.8
Taiyuan	512							4	3.6											148	26.1		152	29.7
Tianjin	528			12	4.6			8	7.1	20	4.5			8	2.2					20	3.5		68	12.9
Ürümqi	224				0.0						0.0												0	
Wuhan	896			8	3.1	4	33.3	4	3.6	20	4.5			16	4.4					40	7.0	16	22.2	
Xi'an	768			32	12.3					72	16.4	4	33.3	8	2.2							4	5.6	
Xining	112																							
Yinchuan	616																							
Zhengzhou	884																							
Total	15,196	64		260		12		112		440		12		364		60		568		72		1964	12.9	

^a Area of city, km².

^b Limit to intervention value established by the Dutch, mg·kg⁻¹ (VROM, 2000).

^c Area more than intervention value, km².

^d Ratio of areas more than intervention value to total areas of all cities.

^e Total areas of all contamination metals in a given city.

^f Ratio of total areas of contamination metals in a given city to its total areas.

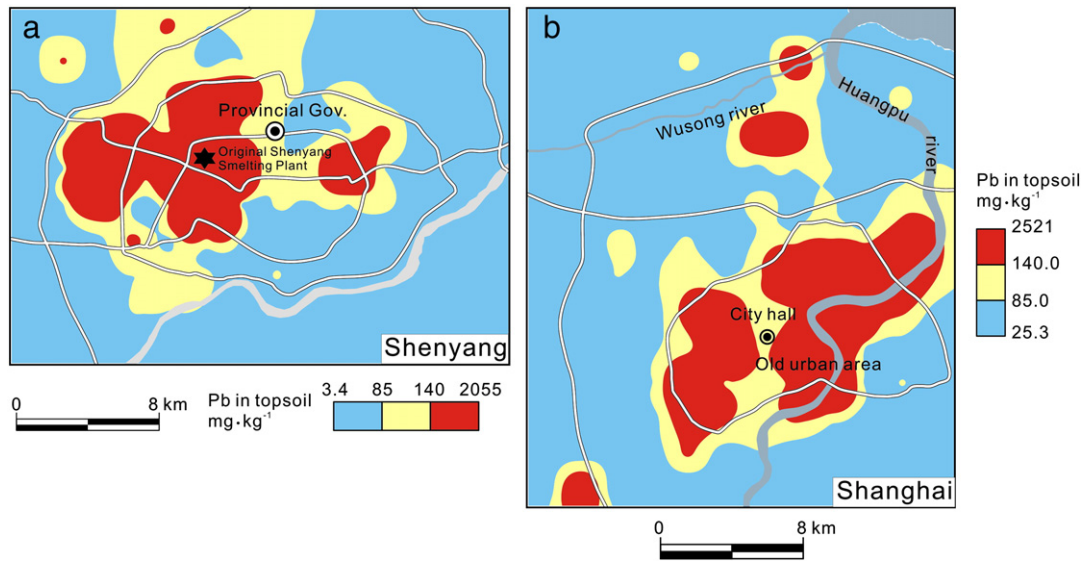


Fig. 7. Distribution of Pb in topsoil in Shenyang (a) and Shanghai (b).

2007 (Tian et al., 2010), anthropogenic sources are likely more important than the natural background (mean Se content: $0.20 \text{ mg} \cdot \text{kg}^{-1}$) in the elevated Se concentration of the urban soils in Taiyuan. In 2007, the Se emission from coal combustion in the Shanxi Province was 142.60 t (Tian et al., 2010).

Moreover, elevated concentrations of Se in the topsoils in Shanghai are clearly caused by long-term industry activities, as shown by both the strong correlation coefficients of the logarithm of Se concentration, the logarithms of Pb ($R^2 = 0.6632$), Zn ($R^2 = 0.6764$), Cu ($R^2 = 0.6478$), Cu ($R^2 = 0.6478$), Sb ($R^2 = 0.6601$), Hg ($R^2 = 0.5381$) and As ($R^2 = 0.6632$) concentrations, and the low concentration of Se in deep soil (Fig. 14).

The average I_{geo} value for Se in urban topsoils of the 31 cities is 1.05 (Table 4). According to I_{geo} , Chengdu, Hefei, Shijiazhuang, and Taiyuan are classified as moderately to heavily contaminated, whereas Beijing, Hangzhou, Jinan, Nanjing, Shanghai, Tianjin, Wuhan, Xi'an, and Zhengzhou are classified as moderately contaminated. Fuzhou, Guangzhou, Haikou, Huhhot, Lhasa, Nanning, Ürümqi, and Xining appear to be uncontaminated by Se.

A total of 568 km² exceeded the guideline value ($1 \text{ mg} \cdot \text{kg}^{-1}$) for residential areas and parkland (CCME, 2009). The Se contamination in Chinese cities is mainly concentrated in Guiyang (140 km²), Shanghai (80 km²), Taiyuan (148 km²), Nanning (44 km²), and Wuhan (40 km²), which accounts for 79.6% of the total contaminated area.

With the implementation of Western development strategies, an increase in coal use in the power plants in Western China will result in an increase in Se emissions. Therefore, Se poses a serious threat to the environment and to human health not only in China but also around the world, and considerable attention should be directed toward its potential negative effects.

