Polybrominated diphenyl ethers (PBDEs) and heavy metals in road dusts from a plastic waste recycling area in north China: implications for human health

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Abstract Road dusts were collected from an area where intense mechanical recycling of plastic wastes occurs in Wen'an, north China. These dusts were investigated for polybrominated diphenyl ethers (PBDEs) and heavy metals contamination to assess the health risk related to these components. Decabromodiphenyl ether (BDE-209) and Σ_{21} PBDE concentrations in these dusts ranged from 2.67 to 10,424 ng g⁻¹ and from 3.23 to 10,640 ng g⁻¹, respectively. These PBDE concentrations were comparable to those observed in road dust from e-waste recycling areas but were 1–2 orders of magnitude higher than concentrations in outdoor or road dusts from other areas. This indicates that road dusts in the study area have high levels of PBDE pollution. BDE-209

was the predominant congener, accounting for 86.3 % of the total PBDE content in dusts. Thus, commercial deca-BDE products were the dominant source. The average concentrations of As, Cd, Cr, Cu, Hg, Pb, Sb, and Zn in these same dust samples were 10.1, 0.495, 112, 54.7, 0.150, 71.8, 10.6, and 186 mg kg⁻¹, respectively. The geoaccumulation index suggests that road dusts in this area are moderately to heavily polluted with Cd, Hg, and Sb. This study shows that plastic waste processing is a major source of toxic pollutants in road dusts in this area. Although the health risk from exposure to dust PBDEs was low, levels of some heavy metals in this dust exceeded acceptable risk levels for children and are of great concern.

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Introduction

The amount of plastic waste generated by households and industries globally has grown rapidly in recent years (Thompson et al. 2009). For example, about 275 million metric tons of plastic waste was generated by 192 coastal countries worldwide in 2010 (Jambeck et al. 2015). Dealing with the substantial quantities of end-of-life plastics generated annually is a critical task worldwide because this waste degrades slowly and can lead to various environmental problems (Rochman et al. 2013; Faure et al. 2015; Ma et al. 2015). Widely used traditional landfill and incineration practices are becoming less desirable disposal methods for plastics because of their negative environmental impacts and waste of resources. Instead, recycling of plastic waste has become the primary policy for treating plastic waste, including primary recycling, mechanical recycling, chemical recycling, and

energy recovery (Al-Salem et al. 2009; Lazarevic et al. 2010). Of these recycling options, mechanical recycling is most popular in many countries because it recovers plastic waste for reuse in manufacturing plastic products; it also has relatively low investment and simple technology requirements (Al-Salem et al. 2009; Lazarevic et al. 2010).

Polybrominated diphenyl ethers (PBDEs) are a class of additive brominated flame retardants with three typical formulations, i.e., penta-, octa-, and deca-BDE mixtures (La Guardia et al. 2006). PBDEs have been used extensively in a number of plastic products, yielding high residual levels of PBDEs in many plastic wastes (Morf et al. 2005; Kajiwara et al. 2008; Chen et al. 2009; Lyu et al. 2015). Morf et al. (2005) analyzed plastics in electrical wastes and found average concentrations of deca- and octa-BDEs of 510 and 530 mg kg⁻¹. Chen et al. (2009) determined maximum PBDE concentrations in hard plastic toys of 5344 mg kg⁻¹. In May 2009, the Stockholm Convention classified penta- and octa-BDE commercial formulations as persistent organic pollutants because of their toxic effects. Both production and use of penta- and octa-BDEs have been banned for several years in the European Union and elsewhere (Labunska et al. 2013). Decabromodiphenyl ether (BDE-209) also exhibits toxic properties in animals and can be metabolized in the environment to form toxic less-brominated PBDEs (La Guardia et al. 2007; Kajiwara et al. 2008; Labunska et al. 2013). Although its use was phased out in North America and Europe, deca-BDE is still being produced and used in many countries, including China (Ni et al. 2013).

Multiclass metals are also added to plastics as additives or fill material (Morf et al. 2007; Li et al. 2011). In particular, high levels of toxic metals are usually found in plastics from electrical waste. For example, Cd concentrations of up to 761 mg kg⁻¹ and Sb concentrations from 1.7 to 21.7 mg kg⁻¹ were recorded in metallized plastic wastes (Soares et al. 2005). Santos et al. (2011) reported 4–12 mg kg⁻¹ of Hg in plastics from electrical and electronic wastes. Some metals are also used as catalysts during plastic synthesis, as well as decoration/coatings for plastic products, resulting in contamination of some plastic wastes by nonadditive metals (Cheng et al. 2010; Guney and Zagury 2012; Kotori et al. 2012).

