Contamination and risk of heavy metals in soils and sediments from a typical plastic waste recycling area in North China

Zhenwu Tang^a, Lianzhen Zhang^a, Qifei Huang^{b,*}, Yufei Yang^b, Zhiqiang Nie^b,
Jiali Cheng^{c,**}, Jun Yang^d, Yuwen Wang^a, Miao Chai^a

^a MOE Key Laboratory of Regional Energy and Environmental Systems Optimization, Environmental Research Academy, North China Electric Power University, Beijing 102206, China

^b State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

^c National Institute for Nutrition and Health, Chinese Center for Disease Control and Prevention, Beijing 100021, China

^d Center for Environmental Remediation, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

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ABSTRACT

Plastic wastes are increasingly being recycled in many countries. However, available information on the metals released into the environment during recycling processes is rare. In this study, the contamination features and risks of eight heavy metals in soils and sediments were investigated in Wen'an, a typical plastic recycling area in North China. The surface soils and sediments have suffered from moderate to high metal pollution and in particular, high Cd and Hg pollution. The mean concentrations of Cd and Hg were 0.355 and 0.408 mg kg⁻¹, respectively, in the soils and 1.53 and 2.10 mg kg⁻¹, respectively, in the sediments. The findings suggested that there is considerable to high potential ecological risks in more than half of the soils and high potential ecological risk in almost all sediments. Although the health risk levels from exposure to soil metals were acceptable for adults, the non-carcinogenic risks to local children exceeded the acceptable level. Source assessment indicated that heavy metals in soils and sediments were mainly derived from inputs from poorly controlled plastic waste recycling operations in this area. The results suggested that the risks associated with heavy metal pollution from plastic waste recycling should be of great concern.

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1. Introduction

The quantities of end-of-life plastics from households and industries have increased markedly in recent decades (Thompson et al., 2009). Plastic waste treatment is becoming a vital task worldwide because these wastes are resistant to degradation and are leading to various environmental problems, including plastic pollution in the ocean (Rochman et al., 2013; Jambeck et al., 2015). In the past, plastic waste was usually landfilled together with municipal solid waste. Low sustainability, increasing cost and decreasing space for landfills are forcing consideration of alternative options for plastic waste disposal. Recycling has become the primary policy for treating plastic waste in many countries (Al-Salem

et al., 2009; Hopewell et al., 2009). Material recycling is becoming the key disposal method for plastic waste in developing countries because of the relatively low investment and simple technology requirements, although energy recovery and chemical recycling are encouraged in some developed countries (Al-Salem et al., 2009; Hopewell et al., 2009).

Many plastic wastes contain or are contaminated with harmful chemicals. It was reported that the chemical ingredients of more than 50% of plastics are hazardous (Lithner et al., 2011). Some hazardous chemicals are also widely used as specific targeted additives in plastic products. For example, brominated organics and phthalate esters (PAEs) have been extensively applied in plastic products as flame retardants and plasticizers, respectively. These additives also include other flame retardants and plasticizers, pigments, stabilizers, and reinforcement components, which are composed of heavy metals or metal compounds (Dimitrakakis et al., 2009; Li and Shi, 2011; Nakashima et al., 2012; Rochman et al., 2013). In some products, the contents of these dangerous additives are high or extremely high (Morf et al., 2007; Dimitrakakis et al., 2009; Stenvall et al., 2013). For example, the upper concentration ranges of Pb and Sb were reported at

* Corresponding author. Fax: +86 10 84913903.

** Corresponding author. Fax: +86 10 83157181.

E-mail addresses: zwtang@ncepu.edu.cn (Z. Tang), 446329773@qq.com (L. Zhang), huangqf@craes.org.cn (Q. Huang), cqyyf@163.com (Y. Yang), niezq@craes.org.cn (Z. Nie), jlcheng77@163.com (J. Cheng), yangji@igsrr.ac.cn (J. Yang), 18810720592@163.com (Y. Wang), 15650798262@163.com (M. Chai).

100–2000 mg kg⁻¹ and 2–13 g kg⁻¹, respectively, in the plastic fraction of waste electrical and electronic equipment (WEEE) (Morf et al., 2007). In addition, plastic wastes can potentially absorb various priority pollutants from the environment to some extent (Nakashima et al., 2012; Rochman et al., 2013). Hence, many toxicants could be released into the environment during the milling, washing and melt processing of material recycling of plastic wastes, especially with the rather primitive and crude facilities used.

Pollutant release from material recycling has not received much attention. Such contamination is traditionally undervalued perhaps because this recycling is only a mechanical process and is conducted at relatively low temperatures (< 300 °C in the extrusion). Several studies have recently reported the emission of harmful compounds from material recycling processes. Dozens of volatile organic compounds were identified in the air and the workers suffered acute and chronic health risks in poly(acrylonitrile-butadiene styrene) (ABS) and polystyrene (PS) recycling workshops (Huang et al., 2013; He et al., 2015). High levels of polycyclic aromatic hydrocarbons and PAEs were also measured in the air from a polyvinyl chloride (PVC) plastic waste recycling plant (Tsai et al., 2009; He et al., 2015). Our previous research reported that high concentrations of polybrominated diphenyl ethers (PBDEs) occurred in the environment and in human hair in a plastic recycling area in China and their potential risks should be of great concern (Tang et al., 2014). Available information about the toxicant emissions and pollution characteristics from these processes is still rare.