4.7. Antimony

Two distribution patterns of Sb exist in the urban soils of the investigated cities. Fig. 15 shows that the high Sb concentration in topsoils is primarily located in the Tiexi Industrial District in Shenyang, Liaoning Province and in the old urban areas of Shanghai. The Sb geochemical background is lower in deep soils of the two cities, however, clearly

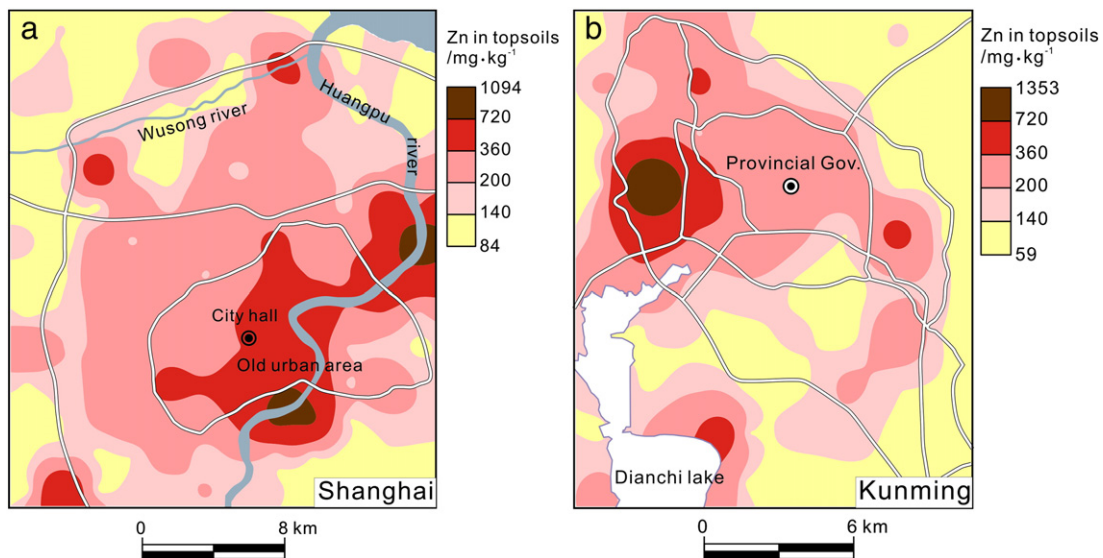


Fig. 8. Distribution of Zn in the topsoil of Shanghai (a) and Kunming (b).

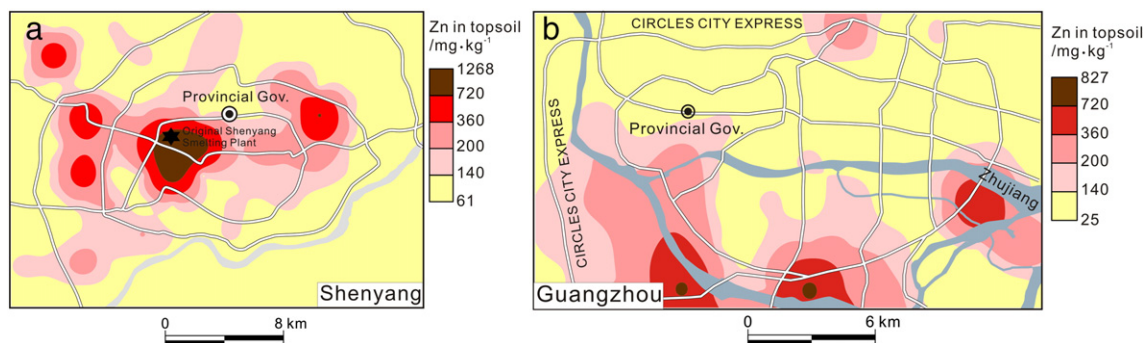


Fig. 9. Distribution of Zn in the topsoil of Shenyang (a) and Guangzhou (b).

indicating an anthropogenic contribution. A similar pattern exists in Changsha, Hunan Province and in Kunming, Yunnan Province.

High Sb-levels are commonly found in all metal sulfide ores, particularly those of Pb and coal (Qi et al., 2008). The significant correlations (R^2) for the concentrations in Sb and Pb are 0.7523 in Kunming, 0.8375 in Shanghai, 0.8036 in Shenyang and 0.5115 in Changsha, indicating a dominant role of industry as a pollutant source.

On the other hand, the spatial distribution of Sb in topsoil and deep soil in Nanning, Guangxi province shows a similar pattern. The average concentration in topsoils is comparable to that in deep soils, demonstrating the dominant role of the parent soil material as a Sb source.

According to I_{geo} , Sb in topsoils is classified as uncontaminated to moderately contaminated. A sharp boundary is observed in some Chinese cities, where the I_{geo} values decrease from moderately to heavily contaminated in Shanghai (2.45), to moderately contaminated in Shenyang (1.46) and Changsha (1.02), and from uncontaminated to moderately contaminated in Kunming, Nanning, Guangzhou, Fuzhou, Hangzhou, Guiyang, Changchun, and Ürümqi. Uncontaminated soil is observed in the remaining 20 cities.