During mechanical recycling of plastics, such hazardous chemicals can be released into the ambient environment, especially when crude recycling methods are used or there are limited pollution control measures (Tang et al. 2014; He et al. 2015). In recent years, mechanical recycling of plastic wastes using environmentally unfriendly and crude technologies has increased in developing countries (Pacheco et al. 2012; Huang et al. 2013; Yang and Buekens 2014). Our previous studies showed that high concentrations of PBDEs (Tang et al. 2014), Cd and Hg (Tang et al. 2015), are present in soils and sediments from a major plastic recycling area in north China. This

constitutes a potential health risk to local inhabitants. To date, evaluation of environmental parameters in these recycling areas is limited.

Road dust is comprised of solid particles deposited on roads that act as a “reservoir” for accumulating short-term environmental materials from the surrounding areas. The particles commonly include materials worn off the pavement, vehicular-related deposition particles, particles from atmospheric deposition, plant matter, and materials produced by erosion of the adjacent soil (Tang et al. 2013). Among the various environmental media, dust has usually been reported to have relatively high levels of toxic pollutants (Leung et al. 2008, 2011; Tang et al. 2013; Xu et al. 2015). Many previous studies have shown that the concentrations of toxic chemicals in the human body are strongly associated with those in dust, suggesting that dust is a significant exposure pathway to toxic substances (Dixon et al. 2009; Watkins et al. 2011).

In the plastic recycling areas, the road dust likely consists of some weathered plastic particles containing or is contaminated with harmful chemicals, which were originated from the transportation of various plastic wastes and the crude recycling of plastic wastes. In these areas, road dust may receive a large number of pollutant inputs from the contaminated surface soils by wind (Tang et al. 2014, 2015). In addition, the atmospheric pollutant emissions from the thermal process of plastic recycling might result in the heavy contamination of road dusts. To our knowledge, no studies have characterized PBDEs and metals in road dusts from plastic waste recycling areas, where crude mechanical methods are used; thus, the health risks at these sites have not been evaluated. We selected a plastic waste recycling area in Wen'an, north China, as our case study site. We carried out a reconnaissance survey to investigate and characterize the levels and contamination patterns of PBDEs and heavy metals in dust from this recycling area. Using these data, we assessed the potential risks of exposure to these two types of pollutants in dusts. Our results contribute to an understanding of the pollution caused by mechanical plastic waste recycling and provide information useful for establishing risk management measures.

Materials and methods

Study area

Wen'an County, situated in the northeast region of the North China Plain, is about 120 km south of Beijing; it has a population of 460,000 and an area of 1028 km². Wen'an has a subhumid warm temperate monsoon climate (Sun et al. 2014). The mean temperature of Wen'an is approximately 12 °C, and its average annual precipitation is approximately 567 mm (Sun et al. 2014). The main soil type in the study area is brown soil (Liu et al. 2001). Plastic recycling has been

carried out intensively in Wen'an for more than 30 years, as one of the largest centers for mechanical recycling of plastics in north China (Liu 2011; Tang et al. 2014). At peak times, there are approximately 100,000 people engaged in this industry in Wen'an (Xu et al. 2010). The recycled wastes include poly(acrylonitrile-butadiene styrene) (ABS), polystyrene, polyvinyl chloride, polyethylene, polypropylene, polyamide, polycarbonate, and others originating from both industrial and household wastes. Recycling operations consist of collection, separation, milling, washing, agglutination, extrusion, quenching, and granulation. These processes were previously performed using crude techniques on a small scale in family workshops within the residential districts. Most had no environmental control and did not treat their gaseous or wastewater emissions. Since late 2011, the crude recycling processes have ceased and have been prohibited by the local government. In the area, the legacy of these previous recycling techniques, including environmental contamination, is still of concern.

Sample collection

Sampling was carried out in December 2011. Twenty composite samples of dust were collected along Road S334 using a random sampling strategy to assess pollution along this road. Road S334 is the main route for waste input and recycling output in the area. Using a modification of the procedure of Tang et al. (2013), each sample was collected by gently sweeping an area about 3 m² adjacent to the curb of the road (within 1 m) with a clean iron dustpan and brush. These dust samples were transferred to clean ground-glass stoppered bottles. An additional 11 composite samples of dust were collected from pathways outside family-run workshops within the residential areas, where long-running recycling activities have occurred in Wen'an. Sampling locations are shown in Fig. 1.

Samples were air dried at room temperature, and coarse debris was removed. Samples were ground using a mortar and pestle, before being passed through a 150-mesh (about 100 μ m) nylon sieve. Road dust was operationally defined in this study as those particles with diameters below 100 μ m because they are easily resuspended and have the higher environment and health risks than the coarser fractions (Charlesworth et al. 2011; Cao et al. 2014). This finer fraction was stored at -20 °C until analysis, in order to alleviate the volatilization and degradation of PBDEs.

