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Pollution and Health Risk Assessment of Heavy Metals in Urban Soil in China

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Abstract

The pollution and potential health risk due to lifetime exposure to heavy metals in urban soil of China were evaluated, based on the urban soil samples collected from published papers from 2005 to 2014. The contamination levels were in the order of Cd > Hg > Cu > Zn > Pb >As > Ni > Cr, and Hg and Cd fell into the category of "moderately contaminated" to "heavily contaminated". The non-carcinogenic for different populations varied greatly, among which children faced high risk, and then the adult female and adult male were followed. The hazard index (non-carcinogenic risk) higher than 1.00 occurred in Shanghai, Gansu, Qinghai, Hunan and Anhui, whereas most of those in the northern and western had low risks. For the carcinogenic risk, Anhui and Ningxia provinces had urban soils exceeding the safe reference (1×10^{-6}-1×10^{-4}). Qinghai and Gansu had high carcinogenic risks since their risk index were much close to the reference, and the others were in low risk.

Key words: Soil heavy metal, pollution, health risk, urban environment, China

Introduction

More than half of people in China are living in cities due to the rapid development of urbanization (Wei and Ye, 2014). Thus it is important to study the influence of urban environment on human health (Cao et al., 2015; Giri and Singh, 2015; Lei et al., 2015; Li et al., 2013). Heavy metals have significant negative effects on human health, ranging from acute reactions to chronic illnesses (Kampa and Castanas, 2008; Pei et al., 2015; Shi et al., 2008; Wang et al., 2015). The mercury (Hg), cadmium (Cd), arsenic (As) and Chromium (Cr), are toxic or carcinogenic even at low concentrations when people are exposed for a long time (Angelone and Udovic, 2014; Khan et al., 2015; Zhang et al., 2015; Zhang et al., 2014). Potentially toxic heavy metal pollution in urban environments has given rise to growing concerns during past decades (Guo et al., 2012; Li et al., 2013; Shi et al., 2008; Wei and Yang, 2010; Xia et al., 2011; Zheng et al., 2015).

Recent studies have focused on monitoring the concentrations, exploring the spatial distribution of heavy metals, identifying the enrichment factors and sources, and assessing the pollution levels in the urban soils. For example, Wei and Yang concluded that about 65% of the investigated cities have high or extremely high contamination levels of heavy metals in urban soils (Wei and Yang, 2010), most of which were in the developed eastern coastal regions and old industrial cities (Yu et al., 2012). The two principle sources of trace metal contamination in urban soil in China were industrial discharges and traffic emissions (Li et al., 2013; Wei and Yang,

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2010). The pollution in urban soils can pose directly a significant health risk through soil ingestion, dust inhalation, or dermal contact (Giri and Singh, 2015; Karim and Qureshi, 2014). Thus, it is critical to quantify the potential human health risk due to the exposure to heavy metals in urban soil.

Soil quality criteria based on total metal concentrations might not accurately estimate the real risks (Baize and van Oort, 2014) and are only appropriate for worst-case scenarios (Angelone and Udovic, 2014). Health risk assessment is an effective approach to determine the risk to human health quantitatively posed by several various contaminants through different exposure pathways (Kampa and Castanas, 2008; Luo et al., 2012). The extent of harmful effects depends on the age and structure of the population, and the type of intensity of contact with contaminated soils (Wcisło et al., 2002; Zhang et al., 2015). Few studies concerned the above questions when assessing the health risk assessment of heavy metal contamination of urban soils in cities but Xiamen (Luo et al., 2012), Huludao city (Zheng et al., 2010), Hangzhou, Changxing and Shangyu cities in Zhejiang (Liu et al., 2013) and Gejiu city (Li et al., 2014a). To enact an effective policy to control further harm on human health risk by heavy metals, a comprehensive health risk assessment in urban soil throughout China is needed.

This study collected heavy metal concentrations in urban soil throughout China, from papers published from 2005 to 2014. The aim is to (1) survey heavy metal concentrations, (2) assess the level of heavy metal pollution, and (3) evaluate the health risk in urban soil throughout China.