The occurrence of material recycling of plastic waste by environmentally unfriendly and primitive technologies in developing countries, especially in Asia, Africa and Latin America, has increased in recent years (Pacheco et al., 2012; Huang et al., 2013; Yang and Buekens, 2014). In China, the total recycling of plastic wastes increased from 16.1 million tonnes in 2008 to 24.9 million tonnes in 2013; of the plastic recycled, primitive material recycling of plastic waste is common (Wei and Zhang, 2014). So far, little is known about the release of heavy metals into the environment in plastic waste recycling areas where primitive methods are used, and the potential ecological and human risks of heavy metals from such sites remain unknown. Using Wen'an, in northern China, as a case study for plastic waste recycling, the objectives of this study were to conduct a reconnaissance survey to investigate and characterize the level and risk of toxic metals in soils and sediments. The results will improve our understanding of the extent of heavy metal pollution caused by plastic waste recycling and provide information to plan risk management measures.

2. Materials and methods

2.1. Study area

Wen'an County, in northeast Hebei Province, China, has a population of 460,000 and an area of 1028 km². The material recycling of plastic wastes has been practiced intensively in 101 villages in eight towns in Wen'an, the largest distribution center of material recycling of plastics in North China, employing nearly 100,000 people (Xu et al., 2010; Liu, 2011). These valuable materials have been reclaimed for more than 20 years in the village of Yincun, located in the central part of Zhaogezhuang, Wen'an. In the town of Daliu, next to Zhaogezhuang, intensive plastic waste recycling processes were also active for over a decade. The recycled wastes include ABS, PS, PVC, polyethylene (PE), polypropylene (PP), polyamide (PA), polycarbonate (PC) and others originating from both industrial and household waste. In Wen'an, recycling operations consist of collection, separation, milling,

washing, agglutination, extrusion, quenching and granulation. It is worth noting that these processes were previously performed using crude techniques in family-run workshops in the residential districts. Previously, the processes were not controlled and gaseous and wastewater emissions were not treated. Since 2011, the crude recycling processes have ceased and have been prohibited by the local government. The recycling process of plastic wastes was concentrated at new industrial parks with strict pollution control in other area. In the area, the legacy of these previous recycling techniques, including environmental contamination, is still of concern.

2.2. Field sampling

Based on the sampling guide and methodologies recommended by Chinese Ministry of Environmental Protection (CMEP, 2010), 65 surface soil samples (0–10 cm) were collected from the typical sites in Zhaogezhuang and the adjacent Daliu in November 2011, with the area sampling pattern. All the sampling sites were selected on the basis of their separate representativeness of contaminated or uncontaminated soils. The site density of soil sampling was increased in Yincun, which has been the site of the longest-running continuous recycling activities in Wen'an. For comparison, 10 soil samples were collected at the same time from reference cropland, located approximately 3 km northeast of Yincun, which was less affected by the waste plastic recycling activities. The sampling locations are shown in Fig. 1. Each soil sample consisted of four sub-samples that were randomly taken from the surroundings of each site, pooled and homogenized to form a representative sample.

In November 2011, 30 surface sediments (the top 10 cm) were collected from the Xiaobaihe River, the Renwen Canal and the Yincun Ditch, according to the Geochemical Survey Specification set by CGS (China Geological Survey) (2009). In Xiaobaihe River and Renwen Canal, the sediment samples were collected from the contact part of the riverbank and river water. Differently, the sediment samples were collected from the bottom of the Yincun Ditch, an intermittent-stream which has received the main wastewater from plastic recycling activities. No sediment was collected in the upper of Renwen Canal (Fig. 1) because of its long-term dry. The sampling sites of sediments were selected according to their possible contamination based on our professional judgments. In the present study, the collection of sediments is difficult to be corresponding to the soil sampling, as a result of the specific distributions of plastic waste recycling facilities in the area. All sediment samples were placed in polyethylene bags and transported to the laboratory.

2.3. Sample analysis

All soil and sediment samples were air-dried at room temperature, the coarse debris was removed and samples were ground using a mortar and pestle and passed through a 100-mesh nylon sieve. Metal concentrations of the samples were measured at the Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences, which is certified by the China National Accreditation Board for laboratories. The results met the accuracy demanded by the China State Bureau of Technical Supervision. The methods of metal extraction and analysis were established by the institute (Yang et al., 2009). The details are described in the Supporting information.

2.4. Geoaccumulation index

In this study, the geoaccumulation index (I_{geo}) has been used to evaluate the pollution level of heavy metals in soils and sediments.

Fig. 1. Map of the soil and sediment sampling sites in Wen'an, northern China.

Table 1

Summary of heavy metal concentrations, with basic statistical parameters, for the soils and sediments collected from a plastic waste recycling area.