Sample analysis

Based on the PBDE congener patterns for commercial formulations (penta-, octa-, and deca-BDE mixtures) and homologue profiles in other media environments in this area (La Guardia et al. 2006; Tang et al. 2014), 21 PBDE congeners were selected to measure in this study. PBDE concentrations

in the dust samples were determined from ultrasonically extracted solutions using a gas chromatography/mass spectrometry (GC/MS), which was a modification of the method recommended by USEPA (EPA 1614, Draft) and reported in our previous study (Tang et al. 2014). Each sample (8.00 g) was spiked with the surrogate standard ¹³C-labeled PCB-141 and extracted with 30 mL of hexane/dichloromethane (1:1, v/v) in an ultrasonic bath. The extracts were cleaned and fractionated using neutral, acid, and alkaline multilayer silica gel columns (silica gel, particle size 100–200 mesh, Sinopharm Chemical Reagent Co., Ltd) eluted with 70 mL of a 1:1 (v/v) mixture of hexane and dichloromethane. The extract was concentrated to 0.2 mL, and ¹³C-labeled PCB-208 was added prior to GC/MS analysis.

The GC oven temperature program was 60 °C for 0.75 min, increasing at 10 °C min⁻¹ to 240 °C, and then at 20 °C min⁻¹ to 290 °C; it was held at this temperature for 10 min. The injector temperature was 250 °C, and the helium mobile phase flow rate was 1.0 mL min⁻¹. For analysis, 1- μ L aliquots of samples were injected. The injector was used in the splitless mode for the less brominated PBDEs (BDE-28, BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-138, BDE-153, BDE-154, BDE-183, and BDE-190) and the split mode for more heavily brominated PBDEs (BDE-196, BDE-197, BDE-201, BDE-202, BDE-203, BDE-205, BDE-206, BDE-207, BDE-208, and BDE-209).

The target PBDE congeners were identified if the GC retention time and ion abundance ratio matched those for the authentic corresponding standard. The levels of PBDEs were quantitatively determined by the internal standard method using peak area of composite standard. Linearity was tested in the concentration range from 2.5 to 100 μ g L⁻¹ (less brominated PBDEs) and 25 to 1000 μ g L⁻¹ (more heavily brominated PBDEs), respectively, by injecting replicate standards at seven different concentration levels. Within our experiments, all target compounds fulfilled the linearity with *R*² higher than 0.994. Relative standard deviations (*RSDs*) for consecutive injections of standard solutions at different concentration levels ranged from 1.00 to 4.50 %. Recoveries were in the range of 81.5–127 %, depending on individual congeners. The limits of detection (*LODs*) were estimated based on a signal to noise ratio of 5. The *LODs* of BDE-203, BDE-206, and BDE-209 were 105, 182, and 227 pg g⁻¹, respectively. Other congeners varied from 1.93 to 77.5 pg g⁻¹. In this study, some samples were re-detected and concentrated to suitable concentrations to inject when their measured areas were beyond the range of calibration curve. Sample concentrations below *LOQ* were replaced with one half of the *LOQ* in statistical analysis (EFSA 2010; Zheng et al. 2015). Reported concentrations were not corrected for surrogate recovery in this study.

In the present study, eight heavy metals were selected as objective metal pollutants, which represent a broad category

background values for heavy metals in Hebei (CNEMC 1990), China, were selected for calculating the I_{geo} values for our samples. The relationship between I_{geo} values and pollution levels is defined as follows (Muller 1969): unpolluted ($I_{geo} \leq 0$), unpolluted to moderately polluted ($0 < I_{geo} \leq 1$), moderately polluted ($1 < I_{geo} \leq 2$), moderately to heavily polluted ($2 < I_{geo} \leq 3$), heavily polluted ($3 < I_{geo} \leq 4$), heavily to extremely polluted ($4 < I_{geo} \leq 5$), or extremely polluted ($I_{geo} > 5$).

Health risk assessment

Children and adults are exposed to dust through three main pathways, i.e., ingestion of dust particles, inhalation of dust particles through the mouth and nose, and dermal contact with dust. The Exposure Factors Handbook (USEPA 2001) provides estimates of the average daily doses (ADDs) through ingestion, dermal contact, and inhalation for both adults and children. Thus, the human noncarcinogenic risk effects from heavy metals in dust can be assessed using the hazard quotient (HQ), which is the ratio of the ADD to the reference dose (RfD) of a given heavy metal for the same exposure pathway (USEPA 1989). If $HQ \leq 1$, adverse health effects are unlikely to be experienced, whereas potential noncarcinogenic effects occur when $HQ > 1$ (USEPA 1989; Leung et al. 2007). In this study, the inhalation RfD and dermal RfD of BDE-47, BDE-99, BDE-153, BDE-209, penta-BDE, and octa-BDE were assumed to be equal to the ingestion RfD, because no reference values for the inhalation RfD and dermal RfD were available. Our estimation methods and values of the parameters used in our risk assessment are given in Tables S1 and S2, as well as in the Supplementary Material.