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Materials and methods

Data collection

This study collected 157 published papers on heavy metal concentrations in urban surface soils (0-20 cm) throughout China from 2005 to 2014. The detailed information on the collection process of the published papers and the heavy metal data can be found in the manuscript (Zhang et al., 2014). Eight heavy metals Cu, Pb, Zn, Cd, Ni, As, Cr and Hg were collected in this study. In total, 297 data records on heavy metal concentrations in urban surface soil were collected. Background information on the investigated urban areas is described in Table S1.

Pollution assessment

The geoaccumulation index (I_{geo}) is employed in assessing the contamination levels and the pollution of heavy metals in urban soils in China. This method has been widely applied in European trace metal studies since the late 1960s and it is also employed in pollution assessment of urban soil (Wei and Yang, 2010), which is calculated using the follow equation (Solgi et al., 2012):

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

where C_n is the measured concentration of every heavy metal in found in urban soil in China, and B_n is the geochemical background value of the heavy metals found in the soil (mg/kg). In this study, the B_n refers to the heavy metal background concentrations of the province that the n-th data record belongs to (CNEMC, 1990). The background concentrations on a provincial scale

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and the 7 classes of I_{geo} are described in Table S2 and S3, respectively (Li et al., 2014b; Wei and Yang, 2010).

Health risk assessment

The risk effects consist of carcinogenic and non-carcinogenic risk assessments for all heavy metals through ingestion and dermal absorption in this study. The average daily intake of heavy metals from soil ingestion (ADI_I) and dermal absorption (ADI_D) are calculated from the equations provided by USEPA (Li et al., 2014b).

A hazard index (HI) approach to assess non-carcinogenic risk is calculated using equation (Yuswir et al., 2014):

$$HI = HQ_i = \sum_{i=1}^{n} \frac{ADI_i}{RFD_i} \qquad (5)$$

where the chronic hazard index (HI) is the sum of hazard quotient (HQ_i) of i-th heavy metal, RFD_i is the chronic reference dose for i-th heavy metal. In this study, RFD for Cu, Pb, Zn, Cd, As, Ni, Cr, Hg were 4×10^{-2}, 1.4×10^{-4}, 3×10^{-1}, 2.5×10^{-5}, 3×10^{-4}, 8×10^{-4}, 1.95×10^{-2}, 1.6×10^{-4}, respectively (Li et al., 2014b).

Carcinogenic risk is estimated (Cao et al., 2015):

$$RISK = ADI \times SF \qquad (8)$$

$$SF_{ABS} = \frac{SF_0}{ABS_{GI}} \qquad (9)$$

where SF is the carcinogenicity slope factor (per mg/kg-day), SF_{ABS} is the dermally adjusted slope factor (per mg/kg-day), SF_0 is the oral slope factor (per mg/kg-day), and ABS_{GI} is the

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gastrointestinal absorption factor(unitless). In this study, only the carcinogenic risk of As is estimated and SF for As is 1.5 (Li et al., 2014b).

Results and discussion

Heavy metal concentrations in urban soil

As shown in Fig. 1, the average concentrations of each heavy metal exceeded the corresponding background values in soils in China (CNEMC, 1990), which were 5.88, 4.15, 2.21, 1.98, 1.83, 1.30, 1.27 and 1.26 times of the background values for Cd, Hg, Cu, Pb, Zn, As, Cr and Ni, respectively. In relative term, the soils in the urban areas were contaminated least by Cr and most by Cd, which was consistent with a previous study (Wei and Yang, 2010). For all metals, the total concentrations showed a large degree of variability, reflecting the non-homogeneous distribution of concentrations of anthropogenically emitted heavy metals (Guo et al., 2012). Firstly, the large urban population generated a large amount of waste, some of which might contained heavy metals (Taghipour et al., 2013). Secondly, vehicle emissions were considered to be the principal source of heavy metal in urban soil (Cheng et al., 2014; Christoforidis and Stamatis, 2009). Thirdly, mining and smelting activities were another source of heavy metal contamination in urban environment, which exposed the minerals to weathering processes during excavation (Yuan et al., 2009).

