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Health risk assessment of soil heavy metals in housing units built on brownfields in a city in China

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Abstract
Purpose The present paper concerns the patterns of heavy metals (As, Hg, Pb, Cd, Ni, Zn, Cr, and Cu) in surface soil of residential areas located on derelict brownfields in a city in China and the health risks to the residents.
Materials and methods Forty-one surface soil samples were collected from 9 housing units built on different brownfields in a city of Henan Province in China, and the concentrations of heavy metals were measured. Based on the health risk models recommended by the US Environmental Protection Agency (US EPA), carcinogenic and non-carcinogenic health risks of soil heavy metals were assessed.
Results and discussion Compared with the original brownfields, soil heavy metal contents and their health risks in housing units built on brownfields have significantly decreased. The contents and health risks of heavy metals in these housing units are all higher than those in non-brownfield housing units. The result of health risk assessment indicates that there are no non-carcinogenic risks and slight carcinogenic risks for the residents in these housing units. The contribution of hazard quotient caused by arsenic (HQ_{As}) to hazard index (*HI*) is approximately 53%, and the contribution of cancer risk caused by arsenic (CR_{AS}) to total carcinogenic risk (*TCR*) is approximately 80%.
Conclusions The obtained results have confirmed the environmental effects of brownfields and that soil remediation is an essential step for the redevelopment of brownfields. Arsenic is the crucial heavy metal for carcinogenic and non-carcinogenic risk in the housing units of the city, and the highest levels of non-carcinogenic and carcinogenic risks are associated with the route of ingestion of soil for all heavy metals.

Keywords Assessment · Brownfield · Health risk · Heavy metals · Housing unit

1 Introduction

With the rapid growth of cities in China, the industrial structure and urban land use are changing sharply. Many enterprises with high pollution emissions originally located within the city center have relocated to the suburbs due to urban planning and redesigning of these areas. These abandoned areas typically contain high contents of heavy metals originating from past industrial activities. These abandoned, idled, or underutilized industrial and commercial facilities, where expansion or redevelopment is complicated by real or perceived environmental contamination, have been defined as brownfields by the *comprehensive Environmental Response, Compensation and Liability Act* passed by the American government (Congress of the United States 1995). Due to their advantageous geographical positions, almost all brownfields have been redeveloped, primarily as housing units. The redevelopment of these brownfields can provide new jobs, build tax bases, and control urban sprawl (Amekudzi and

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Fomunung 2004). Heavy metals are non-degradable elements and are also mobile, in part, in response to land use change (Ha et al. 2014). Without proper remediation, brownfields would remain a potential source of many pollutants (Melymuk et al. 2013; Capobianco et al. 2014), which might cause adverse effects on the environment and health problems among residents (e.g., the US "the Love Canal crisis" (Seuly 2005) and "the Holland Lekkerker event" (Zhao and Yang 2006)). In recent years, China's media has reported many cases in which the reuse of the brownfields has caused public health problems (Xie and Li 2010; Cheng et al. 2011; Yang et al. 2014).

In comparison with Western countries, which have holistic legislation to identify and regulate the remediation of contaminated land (Wcislo et al. 2002), brownfield regeneration is a relatively new urban issue in China. The compatible laws and technical standards have yet to be determined, which leads to an imperfect brownfield management system. In recent years, many studies have analyzed the current pollution situation and assessed the risks induced by brownfields (Julita et al. 2005; Gallagher et al. 2008a; Gallagher et al. 2008b; Sierra et al. 2010; Albanese et al. 2010; Yang et al. 2010; Qian et al. 2012; Chen and Ma, 2013; Sun et al. 2013; Ren et al. 2014; Qian et al. 2014; Mukwaturi and Lin, 2015; Gallego et al. 2015; Wahsha et al. 2016; Enell et al. 2016; Pan and Li 2016). However, few reports have addressed whether health risks exist after the transformation of brownfields. In this paper, a city in Henan Province, China, was chosen as the study area. The objectives of our study are (1) to establish a general understanding of the concentrations of eight heavy metals (As, Hg, Pb, Cd, Ni, Zn, Cr, and Cu) in surface soil of residential areas located on derelict brownfields (brownfield-residential areas); (2) to identify patterns of metal contamination of soils from these sampling sites; (3) to evaluate the human health risks of these heavy metals to both children and adults via ingestion, inhalation, and dermal contact; and (4) to establish guidelines for community residents and references for the management and remediation of brownfield regeneration in China.

2 Materials and methods

2.1 Sample collection

Six brownfield-residential areas, in four different types of industrial areas, as well as three control sites, were selected as sample sites. Site selection was based on an investigation of brownfield-residential areas in this city, the technology used by the original enterprises on these brownfields, and the national economic industry classification of China. General descriptions of the brownfield-residential areas and control sites are listed in Table 1.

A total of 41 soil samples (0–20 cm) were collected from 9 sample sites. At each sample site, except D3 (a single green field, which we considered a single sampling unit), five surface soil samples (0–20 cm) were collected using a stainless steel spade. Each sample was obtained by mixing five subsamples randomly collected within a 2-m^2 area, for a total sample weight of 0.5–1 kg. Samples were placed in polyethylene bags after stones and plant remnants were removed.