Results and discussion

PBDE concentrations

Descriptive statistics of PBDEs in our road dust samples are given in Table 1. PBDE congeners were detected at all sampling sites, indicating that these compounds are ubiquitous throughout the study area. BDE-209 and Σ_{21} PBDE concentrations in the dust samples ranged from 2.67 to 10,424 ng g⁻¹ and from 3.23 to 10,640 ng g⁻¹, respectively. Individual PBDE congeners and the Σ_{21} PBDEs had skewed concentration distributions, according to the Kolmogorov-Smirnov test ($p < 0.05$). Table 1 shows that the concentrations of PBDEs varied widely, having relatively high coefficients of variation. This suggests highly specific sources. In this study, there were no positive correlations between concentrations of TOC and most individual PBDE congeners or with Σ_{21} PBDEs in the dust samples, except for BDE-209 ($p < 0.05$). Relatively high Σ_{21} PBDE concentrations were found at sampling sites W₁₂ (4456 ng g⁻¹), W₁₃ (10,640 ng g⁻¹), and W₁₄ (7089 ng g⁻¹),

collected near Yincun. Tang et al. (2014) also reported relatively high Σ_{21} PBDE concentrations presented at some soil samples which were collected in and near Yincun, the longest-running continuous recycling site in Wen'an. Lowest levels of PBDEs were detected in samples collected from the roadsides in urban areas, which have been swept frequently.

So far, no environmental standards have been established for PBDEs in dusts. To evaluate the PBDE contamination status in the waste plastic recycling area at Wen'an, we compared our PBDE concentrations with those from previous work in other regions (Table S3). Based on PBDE relative potencies reported in the literature (Behnisch et al. 2003), toxic equivalent (TEQ) concentrations for selected indicative PBDE congeners were calculated from our measured dust PBDE concentrations. The Σ_{21} PBDE and TEQ concentrations observed in this study were lower than concentrations reported in outdoor dusts from e-waste recycling regions in Wenling (mean = 64.8 pg TEQ g⁻¹), Qingyuan (mean = 38.7 pg TEQ g⁻¹), and Taizhou (mean = 381 pg TEQ g⁻¹), China (Wang et al. 2010; Jiang et al. 2014; Xu et al. 2015). Typically, PBDE concentrations are much higher in e-wastes than in plastic wastes (Tang et al. 2014). PBDEs released by crude e-waste processing reflect the extremely destructive methods used to recover valuable materials from e-waste, such as acid washing and burning (Leung et al. 2007; Eguchi et al. 2013). However, BDE-209 and Σ_{21} PBDE concentrations in the dusts in our study area were comparable to concentrations observed in road dusts from e-waste recycling sites (Wang et al. 2010; Leung et al. 2011). The level of PBDE contamination in road dusts in Wen'an was an order of magnitude higher than in Suzhou, Nantong, Wuxi, Shantou, and Wenling, China (Shi et al. 2014). Our results were 12 orders of magnitude higher than in regional outdoor dusts from Shantou and Shanghai (Leung et al. 2011; Wu et al. 2015). In addition, the BDE209, Σ_{21} PBDE, and TEQ concentrations in the study road dusts were about double, respectively, those in the soils from this plastic recycling area (Tang et al. 2014). Compared with these results, road dust in the plastic recycling area in Wen'an had relatively high levels of PBDE pollution.

Profile and source of PBDEs

Compositions of PBDE congeners at different sampling sites are shown in Fig. 2. BDE-209 was the predominant congener in all road dust samples, accounting for 44.1–98.9 % (mean = 86.3 %) of their Σ_{21} PBDE concentrations. This reflects the widespread use of commercial deca-BDE products in China, such as Saytex 102E and Bromkal 82-0DE (Xu et al. 2015). In addition, fugitive emissions from the disposal of PBDE-containing plastic wastes could also contribute to BDE-209 in outdoor environments, because BDE-209 has a strong tendency to attach to soil particles. Tang et al. (2014) reported that BDE-209 was the dominant congener (representing

Table 1 Summary of polybrominated diphenyl ether (PBDE) concentrations (ng g^{-1} dry wt), with basic statistical parameters, for the road dust collected from a waste plastic recycling area in Wen'an, northern China