Pollution assessment of heavy metals in urban soils

The classes of the I_{geo} for the heavy metals are presented in Table 1. The I_{geo} values showed that

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all the investigated urban areas for Cr fell below class 3, with 88% falling into class 0, demonstrating that Chinese urban soil was not contaminated by Cr. About 91.72% of Ni soil samples and 76.73% of As fell into class 0, but some of the sampling locations corresponded to heavily contamination by Ni (1.28%) and As (1.99%). More than 85% of the urban soil samples of Cu, Pb and Zn were within class 0 and class 1, indicating moderate level of pollution. However, the I_{geo} values of Cd and Hg were higher than class 3 for 20.76% and 14.07% of urban soil samples respectively, indicating Cd and Hg had been introduced from exterior sources in some regions. Generally, As, Ni and Cr were the least contaminated, with the most of percentage of “uncontaminated” soil samples. Cu, Zn and Pb fell into the category of “uncontaminated” to “moderately contaminated”; Hg and Cd fell into the category of “moderately contaminated” to “heavily contaminated” in China. The contamination levels of these heavy metals were in the order of Cd > Hg > Cu > Zn > Pb >As > Ni > Cr, which was consistent with the results obtained by Wei and Yang (2010). In the study of Wei and Yang (2010), Cr and Ni showed the least contamination in the investigated sites, whereas Cd showed the highest I_{geo} values for most cities.

Among the eight social-economic regions (Fig. 2), Northwest region has the least contaminated area with the lowest I_{geo} for all metals. Middle Yellow River region, south coastal region and Middle Yangtze River had high I_{geo} values, indicating urban cities in these regions were moderately to heavily contaminated, particularly for Hg and Cd. In addition, only in Middle

Yangtze region the I_{geo} of Cu was higher than 3, demonstrating the urban soils in this region were significantly contaminated by Cu. Based on the pollution assessment of I_{geo}, Middle Yellow River region, South Coastal region and Middle Yangtze River appearred to be the most polluted and should be selected as the priority control regions and Hg and Cr should be selected as the priority control heavy metals.

Health risk assessment

Non-carcinogenic assessment

The HI for different populations varied strongly, in the order of children > adult female > adult male. About 23.53% of the 297 soil sample records in the South Coastal region with HI > 1 for children were higher than the other social-economic areas. Adults faced higher health risk exposure from ingestion but lower risk from dermal absorption than children did. Similar results were also obtained in the other studies (Li et al., 2014b; Zheng et al., 2010). This is mainly because that heavy metals could be accumulated in bodies over a long time, and especially the non-carcinogenic adverse effects of Cd, Pb and Hg to the tissues of adults are quite serious (Wcisło et al., 2002). Karim and Qureshi (2014) also found the ratio of a child's cumulative HI to an adult for the combined routes is 8.9 times, indicating the children were more susceptible to non-carcinogenic health effects of trace metals compared to adults (Karim and Qureshi, 2014). Particularly, the non-carcinogenic risk of heavy metals to children via oral ingestion needs special attention.

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When mapping the non-carcinogenic risk level distributions of the examined urban areas, worst-case scenarios (children) were adopted by applying the non-carcinogenic risk (Fig. 3). The HQ values for Zn, As, Cr and Hg were very low and the highest HQ for these metals was below 0.10 and the highest HQ values for Cd, Ni and Pb were all above 1.00. *HI* values higher than 1 shows that non-carcinogenic risk may occur, while the reverse applies when HI lower than 1 (Luo et al., 2012). The HI higher than 1.00 occurred in Shanghai, Gansu, Qinghai, Hunan, Inner Mongolia and Anhui, mainly due to mining and smelting activities (He, 2007; Min et al., 1999; Wang et al., 2009), whereas most of the northern and western urban areas' values were lower than 1.00. These results were consistent with that found by Wei and Yang (2010), which reported that environmental pollution and related health effects were concentrated in Middle Yangtze region and East Coastal region as well as Gansu and Qinghai, whereas low pollution levels existed in southwest of China (Wei and Yang, 2010).

