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Graphical Abstract

Vehicles as outdoor BFR sources: evidence from an investigation of BFR occurrence in road dust

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ABSTRACT

The distribution of brominated flame retardants (BFRs) including $\sum_8$PBDEs, DBDPE, BTBPE, EH-TBB, BEH-TEBP and PBEB in road dust (RD) collected in Xinxiang, China was characterized. Analysis of RD samples indicated that the BFR abundance declined as traffic density decreased, with total mean levels of 292, 184, 163, 104 and 70 ng g^{-1} dust at sites from traffic intersections, main roads, collector streets, bypasses and parks, respectively. A possible explanation for this phenomenon is that the majority of BFRs may be emitted from the interior of vehicles via their ventilation systems. Of the 13 analyzed substances, BDE-209 and BEH-TEBP were the most abundant components in RD from Xinxiang. Similar amounts of ∑BDEs excluding BDE-209 were found at different types of sampling sites, and thus, atmospheric deposition is also a probable source of BFRs in RD which can be subject to air transportation. The main PBDE sources were traced to commercial products including DE-71, Bromkal 79-8DE, Saytex 201E and Bromkal 82 DE mixtures. Our results confirm that the use of deca-BDE commercial mixture is a major source of PBDE contamination in RD. Risk assessment indicated the concentrations of BFRs in RD in this study do not constitute a non-cancer or cancer risk to humans through ingestion. Annual emission fluxes of the commonly detected BFRs via RD in China were estimated to be up to 4980 kg $year^{-1}$.

Keywords: BFRs; road dust; source; exposure; fate

1. Introduction

Brominated flame retardants (BFRs), mainly consisting of polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD), tetrabromobisphenol A (TBBPA), decabromodiphenylethane (DBDPE), 1,2-bis-(2,4,6-tribromophenoxy)ethane (BTBPE), 2-ethyl-1-hexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), bis(2-ethyl-1-hexyl) tetrabromophthalate (BEH-TEBP) and pentabromoethylbenzene (PBEB) are a large group of additives used in numerous products to reduce fire risks. Meanwhile, BFRs are ubiquitous in various environmental media, foods, and biota including humans (Harrad et al., 2010; Fromme et al., 2016). Because of their toxicity and intensive application in urban environments, BFR occurrence, fate, behavior and consequent human health risk have caused increasing concern in recent years (de Boer et al., 2016; Liu et al., 2016; Yu et al., 2016).

Road dust (RD) is formed through sedimentary process of particulate matter which mainly originates from atmospheric precipitation, urban traffic, construction and industrial activities under the action of wind, water and gravity in road surface. RD is simultaneously an important environmental reservoir and source of many contaminants (semi-volatile organic compounds, heavy metals etc.) in urban environments, and fate of those contaminants is closely related to that of RD (Offenberg et al., 2003). RD can enter urban drainage networks, aquatic environment and waste incineration system, as well as undergoing atmospheric transport over a range of spatial scales. Moreover, RD has been identified as constituting potentially over 10% of $PM_{2.5}$ in urban atmospheres (Yu et al., 2013). RD can pose serious risks to human health, especially for street sweepers, pedestrians, street vendors and traffic policemen. Because of rapid urbanization, urban RD is becoming an increasingly serious environmental problem (Zhao and Li, 2013a, b; Zhao et al., 2014).

Evidence suggests that FRs can be released from associated materials and enter the environment through multiple pathways (Cao et al., 2013; Cao et al., 2014; Schreder and La Guardia, 2014; Cao et al., 2015). Although ventilation of indoor air is believed to be the dominant source of PBDEs in urban ambient air (Law et al., 2014), it is plausible that vehicles may constitute significant FR emission sources in light of studies reporting substantial concentrations of FRs in vehicle air and dust (Harrad et al., 2006; Hazrati et al., 2010; Harrad and Abdallah, 2011; Brommer and Harrad, 2015). However, to our knowledge, to date very few studies have investigated FR concentrations in RD (Luo et al., 2009; Tang et al., 2016). Moreover, there is a dearth of evidence about the RD significance of vehicles as a source of BFRs to RD.

To fill those knowledge gaps, the major objectives of the present study were to (1) determine the concentrations and distribution of BFRs in RD; (2) identify important factors influencing the occurrence of BFRs in RD; and (3) examine the proportion of the environmental burden of BFRs associated with RD, and its associated risk.