			As ($\mu\text{g g}^{-1}$)	Cd (ng g^{-1})	Cr ($\mu\text{g g}^{-1}$)	Cu ($\mu\text{g g}^{-1}$)	Hg (ng g^{-1})	Pb ($\mu\text{g g}^{-1}$)	Sb ($\mu\text{g g}^{-1}$)	Zn ($\mu\text{g g}^{-1}$)
Soils	Zhaogezhuang ($n=43$)	Range	7.20–15.8	118–2740	69.0–120	17.0–255	27.0–14,450	18.0–210	1.00–18.3	52.0–956
		Mean $\pm$ SD	10.6 $\pm$ 1.80	418 $\pm$ 547	90.7 $\pm$ 13.2	41.2 $\pm$ 46.8	603 $\pm$ 2224	40.4 $\pm$ 35.5	3.10 $\pm$ 3.80	120 $\pm$ 146
		CV/Skewness	0.171/0.460	1.31/3.19	0.146/0.641	1.13/3.95	3.69/6.05	0.880/3.30	1.25/3.12	1.22/4.98
	Daliu ($n=22$)	Range	7.40–14.9	116–1912	76.0–195	17.0–205	19.0–1750	17.0–589	0.80–33.3	50.0–526
		Mean $\pm$ SD	10.7 $\pm$ 1.80	337 $\pm$ 398	100 $\pm$ 26.7	45.5 $\pm$ 48.0	211 $\pm$ 435	94.0 $\pm$ 134	3.6 $\pm$ 6.90	141 $\pm$ 130
		CV/Skewness	0.170/0.552	1.18/3.40	0.267/2.34	1.05/2.59	2.06/2.91	1.42/2.82	1.92/4.23	0.924/1.95
	Reference area ($n=10$)	Range	9.60–13.7	156–203	81.0–95.0	18.0–34.0	34.5–1935	21.0–29.0	1.00–1.80	62.0–81.0
		Mean $\pm$ SD	11.2 $\pm$ 1.20	180 $\pm$ 18.6	87.5 $\pm$ 4.20	22.1 $\pm$ 4.50	78.1 $\pm$ 57.7	23.4 $\pm$ 2.20	1.10 $\pm$ 0.30	69.5 $\pm$ 6.40
		CV/Skewness	0.111/0.728	0.103/–0.165	0.048/0.249	0.203/2.42	0.739/1.69	0.095/1.95	0.225/2.58	0.092/0.517
	Background values ^a		13.6	94.0	68.3	21.8	36.0	21.5	1.22	78.4
Sediments	Xiaobaihe River ($n=10$)	Range	6.20–14.1	139–1570	75.0–189	23.0–96.0	31.5–2550	22.0–87.0	1.40–8.70	61.0–592
		Mean $\pm$ SD	8.90 $\pm$ 2.30	376 $\pm$ 428	110 $\pm$ 32.2	37.2 $\pm$ 21.6	320 $\pm$ 786	33.2 $\pm$ 19.3	2.70 $\pm$ 2.20	157 $\pm$ 157
		CV/Skewness	0.261/1.36	1.14/2.95	0.293/1.89	0.579/2.71	2.46/3.12	0.581/2.94	0.823/2.62	0.995/2.88
	Renwen Canal ($n=11$)	Range	3.30–10.3	121–5492	60.0–130	22.0–199	24.5–908	17.0–123	1.30–9.60	57.0–257
		Mean $\pm$ SD	6.70 $\pm$ 2.40	1111 $\pm$ 1740	89.1 $\pm$ 18.4	58.4 $\pm$ 51.8	204 $\pm$ 285	50.3 $\pm$ 34.9	3.90 $\pm$ 3.10	134 $\pm$ 80.4
		CV/Skewness	0.351/0.134	1.57/2.14	0.207/0.834	0.888/2.32	1.40/1.94	0.695/1.23	0.801/0.926	0.601/0.910
	Yincun Ditch ($n=9$)	Range	3.70–13.0	248–11,425	98.0–264	37.0–189	60.0–19,400	27.0–336	1.90–19.9	84.0–2452
		Mean $\pm$ SD	9.00 $\pm$ 3.40	33,350 $\pm$ 3551	155 $\pm$ 55.0	107.0 $\pm$ 63.8	6402 $\pm$ 6951	153 $\pm$ 113	8.80 $\pm$ 6.80	895 $\pm$ 783
		CV/Skewness	0.379/–0.618	1.07/1.65	0.354/1.06	0.597/0.120	1.09/0.898	0.735/0.199	0.772/0.421	0.874/0.947
	TEC ^c		9.79	990	43.4	31.6	180	35.8	–	121
	PEC ^c		33.0	4980	111	149	1060	128	–	459

^a Soil background values for heavy metals in Hebei Province, China (CNEMC, 1990).

^b Grade I/II guideline level for soil in China (GB 15618–1995) aims to protect the crop yield and public health (CMEP, 1995).

^c TEC, threshold effect concentration; PEC, probable effect concentration (MacDonald et al., 2000).

It can be calculated using $I_{\text{geo}} = \log_2 (C_n/1.5B_n)$, where C_n is the measured concentration of the metal ions in soil, B_n is the geochemical background value of the corresponding metal and the coefficient 1.5 is used to detect very small anthropogenic influences (Muller, 1969). In this study, soil background values for heavy metals in Hebei Province, China (CNEMC, 1990), which are given in Table 1, were chosen as the geochemical reference values

to calculate the I_{geo} values.