A correlation between traffic density and heavy metal deposited in urban soil was found (Li et al., 2013; Wang et al., 2012). Moreover, there was a postive relationship between population and heavy metal contents in urban soil (Charlesworth et al., 2003; Zheng et al., 2010). The main roads of Shanghai city were marked by traffic and heavy commercial verhicles subjected to frequent stop-and-go situations (Karim and Qureshi, 2014). Moreover, the population of Shanghai city was higer than other cities in Gansu, Qinghai, Hunan and Anhui. This suggested that the major contribution to high HI values in Shanghai city was from atmospheric deposition, which traffic denisty and population made relatively significant contributions. However, this was not the case for Hunan, Gansu, Qinghai, Hunan, and Anhui, where the high HI values in the cities of these provinces were much higher than those of other cities with similar size of population. In smelting district, the heavy metal contamination due to semlting activities pose much higher risk than that from automobiles (Cai et al., 2013; Karim et al., 2014).

Carcinogenic risk assessment

The carcinogenic risks of As for different populations varied greatly for different people groups. The children faced the highest risk, and then adult females and adult males were followed (Fig. 4). The most susceptible subpopulation was young children, through dermal absorption because of their hand-to-mouth activity (Zheng et al., 2010). For adult males and adult females, dermal absorption was the primary pathway of exposure, whereas for children ingestion and dermal absorption both acted as common routes of exposure (Li et al., 2014b). Therefore, the exposure of the heavy metals to children could create more potential health risks.

The acceptable or tolerable risk for regulatory purposes is within the range of 10^{-6}–10^{-4} (Luo et al., 2012). For children, only 0.62% of the soil samples had carcinogenic risks lower than 1×10^{-6}, indicating these areas were within the absolute safe level. 96.44% of the soil samples in the investigated regions were within the safe range. The remaining 2.47% of the soil samples had the carcinogenic risk values exceeding 1×10^{-4} in the investigated regions. Compared with children, adult health risk due to heavy metals exposure from urban soils is lower. The ingestion resulted in a carcinogenic risk for As, followed by dermal contact. Moreover, adult health risk due to dermal contact as the route of exposure to urban soils was lower.

To map the carcinogenic risk level of the examined urban areas in China, the worst-case scenario (children) was selected. On a carcinogenic risk map (Fig. 5), Anhui and Ningxia provinces had urban soils exceeding the safe reference, indicating people in the two provinces faced a high carcinogenic risk by As through ingestion and dermal absorption, mainly due to sewage irrigation (Wang et al., 2010), traffic emissions (vehicle exhaust particles, tire wire particles, weathered street surface particles, brake lining wear particles) and urban and industry development (Zhang et al., 1999). Qinghai and Gansu had high carcinogenic risks with the index values close to the reference of 1×10^{-4}, mainly due to irrigation by sewage from smelting or industry plant (Liu, 2008; Nan and Zhao, 2000) and industrial emissions (power plants, coal combustion, metallurgical industry, auto repair shop, chemical plant, etc.) (Nan and Zhao, 2000). But it should be noticed that Hunan, Guangzhou, Jiangxi and Fujian also had the possibility of high carcinogenic risks, while the other provinces were within the low range of carcinogenic risks.

The findings of this study would facilitate the decision-makers to manage and dispose of contaminated soils, and minimize health risks on urban inhabitants. However, there is a need for further study of urban soil exposure parameters that would help reduce the uncertainties associated with the risk calculations. In fact, most of the urban soil samples selected in these

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published papers in China were distributed in the developed regions, which covered a disproportionately small fraction of China. In conclusion, there were large uncertainties in the assessment of health risk assessment in China, which resulting from sampling procedures, analysis method and parameters used to calculate the risk index.