2. Materials and methods

2.1. Sampling strategy and methods

Individual RD samples were obtained from 4 traffic intersections, 14 sites on main roads, 11 sites on collector streets and 7 sites on bypasses in Xinxiang, China on sunny, windless days in October, 2014. In addition, 4 RD samples were collected from paths in parks as reference “urban background” sites. All sampling sites on roads or streets were located between two crossings and kept away from construction activities. At each site, sampling was performed with bristle brushes on an area of about 4 m^2 along road curbs. Between collecting each sample, the brushes were cleaned with water and dried with a clean electric blower. After collection, samples were sieved

through a stainless steel mesh to < 25 μm, during which the mesh were cleaned in ultrasonic water bath and dried with a clean electric blower between each sample. All 40 RD samples were packed with aluminum foil, sealed in clean polyethylene zip bags and stored in the dark at −20 °C until analysis was performed.

2.2. Chemicals

BDE-77, BDE-128, ^{13}C-BTBPE, ^{13}C-BDE-209 and PCB-129 purchased from Wellington Laboratories Inc. were used as internal standards (IS). All solvents used (acetone, n-hexane, iso-octane (2,2,4-Trimethylpentane) and DCM (dichloromethane)) were HPLC grade.

2.2. Analytical methods

In summary, a sample aliquot (~100 mg of dust or 30 mg for SRM 2585) was accurately weighed and spiked with known amounts of IS. Two mL of solvent mixture n-hexane/acetone (3:1, v/v) was employed for the extraction. The process consisted of consecutive steps of vortexing (1 min), ultrasonication (5 min) and centrifugation (2 min, 2000 g) for one cycle repeated three times. After each cycle, the supernatant was transferred to a clean tube. All tubes were baked at 420 °C for 6 h before use. The extracts were then evaporated to 0.1~0.2 mL under a gentle nitrogen stream and further purified on Florisil cartridges (Florisil ENVI, 500 mg, 3 mL, Supleco, Bellefonte, PA, USA). Prior to use, all cartridges were pre-cleaned and conditioned with 10 mL n-hexane. BFRs were eluted with 10 mL of n-hexane/DCM (1:1, v/v). The eluate was concentrated to dryness and resolubilized in 100 μL isooctane with a known amount of PCB-129 as a recovery determination standard ready for GC-NCI-MS analysis.

The analysis procedure is similar to a recent publication (Kuang et al., 2016). Analysis of eight PBDEs (BDE-28, -47, -100, -99, -154, -153, -183 and -209) and five NBFRs (novel brominated flame retardants, EH-TBB, BEH-TEBP, BTBPE, DBDPE, PBEB) was performed. Under

electron capture negative ionization (ECNI) mode, a Thermo Trace 1310 GC coupled with an ISQ single quadrupole MS equipped with a programmable-temperature vaporizer (PTV) was employed to conduct the analysis. Two μL of cleaned extract were injected on a Thermo TG-SQC column (15 m×0.25 mm×0.25 mm). The injection temperature was set at 92 °C, hold 0.04 min, ramp 700 °C min^{-1} to 295 °C. The GC temperature program was initially 50 °C, hold 0.50 min, ramp 20 °C min^{-1} to 240 °C, hold 5 min, ramp 5 °C min^{-1} to 270 °C and then ramp 20 °C min^{-1} to 305 °C, hold 16 min. Helium was used as a carrier gas with a flow rate of 1.5 mL min^{-1} for the first 22.00 min, then ramp 1.0 mL min^{-2} to 2.5 mL min^{-1}, hold 13.00 min. The mass spectrometer was employed in selected ion monitoring (SIM) mode with measured ions for each compound listed in Table SI-1. Dwell times for each ion were 30 ms. Ion source and transfer line temperatures were 300 and 320 °C, respectively and the electron multiplier voltage was 1400 V. Methane was used as moderating gas.

2.3. QA/QC

Average±σ_{n-1} recoveries of BDE-77, BDE-128 and ^{13}C-BDE-209 were 84±22%, 122±25% and 121±19%, respectively. The results of all analyses in 6 replicates of SRM 2585 (NIST, Gaithersburg, MD, US) demonstrated both good repeatability and good agreement with the certified values reported elsewhere (Van den Eede et al., 2012). However, compared with the literature data it's clear that uncertainty also existed for the determination of BDE-209 and BEH-TEBP because their concentrations in SRM 2585 detected from different researches were not precisely consistent (Table SI-2).