According to Muller (1969), the corresponding relationships between I_{geo} and the pollution level are as follows: unpolluted ($I_{\text{geo}} \leq 0$), unpolluted to moderately polluted ($0 < I_{\text{geo}} \leq 1$), moderately polluted ($1 < I_{\text{geo}} \leq 2$), moderately to heavily polluted ($2 < I_{\text{geo}} \leq 3$), heavily polluted ($3 < I_{\text{geo}} \leq 4$), heavily to extremely polluted ($4 < I_{\text{geo}} \leq 5$), or extremely polluted ($I_{\text{geo}} > 5$).

2.5. Potential ecological risk index

The potential ecological risk index method was developed by Håkanson (1980). In this study, the monomial potential ecological risk index (PERI) was used to assess the potential ecological risk of individual metals in soils and sediments. According to this methodology, the PERI of each heavy metal (E_r^i) is defined as: $E_r^i = T_r^i \times C_f^i = T_r^i \times C_0^i/C_n^i$ where T_r^i is the toxic response factor of each metal (e.g., Hg=40, Cd=30, As=Sb=10, Pb=Cu=5, Cr=2, Zn=1) (Xu et al., 2008); C_f^i is the contamination factor; C_0^i is the concentration of each metal; and C_n^i is the corresponding background value. In this study, the heavy metal content of soils in Hebei Province was used as the regional background value (CNEMC, 1990).

According to Håkanson (1980), the following E_r^i values give an indication of ecological risk: $E_r^i < 40$, low risk; $40 \leq E_r^i < 80$, moderate risk; $80 \leq E_r^i < 160$, considerable risk; $160 \leq E_r^i < 320$, high risk; $E_r^i \geq 320$, very high risk.

2.6. Health risk assessment

In general, individuals are exposed to soil metals through three main pathways: ingestion, inhalation and dermal contact. According to the Exposure Factors Handbook (USEPA, 2001), the average daily doses (ADDs) through ingestion, dermal contact and inhalation for both adults and children were estimated. The human non-carcinogenic risk effects from metals can be assessed using the hazard quotient (HQ), which is the ratio of the ADD to the reference dose (RfD) of a heavy metal for the same exposure pathway (USEPA, 1989). If $HQ \leq 1$, adverse health effects would be unlikely to be experienced, whereas potential non-carcinogenic effects would occur when $HQ > 1$ (USEPA, 1989; Leung et al., 2007).

To assess the overall potential non-carcinogenic effects posed by many chemicals, the hazard index (HI) was calculated using the sum of HQ values of all chemicals in each site (assuming additive effects). For carcinogens, the ADDs were multiplied by the corresponding slope factor (SF, per $\text{mg kg}^{-1} \text{day}^{-1}$) to produce a level of cancer risk (CR) (Wei et al., 2015). If multiple carcinogenic contaminants were present, the cancer risks from all carcinogens and exposure routes were summed (assuming additive effects). The acceptable or tolerable risk for regulatory purposes is within the range of 1.0×10^{-6} – 1.0×10^{-4} (USEPA, 1989; Ferreira-Baptista and De Miguel, 2005).

The estimation methods and values of parameters used are shown in Tables S1 and S2, Supplementary information.

3. Results and discussion

3.1. Heavy metals in soils

The descriptive statistics of heavy metal concentrations in the soil samples from the plastic waste recycling area and reference area are summarized in Table 1. While As and Sb are metalloids, an extensive definition of “heavy metal” is used in the present study to simplify the expression of the whole manuscript. In Zhaogezhuang and Daliu, the Kolmogorov–Smirnov (K–S) test indicated that Cd, Cr, Cu, Hg, Pb, Sb and Zn had skewed concentration distributions and As had normal concentration distributions ($p > 0.05$). As shown in Table 1, the concentrations of Cd, Cu, Hg, Pb, Sb and Zn varied widely and had relatively high coefficients of variation, suggesting that their inputs into soils in the study area might be, to a considerable degree, attributable to anthropogenic sources (Yongming et al., 2006). In the reference area, most of the

metals had relatively narrow concentration ranges and low coefficients of variation, although only As, Cr and Zn passed the K–S test for normality ($p > 0.05$).

The mean concentrations of all metals, excluding arsenic, exceeded the corresponding natural background levels for Hebei Province. This was especially true for Cd, Hg, Pb and Sb, which were about 4.45, 16.8, 1.88 and 2.51 times greater in Zhaogezhuang and 3.59, 5.87, 4.37 and 2.93 times greater in Daliu than their respective background values. In the soil quality standards of China, the Grade I level represents the average natural levels for uncontaminated soil, and Grade II can be used as the threshold values for protecting human health (CMEP, 1995). In this plastic recycling area, 61.5, 47.7, 29.2, 33.8 and 29.2% of soil samples for Cd, Cr, Hg, Pb and Zn, respectively, surpassed their corresponding Grade I values, whereas almost all samples for As and Cu were comparable to their Grade I values. In particular, approximately 33.8% of Cd samples and 12.3% of Hg samples exceeded their Grade II values. In contrast, the metal concentrations in soils from the reference area were generally comparable to their corresponding Grade I values, reflecting little influence from anthropogenic emissions of these metals. In the plastic recycling area, soils were contaminated with Cd, Hg, Pb and Sb and the pollution levels of Cd and Hg were relatively high.