Statistics show that the region with $Sb > 20 \text{ mg} \cdot \text{kg}^{-1}$ has an area of 60 km^2 that is distributed in Shanghai (24 km^2), Nanning (16 km^2), Shenyang (12 km^2), Guangzhou (4 km^2), and Kunming (4 km^2). The heaviest Sb contamination was observed in Shanghai.

4.8. Nickel

The average I_{geo} value for Ni in urban topsoils is -0.59 , which indicates that urban soils are uncontaminated by Ni at a national scale.

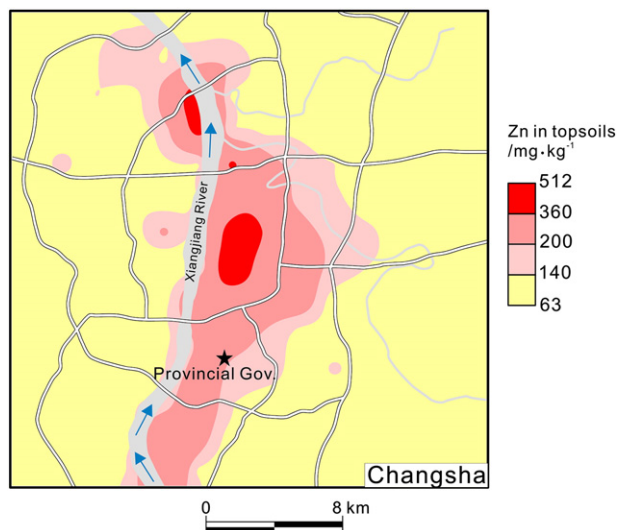


Fig. 10. Distribution of Zn in the topsoil of Changsha.

Moreover, only urban soils of 12 km^2 , of which 8 km^2 in Haikou and 4 km^2 in Xi'an, are unsuitable for residents according to the SVGs drawn up by the Environment Agency (2009c), UK. Compared with other metal contaminations, Ni contaminations in urban soils of Chinese cities are not serious.

4.9. Chromium

The contour map for Cr in urban topsoils and deep soils in Shanghai is illustrated in Fig. 16.

Overall, Cr has a different distribution pattern from Pb, Zn, Hg, Cd, Cu, and Sb in Shanghai, which implies a different main source. Two relatively high concentrations or hotspots for Cr were found in the city, with elevated levels compared with those in deep soils. The highest level occurred in a hotspot in the northwestern region of the city, crossing the Outer Ring Highway and the Wusong River, where Baoshan Steel Company Limited, the biggest steel company in China, is located. This spatial distribution of Cr indicates that industrial activities are the dominant pollutant source.

Another hotspot is located in the southwestern region of the city where the high Cr concentration stretches along the Huangpu River.

An overall Cr contamination with an average I_{geo} of -0.21 for Shanghai can be defined and classified as uncontaminated according to the classifications for contamination by Muller (1969).

However, some isolated Cr contaminations with a Cr concentration $\geq 380 \text{ mg} \cdot \text{kg}^{-1}$ still exist in Shanghai (4 km^2) and other cities such as Jinan (4 km^2) and Wuhan (4 km^2). Compared to that of other metals, the total area of Cr contamination is the smallest.

4.10. Arsenic

Two isolated areas with high As concentrations in the topsoils are located in an old metallic smelting factory in Shenyang (Fig. 17a). The larger region with high As concentration was also found in the northern urban regions and near Dianchi Lake in Kunming (Fig. 17b), which is consistent with the spatial distribution of Sb, Cu, Pb, Zn, Cd, Ni, and Cr. Moreover, the As concentration in deep soils was low in these two cities; both of these observations indicate an anthropogenic source for As.

An As pattern similar to that distributed along the Xiangjiang River was discovered in Changsha, Hunan Province (Fig. 18). An elevated As concentration in the topsoils was traced to numerous Pb–Zn and Sb ore deposits and the corresponding metallic smelting factories located in the upstream regions of Xiangjiang (Chen et al., 1999; Wei et al., 2009a; Zhang et al., 2012).

Contamination with an average I_{geo} of -0.39 for As is classified as uncontaminated. However, some local areas of contamination with As concentrations $\geq 55 \text{ mg} \cdot \text{kg}^{-1}$ still exist in Changsha (20 km^2), Guiyang (16 km^2), Kunming (12 km^2), Shenyang (12 km^2), and Nanjing (4 km^2), although each has a small area of contamination.