3. Results and discussion

3.1 BFR concentrations and spatial distribution

Except for BDE-28, PBEB, BTBPE and DBDPE, all seven PBDEs, EH-TBB and BEH-TEBP were commonly detected in all 40 RD samples (Table SI-3). A summary of the concentrations of BFRs in RD samples from each location category is provided in Table 1. Concentrations of $\sum_6$PBDEs (excluding BDE-209) and EH-TBB varied from 3.2 to 15.5 ng g^{-1} and from 0.7 to 19.1 ng g^{-1}, respectively. BEH-TEBP and BDE-209 concentrations ranged from 1.5 to 189 ng g^{-1} and from 5.7 to 261 ng g^{-1}. Concentrations of $\sum_9$BFRs ranged from 17.0 to 458 ng g^{-1}, with a geometric mean of 139 ng g^{-1}, at levels 1-3 orders of magnitude lower than in vehicle dust reported elsewhere (Besis and Samara, 2012; Coelho et al., 2014). PBDE concentrations in RD in this study are comparable with those in urban RD from Suzhou, Wuxi and Nantong in the Yangtze River Delta, China (geometric mean concentration of $\sum_8$PBDEs including BDE-209 was 169 ng g^{-1}) (Shi et al., 2014), but was much lower than that in urban RD from Beijing, China (concentration of $\sum_9$PBDEs including BDE-209 were 23700 ng g^{-1} in a pooled sample) (Cao et al., 2014), which indicates substantial variation between cities in China.

The mean concentrations of $\sum_9$BFRs in RD from different sampling areas decreased in the following sequence: traffic intersections > main roads > collector streets > bypasses > parks (Fig. 1), revealing the significant influence of traffic density on BFR abundance in RD. Moreover, for all monitored BFRs, concentrations in RD from traffic intersections and streets exceeded significantly those in parks. Because previous studies have identified high concentrations of BFRs in air and dust samples inside vehicles (Besis and Samara, 2012), and demonstrated vehicle exhaust to be an important PBDE emission source (Wang et al., 2010, 2011), it is plausible that emissions from vehicles via ventilation or exhaust constitutes an important source of BFRs in RD. Interestingly, while there were no significant differences in concentrations of Σ_6BDEs between trafficked locations and parks; concentrations of less volatile BFRs including

BDE-209 and BEH-TEBP at trafficked locations exceeded significantly those at parks (Fig. 1). This suggests that the influence of vehicle emissions on BFRs in RD is greater for these less volatile BFRs.

As RD is a mixture of soil, sand, and deposited particles, the mean organic content of RD in this study was 9.9±2.7%. By comparison, organic content in indoor dust generally exceeds 50% (Morawska and Salthammer, 2003; Cao et al., 2015). Thus, while dry mass concentrations of BFRs were typically an order of magnitude lower than those in indoor settled dust from China generally (Yu et al., 2012; Zhu et al., 2015); when normalized to organic content of RD, BFR concentrations in RD were comparable to those in indoor dust (Fig. SI-1). Normalized mean Σ_9BFR concentrations were 2670, 1830, 1420, 1140 and 556 ng g^{-1} in RD from traffic intersections, main roads, collector streets, bypasses and parks, respectively, which exceed BFR concentrations in indoor dust from Germany (median: 74 ng g^{-1}) (Sjodin et al., 2008) and Australia (median: 469 ng g^{-1}) (Toms et al., 2015).

3.2 Component profiles and global comparison

The relative contributions of individual BFRs to total BFR concentrations in RD are presented in Fig. 2 and Fig. SI-2. BDE-209 accounted for 56.4±18.5% (mean ± standard deviation) of the total BFR concentrations in all samples. The next most important contributors were BEH-TEBP at 31.9±18.4% Σ_9BFRs, followed by BDE-183 (3.0±4.6%) and BDE-99 (2.2±1.3%). Other BFRs were present only at low abundances.