A comparison of heavy metal concentrations in soils from Wen'an with those from other regions is given in Table S3, Supporting information. The metal concentrations in the soils in our study were much lower than the concentrations found in soils from China's mining areas and from the e-waste recycling sites (Li et al., 2011; 2014; Luo et al., 2011; Zhang et al., 2014). Generally, Pb, Cd, Cu, Sb and Zn were the most enriched metals in the soils in the e-waste recycling area (Song and Li, 2014). Differently, the pollutions of Cu and Zn were relatively lower in the study soils. The concentrations of As, Cr, Cu and Zn in the soils in the study area were comparable to concentrations observed in the soils from 31 cities in China (Cheng et al., 2014). However, the Cd, Hg, Pb and Sb concentrations were 2–5 times higher than those found in the contaminated soils from the Chinese metropolises (Cheng et al., 2014). Compared with these areas, the surface soils in the plastic recycling area in Wen'an have suffered from moderate to high metal pollution.

To further characterize the soil metal pollution, the I_{geo} values for heavy metals in the study soils were calculated and the results are described in Fig. 2(A). The I_{geo} values of Cd, Hg, Pb and Sb varied the most, ranging from unpolluted to heavily or extremely polluted levels. In the plastic recycling area, the I_{geo} values for Cd in 35.4 and 12.3% of samples, and for Hg in 40 and 23.1% of samples were larger than 1.0 and 2.0, respectively, suggesting that the soils were moderately to heavily polluted with Cd and Hg. Approximately one fifth of samples were moderately polluted with Pb and Sb but there was no obvious geo-accumulation of other metals. Based on I_{geo} values the reference area was not polluted with metals except for slight Hg pollution.

There were no significant Pearson's correlations between TOC and metals concentrations ($p > 0.05$), except for Cr and Sb. Several independent samples tests (Kruskal–Wallis H) showed that the concentrations of Cd, Cu, Hg and Sb were significantly different among Zhaogezhuang, Daliu and the reference area ($p < 0.05$), which suggested that different human activities had significant effects on the distribution of heavy metals in soils. The concentrations of Cd, Cu, Pb, Sb and Zn in soils were significantly higher in the plastic recycling area than in the reference area ($p < 0.05$), as tested by the non-parametric test method (Mann–Whitney U). The mean levels of As and Cr were similar in the three sites and there were no significant differences ($p > 0.05$) in concentration distributions. Although the p value (Mann–Whitney U) for Hg surpassed 0.05, the mean levels of Hg were about eight

Fig. 2. Box plots of the geoaccumulation index for heavy metals in soils (A) from Zhaogezhuang (red), Daliu (black), Reference area (blue) and in sediments (B) from Renwen Canal (red), Yincun Ditch (black) and Xiaobaihe River (blue) in Wen'an, northern China. The lower and upper limits of the whiskers indicate 5% and 95% values, respectively; boxes extend from 25th to 75th percentiles; horizontal lines within the boxes represent medians; asterisks below or above the whiskers indicate outlier values. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

times greater in Zhaogezhuang and three times greater in Daliu than those in the reference area. In particular, extremely high Hg concentrations were found at sampling sites Y₄₃ (14.45 mg kg⁻¹), Y₁₃ (2.40 mg kg⁻¹) and Y₁₅ (2.52 mg kg⁻¹) in Zhaogezhuang and D₈ (1.75 mg kg⁻¹) and D₉ (1.19 mg kg⁻¹) in Daliu, where the intensive plastic waste recycling processes were active and had existed for more than a decade. The Hg levels in the soils were statistically significantly higher in Zhaogezhuang than in Daliu, which was perhaps related to the recycling history of the two towns (Tang et al., 2014).

3.2. Heavy metals in sediments

The basic statistics related to heavy metals in sediments from the three waterways were summarized in Table 1. The metal concentrations of As, Cr and Pb fit the normal distributions (K-S test, $p > 0.05$) in the sediments from the Renwen Canal, while the concentrations of other metals presented skewed distributions in the three waterways. The heavy metals in surface sediments presented relatively wide concentration ranges and high coefficients

of variation, implying that heavy metals could have been strongly influenced by anthropogenic inputs in the area.

The metal concentrations in sediments in this study were compared with those in other rivers, which are shown in Table S3. The Cd, Cu, Pb, Sb and Zn concentrations in sediments from e-waste recycling areas were much higher than the concentrations found in sediments from the plastic recycling area in this study (Wang et al., 2009; Chen et al., 2010; Quan et al., 2014; Nie et al., 2015). However, the metal concentrations in the sediments in the study area were comparable to concentrations observed in sediments from the Lianjiang River, flowing across Guiyu where large-scale e-waste dismantling activities have been conducted (Wang et al., 2009). The contamination levels of metals (excluding As) in the sediments from Wen'an were higher than those found in the sediments from the main river systems in China (Zhu and Wang, 2012).