2.2 Laboratory analysis

The soil samples were dried; ground, through a nylon 100 mesh sieve; and digested in a graphite digestion instrument using the mixture solution of concentrated HCl–HNO_3–HF–$HClO_4$. Contents of Cr, Cu, Ni, and Zn in soils were measured by atomic absorption spectrometry (AA-6601F Model, Shimadzu Ltd., Japan), and Cd and Pb were measured using ICP-MS (X-Series II Model, Thermo Fisher Scientific, USA). The contents of As and Hg were determined by atomic fluorescence spectrometry (AFS-930 Model, Haiguang, China) with an HNO_3–HCl digestion system from DB51/T 836-2008 of China. All analytical data were subject to strict quality control. The instruments were calibrated daily using calibration standards. Precision and accuracy were verified using standard reference materials from the National Research Center for Geoanalysis of China [soil, GBW07422 (GSS-8)]. Satisfactory recoveries were obtained for 95.2–105.1%. Differences in heavy metal contents between this study and certified values were <10%. Blank samples for digestion and analysis methods were evaluated in duplicate with each set of samples. The relative deviation of the duplicate samples was <5% in all batch treatments.

In addition, the pH of each soil sample was measured using a Mettler-Toledo pH meter after soil was mixed with deionized water free of CO_2 at a soil to water ratio of 1:2.5 (*w*/*v*).

2.3 Potential health risk assessment

2.3.1 Exposure dose

Resident exposure to the heavy metals in soil can occur via three main pathways: (a) direct ingestion of soil particles, (b) inhalation of resuspended soil particles through the mouth and nose, and (c) dermal absorption of metals in soil particles adhered to exposed skin. For non-carcinogens, the average daily dose (*ADD*) (mg·kg^{-1}·day^{-1}) of potentially toxic heavy metals (As, Hg, Pb, Cd, Ni, Zn, Cr, and Cu) received through

Table 1 Information on the sample sites

Sample sites		Factory establishment time	Abbr. name	Residential areas' establishment time	Area/ha	Remediation method
Textile industry	Textile factory	1981	TF	2006	7.94	OSC
	Dyeing factory	1962	DF	2009	1.93	CF
Printing industry	Printing factory	1978	PF	2011	4.00	OSC
Chemical industry	Daily chemical factory	1955	DCF	2010	7.85	CF
Equipment manufacture industry	Boiler factory	1968	BF	2008	3.40	OSC
	Machinery factory	1953	MF	2003	1.53	OSC
Control sites[a]		–	D1	2009	6.70	NR
		–	D2	2006	2.58	NR
		1953	D3	–	1.13	NR

OSC out-soil covering, *CF* clean fill, *NR* no remediation

[a] D1 is a residential area built on farmland, D2 is a residential area built on land previously used as residential land, D3 is now the location of a warehouse and was originally part of the same brownfield as XT

each of the three paths were calculated using Eqs. 1–3 (US EPA 1989, 1996):

$$ADD_{ing} = \frac{c \cdot \mathrm{Ing}R \cdot CF \cdot EF \cdot ED}{BW \cdot AT} \quad (1)$$

$$ADD_{\mathrm{inh}} = \frac{c \cdot \mathrm{Inh}R \cdot EF \cdot ED}{PEF \cdot BW \cdot AT} \quad (2)$$

$$ADD_{\mathrm{derm}} = \frac{c \cdot SA \cdot CF \cdot SL \cdot ABS \cdot EF \cdot ED}{BW \cdot AT} \quad (3)$$

For carcinogens, the lifetime average daily dose (mg·kg^{-1}·day^{-1}) for As, Cd, Ni, and Cr, used to assess cancer risk in children, has been calculated as a weighted average daily dose for each exposure route as shown in Eqs. 4–6 (US EPA 1996, 2002; Ferreira Baptista and De Miguel 2005):

$$\mathrm{LADD}_{\mathrm{ing}} = \frac{c \cdot CF \cdot EF}{AT} \times \left(\frac{\mathrm{Ing}R_{\mathrm{child}} \cdot ED_{\mathrm{child}}}{BW_{\mathrm{child}}} + \frac{\mathrm{Ing}R_{\mathrm{adult}} \cdot ED_{\mathrm{adult}}}{BW_{\mathrm{adult}}} \right) \quad (4)$$

$$\mathrm{LADD}_{\mathrm{inh}} = \frac{c \cdot EF}{PEF \cdot AT} \times \left(\frac{\mathrm{Inh}R_{\mathrm{child}} \cdot ED_{\mathrm{child}}}{BW_{\mathrm{child}}} + \frac{\mathrm{Inh}R_{\mathrm{adult}} \cdot ED_{\mathrm{adult}}}{BW_{\mathrm{adult}}} \right) \quad (5)$$

$$\mathrm{LADD}_{\mathrm{derm}} = \frac{c \cdot CF \cdot EF \cdot SL \cdot ABS}{AT} \times \left(\frac{SA_{\mathrm{child}} \cdot ED_{\mathrm{child}}}{BW_{\mathrm{child}}} + \frac{SA_{\mathrm{adult}} \cdot ED_{\mathrm{adult}}}{BW_{\mathrm{adult}}} \right) \quad (6)$$

where c is the content of heavy metals in soil (mg·kg^{-1}).