Several reports exist of elevated concentrations of PBDEs in air and dust samples from vehicles (Betts, 2008; Batterman et al., 2009; Abdallah and Harrad, 2010; Kalachova et al., 2012), that exceed those reported in houses (Besis and Samara, 2012; Coelho et al., 2014), indicating

182 that vehicles are possible emission sources of PBDEs. In contrast, data on the presence of
183 NBFRs in vehicles are scarce and we are aware of only two such studies that have reported the
184 presence of DBDPE at much lower levels compared to PBDEs (Harrad et al., 2008; Kalachova et
185 al., 2012).

186 The spatial variation of BFRs in this study suggests that vehicle emissions constitute a
187 substantial source of BFRs in RD. The available global database on concentrations of BFRs in
188 vehicle and road dust is summarized in Fig. 3 and Fig. SI-3. While absolute concentrations
189 display international variation (UK>US>other EU countries), PBDE congener patterns are
190 similar in vehicle dust from different countries. Combined, this suggests that while the
191 commercial PBDE mixtures used in vehicles are universal, the amounts applied vary between
192 jurisdictions. Apparent dominance of BDE-209 (typically showing present proportion higher
193 than 90%) are consistent in RD from China and vehicle dust from abroad, indicating PBDE
194 application patterns might be similar in China with abroad. And only in some US vehicles,
195 penta-BDE congeners represented relatively higher proportion than other countries/regions,
196 representing higher application amount of Penta-BDE in US. Meanwhile, compared to vehicle
197 dust abroad, PBDEs in RD from China showed remarkable higher proportion of BDE-183,
198 possibly implicating Octa-BDE might be applied in China vehicle industry more widely than
199 abroad.

200 Similar to the former reported dominance of PBDEs in vehicle dust, this study verified the
201 dominance of PBDEs in RD, greater than other BFRs. Further, with BDE-209 as the dominant
202 congener, the PBDE congener profiles in this study are similar to previous results found for
203 urban RD (Cao et al., 2014; Shi et al., 2014) and road soils from e-waste (Luo et al., 2009) and
204 plastic waste (Tang et al., 2016) recycling region from China, probably due to more extensive

application of Deca-BDE than Penta- and Octa-BDE in China (Yu et al., 2016). Moreover, less brominated BDEs are more volatile than BDE-209, which enhanced the dominance of BDE-209 in RD.

In contrast to previous reports of the presence of DBDPE in vehicle dust (Stuart et al., 2008; Kalachova et al., 2012), DBDPE and BTBPE were rarely detected in RD in Xinxiang. Nevertheless, this study firstly reported the contamination of EH-TBB and BEH-TEBP in RD, suggesting that as vehicles containing PBDEs become obsolete, more attention should be paid to the occurrence of NBFRs in vehicle and road dust. It is demonstrated Deca-BDE and BEH-TEBP might be the most important BFR components in vehicles. And this revealed that BFR application patterns in vehicles were different from that indoors in China because DBDPE and BTBPE have been detected in considerable levels in indoor dust recently (Cao et al., 2014; Yu et al., 2016).

3.3 Source appointment with diagnostic ratios

As specific PBDE commercial formula shows distinctive congener pattern (La Guardia et al., 2006; Li et al., 2015), a diagnostic ratio model is proposed and performed on concentration distribution of the 7 PBDE congeners to analyze the possible sources of PBDEs. With similar physical-chemical properties, two pairs of BDE congeners including BDE-100 and BDE-99, BDE-154 and BDE-153 were employed as principle ratios to conduct source apportionment, with the ratio of BDE-183 and BDE-209 as an auxiliary parameter. In this study, the geometric mean ratio of BDE-100 to BDE-99 were 0.25, which is closer to that in the penta-BDE mixture DE-71 (0.27) than Bromkal 70-5DE (0.17). The geometric mean ratio of BDE-154 to BDE-153 were 1.21, which is closer to that in the penta-BDE mixture DE-71 (0.83) than penta-BDE

mixture Bromkal 70-5DE (0.50) and octa-BDE mixture DE-79 (0.12). Thus it's concluded Bromkal 70-5DE and DE-79 could not be the possible sources of penta- and octa-BDEs, and BDE-183 can only origin from octa-BDE mixture Bromkal 79-8DE probably. The geometric mean ratio of BDE-183 to BDE-209 was 0.03, which is essentially different from that in the octa-BDE mixture Bromkal 79-8DE (0.25) and DE-79 (32.1), indicating BDE-209 can primarily origin from Saytex 201E or Bromkal 82 DE. Consequently, comparison between the congeners in RD and the PBDE pattern in commercial products suggested the commercial formulas including DE-71, Bromkal 79-8DE, Saytex 201E and Bromkal 82 DE were the possible origin of PBDEs in these RD samples. According to limited data, EH-TBB and BEH-TEBP were produced as the replacements of Penta-BDE, possibly originating from the commercial mixture Firemaster 550 (Stapleton et al., 2008; Covaci et al., 2011).

