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Concentrations of "legacy" and novel brominated flame retardants in matched samples of UK kitchen and living room/bedroom dust

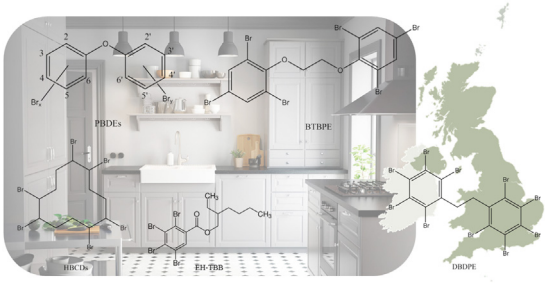
Jiangmeng Kuang*, Yuning Ma, Stuart Harrad

School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham B15 2TT, UK

HIGHLIGHTS

- First report of BFRs in domestic kitchen dust.
- Levels of most BFRs significantly lower in kitchen than living room/bedroom dust.
- Lower levels in kitchens may be due to more frequent cleaning and fewer BFR sources.
- BDE-209 and DBDPE in house dust respectively decreased and increased since 2006–07.

GRAPHICAL ABSTRACT



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ABSTRACT

Concentrations of polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDDs) and 5 novel brominated flame retardants (NBFRs) were measured in paired samples of kitchen and living room/bedroom dust sampled in 2015 from 30 UK homes. BDE-209 was most abundant (22–170,000 ng/g), followed by γ-HBCDD (1.7–21,000 ng/g), α-HBCDD (5.2–4,900 ng/g), β-HBCDD (2.3–1,600 ng/g), BDE-99 (2.6–1,440 ng/g), BDE-47 (0.4–940 ng/g), decabromodiphenyl ethane (DBDPE) (nd-680 ng/g) and bis(2-ethylhexyl)-3,4,5,6-tetrabromo-phthalate (BEH-TEBP) (2.7–630 ng/g). The concentrations in kitchens and living rooms/bedrooms are moderate compared with previous studies. Concentrations of BDE-209 in living room/bedroom dust were significantly lower and those of DBDPE significantly higher ($p < 0.05$) compared to concentrations recorded in UK house dust in 2006 and 2007. This may reflect changes in UK usage of these BFRs. All target BFRs were present at higher concentrations in living rooms/bedrooms than kitchens. With the exception of BDE-28, pentabromoethylbenzene (PBEB) and DBDPE, these differences were significant ($p < 0.05$). No specific source was found that could account for the higher concentrations in living rooms/bedrooms.

1. Introduction

In order to comply with flame retardancy regulations in many jurisdictions, flame retardants (FRs) are widely added to textiles, plastics and building materials. At the current time, brominated flame retardants (BFRs) remain the most widely used class of FRs across the world, including: polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDDs), tetrabromobisphenol A (TBBPA), decabromodiphenyl ethane (DBDPE) and 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE) (Alaee et al., 2003; Covaci et al., 2011). To date, a number of studies have reported potential adverse human health impacts for some BFRs, including

* Corresponding author.
E-mail address: kuangjiangmeng@163.com (J. Kuang).

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thyroid toxicity (Meerts et al., 2000), neurotoxicity (Dingemans et al., 2011), reproductive toxicity (Meeker et al., 2009) and carcinogenicity (Darnerud, 2003). In addition, BFRs like PBDEs and HBCDDs are persistent, bioaccumulative and capable of undergoing long range environmental transport (Dickhut et al., 2012; Marvin et al., 2011; Wu et al., 2011; Zhang et al., 2009; Zhu et al., 2013). Owing to emissions from the myriad range of goods within which they have been incorporated, BFRs are ubiquitous in the environment and have been detected in nearly all abiotic environmental compartments (including water, air, soil, sediments, sewage sludge and dust) (Besis and Samara, 2012; Cristale et al., 2013; Gorga et al., 2013; Luo et al., 2013; Zhu et al., 2008). Such contamination has led to the widespread presence of BFRs in biota such as insects, birds and mammals (Gaylor et al., 2012; Guo et al., 2012; Jorundsdottir et al., 2013), as well as human tissues like hair, breast milk and blood serum (Aleksa et al., 2012; Kim and Oh, 2014; Lee et al., 2013; Sjödin et al., 2014; Tang et al., 2013).