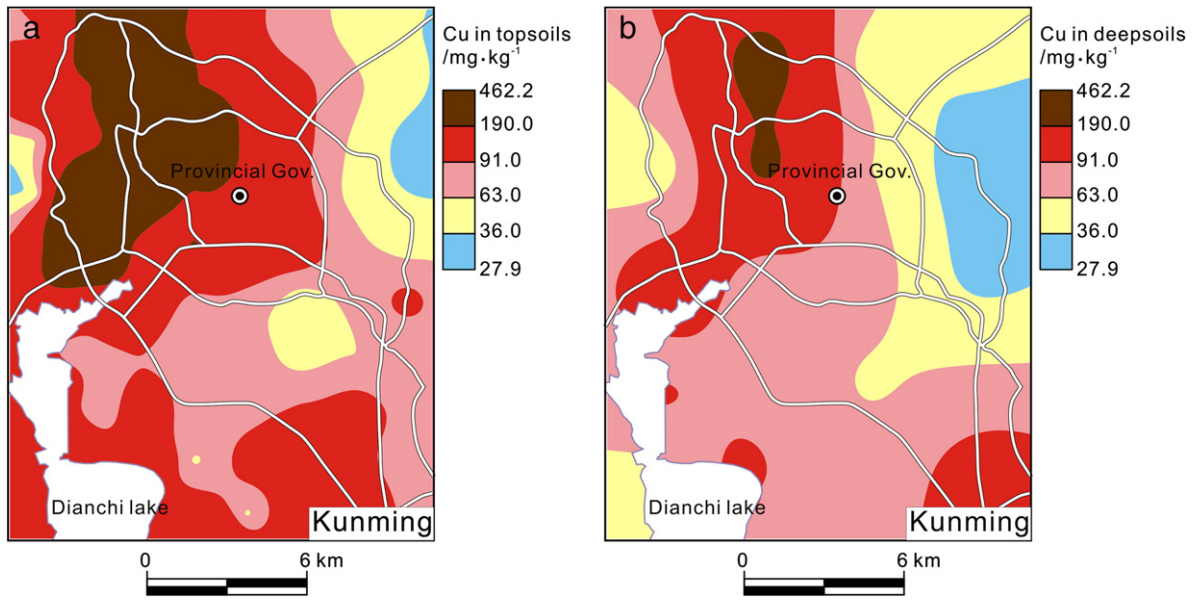


Fig. 11. Geochemical maps of Cu in topsoil (a) and deep soil (b) in Kunming.

5. Rank of contamination metals

The I_{geo} values of different metals in urban topsoils vary greatly in different cities. Overall, the different elements can be arranged in descending I_{geo} values, as follows: Hg (1.96) > Cd (1.26) > Se (1.05) > Sb (0.45) > Pb (0.37) > Zn (0.13) > Cu (0.07) > As (-0.39) > Cr (-0.53) > Ni (-0.59). These results imply that, on a national scale, urban soils in China are moderately contaminated by Hg, Cd, and Se, and the contamination levels of Cu, Sb, Pb, and Zn range from uncontaminated to moderately contaminated. Urban soils may be uncontaminated by As, Cr, and Ni, indicating that these three metals are commonly derived from natural (geogenic) processes. The number of cases of $I_{geo} > 0$ in the 31 cities is as follows: 29 for Hg, 26 for Cd, 23 for Se, 16 for Pb, 14 for Zn, 11 for Cu and Sb, 1 for As, and 0 for Cr and Ni (Table 4). The I_{geo} of Hg is ranked first, followed by Cd and Se, among all metal contaminants in the urban soils of China. The area of metal contamination is, respectively, as follows: 568 km² for Se,

440 km² for Hg, 364 km² for Pb, 260 km² for Cd, 112 km² for Cu, 64 km² for As, 60 km² for Sb, 12 km² for Cr, and 12 km² for Ni (Table 6). The areas of metal contamination are higher than the adopted intervention values. This finding disagrees with previous studies on Pb, Zn, and Cu in urban soils (Gu et al., 2009; Guo et al., 2012; Li and Huang, 2007). Therefore, considerable attention should be given to long-term Hg, Cd, and Se contamination in urban soils in China.

6. Association of metal contaminations in different cities

The type of major industry in an urban area directly affects the contamination level of metals and their association in soils. To learn more about the associations among the metal contaminants in each city, the statistics on the number of metals with $I_{geo} > 0$ are presented in Table 4.

There are complex associations among the metal contaminants, which varies greatly from one city to another and can be classified as