However, all these analysis is not deterministic because: firstly, BFRs were applied in various materials where BFRs own different migration pathways; secondly, different BFR components have different volatility and partition characteristics among environmental matrix, which result in different environmental fates; thirdly, the different compounds may have undergone different reductive debromination processes, from which their occurrence proportions may increase or decrease in RD compared with the commercial products, especially for lower brominated BDEs; fourthly, unknown mixtures which contain different relative compositions are likely to be applied.

3.4 Exposure and risk assessment

Ingestion may be important contribution of human exposure to BFRs in settled dust. Daily Intake (DI, mg kg^{-1} d^{-1}), hazard index (non-cancer) and cancer risk were estimated using the following equations (Li et al., 2015):

$$DI = \frac{C \times IR \times ET \times EF \times ED}{BW \times AT \times 10^{9}}$$

$$Hazard\ index = \frac{DI}{RfD}$$

$$Cancer\ risk = DI \times CSF$$

where C is the concentration of ∑BFRs in RD (ng g^{-1}). IR is the intake rate of dust (Harrad et al., 2006; Hazrati et al., 2010). For children, adults, and professional street sweepers high-end IR were assumed to be 200 mg d^{-1}, 50 mg d^{-1} and 500 mg d^{-1} (10 times of normal adults), respectively. Due to a lack of data on human absorption efficiency of PBDEs in dust, a 100% absorption efficiency was used, representing an upper limit of the uptake rates. Exposure time (ET) is 3 hours (1/8 d) for children and adults, 12 hours (1/2 d, 9 h for work and 3 h for other activities) for professional street sweepers, exposure frequency (EF) is 365 days $year^{-1}$, exposure duration (ED) is 6 years for children and 30 years for adults, body weight (BW) is 15 kg for children and 70 kg for adults and professional street sweepers, averaging time (AT) is 2,190 days (6 years) for children and 10,950 days (30 years) for adults. The reference dose (RfD) varies with the types of congeners. Specifically, the RfD values promulgated by the USEPA are 0.002 mg kg^{-1} d^{-1} for penta-BDEs, 0.003 mg kg^{-1} d^{-1} for octa-BDEs, and 0.007 mg kg^{-1} d^{-1} for BDE-209 (for EH-TBB and BEH-TEBP, no data are available) (Krol et al., 2012). Here, the most conservative RfD (0.002 mg kg^{-1} d^{-1}) was employed in the calculation of the aggregate exposure risk from ∑BFRs. Cancer slope factor (CSF) was assumed to be that of BDE-209 (7×10^{-4} mg day kg^{-1}) (Ni et al., 2012).

With the BFR data in this study, DI, hazard index and cancer risk were derived in Table 2. When the DI of BFRs ranged from 28.3 to 764 pg kg^{-1} d^{-1}, from 1.4 to 38.2 pg kg^{-1} d^{-1} and from 56.5 to 1530 pg kg^{-1} d^{-1} for children, adults, and professional street sweepers, the hazard index increased from 1.4×10^{-5} to 3.8×10^{-4}, from 7.1×10^{-7} to 1.9×10^{-5} and from 2.8×10^{-5} to 7.6×10^{-4}.

While the values for cancer risk in the range of 2.0×10^{-11} to 5.3×10^{-10}, 9.9×10^{-13} to 2.7×10^{-11} and 4.0×10^{-11} to 1.1×10^{-9}, indicated lower risk for both non-cancer (hazard index <1) and caner (the threshold level 10^{-6}). Conclusively, risk derived by BFR exposure through RD is negligible, however, it is obvious that the exposure level and cancer risk for professional street sweepers were approximately twice as high as the level found for children and one order of magnitude higher than that for adults.