Conclusion

In this study, data on heavy metals in urban soils throughout China were collected and heavy metal soil pollution and health risk assessment were evaluated. Northwest region showed the least contaminated, while Middle Yellow River region, south coastal region and Middle Yangtze River were moderately contaminated, especially for Hg and Cd. The urban soils in Middle Yangtze region were significantly contaminated by Cu. Soil pollution by heavy metals continues to pose high carcinogenic and non-carcinogenic risks to the public in some urban areas in China, especially to children and those living in the most heavily polluted regions. Special attention should be paid to the priority control in East Coastal and Middle Yangtze region in order to target the biggest threats to human health. In addition, the protection of vulnerable populations, especially children living in urban areas, should be prioritized. These results would provide valuable information not only for soil quality assessment, but also to enact effective polices to protect human's health.

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Table 1 Class distribution of geoaccumulation index for heavy metals found in the urban areas in China (%).

	Cu	Pb	Zn	Cd	As	Ni	Cr	Hg
0	50.42	54.58	55.73	27.97	76.73	91.72	87.50	31.11
1	41.18	34.86	33.60	25.42	18.87	5.73	10.08	30.37
2	6.30	7.39	7.51	25.85	2.52	1.27	0.40	24.44
3	0.00	2.11	1.98	11.02	1.26	0.64	2.02	10.37
4	0.84	0.70	0.79	5.93	0.63	0.00	0.00	2.22
5	0.84	0.35	0.40	2.12	0.00	0.64	0.00	0.74
6	0.42	0.00	0.00	1.69	0.00	0.00	0.00	0.74

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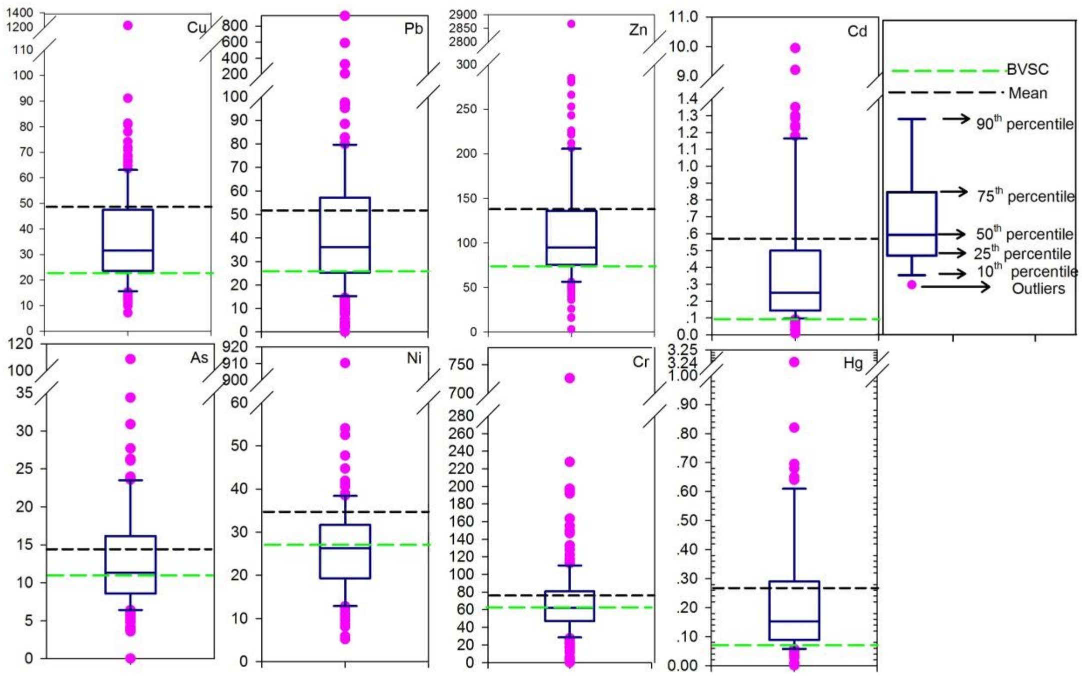

Fig. 1 Boxplots of the heavy metal concentrations (mg/kg) for the urban areas (BVSC: Background values for soils in China).