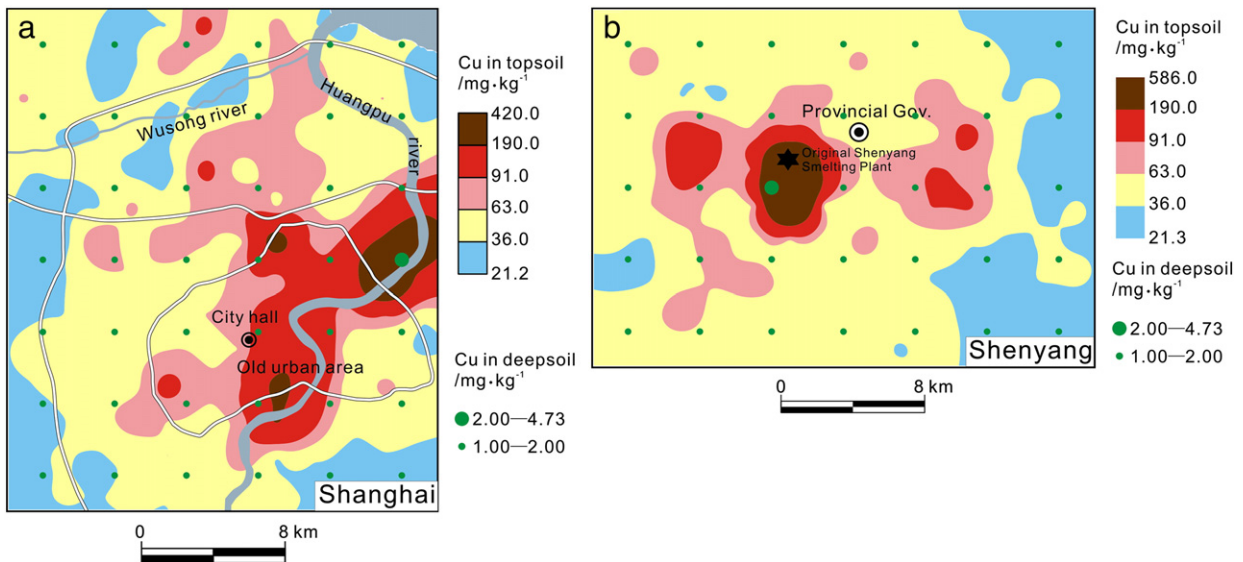


Fig. 12. Geochemical maps of Cu in the urban soil of Shanghai (a) and Shenyang (b).

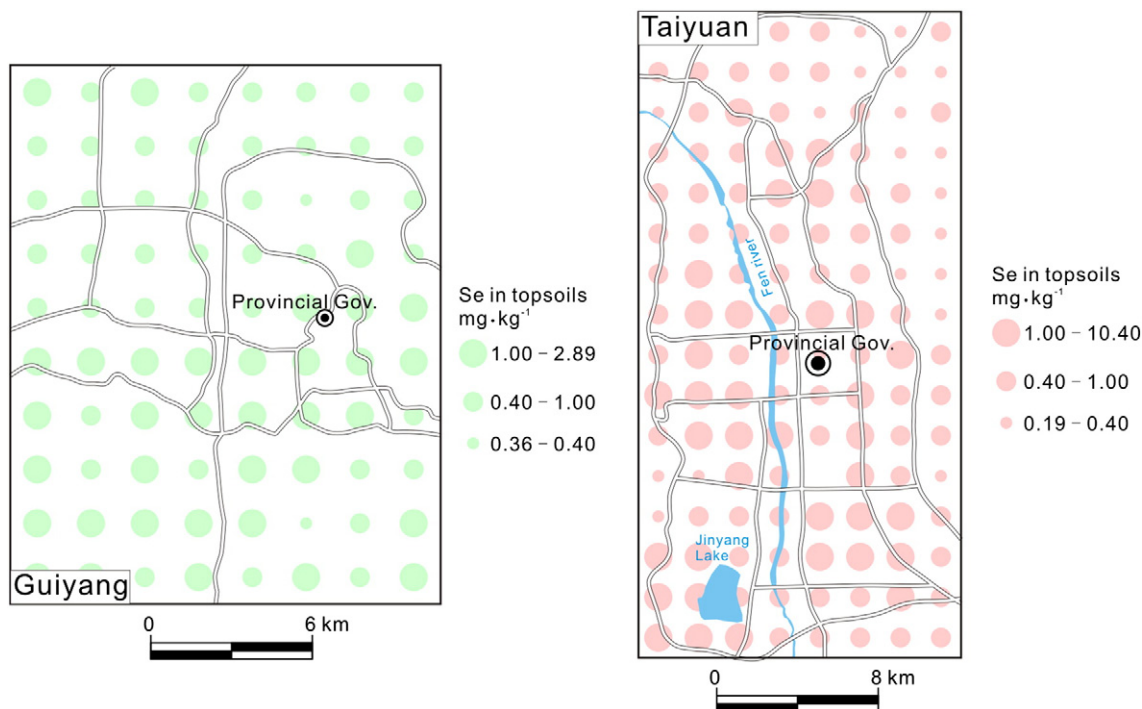


Fig. 13. Distribution of Se in the topsoil in Guiyang (a) and Taiyuan (b).

follows: As, Cd, Cu, Pb, Zn, Hg, Se, and, Sb in Kunming; Cd, Cu, Hg, Pb, Sb, Se, and Zn in Changsha, Hangzhou, Shanghai, and Shenyang; Cd, Cu, Hg, Pb, Se, and Zn in Beijing, Chengdu, Tianjin, and Wuhan; Cd, Cu, Hg, Pb, Sb, and Zn in Guangzhou; Cd, Pb, Zn, Hg, and Se in Hefei, Shijiazhuang, and Xi'an; Cd, Hg, Sb, and Se in Changchun and Guiyang; Cd, Hg, Pb, and Se in Jinan, Lanzhou, and Nanning; Cd, Hg, Cu, and Se in Taiyuan; Cd, Hg, and Se in Chongqing, Harbin, Nanchang, and Zhengzhou; Cd, Hg, and Zn in Haikou; Hg and Sb in Fuzhou and Ürümqi; Cd and Sb in Nanning; Cd and Hg in Xining; Hg and Se in Yinchuan; Hg in Lhasa; and no contaminant metals were found in Huhhot.

