



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



Zhiguo Cao^{a, b}, Leicheng Zhao^a, Jiangmeng Kuang^{b, c}, Qiaoying Chen^a, Guifen Zhu^a, Kunlun Zhang^{b, c}, Shihua Wang^a, Peipei Wu^a, Xin Zhang^a, Xuefeng Wang^a, Stuart Harrad^{c, *}, Jianhui Sun^{a, **}

^a School of Environment, Henan Normal University, Key Laboratory for Yellow River and Huai River Water Environment and Pollution Control, Ministry of Education, Henan Key Laboratory for Environmental Pollution Control, Xinxiang 453007, China

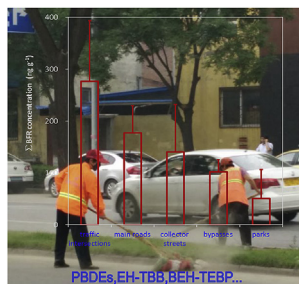
^b Beijing Key Laboratory for Emerging Organic Contaminants Control, School of Environment, Tsinghua University, Beijing 100084, China

^c School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham B152TT, United Kingdom

HIGHLIGHTS

- 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.

GRAPHICAL ABSTRACT



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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 \sum 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.

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* Corresponding author.

** Corresponding author.

E-mail addresses: S.J.Harrad@bham.ac.uk (S. Harrad), sunjh@htu.cn (J. Sun).

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 pentabromoehtylbenzene (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, 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 m² 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, ¹³C-BTBPE, ¹³C-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.

2.3. 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 iso-octane 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 Tables 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.

2.4. QA/QC

Average ± σ_{n-1} recoveries of BDE-77, BDE-128 and ¹³C-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.,

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