Other variables include Ing$R$, ingestion rate, in this study, 200 mg day^{-1} for children and 100 mg day^{-1} for adults (Ferreira Baptista and De Miguel 2005); Inh$R$, inhalation rate, in this study, 5 m^3 day^{-1} for children and 15 m^3 day^{-1} for adults (Environmental site assessment guideline 2009); CF, conversion factor, in this study, 1×10^{-6} kg mg^{-1} (Lim et al. 2008); *EF*, exposure frequency, in this study, 365 day a^{-1} (Ferreira Baptista and De Miguel 2005); *ED*, exposure duration, in this study, 6 a for children and 24 a for adults (Ferreira Baptista and De Miguel 2005; Li et al. 2013); *BW*, average body weight, in this study, 15 kg for children and 60 kg for adults (Environmental site assessment guideline 2009); *AT*, averaging time, in this study, *ED* × 365 days for non-carcinogens and 70 × 365 days for carcinogens (Ferreira Baptista and De Miguel 2005); *PEF*, particle emission factor, in this study, 1.36×10^9 m^3 kg^{-1} (Li et al. 2013); *SA*, exposed skin area, in this study, 1600 cm^2 for children and 4350 cm^2 for adults (Environmental site assessment guideline 2009); *SL*, skin adherence factor, in this study, 0.2 mg (cm^2 day)$^{-1}$ (Ferreira Baptista and De Miguel 2005); and *ABS*, dermal absorption factor, in this study, 0.001 (Ferreira Baptista and De Miguel 2005; Lim et al. 2008; Li et al. 2013).

2.3.2 Risk characterization

The *ADD* for the three exposure pathways (ADD_{ing}, ADD_{inh}, and ADD_{derm}) were calculated as above, and the potential non-carcinogenic and carcinogenic risks for individual metals were calculated as follows (Lim et al. 2008; Man et al. 2010; Zheng et al. 2010):

$$\mathrm{HQ_i} = \sum_{j=1}^{3} \frac{ADD_{ij}}{RfD_{ij}} \quad (7)$$

$$HI = \sum_{i=1}^{8} HQ_i \quad (8)$$

Table 2 Reference doses for non-carcinogenic metals and slope factors for carcinogenic metals

Element	*RfD*/mg·(kg·day) $^{-1}$			*SF*/(kg·day)·mg^{-1}		
	Oral	Dermal	Inhal.	Oral	Dermal	Inhal.
As	3.00×10^{-4}	3.00×10^{-4}	1.23×10^{-4}	1.50	1.50	4.30×10^{-3}
Hg	3.00×10^{-4}	2.40×10^{-5}	3.00×10^{-4}	n.d.	n.d.	n.d.
Cd	1.00×10^{-3}	1.00×10^{-5}	1.00×10^{-3}	6.10	6.10	1.80×10^{-3}
Pb	3.50×10^{-3}	5.25×10^{-4}	3.52×10^{-3}	n.d.	n.d.	n.d.
Zn	0.30	6.00×10^{-2}	0.30	n.d.	n.d.	n.d.
Cu	4.00×10^{-2}	1.20×10^{-2}	4.00×10^{-2}	n.d.	n.d.	n.d.
Ni	2.00×10^{-2}	5.40×10^{-3}	2.06×10^{-2}	n.d.	n.d.	0.84
Cr(VI)	3.00×10^{-3}	6.00×10^{-5}	2.86×10^{-5}	n.d.	n.d.	42.0

n.d. not determined

Table 3 Statistics of soil metal contents ($n = 5$) in different communities/mg·kg^{-1}