	REP ^a	Range	Mean $\pm$ SD	CV/skewness
BDE-28	–	0.001–1.52	0.117 $\pm$ 0.267	2.27/5.13
BDE-47	1.1×10^{-6}	0.0004–7.74	0.806 $\pm$ 1.49	1.85/3.74
BDE-66	2.0×10^{-6}	0.002–0.75	0.121 $\pm$ 0.149	1.23/2.67
BDE-85	2.1×10^{-6}	0.001–1.55	0.117 $\pm$ 0.290	2.49/4.41
BDE-99	5.6×10^{-7}	0.001–10.2	0.836 $\pm$ 1.92	2.30/4.33
BDE-100	1.2×10^{-6}	0.002–3.14	0.311 $\pm$ 0.627	2.01/3.80
BDE-138	–	0.002–8.35	0.829 $\pm$ 2.13	2.57/2.93
BDE-153	2.3×10^{-6}	0.002–101.4	4.67 $\pm$ 18.1	3.87/5.47
BDE-154	–	0.002–39.9	2.82 $\pm$ 7.52	2.67/4.37
BDE-183	1.5×10^{-6}	0.003–988	40.3 $\pm$ 177	4.39/5.48
BDE-190	4.6×10^{-5}	0.003–37.9	2.12 $\pm$ 7.16	3.38/4.62
BDE-196	–	0.008–61.9	4.47 $\pm$ 11.3	2.53/4.72
BDE-197	–	0.003–180	8.72 $\pm$ 32.2	3.69/5.38
BDE-201	–	0.007–12.3	1.54 $\pm$ 2.68	1.74/2.97
BDE-202	–	0.007–4.05	0.479 $\pm$ 0.803	1.68/3.33
BDE-203	–	0.053–75.7	6.29 $\pm$ 14.0	2.23/4.36
BDE-205	–	0.011–4.18	0.417 $\pm$ 1.03	2.48/2.75
BDE-206	–	0.091–103	13.3 $\pm$ 21.5	1.62/2.96
BDE-207	–	0.057–110	9.95 $\pm$ 20.8	2.09/4.10
BDE-208	–	0.039–293	12.8 $\pm$ 52.2	4.08/5.49
BDE-209	1.6×10^{-5}	2.67–10424	1430 $\pm$ 2253	1.58/2.73
Σ_{21} PBDEs		3.23–10640	1541 $\pm$ 2348	1.52/2.56
Σ TEQ (pg TEQ g^{-1})		0.043–167	23.1 $\pm$ 36.1	1.57/2.71

SD standard deviation, CV coefficient of variation

^aRelative potencies (REPs), derived from a chemically activated luciferase gene expression (CALUX) cell bioassay ($\text{EC}_{50\text{TCDD}}$), were used to calculate the toxic equivalents (TEQs) (Behnisch et al. 2003). Σ TEQ concentrations were calculated as the mean of the sum of BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-153, BDE-183, BDE-190, and BDE-209

91.2 % of the total PBDEs) in soils from the study area. BDE-183, one of the primary congeners found in octa-BDE products (La Guardia et al. 2006), had the second highest

proportion, contributing on average 5.25 % to the Σ_{21} PBDE concentrations in our dust samples. Similar results for outdoor or road dusts were reported elsewhere in China (Yu et al.

Fig. 2 Box plots of the relative abundances of different polybrominated diphenyl ether (PBDE) congeners in the dusts from Wen'an, northern China

2012; Shi et al. 2014). Relatively high concentrations of the higher molecular weight PBDE congeners (BDE-206, BDE-207, and BDE-208) were also found in our samples. The tri-, tetra-, and penta-BDE homolog groups each represented less than 1 % of the Σ_{21} PBDE concentration, indicating that the congener patterns in dusts from the waste plastic recycling area were dominated by highly brominated PBDEs.

The congener profiles of PBDEs in our dust samples were different from those found in other studies. In some e-waste recycling areas in China, in addition to BDE-209 and BDE-183, BDE-47 and BDE-99 were clearly enriched in road dusts, reflecting the previous use of these commercial PBDEs in electronic products (Leung et al. 2011; Shi et al. 2014; Xu et al. 2015). Xu et al. (2015) reported that BDE-47 and BDE-99 contributed 24.4 and 26.5 % to PBDE concentrations in outdoor dust samples from e-waste workshops in Taizhou. These differences in pollution patterns can be attributed to the different PBDEs used in electronics (e.g., printed circuit boards) compared with plastics (Leung et al. 2007). Furthermore, BDE-47 was the dominant congener, aside from BDE-209, in indoor dusts in many countries (Besis and Samara 2012). Thus, PBDE products used in China differ from other countries, contributing to a different congener profile.