3.5 Contamination load and implications for BFR fate

According to the Chinese Standard for Quality and Assessment of City Road Sweeping and Cleaning (CJJ/T126-2008), the deposition rate of urban road dust in China is 100 g $m^{-2}\cdot d^{-1}$ (Zhao et al., 2014) of which 10% (w/w) is <25 μm (Zhao et al., 2010; Wang and Feng, 2011). The mechanical cleaning area in Xinxiang City is about 7.33×10^{6} m^2, and thus the total amount of RD with particle size <25 μm is approximately 73.3 tons d^{-1}. Based on the measured BFR concentrations, the estimated mass of BFRs associated with RD in Xianxiang City alone ranges between 0.09 to 0.4, from 0.2 to 7.0, from 0.02 to 0.5, from 0.04 to 5.1, from 0.5 to 12.3 kg $year^{-1}$ for ∑6BDEs (excluding BDE-209), BDE-209, EH-TBB, BEH-TEBP and ∑9BFRs, respectively. Further, the vehicle amount of China was 162 million in 2015 and Xinxiang owned 0.4 million, from which it is deduced mass of BFRs associated with RD in China may be up to 162, 2840, 203, 2070 and 4980 kg $year^{-1}$ for ∑6BDEs (excluding BDE-209), BDE-209, EH-TBB, BEH-TEBP and ∑9BFRs, respectively. RD is thus an important sink of BFRs and also an important source of BFRs to the environment. Large proportion of RD is possible to enter into waste water treatment plants, to go through long range atmospheric transmission, or to be treated via incineration and landfill, which will contribute to the formation of atmospheric BFRs or

296 PBDD/F (Zhang et al., 2016). Because Xinxiang is only a middle-scale city in China, it is
297 deduced that with much higher traffic density in metropolises, BFR contaminations in RD would
298 be much higher. As a result, from a national or global perspective, considering the tremendous
299 urban areas and dust load on roads, RD should be a significant source of BFRs in the
300 environment. Systematic monitoring and risk assessment programs should be instituted in future.
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302

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313 **Supporting Information Available**

314 Further detailed information is available free of charge via the Internet at xx.
315

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453 **Table 1**. Summary of the BFR levels in those road dust samples

		Mean	Geomean	SD	RSD	Min	Max
traffic intersections (N=4, LOI=10.4±1.6%)	∑BDEs	10.7	10.4	3.0	28.2%	6.9	13.9
	BDE-209	209	207	32.3	15.4%	173	252
	EH-TBB	4.3	3.4	3.3	78.6%	1.5	9.1
	BEH-TEBP	67.6	32.3	84.6	125%	9.0	189
	BFRs	292	276	117	40.1%	196	458
main roads (N=14, LOI=9.8±1.9%)	∑BDEs	9.8	9.5	2.4	24.6%	6.1	12.7
	BDE-209	102	94.5	40.9	40.2%	51.6	206
	EH-TBB	8.9	6.3	6.0	68.1%	0.9	19.1
	BEH-TEBP	63.3	60.2	22.6	35.7%	40.4	122
	BFRs	184	177	52.7	28.7%	101	306
collector streets (N=11, LOI=10.3±3.9%)	∑BDEs	9.1	8.4	3.6	39.8%	3.2	15.1
	BDE-209	79.6	55.0	72.5	91.1%	10.6	261
	EH-TBB	3.4	3.0	2.0	56.8%	1.5	7.9
	BEH-TEBP	70.8	59.0	41.9	59.2%	20.1	165
	BFRs	163	140	91.7	56.3%	43.1	353
Bypasses (N=7, LOI=9.2±2.5%)	∑BDEs	11.2	11.0	2.3	20.7%	9.1	15.5
	BDE-209	64.8	61.0	22.2	34.2%	31.3	88.9
	EH-TBB	3.8	3.1	2.0	54.0%	0.7	6.1
	BEH-TEBP	24.2	22.5	8.7	35.9%	10.6	33.7
	BFRs	104	102	23.6	22.7%	77.0	141
Parks (N=4, LOI=9.6±3.7%)	∑BDEs	7.1	7.1	0.5	7.2%	6.4	7.5
	BDE-209	58.3	31.8	55.2	94.8%	5.7	114
	EH-TBB	1.6	1.5	0.5	29.9%	1.3	2.3
	BEH-TEBP	3.3	3.0	1.3	38.4%	1.5	4.4
	BFRs	70.2	50.4	56.2	80.0%	17.0	127