Element		Textile industry		Printing industry	Chemical industry	Equipment manufacture industry		Control sites			Reference background values [a]
		TF	DF	PF	DCF	BF	MF	D1	D2	D3[b]	
As	Max	20.19	9.95	9.76	9.93	10.69	9.39	7.06	7.17	–	4.93
	Min	8.40	4.70	7.17	6.67	8.70	5.91	4.78	5.00	–	
	Mean	11.53	7.31	8.36	8.42	9.51	7.40	5.72	5.96	8.19	
	SD	4.90	1.89	1.12	1.21	0.81	1.18	0.88	0.87	–	
	Cv (%)	42.50	25.86	13.40	14.37	8.52	15.95	15.38	14.60	–	
Hg	Max	0.14	0.05	0.10	2.07	0.61	0.26	0.31	0.18	–	0.04
	Min	0.02	0.01	0.02	0.03	0.02	0.01	0.01	0.04	–	
	Mean	0.06	0.02	0.06	0.33	0.06	0.09	0.09	0.10	0.68	
	SD	0.04	0.02	0.03	0.10	0.23	0.82	0.12	0.05	–	
	Cv (%)	66.67	100.00	50.00	30.30	383.33	248.48	133.33	50.00	–	
Cd	Max	0.61	0.49	0.93	0.62	0.75	0.47	0.58	0.47	–	0.24
	Min	0.35	0.28	0.42	0.38	0.40	0.36	0.33	0.26	–	
	Mean	0.44	0.41	0.63	0.50	0.48	0.42	0.43	0.36	0.74	
	SD	0.11	0.08	0.19	0.05	0.14	0.11	0.09	0.07	–	
	Cv (%)	25.00	19.51	30.16	10.00	29.17	22.00	20.93	19.44	–	
Pb	Max	24.18	13.23	29.82	22.25	52.81	130.28	11.24	49.89	–	16.70
	Min	12.15	4.14	8.70	7.71	13.03	11.22	3.41	13.99	–	
	Mean	17.15	7.35	18.10	12.62	25.37	39.02	8.09	25.57	60.42	
	SD	5.05	3.18	7.61	4.98	15.33	47.22	3.10	15.74	–	
	Cv (%)	29.45	43.27	42.04	39.46	60.43	121.01	38.32	61.56	–	
Ni	Max	28.58	23.22	18.49	29.90	22.23	21.51	23.44	18.33	–	15.77
	Min	16.85	14.03	13.99	16.65	18.67	16.14	16.83	14.14	–	
	Mean	21.88	19.79	16.96	23.26	20.33	18.86	20.37	16.43	26.49	
	SD	4.22	3.64	1.71	4.71	4.71	2.19	2.83	1.75	–	
	Cv (%)	19.28	18.42	10.08	20.24	20.24	11.60	13.88	10.68	–	
Zn	Max	93.71	59.46	84.25	151.74	109.92	84.61	68.53	58.43	–	34.45
	Min	43.82	38.60	46.87	36.54	45.47	45.80	38.60	30.90	–	
	Mean	60.30	47.06	63.74	63.51	64.81	65.73	49.00	42.21	117.17	
	SD	17.90	7.74	13.75	43.61	24.04	16.19	10.60	9.48	–	
	Cv (%)	29.69	16.44	21.57	68.66	37.10	24.63	21.63	22.45	–	
Cr	Max	51.52	45.30	44.11	357.57	51.49	77.31	85.88	92.42	–	17.47
	Min	28.54	9.27	25.36	30.25	26.23	27.32	21.68	17.54	–	
	Mean	38.87	29.96	36.82	97.18	36.90	53.39	46.27	38.08	67.51	
	SD	10.34	14.55	7.23	128.75	9.66	21.55	21.20	27.26	–	
	Cv (%)	26.61	48.57	19.63	132.48	26.19	40.37	45.82	71.58	–	
Cu	Max	32.81	119.28	30.56	25.43	75.59	44.99	48.74	17.06	–	13.69
	Min	13.29	9.96	12.62	17.27	17.26	13.99	10.36	10.99	–	
	Mean	21.59	31.64	20.28	20.67	27.95	28.21	21.75	13.59	71.04	
	SD	7.19	43.03	6.13	2.81	23.37	12.14	13.77	2.49	–	
	Cv (%)	33.32	135.99	30.24	13.61	83.61	43.05	63.31	18.30	–	

CV coefficient of variation

[a] The reference background values were calculated based on the references of Du et al. 2008; Ma et al. 2011; Tong 2013; and the number of samples of As, Hg, Cd, Pb, Ni, Zn, Cr, and Cu used for statistics are 6, 4, 5, 5, 3, 3, 3, and 3, respectively

[b] There were not sufficient data from D3 for statistics, as there was only one greenfield in D3

$$CR_i = \sum_{j=1}^{3} ADD_{ij} \cdot SF_{ij} \quad (9)$$

$$TCR = \sum_{i=1}^{4} CR_i \quad (10)$$

The reference dose (*RfD*) (mg·kg^{-1}·day^{-1}) is an estimation of maximum permissible risks to human populations through daily exposure. This dose is calculated by considering a sensitive group (children) during their lifetime. Non-carcinogenic risk is represented in terms of hazard quotient (*HQ*) for a single substance or hazard index (*HI*) for multiple substances and/or exposure pathways. If the exposure level of a substance exceeds the corresponding *RfD*, i.e., *HQ* > 1, there may be concern for potential non-carcinogenic effects (Kong et al. 2011). The estimated value for the carcinogenic risk (*CR*) and total carcinogenic risk (*TCR*) is the probability that an individual will develop any type of cancer from a lifetime exposure to carcinogenic hazards. For carcinogenic risk, the dose is multiplied by the corresponding slope factor (*SF*) to produce an estimate of cancer risk. In general, the US EPA recommends that a *CR* and *TCR* lower than 1×10^{-6} be regarded as negligible, whereas a *CR* and *TCR* above 1×10^{-4} is likely to be harmful to human beings. Some experts propose that the acceptable or tolerable risk for regulatory purposes is in the range of 1×10^{-6}–1×10^{-4} (Ferreira Baptista and De Miguel 2005; US EPA 1989, 2002).

Based on data from Chinese locations (Environmental site assessment guideline 2009) and previously published results (Lim et al. 2008), the values for *RfD* and *SF* are listed in Table 2. Most of the data does not need further comments but additional argumentation is required in the case of Cr. The presence of Cr(VI) in natural environments requires a rather high redox potential, over 700 mV for a pH of around 5.0, but a redox potential of 400 mV for pH 7.0 to 8.0 is sufficient for Cr(VI) to dominate in the system (Gržetić and Ghariani 2008). The redox potential in soil usually varies from a minimum of −550 to maximum of 700 mV, but aerated soil most frequently has a redox potential up to 400 mV (Michel et al. 2004). Therefore, it is assumed that Cr (VI) in the soils of these housing units was the dominating chromium species since the measured soil pH was around 7.86 (Table S1, Electronic Supplementary Material).