7. Degree of contamination with trace metals

The 31 cities can be arranged in descending IPI_N values as follows: Kunming (2.69) > Chengdu (1.70) > Nanning (1.54) > Shanghai (1.51) > Guangzhou (1.40) > Shenyang (1.36) > Guiyang (1.31) > Fuzhou (1.30) > Tianjin (1.13) > Wuhan (1.11) > Xi'an (1.09) > Beijing (1.04) > Changsha (0.99) > Nanjing (0.94) > Jinan (0.87) > Hangzhou (0.86) > Hefei (0.85) > Ürümqi (0.78) > Taiyuan (0.76) > Haikou and Xining (0.73) > Chongqing (0.71) > Shijiazhuang (0.69) > Changchun (0.68) > Lanzhou and Nanchang (0.66) > Lhasa (0.60) > Yinchuan (0.59) > Harbin (0.57) > Zhengzhou (0.55) > Huhhot (0.44) (Table 4). These results imply that Kunming is categorized at a moderate pollution level, whereas Chengdu, Nanning, Shanghai, Guangzhou, Shenyang, Guiyang, Fuzhou, Tianjin, Wuhan, Xi'an, and Beijing are categorized at a slight pollution level. Changsha, Nanjing, Jinan, Hangzhou, Hefei, Ürümqi, Taiyuan, Haikou, Xining, and Chongqing are classified as under precaution, and Shijiazhuang, Changchun, Lanzhou, Nanchang, Lhasa, Yinchuan, Harbin, Zhengzhou, and Huhhot are within the safe range of trace metal pollution.

However, IPI_N is not the only indicator for assessing the degree of metal contamination in a city. A higher weight of a given metal corresponds to a higher concentration of the metal. The area of metal contamination should be considered as an important parameter in assessing whether the metal concentration in a city should be taken as the intervention values or soil environmental quality guidelines. The computed contamination area of each metal and the total area of contamination for each city (including areas with metal co-contamination) are listed in Table 6. The table shows that the total area of metal contamination and the percentage of the urban area are as follows: Shanghai (308 km², 46.1%), Shenyang (216 km², 38.6%), Guiyang (176 km², 55.0%), Kunming (176 km², 51.2%), Taiyuan (152 km², 29.7%), Xi'an (120 km², 15.6%), Changsha (112 km², 14.3%), Wuhan (108 km², 12.1%), and Guangzhou (100 km², 29.1%). Other cities had less than 100 km² of contaminated area.

Thus, cities with a total area of metal contamination that exceeds 100 km² should be paid attention when assessing the degree of

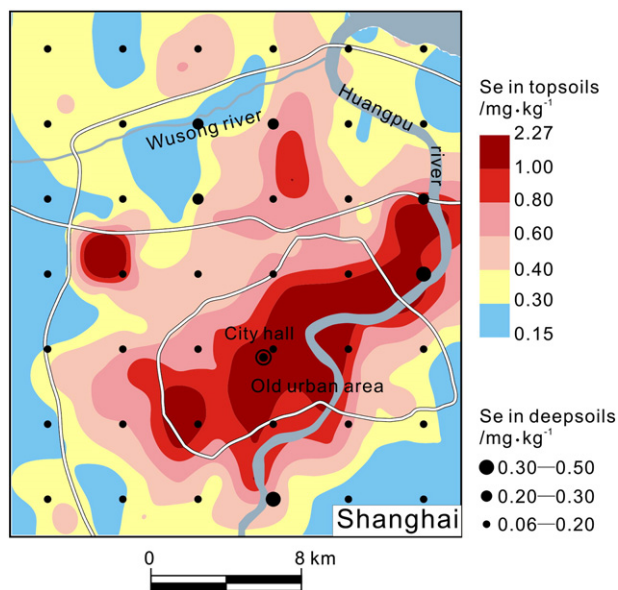


Fig. 14. Distribution of Se in the topsoils and deep soils in Shanghai.