In the studied sediments, the levels of all heavy metals (except As) were higher than their soil background values for Hebei Province (CNEMC, 1990); in particular, Cd and Hg were 15.3 and 54.7 times their background values, respectively. The sediment quality guidelines include a threshold effect concentration (TEC) and a probable effect concentration (PEC); below the TEC, adverse biological effects are rarely expected to occur whereas the PEC is defined as the level above which adverse biological effects are expected to occur more often than not (MacDonald et al., 2000). In this study, the concentrations of Cr in all samples, As and Cd in about one third of samples, Hg, Pb and Zn in half of the samples, and Cu in 70.0% of samples exceeded the TECs. Moreover, 43.3% (Cr), one fifth (Hg, Pb and Zn) and about 10% (Cd and Cu) of the samples had concentrations above their corresponding PECs. That is, adverse biological effects were probable because of the concentrations of these metals in the study sediments. This finding also suggests that further investigation of sources and ecological risk assessment of heavy metals is warranted.

The boxplots of the I_{geo} values for the individual metals in the sediments are shown in Fig. 2(B). The I_{geo} value of As was negative with no contamination in most sites. There was no or only very light Cr pollution in the study sediments. The I_{geo} values of Pb, Sb, Cu and Zn in sediments ranged from unpolluted to lightly unpolluted levels in the Xiaobaihe River and the Renwen Canal. In contrast, moderately to heavily polluted levels of Pb, Sb, Cu and Zn were found in the Yincun Ditch. The I_{geo} values of Cd and Hg were relatively high and more than half of the total sediment samples were classified as moderately to heavily polluted. In particular, I_{geo} values greater than 5 were recorded at some sites from the Renwen Canal and the Yincun Ditch for Cd and Hg. This indicated that the accumulation of Cd and Hg was relatively serious in the sediments and that the metal contamination at some sites should be of concern.

Excluding As, the heavy metals in sediments presented significantly different concentration distribution trends in the three waterways (Kruskal–Wallis H test, $p < 0.05$). There was no statistically significant difference (Mann–Whitney U test, $p > 0.05$) in metal concentrations between the Renwen Canal and the Xiaobaihe River. High levels of seven metals were found in the sediments from the Yincun Ditch, which were significantly higher ($p < 0.05$) than those in the other two waterways. For example, the mean concentrations of Cd (3.34 mg kg⁻¹) and Hg (6.40 mg kg⁻¹) in the sediment from Yincun Ditch were almost 3–20 times higher than those in the other two waterways. Yincun Ditch was the key outlet of wastewater from the plastic recycling area, reflecting that the metal contamination was mainly derived from waste discharges from the waste plastic recycling facilities. On the other hand, relatively larger water volume and flow rate might influence the metal accumulation in sediments from the Renwen Canal and the Xiaobaihe River.

Fig. 3. Biplots of soil samples (A), sediment samples (B) and heavy metal concentrations, produced from principal component analysis.

3.3. Source identification of heavy metals

Principal component analysis (PCA) was conducted on the metal concentrations in soils to identify the possible sources of the eight metals. The results of the Kaiser–Meyer–Olkin test ($KMO=0.824$) and Bartlett's test of sphericity ($p < 0.001$) indicated that metal concentrations were suitable for PCA. All the heavy metals were well represented by principal component 1 (PC1) and principal component 2 (PC2), which accounted for 72.8% of the total variance. In the loading plot of PCA shown in Fig. 3(A), two groups were identified. Zn, Cu, Cd, Sb, Cr, Hg and Pb formed a group, which showed similar loading for both principal components, indicating that they were likely to be mainly from the same source. A second group consisted only of As, which had a higher loading in PC2.

Various metals are used in different plastics as additives or filling materials (Dimitrakakis et al., 2009; Li and Shi, 2011; Nakashima et al., 2012; Rochman et al., 2013). These additives include pigments (e.g., ZnO and Cr₂O₃), flame retardants (often brominated organics combined with Sb₂O₃) and stabilizers or plasticizers (e.g., compounds of Cd, Pb and Zn) (Morf et al., 2007; Li and Shi, 2011). The amounts and types of metal compounds applied to plastic products constantly increase, because a range of different properties are achieved by the incorporation of these metal

compounds (Yu et al., 2008; Kotori et al., 2012). Some metals are also used as catalysts during plastic syntheses, particularly Sb and Hg (Cheng et al., 2010; Kotori et al., 2012). Hg compounds are customarily applied as decoration or coating in plastic products such as plastic toys (Sun, 2005; Guney and Zagury, 2012). Some plastics from WEEE could contain or be contaminated by Hg and its compounds that are normally used in fluorescent lamps, relays, or temperature switches in electric and electronic equipment (Li and Shi, 2011; Ni et al., 2014). High metal levels in plastic wastes have been reported in many previous studies (Soares et al., 2005; Morf et al., 2007; Dimitrakakis et al., 2009; Stenvall et al., 2013). For example, the Cd concentrations were up to 761 mg kg⁻¹ and Sb concentrations ranged from 1.7 to 21.7 mg kg⁻¹ in some metallized plastic wastes (Soares et al., 2005). Lead was detected at levels up to 200 mg kg⁻¹ in the recyclable waste plastics from the WEEE (Stenvall et al., 2013). Santos et al. also reported 4–12 mg kg⁻¹ of Hg in WEEE plastics (Santos et al., 2011). These heavy metals could be released during uncontrolled plastic waste recycling activities, particularly during milling, washing and extrusion, and become a major contributor of heavy metals to the environment. These facts suggested that the Zn, Cu, Cd, Sb, Cr, Hg and Pb contents that were represented by PC1 were related to the discharge from plastic waste recycling.