3 Results and discussion

3.1 Heavy metal contamination of soil

Table 3 summarizes the contents of soil heavy metals in these sampling sites. The average contents of soil heavy metals in different sampling sites are shown in Fig. 1. As summarized in Table 3 and Fig. 1, the heavy metal contents of these sites have the following characteristics: (1) the average contents of soil heavy metals in all housing units are higher than their reference background values. There are exceptions, including the Pb contents at DF, DCF, and D1; the Hg content at DF; and the Cu content at D2, which are lower than their background

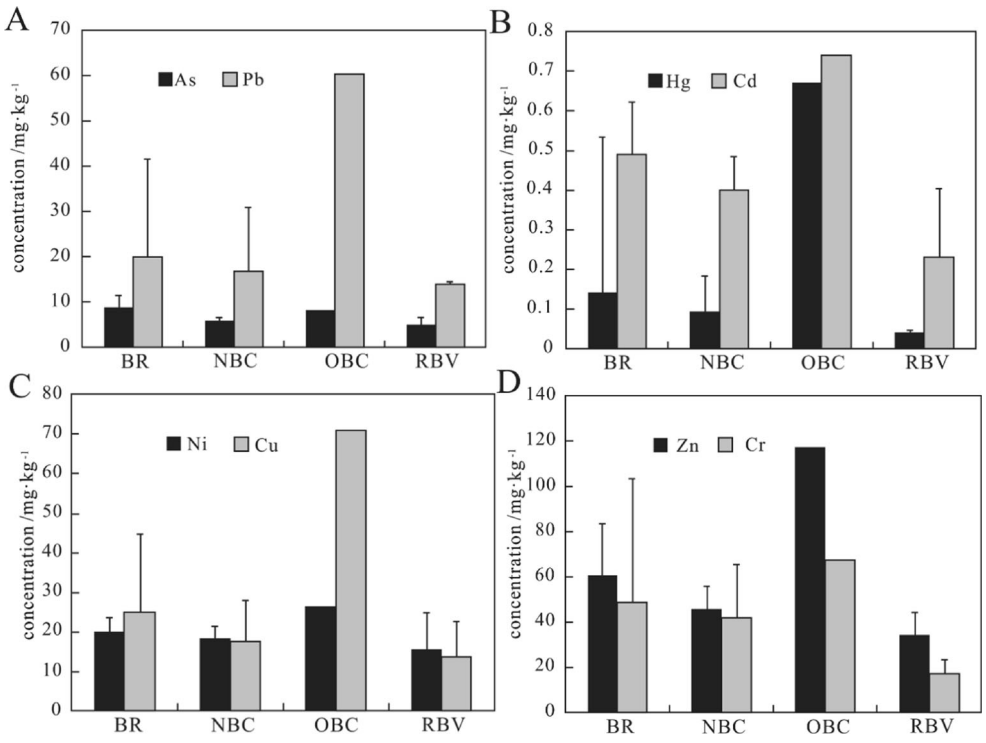

Fig. 1 Average contents of soil heavy metals in different sample sites: **a** As and Pb, **b** Hg and Cd, **c** Ni and Cu, **d** Zn and Cr. BR=Brownfield-residential; NBC=Non-brownfield control site; OBC=Original brownfield control site; RBV=Reference background value; Bars stand for standard deviation

Table 4 Indexes of non-carcinogenic risk for children

Residential areas	*HQ*								*HI*
	As	Hg	Cd	Pb	Ni	Zn	Cr	Cu	
DCF	0.39	4.01×10^{-3}	6.54×10^{-3}	4.86×10^{-2}	1.56×10^{-2}	2.85×10^{-3}	0.33	6.93×10^{-3}	0.80
BF	0.42	2.78×10^{-3}	7.37×10^{-3}	9.77×10^{-2}	1.36×10^{-2}	2.90×10^{-3}	0.12	9.37×10^{-3}	0.68
TF	0.51	2.51×10^{-3}	6.84×10^{-3}	6.60×10^{-2}	1.47×10^{-2}	2.70×10^{-3}	0.13	7.23×10^{-3}	0.74
PF	0.37	2.57×10^{-3}	9.81×10^{-3}	6.97×10^{-2}	1.14×10^{-2}	2.86×10^{-3}	0.12	6.80×10^{-3}	0.60
MF	0.33	1.51×10^{-2}	7.67×10^{-3}	0.15	1.26×10^{-2}	2.94×10^{-3}	0.18	9.45×10^{-3}	0.71
DF	0.33	1.00×10^{-3}	6.26×10^{-3}	2.83×10^{-2}	1.33×10^{-2}	2.11×10^{-3}	0.10	1.06×10^{-2}	0.49
D1	0.26	4.11×10^{-3}	6.58×10^{-3}	3.11×10^{-3}	1.37×10^{-2}	2.20×10^{-3}	0.16	7.29×10^{-3}	0.48
D2	0.27	4.70×10^{-3}	5.64×10^{-3}	9.84×10^{-2}	1.10×10^{-2}	1.89×10^{-3}	0.13	4.55×10^{-3}	0.52
D3	0.36	3.04×10^{-2}	1.15×10^{-2}	0.23	1.78×10^{-2}	5.25×10^{-3}	0.23	2.38×10^{-2}	0.84