Current understanding is that human exposure to PBDEs and HBCDDs occurs via a combination of diet, indoor dust ingestion, dermal exposure, and inhalation of (largely indoor) air (Abdallah et al., 2008; Besis and Samara, 2012; Daso et al., 2010; Johnson-Restrepo and Kannan, 2009; Trudel et al., 2011). The suspected ecological and human health risks of BFRs have driven international regulation of production and use of some. Specifically, the commercial Penta- and Octa-BDE formulations have been banned worldwide and listed under the UNEP Stockholm Convention on persistent organic pollutants (POPs) since 2009 (Ashton et al., 2009). Moreover, the commercial Deca-BDE formulation has also been restricted severely in Europe since July 2008 (European Court of Justice (2008)), and is currently under active consideration for listing under the Stockholm Convention. In addition, HBCDDs was listed under Annex A of the Stockholm Convention in 2013 (Report of COP6, Stockholm Convention, 2013). Such restrictions and bans on PBDEs and HBCDDs, when coupled with the fixed or even increasing market demand for flame retardants is inevitably leading to increased production of alternatives. While organophosphate flame retardants (PFRs) are one alternative, others include the so-called "novel" BFRs (NBFRs) such as: DBDPE, BTBPE, pentabromoethylbenzene (PBEB), bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (BEH-TEBP) and 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB). However, despite their perceived low cost and high performance, there exist substantial concerns about the environmental impacts of these and other NBFRs. Combined with the substantial remaining inventory of goods containing banned (or "legacy") BFRs and their persistence in the environment, this increased use of NBFRs means that environmental concerns about BFRs will remain an important issue for a considerable time.

With respect to the contamination of indoor dust with BFRs, most attention has been paid to house dust, with offices, cars and schools also featuring in some studies (Harrad et al., 2010). Within homes, the majority of studies have examined living room dust, with a smaller proportion studying bedrooms. To our knowledge however, no data exist about concentrations of BFRs in dust from domestic kitchens. This is a surprising omission, given that people may spend a substantial proportion of time in this microenvironment, and that kitchens contain a substantial number of goods such as microwave ovens, dishwashers, food processors, fridges, and freezers etc. that because their plastic components represent a fuel source in the event of fire, are likely to be flame-retarded.

Given this background, the objectives of this study are: 1. to report for the first time the concentrations of selected BFRs in kitchen dust; 2. to test the hypothesis that concentrations of BFRs in domestic kitchen dust exceed those in dust sampled simultaneously from other areas (living rooms/bedrooms) in the same houses, and 3. to test the hypothesis that restrictions on PBDEs in the EU, have led to reductions in concentrations of PBDEs in dust from UK living rooms, accompanied by concomitant increases in concentrations of NBFRs.

To achieve these objectives, we determined concentrations of 8 PBDEs (BDEs-28, 47, 99, 100, 153, 154, 183 and 209), 5 NBFRs (PBEB, EH-TBB, BTBPE, BEH-TEBP and DBDPE) and HBCDDs (α-, β-, γ-) in paired UK kitchen and living room (or bedroom) dust samples taken from 30 homes in the UK West Midlands conurbation in 2015. Data from kitchens are compared with those from living rooms and bedrooms; with those from living rooms/bedrooms in this study compared with those recorded in an earlier study conducted by our research group of dust from living rooms sampled in the UK West Midlands conurbation in 2006–07.

2. Material and methods

2.1. Sampling

In total, 30 homes from the West Midlands conurbation in the UK (of which Birmingham is the main city) were sampled in 2015. For each home, a dust sample from the kitchen floor was collected with a floor dust sample collected from the living room in the same house for comparison. For the 11 homes in which the living room and kitchen were in the same room, dust in the bedroom was collected instead. For carpeted floor, dust was collected by vacuuming a 1 m^2 area for 2 min; while for bare floors, the vacuuming area and time were 4 m^2 and 4 min, respectively. More details about dust collection and storage protocols have been described in our previous studies (Harrad et al., 2008). An aliquot of 2–3 g pre-baked sodium sulfate vacuumed from a clean Al foil surface served as a field blank.

2.2. Chemicals

Native BDEs 77 and 128, ^{13}C-BTBPE, ^{13}C-BEH-TEBP, ^{13}C-BDE-209 and ^{13}C- α-, β-, γ-HBCDDs were used as internal standards. All standards above were purchased from Wellington Laboratories Inc. All solvents used (acetone, hexane, iso-octane and methanol) were HPLC grade.

2.3. Clean-up

First, 50–100 mg dust was accurately weighed and spiked with 25 ng internal (surrogate) standards. Hexane: acetone (3:1) (2 mL) was added to the sample, which was vortexed for 60 s, sonicated for 5 min and centrifuged at 2000 g for 2 min. After collecting the supernatant, the residues were subjected to the same extraction process twice more. The combined supernatants were reduced in volume to ~2 mL under a gentle stream of nitrogen gas, before mixing with 3–4 mL 98% sulfuric acid. The mixture was then vortexed for 20 s followed by centrifugation at 2000 g for 5 min. The supernatant was then collected. To ensure complete transfer, the residue was rinsed with hexane (2 mL) three times. The combined supernatant was then reduced to incipient dryness under a gentle stream of nitrogen gas. The final concentrate was re-dissolved in 200 μL iso-octane prior to analysis of PBDEs and NBFRs by GC–MS. Following GC–MS analysis, solvent exchange from iso-octane to methanol was conducted to facilitate determination of HBCDDs on LC-MS-MS.

2.4. Analytical methods

2.4.1. GC–MS

A Thermo Trace 1310 gas chromatography interfaced with an ISQ