The contamination pattern was further studied with Pearson's correlation analyses. The concentration of As was not significantly correlated with any of the other heavy metals in soils, as shown in Table S4. The other seven metals were positively correlated among each other at $p < 0.01$, excluding Pb and Hg that were significantly positively correlated at $p < 0.05$, suggesting that these elements had almost the same source. PBDEs are a class of pollutants characteristic of plastic waste recycling because of their extensive usage as flame retardants in plastic products (Leung et al., 2007; Tang et al., 2014). Correlations between concentrations of metals and PBDEs were analyzed based on our previously published data (Tang et al., 2014). There was no significant correlation between As and PBDEs. Zn, Cu, Cd, Sb, Cr, Hg and Pb were all significantly correlated with the major components of technical penta-, octa- and/or deca-BDE products at $p < 0.05$. In particular, significant positive correlations ($p < 0.01$) were found between all seven metals and BDE-209, the predominant congener, contributing an average of about 90% of the Σ PBDE concentrations in surface soils (Tang et al., 2014). These results intensively supported that conclusion that heavy metals in the soils were mainly derived from plastic waste recycling.

Most of the studied sites coordinated near the origin of the PCA plot (Fig. 3(A)), which represented the mean concentrations of all sampling sites (Zhao et al., 2014). Sites from the plastic waste recycling area had a scattering concentration trend along the right direction of PC1, reflecting the main influence from waste plastic recycling activities. Sites Y₁₃, Y₁₅ and Y₄₃ from Zhaogezhuang and D₈, D₉, D₁₉ and D₂₀ from Daliu, with high levels of Hg and Cd, could be easily recognized among all of the sampling sites. The metal emissions from the low-tech plastic waste recycling methods used in such sites could have caused the high metal concentrations found in those soil samples. However, some sites from both the plastic recycling and the reference area had relatively higher loading in PC2, which might imply pollution from municipal wastewater irrigation or other agricultural sources (Wang et al., 2014).

PCA was also used to identify the sources of heavy metal contamination in the sediments ($KMO=0.788$; Bartlett's sphericity tests at $p < 0.001$), and the results are shown in Fig. 3(B). The first two components explained 83.3% of the total variance in the metal concentrations in the sediment samples, and the PC1 and PC2 loading plots were similar to those for the soils. PC1 was dominated by Zn, Cu, Cd, Sb, Cr, Hg and Pb. The concentrations of the

seven metals were significantly correlated with one another, except for Hg and Cr (Table S5), suggesting that these elements had common sources. Similar to the soil samples, the sediment metal concentrations were significantly positively correlated, in general, with those of the major components of technical penta-, octa- and/or deca-BDE products (Tang et al., 2014). These results showed that the seven metals in the sediments mostly originated from waste plastic recycling. Of particular note is that Cd had a relatively high negative loading in PC2, implying that Cd in the sediments likely had other sources. Arsenic had a high positive loading in PC2, which was similar to the result from the soils. From the Pearson's correlation matrix (Table S5), no significant correlations were found between As and the other metals or PBDEs, indicating a specific source for As. The sources for arsenic might be geochemical and/or anthropogenic rather than originating from plastic recycling.

The plot of PC1 versus PC2 scores calculated for each sediment sample was shown in Fig. 3(B). Nearly all of the sediments from the Xiaobaihe River, half of the sediments from the Renwen Canal and a few of the sediments from the Yincun Ditch had low scores in both PC1 and PC2 and grouped together distinctly at the left corner of the figure, suggesting the occurrence of moderate pollution at these locations. The other sites from the Yincun Ditch displayed high scores in PC1, such as sites P₂, P₃, P₄, P₅ and P₆, which clearly contained the highest concentrations of Hg and Cd, indicating the major influence of the metals derived from wastewater discharges from the waste plastic recycling facilities. Other seriously polluted sites, such as R₂, R₃ and P₂, were clearly separated on the PC1 axis. At these sites, large quantities of municipal wastewater are discharged without treatment. The loading plots of these sites in PC2 might imply that heavy metals in the sediments were also related to the municipal wastewater discharge.