values. (2) The As and Zn contents at brownfield-residential areas are generally higher than at non-brownfield control sites (D1 and D2). The Cd, Pb, Ni, and Cu contents at most brownfield-residential areas are higher as well. Nevertheless, though the contents of Hg and Cr at some brownfield-residential areas are lower than those at control sites, their average contents are still higher than at control sites. (3) Site D3, now used as a warehouse, was originally part of the same brownfield as MF. D3 has higher heavy metal contents than do any other brownfield-residential and non-brownfield control sites, except that it has lower As and Cr contents than do a few brownfield-residential areas. (4) The two residential control sites possess similar heavy metal contents, except that the Pb content of D2 (a residential area built on old residential land) is higher than that of D1 (a residential area built on farmland). (5) Of the two sites originally from the same brownfield, D3 (now used as a warehouse) has higher heavy metal content than MF (now used as a residential quarter).

When residential areas were built on brownfields, out-soil backfill and covering were applied. Nevertheless, there was still the possibility that the contaminated soil had not been removed thoroughly. Heavy metals remaining in the original brownfields could reach the surface soil, accompanying the rise of pore water, and become concentrated there. Therefore, the heavy metal contents in the brownfield-residential areas are higher than those in the non-contaminated residential area. Due to different properties of factories on the original brownfield, the heavy metal contents in different residential areas vary accordingly. For example, the soil underlying a textile factory should be contaminated by arsenic-containing dyes. This condition may explain why the arsenic content in the TF residential area is higher. Because residential area D2 was built on old residential land, the elevated Pb content may have been caused by household waste and Pb-containing house paints (Mielke et al. 1999).

3.2 Non-carcinogenic risk assessment of soil heavy metals

The non-carcinogenic risks (*HQ* and *HI*) of all heavy metals through three exposure routes for local residents (children and adults), calculated in accordance with Eqs. 1–8, were determined (Tables 4 and 5). According to Tables 4 and 5, the *HQ*

Table 5 Indexes of non-carcinogenic risk for adults

Residential areas	*HQ*								*HI*
	As	Hg	Cd	Pb	Ni	Zn	Cr	Cu	
DCF	5.49×10^{-2}	6.16×10^{-4}	1.49×10^{-3}	7.19×10^{-3}	2.26×10^{-3}	4.16×10^{-4}	6.18×10^{-2}	1.00×10^{-3}	0.13
BF	6.03×10^{-2}	4.26×10^{-4}	1.68×10^{-3}	1.44×10^{-2}	1.98×10^{-3}	4.25×10^{-4}	2.35×10^{-2}	1.35×10^{-3}	0.10
TF	7.31×10^{-2}	3.85×10^{-4}	1.56×10^{-3}	9.76×10^{-3}	2.13×10^{-3}	3.95×10^{-4}	2.47×10^{-2}	1.05×10^{-3}	0.11
PF	5.29×10^{-2}	3.95×10^{-4}	2.23×10^{-3}	1.03×10^{-2}	1.65×10^{-3}	4.18×10^{-4}	2.34×10^{-2}	0.98×10^{-3}	0.09
MF	4.68×10^{-2}	2.32×10^{-3}	1.75×10^{-3}	2.22×10^{-2}	1.83×10^{-3}	4.31×10^{-4}	3.39×10^{-2}	1.37×10^{-3}	0.11
DF	4.63×10^{-2}	1.54×10^{-4}	1.43×10^{-3}	4.19×10^{-3}	1.92×10^{-3}	3.08×10^{-4}	1.90×10^{-2}	1.53×10^{-3}	0.08
D1	3.63×10^{-2}	6.31×10^{-4}	1.50×10^{-3}	4.60×10^{-3}	1.98×10^{-3}	3.21×10^{-4}	2.94×10^{-2}	1.05×10^{-3}	0.07
D2	3.78×10^{-2}	7.23×10^{-4}	1.28×10^{-3}	1.46×10^{-2}	1.60×10^{-3}	2.76×10^{-4}	2.42×10^{-2}	0.66×10^{-3}	0.08
D3	5.14×10^{-2}	4.67×10^{-3}	2.61×10^{-3}	3.44×10^{-2}	2.58×10^{-3}	7.68×10^{-4}	4.29×10^{-2}	3.44×10^{-3}	0.14

Table 6 Relative contribution of each exposure pathway to non-carcinogenic risks for children and adults

Residential areas	Children/%			Adults /%		
	Oral	Dermal	Inhal.	Oral	Dermal	Inhal.
DCF	96.50	3.13	0.19	83.94	15.67	0.39
BF	98.08	1.78	0.15	90.86	8.95	0.20
TF	98.19	1.65	0.16	91.43	8.38	0.19
PF	97.84	2.00	0.15	89.78	10.00	0.22
MF	97.47	2.39	0.14	88.01	11.73	0.26
DF	97.93	1.91	0.16	90.23	9.55	0.22
D1	97.02	2.81	0.17	86.11	13.56	0.32
D2	97.56	2.29	0.15	88.46	11.29	0.25
D3	97.42	2.45	0.13	87.76	11.99	0.25

and *HI* values of soil heavy metals in each sampling site were lower than 1, which indicates that there were no non-carcinogenic risks in these sites. Non-carcinogenic risk is higher for children because of their low tolerance to toxins as well as their inadvertent ingestion of considerable quantities of soil through oral pathways (Zhao et al. 2014). The *HI* value of heavy metals for children is exactly six times higher than for adults, and similar observations have been reported in other places (Zheng et al. 2010; Morra et al. 2006).