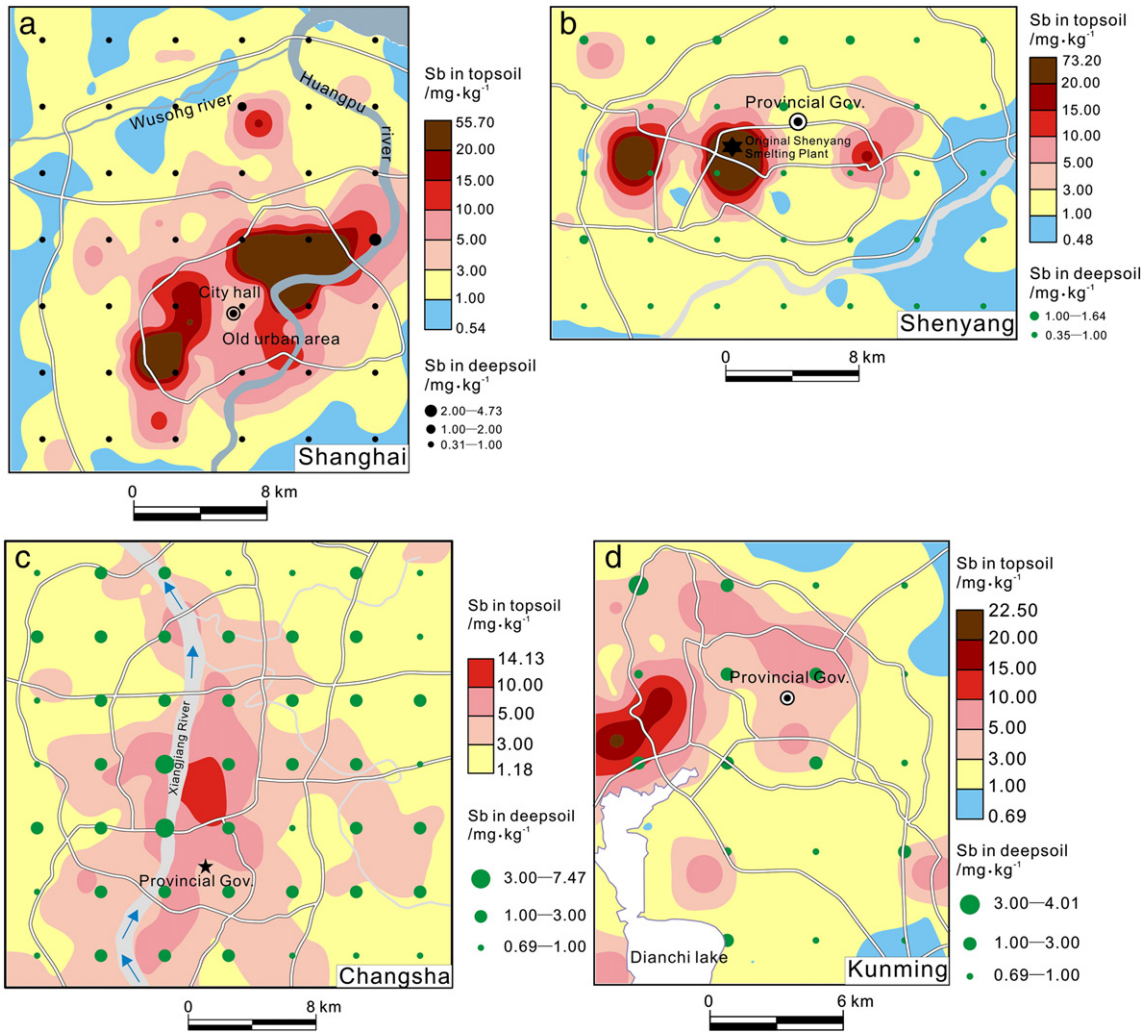


Fig. 15. Geochemical maps of Sb in Shanghai (a), Shenyang (b), Changsha (c), and Kunming (d).

contamination. Information on the quality of the environment is important in evaluating the sustainability of urban areas (van Kamp et al., 2003). The information provided in this study will aid in the

decision-making and sustainable urban planning of the various local governments.

8. Concluding remarks

The NMPRGS, sponsored by the China Geological Survey, has been conducted since 1999. Up to 2012, the survey has already covered an area of 1,650,000 km² (Xi, 2009). The high-quality data collected on trace metals has become the most systematic source of information for the assessment of trace metal contamination in urban soils in China. Only 10 trace metals and 31 metropolises were selected in this study, but within the regions covered by the NMPRGS, other trace metals and medium and small-sized cities (towns) will be selected for similar studies. New contaminant metals and the influence of other factors will be explored, and the degree of contamination of various types and sizes of cities will be assessed. These studies will provide basic information for the improvement of urban land use management and pollution prevention and control in China.

As shown by our data, serious contaminations in urban area originated from factories that produced serious pollution, although many of these plants have been closed or moved to other locations. However, the inventories of factories that moved from an urban area are incomplete, both in detail and extent. Improving urban soil quality requires understanding and identifying the pollution source and integrating information processing. Overall, a transparent system of urban land use management is urgently needed.

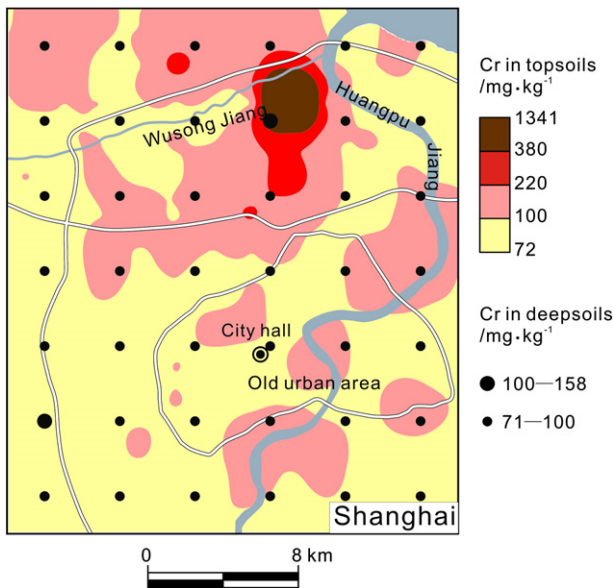


Fig. 16. Distribution of Cr in the topsoil and deep soil in Shanghai.