3.4. Risk characteristics of contamination

Potential ecological risks related to heavy metals in the study soils were calculated and the E_r^i values of individual elements are described in Fig. 4(A). In the reference area, the E_r^i values for As, Cr, Cu, Pb, Sb and Zn were less than 40 and for Cd and Hg were less than 80 in most samples, implying low potential ecological risk. In the plastic recycling area, the E_r^i values for As, Cr, Cu and Zn ranged within the low risk levels. The E_r^i of Pb and Sb were generally low, while the E_r^i of Sb was greater than 80 in several sites. The mean E_r^i values for Cd and Hg were 125 and 523, respectively, much higher than those of the other metals. About 40.0 and 52.3% of the sites had E_r^i values greater than 80 for Cd and Hg, respectively. In particular, about one sixth of samples based on Cd contamination and one third of samples based on Hg contamination were classified as high risk in the plastic recycling area.

The E_r^i values for metals in the sediments and their ecological risk levels are described in Fig. 4(B). Low E_r^i values for As, Cr, Cu and Zn were observed for the three study waterways. The E_r^i values for Pb and Sb ranged from low to moderate levels, with half of the sites in the Yincun Ditch showing considerable risks for Sb. The E_r^i values for Cd and Hg ranged from moderate to very high risk for almost all sampling sites. In the Xiaobai River, sites X₆ and X₈ were classified as very high ecological risk. About one third of the sites in the Renwen Canal and 77.8% of the samples from the Yincun Ditch were classified as causing a very high potential ecological risk because of their Cd and Hg concentrations. The sediment pollution by Cd and Hg and their potential ecological risks should be of great concern.

In this study, the non-carcinogenic health risks to adults and children from exposure to soil metals in the plastic waste recycling area were estimated and the results are shown in Table 2. The non-

Fig. 4. Monomial potential ecological risk index (E_r^i) for heavy metals in soil (A) and sediment (B) from the waste plastic recycling area. Contamination classes: $E_r^i < 40$, low risk; $40 \leq E_r^i < 80$, moderate risk; $80 \leq E_r^i < 160$, considerable risk; $160 \leq E_r^i < 320$, high risk; $E_r^i \geq 320$, very high risk, using the method developed by Håkanson (1980).

carcinogenic health risks were not calculated in the reference area, because of the relatively low levels of metals in the soils. For adults, the total exposure HI from ingestion, dermal contact, and inhalation for all of the eight metals generally ranged from 0.20 to 1.0. The HI was higher than 1.0 in only 3.1% of soils indicating that there would be, in general, no potential human health risks to adults in this area. For children the total hazard quotients of Cd, Pb and Sb were 2.01, 1.11, and 1.88, respectively, at the 95th percentile. The combined HI values from the eight heavy metals exceeded 1 for almost all of the soils in the plastic recycling area. The mean total exposure HI from the eight metals was 2.19 and the combined HI was 5.07 at the 95th percentile, indicating a high potential non-carcinogenic risk to the local children. Children are more susceptible to a given dose of toxin and are likely to inadvertently ingest significant quantities of soil by virtue of their pica behavior and hand or finger sucking (Rasmussen et al., 2001). Therefore, there might be significant non-carcinogenic risks to the local population associated with soil in the plastic recycling area.

In the plastic recycling area, the carcinogenic risks of As and Cd in the soils were estimated through the three exposure modes and those of Cr were considered only via inhalation because of the lack of corresponding carcinogenic slope factors. In the plastic recycling area, the total carcinogenic risk values for the three metals ranged from 9.94×10^{-6} to 2.25×10^{-5} (mean, 1.43×10^{-5}) for adults and from 1.45×10^{-5} to 3.27×10^{-5} (mean, 2.09×10^{-5}) for children, which were within the acceptable levels based on USEPA guidelines (USEPA, 1989). The majority of the carcinogenic risk values for both adults and children were between 1.0×10^{-5} and

Table 2

Non-carcinogenic risks for adults and children because of environmental exposure to soil metals in Wen'an, northern China.

	Adults			Children		
	5th percentile	Median	95th percentile	5th percentile	Median	95th percentile
As	0.065	0.091	0.127	0.447	0.625	0.869
Cd	0.024	0.043	0.389	0.125	0.221	2.01
Cr	0.040	0.048	0.066	0.319	0.380	0.525
Cu	0.003	0.004	0.030	0.016	0.025	0.174
Hg	0.001	0.003	0.095	0.006	0.019	0.552
Pb	0.011	0.018	0.154	0.080	0.130	1.11
Sb	0.021	0.037	0.331	0.118	0.209	1.88
Zn	0.001	0.001	0.004	0.004	0.005	0.028
Total	0.181	0.255	0.859	1.21	1.67	5.07

2.0×10^{-5} , which were higher than 1.0×10^{-6} , showing a requirement for pollution control based on the carcinogenic risk of soil metals.

Our results confirmed that exposure to toxic metals from soils and sediments would cause high potential ecological risks and health risks in this area, although the results would inevitably be affected by the inherent uncertainty in risk assessment (USEPA, 1989; Cao et al., 2015). In this study, bioavailability of the metals in soils and sediments was not considered, which might result in an overestimation of the potential risks. However, the risks might be underestimated if synergistic harmful effects on humans and other organisms occurred with the coexisting metal pollution in soils and sediments. This study has shown that poorly controlled material recycling of plastic wastes is a significant source of heavy metals to the environment and further research on risk assessments and risk controls are warranted.

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Appendix A. Supplementary information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.ecoenv.2015.08.006>.

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