For children, the contribution of HQ_{As} to *HI* ranged from 42.86 to 68.92%, with an average of 55.29%. For adults, the contribution of HQ_{As} to *HI* ranged from 36.71 to 66.45%, with an average of 50.53%. Arsenic posed the greatest non-carcinogenic risk in the residential areas of this city. As illustrated in Table 6, for non-carcinogenic risks, the risks with different exposure pathways vary greatly, generally in the order of ingestion > dermal absorption > inhalation. The contribution of ingestion exposure to *HI* for children and adults was approximately 97.56 and 88.51%, respectively. The highest levels of non-carcinogenic risks caused by all eight heavy metals were associated with the route of ingestion, which has been widely regarded as one of the key metal exposure pathways (Mielke et al. 1999; Rasmussen et al. 2001).

Except for the DF, the *HI* values of soil heavy metals in brownfield-residential areas are higher than those in non-brownfield control sites. Due to different main pollutants and remediation methods at brownfield-residential areas, the *HI* values in different types of brownfield-residential areas vary accordingly, generally in the order of chemical industry > equipment manufacture industry > textile industry > printing industry. In a comparison of two sites originally from the same brownfield, MF (now used for as a residential quarter) has a lower *HI* value than D3 (now used as a warehouse). For instance, the *HI* value for children and adults of MF amounts to 84.52 and 78.57% of D3, respectively. This finding indicates that the restoration activities carried out at the brownfield site have been beneficial in terms of achieving a risk reduction.

3.3 Carcinogenic risk assessment of soil heavy metals

The carcinogenic risks (*CR* and *TCR*) of As, Cd, Ni, and Cr through three exposure routes for local residents, calculated in accordance with Eqs. (9) and (10), were determined (Tables 7 and 8). As shown in Tables 7 and 8, for each sampling site, the *CR* values of Ni and Cr for adults and children are all lower than the soil remediation criteria recommended by the US EPA (10^{-6}). This finding suggests that the carcinogenic risk exposure from Ni and Cr in soils may be negligible. However, the *CR* values for As and Cd and the *TCR* values for As, Cd, Ni, and Cr for adults and children met the soil remediation criteria, even though they were lower than the lenient standard proposed by some experts (10^{-6}–10^{-4}). However, the carcinogenic risks of heavy metals are in need of attention for pollution control. Compared to children, the carcinogenic risk for adults due to heavy metal exposure from soil is lower. However, for children, the contribution of CR_{As} to *TCR* ranged from 71.80 to 85.84%, with the average of 78.90%;

Table 7 Indexes of carcinogenic risk for children

Residential areas	*CR*				*TCR*
	As	Cd	Ni	Cr	
DCF	2.34×10^{-5}	4.63×10^{-6}	4.68×10^{-9}	6.85×10^{-7}	2.87×10^{-5}
BF	2.56×10^{-5}	5.22×10^{-6}	4.09×10^{-9}	2.60×10^{-7}	3.11×10^{-5}
TF	3.11×10^{-5}	4.85×10^{-6}	4.41×10^{-9}	2.74×10^{-7}	3.62×10^{-5}
PF	2.25×10^{-5}	6.95×10^{-6}	3.41×10^{-9}	2.59×10^{-7}	2.97×10^{-5}
MF	1.99×10^{-5}	5.43×10^{-6}	3.80×10^{-9}	3.76×10^{-7}	2.57×10^{-5}
DF	1.97×10^{-5}	4.44×10^{-6}	3.98×10^{-9}	2.11×10^{-7}	2.43×10^{-5}
D1	1.54×10^{-5}	4.66×10^{-6}	4.10×10^{-9}	3.26×10^{-7}	2.04×10^{-5}
D2	1.61×10^{-5}	3.99×10^{-6}	3.31×10^{-9}	2.68×10^{-7}	2.03×10^{-5}
D3	2.19×10^{-5}	8.12×10^{-6}	5.33×10^{-9}	4.76×10^{-7}	3.05×10^{-5}