454 LOI: loss of ignition, which reflects the organic content of dust samples.

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456 **Table 2**. Estimated DI, hazard index and cancer risk of BFRs for the three types of populations

		Min	Mean	Geomean	Median	Max
DI ($pg\ kg^{-1}\ d^{-1}$)	children	28.3	273	231	245	764
	adults	1.4	13.6	11.6	12.2	38.2
	street sweepers	56.5	545	463	490	1530
Hazard index	children	1.4×10^{-5}	1.4×10^{-4}	1.2×10^{-4}	1.2×10^{-4}	3.8×10^{-4}
	adults	7.1×10^{-7}	6.8×10^{-6}	5.8×10^{-6}	6.1×10^{-6}	1.9×10^{-5}
	street sweepers	2.8×10^{-5}	2.7×10^{-4}	2.3×10^{-4}	2.5×10^{-4}	7.6×10^{-4}
Cancer risk	children	2.0×10^{-11}	1.9×10^{-10}	1.6×10^{-10}	1.7×10^{-10}	5.3×10^{-10}
	adults	9.9×10^{-13}	9.5×10^{-12}	8.1×10^{-12}	8.6×10^{-12}	2.7×10^{-11}
	street sweepers	4.0×10^{-11}	3.8×10^{-10}	3.2×10^{-10}	3.4×10^{-10}	1.1×10^{-9}

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Figure Captions

Fig. 1. Concentration variations of BFRs in RD with sampling locations (∑BDEs contain all PBDE congeners except for BDE209).

Fig. 2. Average congener profiles of BFRs in dust samples. Whiskers on the bars represent standard deviations for BFR congener.

Fig. 3. PBDE concentrations and profiles (median values were adopted for all these data) in car dust and urban RD. Different symbols are used to differentiate the countries/regions (♠ for US, □ for EU countries and ☼ for China) (Gearhart and Posselt, 2006; Stuart et al., 2008; Batterman et al., 2009; Lagalante et al., 2009; Cunha et al., 2010; Harrad and Abdallah, 2011; Lagalante et al., 2011; Kalachova et al., 2012; Thuresson et al., 2012; Shi et al., 2014).

477 **Fig. 1**

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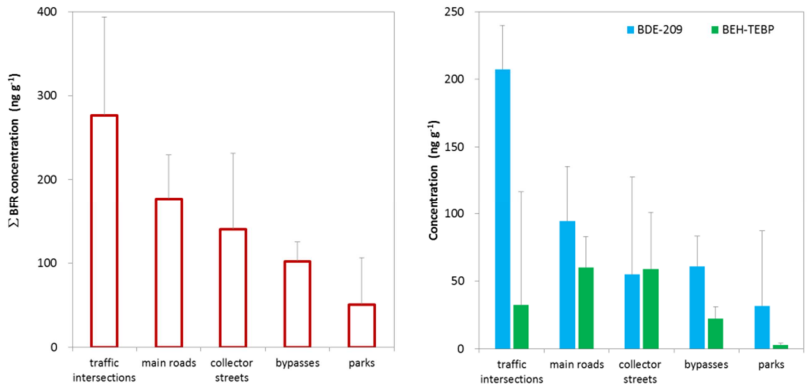

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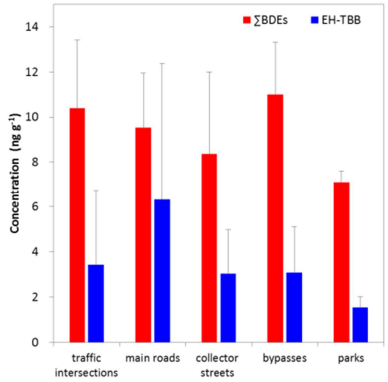