Differences and similarities among the PBDE profiles of the dust samples were examined using principal component analysis (PCA) to evaluate source characteristics and subsequent environmental behaviors. Commercial penta-, octa-, and deca-BDE formulations (La Guardia et al. 2006) were explored as the major PBDE sources in the study area. Loading plots for individual congeners are given in Fig. 3. BDE-209 had a high negative loading for principal component 1 (PC1), indicating strong contributions from commercial deca-BDE formulations. BDE-47, BDE-85, BDE-99, BDE-100, BDE-153, and BDE-154 are major components of commercial penta-BDE mixtures, with high positive loadings in PC1. They are clustered in one group, confirming their similar origin from technical penta-BDE products. Principal component 2 (PC2) accounted for 22.9 % of the total variance, predominated by BDE-183, BDE-196, BDE-197, BDE-201, BDE-203, BDE-206, and BDE-207, which are all major components of technical octa-BDE products, suggesting that their common source was octa-BDE products. Figure 3 shows that some octa- and nona-BDE congeners, such as BDE-202, BDE-205, BDE-206, and BDE-208, are separated from BDE-209 and BDE-183. These results signal that some degradation/transformation of higher molecular weight PBDEs has occurred. Deca-BDE in dusts is degraded under light to form nona-BDE or other species (Kajiwara et al. 2008). Likewise, deca-BDE is confirmed to undergo degradation under hydrothermal treatment to form lower brominated PBDEs (Nose et al. 2007). These highly brominated PBDEs may have been subject to degradation by the physical and

chemical processes occurring during plastic manufacturing and/or waste plastic recycling.

Figure S1 shows a plot of PC1 versus PC2 scores calculated for each sample as well as for six commercial PBDE formulations. Most of the dust samples clustered with deca-BDE mixtures (Saytex 102E and Bromkal 82-0DE), indicating that deca-BDE products were the major PBDE source for dusts in the study area. In China, commercial deca-BDE mixtures account for most of the PBDEs used as flame retardants in plastics (Chen et al. 2012). Approximately 80 % of the total deca-BDE product produced in China is used in ABS and high impact polystyrene, as well as in polybutylene terephthalate and polyamide polymers (Chen et al. 2012). Because some dust samples (e.g., W₆, W₁₀, W₈, and W₉) had relatively high positive PC1 scores and were closely related to DE-71 and Bromkal 70-5DE, this indicates that penta-BDE formulations also influenced these samples.

Contamination levels of metals

Descriptive statistics for metal concentrations in the road dust samples from Wen'an are given in Table 2. Background soil values for Hebei as well as Chinese soil quality standards were used to evaluate metal contamination in our samples, because there is limited information for dust (CNEMC 1990; CEPA 1995). In our study area, Cd, Cu, Hg, Pb, Sb, and Zn had nonnormal concentration distributions (Kolmogorov-Smirnov test, $p < 0.05$), with relatively wide concentration ranges and high coefficients of variation. This suggests that heavy metal concentrations were strongly influenced by anthropogenic input. In the study, metal concentrations in the road dusts were generally comparable to concentrations observed in soils in this area (Tang et al. 2015). Aside from arsenic, the mean concentrations of all metals measured in our dust samples were higher than their background values in local soils. In particular, Cd, Hg, Pb, and Sb were 5.27, 4.18, 3.34, and 8.65 times their background values, respectively. In this plastic recycling area, Cd, Cr, Cu, and Pb levels in dust samples surpassed their corresponding grade I guidelines for soil by 71.0, 90.3, 51.6, 61.3, and 29.2 %. In addition, approximately 54.8 % of Cd samples exceeded their grade II guideline values.

A comparison of heavy metal concentrations in road dusts from Wen'an with those from other regions is given in Table S4. Compared with metal concentrations in street dusts from other Chinese cities, the road dusts from the plastic recycling area had relatively low to moderate levels of heavy metals. This suggests that higher levels of heavy metals are ubiquitous in urban dusts, as a result of a wide range of industrial and traffic sources (Lei et al. 2012; Lu et al. 2014). Concentrations of As, Zn, Cd, and Cu in our road dusts also were relatively low compared with road dusts from e-waste recycling sites (Leung et al. 2008; Xu et al. 2015). In contrast,

Fig. 3 Loading plots from the principal component analyses (PCA) for PBDEs in the dust samples collected from the waste plastic recycling area studied

our Cd and Pb concentrations were comparable to those from the e-waste site at Wenling, China (Xu et al. 2015). Thus, moderate heavy metal pollution was recorded in the road dusts from the waste plastic recycling area.

To characterize the dust metal pollution, I_{geo} values for heavy metals in dust samples were calculated (Fig. 4). The I_{geo} values for Cd, Hg, Pb, and Sb varied most, ranging from unpolluted to heavily or extremely polluted levels. This variability in road dust properties suggests different pollution sources for these metals. In the plastic recycling area, the I_{geo} values exceeded 1.0 and 2.0 for Cd in 54.8 and 22.6 % of samples, for Hg in 41.9 and 16.1 % of samples, and for Sb in 32.3 and 19.4 % of samples. This suggests that road dusts were moderately to heavily contaminated with Cd, Hg, and Sb. Approximately one third of samples were moderately

contaminated with Pb. No geo-accumulation of other metals was observed.