Table 8 Indexes of carcinogenic risk for adults

Residential areas	*CR*				*TCR*
	As	Cd	Ni	Cr	
DCF	8.74×10^{-6}	1.68×10^{-6}	1.39×10^{-9}	2.03×10^{-7}	1.04×10^{-5}
BF	9.30×10^{-6}	1.89×10^{-6}	1.22×10^{-9}	7.73×10^{-8}	1.13×10^{-5}
TF	1.13×10^{-5}	1.76×10^{-6}	1.31×10^{-9}	8.14×10^{-8}	1.31×10^{-5}
PF	8.17×10^{-6}	2.52×10^{-6}	1.01×10^{-9}	7.71×10^{-8}	1.08×10^{-5}
MF	7.22×10^{-6}	1.97×10^{-6}	1.13×10^{-9}	1.12×10^{-7}	9.31×10^{-6}
DF	7.14×10^{-6}	1.61×10^{-6}	1.18×10^{-9}	6.27×10^{-8}	8.81×10^{-6}
D1	5.59×10^{-6}	1.69×10^{-6}	1.22×10^{-9}	9.69×10^{-8}	7.38×10^{-6}
D2	5.83×10^{-6}	1.45×10^{-6}	9.83×10^{-10}	7.97×10^{-8}	7.36×10^{-6}
D3	7.93×10^{-6}	2.94×10^{-6}	1.58×10^{-9}	1.41×10^{-7}	1.10×10^{-5}

for adults, the contribution of CR_{As} to *TCR* ranged from 72.09 to 85.98%, with an average of 79.14%. Therefore, arsenic poses the greatest carcinogenic risk in these residential areas. As summarized in Table 9, for carcinogenic risks, the contribution of ingestion exposure accounted for 98.28 and 98.07% of the *TCR*, respectively. The highest levels of carcinogenic risks were associated with the route of ingestion of soil for all the heavy metals.

For both children and adults, the carcinogenic risks of As, Cd, Ni, and Cr in brownfield-residential areas remain higher than those at the non-brownfield control sites. *TCR* values for different types of brownfield-residential areas decreased in the order of textile industry > printing industry > chemical industry > equipment manufacturing industry. A comparison of two sites originally from the same brownfield revealed that MF (now used as a residential quarter) has a lower *TCR* value than D3 (now used as a warehouse). For instance, the *TCR* value for residents of MF amounts to 84% found at D3.

The elevated health risk of soil arsenic is most likely a consequence of coal combustion. Arsenic in coal is typically 2–82 mg kg^{-1} but can reach as high as 1500 mg kg^{-1}. Combustion releases approximately 50% of this arsenic into the atmosphere (Bertine and Goldberg 1971). Airborne ash and residues contain large amounts of arsenic (Smith et al. 1998). This city is heavily dependent on burning coal for home heating and industrial power. From 2008 to 2011, the coal consumption of the city increased from 286×10^4 t to 619×10^4 t, with an average annual growth rate of 21.33%. Arsenic emitted from the industrial process may be enriched in soil by atmospheric deposition (dry and wet). Additionally, the fertilizer plants of this city discharge a large amount of arsenic as waste. For example, the average concentration of arsenic in discharged wastewater from a plant was 0.5 mg L^{-1} in the last decade, far beyond the fifth grade criteria (0.1 mg L^{-1}) of the national standard for surface water quality of China (GB 3838–2002). Therefore, soil in the sewage irrigation area formed by the wastewater from the plant is seriously polluted with arsenic (Han et al. 2006). Finally, arsenic aerosols discharged from the fertilizer plant and the topsoil of the sewage irrigation area can migrate to urban areas, which to some extent aggravates soil arsenic pollution.

Table 9 Relative contribution of each exposure pathway to carcinogenic risks for children and adults

Residential areas	Children/%			Adults /%		
	Oral	Dermal	Inhal.	Oral	Dermal	Inhal.
DCF	97.19	0.40	2.40	97.18	0.85	1.98
BF	98.74	0.41	0.85	98.45	0.86	0.70
TF	98.82	0.41	0.77	98.51	0.86	0.63
PF	98.70	0.41	0.88	98.42	0.86	0.73
MF	98.11	0.41	1.48	97.93	0.85	1.21
DF	98.71	0.41	0.88	98.42	0.86	0.73
D1	98.36	0.41	1.23	97.82	0.85	1.33
D2	98.21	0.41	1.38	98.05	0.85	1.10
D3	97.64	0.41	1.95	97.85	0.85	1.30

4 Conclusions

The contents and health risk assessments of heavy metals (As, Hg, Pb, Cd, Ni, Zn, Cr, and Cu) in surface soils, collected from housing units built on brownfields in a city of Henan Province, were investigated and assessed. Heavy metal contents of brownfield-residential areas are higher than in the non-brownfield control sites. Brownfield-residential areas were found comparatively less contaminated after soil remediation.

With respect to non-carcinogenic effects, the *HI* values for eight soil heavy metals in each residential area are all lower than the safe level of 1. However, arsenic poses higher risk values for the residents, especially for children than adults.

The exposure pathway that resulted in the highest levels of non-carcinogenic risk for the residents is ingestion. The carcinogenic risk levels for As, Cd, Cr, and Ni were slightly higher than the standards suggested by the US EPA, which indicates that slight carcinogenic risks might exist for the residents in these residential areas. The highest level of carcinogenic risk is associated with the ingestion of soil for all the heavy metals.

In summary, heavy metals in the soil of brownfield-residential areas may bring a certain degree of carcinogenic health risk, and arsenic poses the highest potential risk. Therefore, the results suggest that soil remediation is an essential step for the redevelopment of brownfields.

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