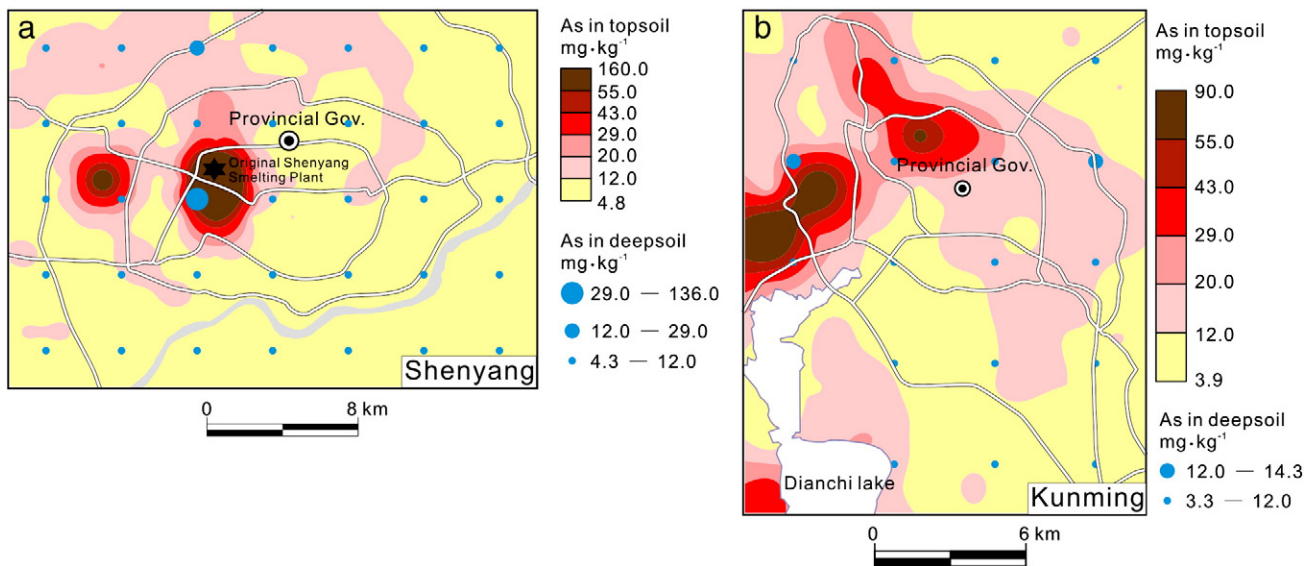


Fig. 17. Distribution of As in the urban soil of Shenyang (a) and Kunming (b).

Complex co-contaminations of trace metals exist in many cities. The current and potential threats need to be understood so that suitable technologies can be developed to repair contaminated soil. However, changes in the concentrations of serious co-contaminating trace metals in some cities such as, Shanghai, Kunming, Shenyang, and Changsha need to be monitored before finding a cost-effective remediation technology for soils that are heavily polluted with metals.

The priority pollutants and limits for land use vary greatly in different countries. Therefore, an international agreement on a guideline for a common range of limits of soil pollutants is urgently needed. Although a unique guideline for agricultural soils was issued in 1995 (CEPA, 1995), little legislation on urban soils and soil protection has been established in China, whereas soil management legislation was already enacted in Canada (CCME, 1999), the Netherlands (VROM, 2000), the UK (Environment Agency, 2009a, 2009b, 2009c) and the USA (USEPA, 2012).

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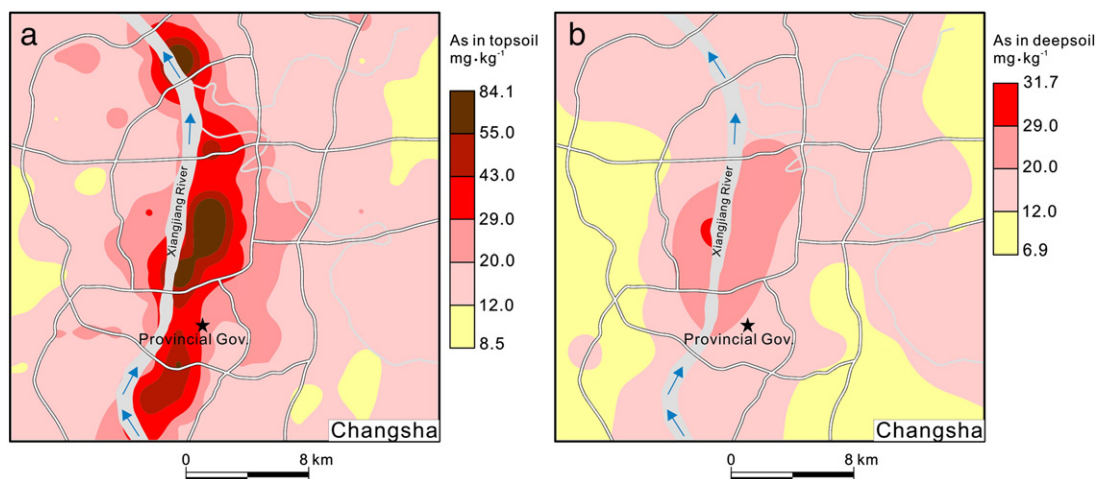


Fig. 18. Distribution of As in the topsoil (a) and deep soil (b) at Changsha.

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