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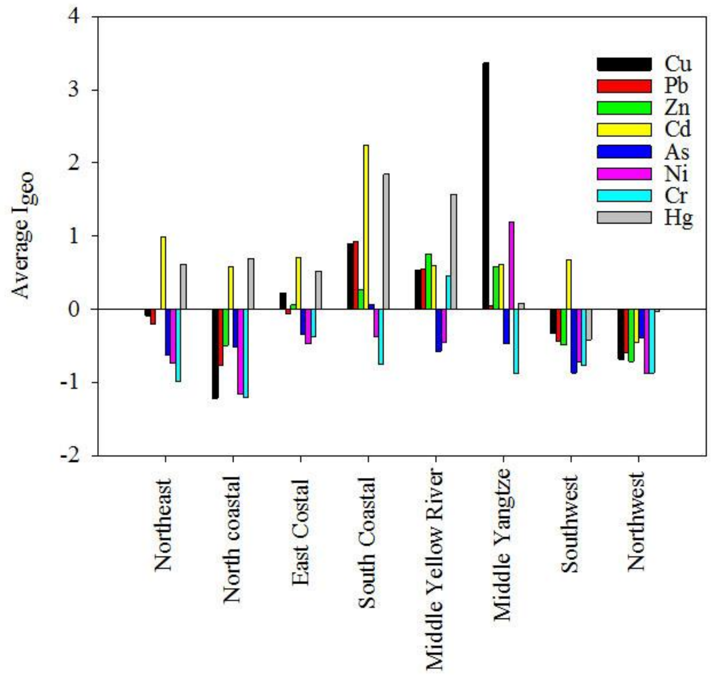

Fig. 2. Average I_{geo} values of heavy metals for different types of urban areas.

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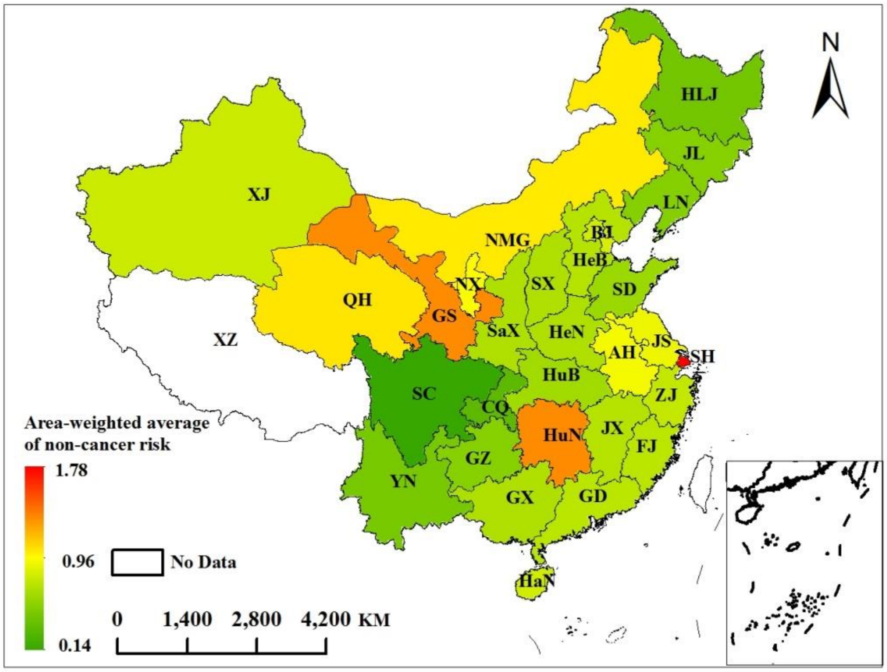

Fig. 3 Spatial distribution of HI in different types of the examined urban areas. **Notes**: The full names of the provinces are Anhui, Beijing, Chongqing, Fujian, Gansu, Guangdong, Guangxi, Guizhou, Hainan, Hebei, Henan, Heilongjiang, Hubei, Hunan, Jilin, Jiangsu, Jiangxi, Liaoning, Neimenggu, Ningxia, Qinghai, Shandong, Shanxi, Shannxi, Shanghai, Sichuan, Tianjin, Xizang, Xinjiang, Yunnan, Zhejiang. Due to lack of data from Taiwan, Hong Kong, Xizang and Macao, these provinces were not considered