Source identification of metals

PCA was used to identify the sources of heavy metal contamination in the road dusts. Results with a Kaiser-Meyer-Olkin Index=0.823 and Bartlett's sphericity tests at $p<0.001$ are shown in Fig. 5. The first two components explained 68.7 % of the total variance in metal concentrations in the road dust samples. PC1 was dominated by Cd, Cu, Sb, and Zn, accounting for 53.2 % of the total variance. As and Hg were highly loaded in PC2, which explained 15.5 % of the total variance.

In the loading plot (Fig. 5), Cd, Cu, Sb, and Zn formed a group with similar loading for both principal components,

Table 2 Heavy metal concentrations (mg kg⁻¹ dry wt) of the road dust from the waste plastic recycling area

	Concentration			Background values ^a	Chinese soil guidelines (grade I) ^b	Chinese soil guidelines (grade II) ^b
	Range	Mean±SD	CV/Skewness			
As	5.36–13.7	10.1±1.96	0.19/–0.15	13.6	15	30
Cd	0.15–3.04	0.50±0.60	1.20/3.39	0.094	0.2	0.3
Cr	68.9–160	112±22.1	0.20/0.44	68.3	90	200
Cu	20.7–554	54.7±93.9	1.72/5.35	21.8	35	100
Hg	0.02–0.90	0.15±0.19	1.26/2.91	0.036	0.15	0.50
Pb	18.7–589	71.8±106	1.47/4.35	21.5	35	300
Sb	1.19–196	10.6±34.9	3.31/5.38	1.22	–	–
Zn	58.3–2025	186±346	1.86/5.34	78.4	100	250

SD standard deviation, CV coefficient of variation

^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 (CEPA 1995)

Fig. 4 Box plots of the geoaccumulation index for heavy metals in dusts from Wen'an, northern China. Contamination classes: unpolluted ($I_{geo} \leq 0$) (I), unpolluted to moderately polluted ($0 < I_{geo} \leq 1$) (II), moderately polluted ($1 < I_{geo} \leq 2$) (III), moderately to heavily polluted ($2 < I_{geo} \leq 3$) (IV), heavily polluted ($3 < I_{geo} \leq 4$) (V), heavily to extremely polluted ($4 < I_{geo} \leq 5$) (VI), and extremely polluted ($I_{geo} > 5$) (VII), using the method developed by Muller (1969)

suggesting that these elements were from the same source. In plastics, various metals are used as additives or fill materials, including pigments (e.g., ZnO and Cr_2O_3), flame retardants (Sb_2O_3), stabilizers, and plasticizers (e.g., compounds of Cd and Zn). High heavy metal levels in plastic wastes have been reported in many previous studies (Soares et al. 2005; Santos et al. 2011). In addition, these four metals are positively correlated with each other at $p < 0.01$ (Table S5). Concentrations of these four metals in most of the dust samples were higher than their soil background values. Thus, these four metals were mostly derived from plastic waste recycling. Cr also had a relatively high positive loading in PC1 and was positively correlated with Cd, Cu, Sb, and Zn, suggesting that the Cr pollution also was related to plastic recycling.

The Pb loading in the PCA was different from other metals, despite Pb being added to different plastic products as a stabilizer (Morf et al. 2007; Li et al. 2011). There were no significant correlations found between Pb and other metals, except for Pb and Zn, indicating other sources for Pb. Previous studies have found that accumulations of Pb in road dusts are attributed to the use of fossil fuels (Chen et al. 2010). However, given that leaded gasoline has been banned since 2000 in China, vehicle-exhaust emissions should contribute little to Pb pollution in road dusts (Cheng and Hu 2010). Large coal consumption (2.64×10^8 t in 2011) and its associated industrial smoke discharge (1.3×10^6 t in 2011) in Hebei suggest that coal combustion could be a major source of Pb pollution in road dusts in this area (CSB 2012).

Fig. 5 Loading plots of the principal component analysis (PCA) for metals in the road dust samples

As and Hg had relatively high loadings in PC2. Almost all dust samples gave As concentrations comparable to the Hebei soil background values, indicating a natural source for As in samples. In Fig. 5, As and Hg are separated in the PCA loading plot, indicating that the two elements are poorly correlated. Hg loading (0.070) in PC1 was obviously lower than for other metals. Hg was not significantly correlated with any of the other metals, pointing to quasi-independent behavior of Hg.