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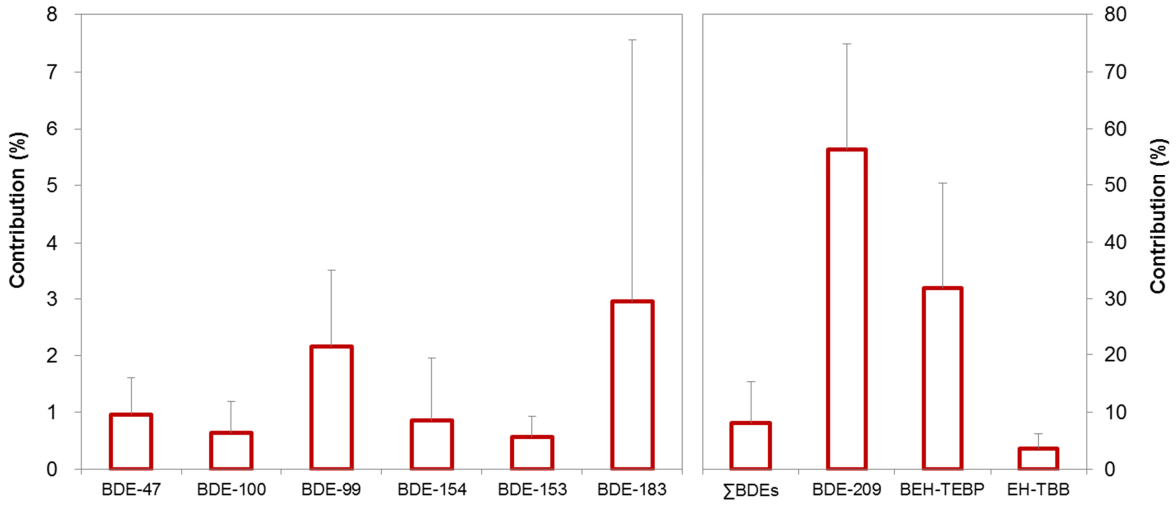
481 **Fig. 2**



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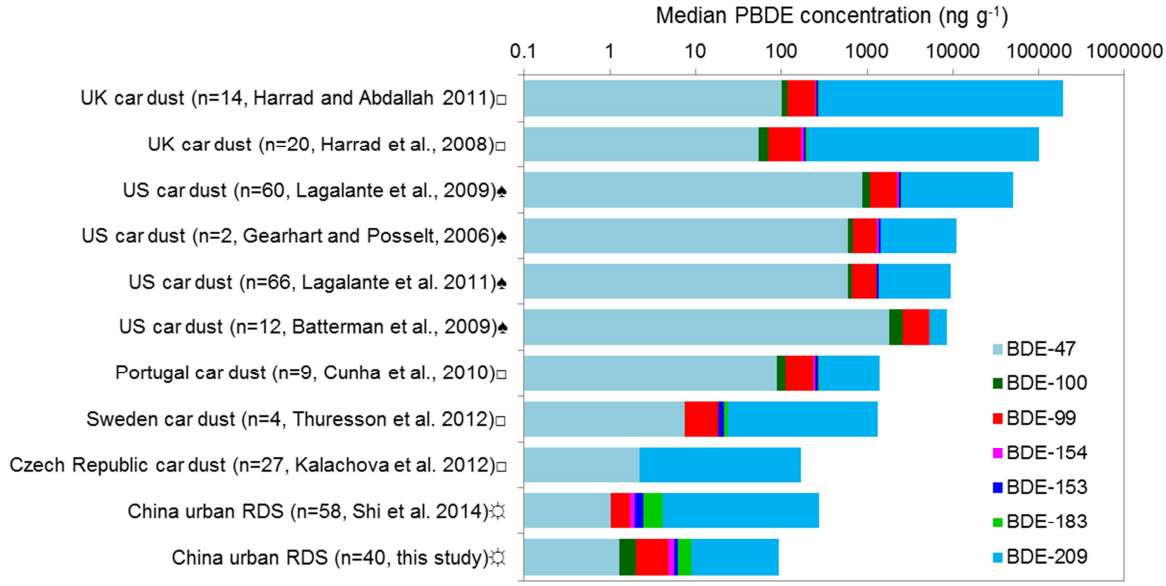
484 **Fig. 3**



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BDE-209 and BEH-TEBP dominated in the road dust samples.

BFR abundance in road dust declined as traffic density increased.

Traffic was deduced to be an important outdoor emission source of BFRs.

BFRs associated with road dust in China were estimated up to be 4980 kg $year^{-1}$.