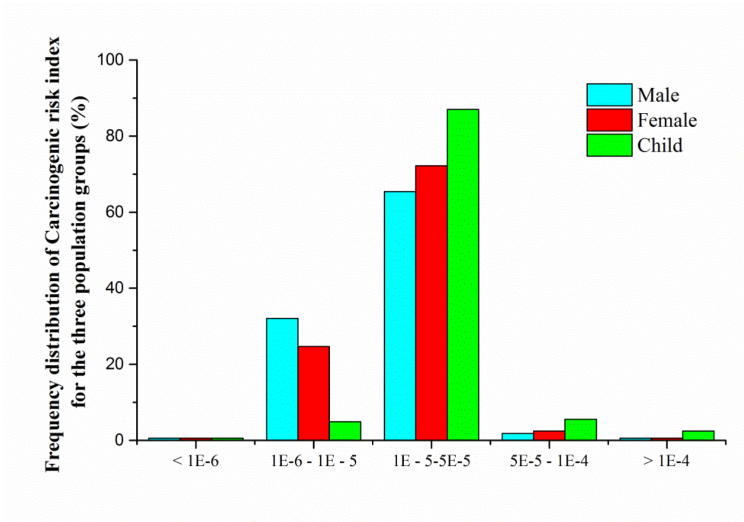

Fig. 4 Frequency distribution of Carcinogenic risk index for the three population groups

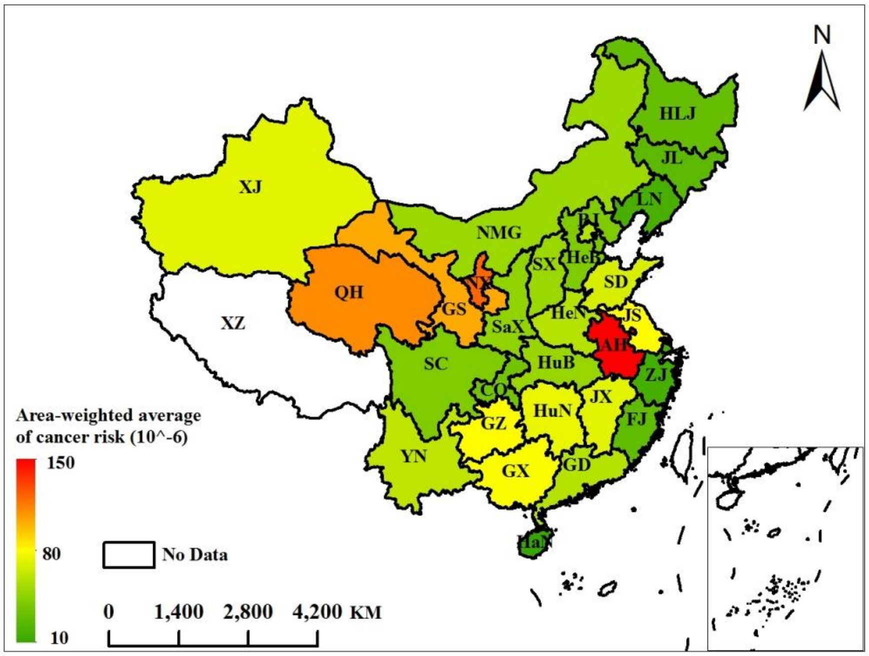

Fig. 5 Spatial distribution of risks of As for the examined urban areas in China. **Notes**: The full names of the provinces are Anhui, Beijing, Chongqing, Fujian, Gansu, Guangdong, Guangxi, Guizhou, Hainan, Hebei, Henan, Heilongjiang, Hubei, Hunan, Jilin, Jiangsu, Jiangxi, Liaoning, Neimenggu, Ningxia, Qinghai, Shandong, Shanxi, Shannxi, Shanghai, Sichuan, Tianjin, Xizang, Xinjiang, Yunnan, Zhejiang. Due to lack of data from Taiwan, Hong Kong, Xinjiang and Macao, these provinces were not considered.