Health risk assessment

As part of a preliminary evaluation of human exposure to PBDEs in road dusts, the noncarcinogenic health risks to adults and children living in the vicinity of the plastic waste recycling area were calculated (Table 3). The exposures to BDE-47, BDE-99, BDE-153, BDE-209, penta-, octa-, deca-BDE, and Σ_{21} PBDEs via dust ingestion, inhalation, and dermal contact for both adults and children were several orders of magnitude lower than their corresponding RfD values. Thus, there is an overall low health risk for local PBDE exposure. However, the HQ for children for BDE-209 was 0.616 at the 95th percentile, indicating that potential noncarcinogenic risk for local children may deserve attention.

The HQs for metals for adults and children are shown in Table 3. The exposure HQs for adults for all eight metals was generally less than 1.0. HQs were higher than 1.0 in only a few dust samples, indicating no human health risks to adults in this area. For children, adverse health effects were predicted when

using the 95th percentile concentrations for Cd, Pb, and Sb, with HQs of 2.50, 1.77, and 11.5, respectively. The combined HQs for all eight heavy metals exceeded 1.0 for all road dust samples, indicating a high potential noncarcinogenic risk to local children within the plastic recycling area.

The model used in this investigation provides a useful tool for risk assessment, identifying health risks related to PBDEs and metals in local dusts. However, there are a number of inherent uncertainties. We did not consider the pollution variation over time nor the bioavailability of the pollutants in these dusts. Risks related to consumption of contaminated food and water that also contribute to ADDs were not taken into account. Last, the toxicity effect of PBDEs and metals are likely to be weakened or enhanced by other pollutants.

Conclusions

To the best of our knowledge, this is the first study on heavy metals and PBDEs in road dust from a plastic recycling area. This study shows that the crude mechanical recycling of plastic waste is a significant contributor of PBDEs and heavy metals to road dust. The levels of PBDEs in this study area were comparable with some e-waste recycling areas but much higher than those in the road dusts or outdoor dusts from other urban areas. This indicates that the road dust in this area was highly polluted with PBDEs. BDE-209 was the predominated congener in the study dusts, indicating that the commercial

Table 3 Hazard quotient for polybrominated diphenyl ether (PBDE) and metals in the road dust from the waste plastic recycling area

	Adults			Children		
	5th percentile	Median	95th percentile	5th percentile	Median	95th percentile
As	5.60E-02	8.90E-02	1.18E-01	3.86E-01	6.13E-01	8.08E-01
Cd	3.10E-02	6.80E-02	4.82E-01	1.61E-01	3.54E-01	2.50E+00
Cr	4.10E-02	5.90E-02	8.60E-02	3.22E-01	4.65E-01	6.84E-01
Cu	4.00E-03	5.00E-03	4.30E-02	2.00E-02	3.20E-02	2.54E-01
Hg	1.00E-03	4.00E-03	3.30E-02	6.00E-03	2.30E-02	1.91E-01
Pb	1.40E-02	2.40E-02	2.43E-01	1.04E-01	1.74E-01	1.77E+00
Sb	2.60E-02	5.30E-02	2.03E+00	1.49E-01	3.04E-01	1.15E+01
Zn	1.00E-03	1.00E-03	1.00E-02	5.00E-03	7.00E-03	6.50E-02
Metals ^a	2.14E-01	3.19E-01	2.60E+00	1.43E+00	2.06E+00	1.50E+01
BDE-47	6.20E-08	3.82E-05	7.93E-04	3.38E-07	2.08E-04	4.32E-03
BDE-99	1.71E-07	3.86E-05	1.02E-03	9.28E-07	2.10E-04	5.54E-03
BDE-153	8.56E-08	5.13E-05	3.52E-03	4.64E-07	2.79E-04	1.92E-02
BDE-209	3.24E-05	4.93E-03	8.28E-02	2.41E-04	3.66E-02	6.16E-01
Penta-BDE	2.71E-08	3.74E-06	6.11E-05	1.48E-07	2.04E-05	3.32E-04
Octa-BDE	5.95E-07	3.67E-05	1.15E-03	3.59E-06	2.21E-04	6.96E-03
PBDEs ^b	4.34E-05	5.77E-03	8.29E-02	3.11E-04	4.03E-02	6.17E-01

^a Sum of the eight metals

^b Sum of BDE-47, BDE-99, BDE-153, and BDE-209

deca-BDE product was dominant. The road dusts have suffered from moderate to high metal pollution and in particular, high Cd, Hg, and Sb pollution. Source assessment indicated that Cd, Cu, Sb, Zn, and Cr in dusts were mainly derived from inputs from poorly controlled plastic recycling operations in this area. Although the health risk from exposure to dust PBDEs was low, the noncarcinogenic risks for certain metals in this dust to local children exceeded an acceptable level. Further research on risk assessments and control measures for harmful chemicals arising from mechanical recycling of plastic wastes is warranted.

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