Chemosphere 146 (2016) 346–353

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Polybrominated diphenyl ethers and polychlorinated biphenyls in dust from cars, homes, and offices in Lagos, Nigeria

Stuart Harrad [a, *], Mohamed Abou-Elwafa Abdallah [a, b], Temilola Oluseyi [c]

[a] School of Geography, Earth, and Environmental Sciences, University of Birmingham, Birmingham, B15 2TT, UK
[b] Department of Analytical Chemistry, Faculty of Pharmacy, Assiut University, 71526 Assiut, Egypt
[c] Department of Chemistry, Faculty of Science, University of Lagos, Lagos, Nigeria

HIGHLIGHTS

- PBDEs and PCBs measured in Nigerian car, home & office dust.
- Penta-BDE levels in 2 cars amongst highest ever reported.
- Only second ever report of PCBs in cars.
- PCB 180 in Nigerian house dust at high end of global range.
- Levels of all target PCBs in Nigerian offices exceed those in cars.

ARTICLE INFO

Article history:
Received 16 October 2015
Received in revised form
9 December 2015
Accepted 13 December 2015
Available online 29 December 2015

Handling Editor: Myrto Petreas

Keywords:
POPs
BFRs
PCBs
Africa
Nigeria
Indoor dust

ABSTRACT

Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) were measured in dust from 16 cars, 12 homes, and 18 offices in Lagos, Nigeria. These represent the first and second reports respectively of contamination of Nigerian indoor dust with these contaminants, and the second report on PCBs in car dust worldwide. Concentrations of BDE-47 and BDE-99 in two car dust samples (9300 and 3700 ng g^{-1} for BDE-47 and 4200 and 19,000 ng g^{-1} for BDE-99), are amongst the highest ever reported in car dust. ANOVA comparison with Canada, New Zealand, the UK, and the USA; reveals concentrations of BDEs-28, 49, 47, 66, 100, 99, 154, and 153 in Nigerian house dust, to be significantly lower than in Canada and the USA, with those of BDE-49 and 154 significantly lower than in New Zealand and the UK. Concentrations of BDE-209 in Nigeria were significantly lower than concentrations in the UK and the USA; while concentrations of PCB-180 were significantly greater than those in New Zealand, the UK, and the USA. Median concentrations of PCBs in cars were substantially higher than in the only previous study (in Kuwait and Pakistan). While median concentrations of PBDEs in cars generally exceeded those in homes, this was significant only for BDEs-49, 154, and 197, with concentrations in cars significantly greater than those in offices for BDEs-49 and 154. Contrastingly, concentrations of all target PCBs in offices exceeded significantly those in cars. This study underlines the truly global distribution of indoor contamination with PBDEs and PCBs.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are chemicals added to a wide range of consumer products (electrical and electronic equipment, textiles, polyurethane and polystyrene foams) to meet flame retardancy standards set by various jurisdictions worldwide (Alaee et al., 2003). Since these chemicals are used additively in most applications – i.e. they are not covalently bound to the products to which they are added – they can transfer from such products into the environment. An extensive body of evidence exists concerning the presence of PBDEs in indoor air (Allen et al., 2007; Harrad et al., 2004; Newton et al., 2015) and indoor dust (Harrad et al., 2008a, b; Jones-Otazo et al., 2005; Stapleton et al., 2005). Evidence of their persistence and capacity for bio-accumulation, coupled with concerns about their adverse health effects (Birnbaum and Staskal, 2004), have led to widespread bans and restrictions on the manufacture and use of both the Penta- and Octa-BDE mixtures and their listing under the Stockholm

* Corresponding author.
E-mail address: S.J.Harrad@bham.ac.uk (S. Harrad).

http://dx.doi.org/10.1016/j.chemosphere.2015.12.045
0045-6535/

Convention on Persistent Organic Pollutants (POPs) (UNEP, 2007). Moreover, manufacture and use of Deca-BDE has been progressively restricted and it is currently under consideration for listing under the Stockholm Convention (UNEP, 2013).

Human exposure to PBDEs occurs via the diet, and via inhalation of (primarily indoor) air, as well as ingestion of indoor dust (Harrad et al., 2004, 2006; Jones-Otazo et al., 2005; Lorber, 2008). The relative significance of each pathway varies considerably according to factors such as: geographical location (dust ingestion appears more important in North America than elsewhere (Harrad et al., 2008b)), age (dust ingestion is considered of greater magnitude for young children than adults (Jones-Otazo et al., 2005)), and the physicochemical properties of a given PBDE congener (exposure to decabromodiphenyl ether (BDE-209) is dominated by dust ingestion as a consequence of its very low vapour pressure and comparatively low capacity for bioaccumulation).

To date, the vast majority of exposure assessments conducted for PBDEs, have been conducted in East Asia (China, Korea, and Japan), Europe, and North America (Besis and Samara, 2012; Harrad et al., 2010). While data is emerging for other regions (including Egypt (Hassan and Shoeib, 2015), Kuwait (Ali et al., 2013; Gevao et al., 2006), and South Africa (Kefeni and Okonkwo, 2012; Abafe and Martincigh, 2015)), to our knowledge only two previous studies exist concerning the presence of PBDEs in indoor dust in Nigerian car and house dust respectively (Olukunle et al., 2015a, b).

Another class of POPs listed under the Stockholm Convention are polychlorinated biphenyls (PCBs). Despite almost universal cessation of their manufacture and new use in the late 1970s, their extensive use in applications such as plasticisers in building sealants and dielectric fluids in capacitors and transformers (Harrad et al., 1994), coupled with their persistence, means that they maintain a discernible environmental presence even today. Their substantial use in indoor applications is manifested by numerous reports of elevated concentrations of PCBs in indoor environments (Currado and Harrad, 1998; Harrad et al., 2006; Herrick et al., 2004; Kohler et al., 2005). Given their comparatively higher vapour pressures than PBDEs, far fewer data exist about concentrations of PCBs in indoor dust than for PBDEs. However, a previous study by our group suggests that although inhalation is the principal indoor exposure pathway under a typical dust ingestion scenario, exposure via dust ingestion exceeds that from either inhalation or diet for a small proportion of North American toddlers (Harrad et al., 2009). To our knowledge, this study constitutes the first measurements of PCBs in indoor dust in Nigeria. Moreover, PCBs in car dust have only been reported in one previous study conducted in Kuwait and Pakistan (Ali et al., 2013).

Against this background, this study seeks to corroborate the recently reported presence of PBDEs in indoor dust from various microenvironments in Makurdi, Benue State, Nigeria (Olukunle et al., 2015a, b), and to provide the first data on concentrations of PCBs in Nigerian indoor dust. We examine exposure in homes, offices and cars because they are oft-frequented environments. Moreover, we provide only the second such report worldwide on concentrations of PCBs in cars. We place our data for Nigerian indoor dust in an international context by comparing the levels found with those reported previously elsewhere. While the Nigerian economy is growing, it is not yet at the level of countries in North America and the EU for example, and thus our overarching hypothesis was that concentrations of PBDEs in Nigeria would be lower than those in more developed countries. However, it has been suggested that import of older electrical and electronic equipment may be an important source of BFRs like PBDEs in countries such as Nigeria (Nnorom and Osibanjo, 2008). As similar considerations may apply to PCBs, we tested our hypothesis by determining concentrations of PBDEs and PCBs in samples of settled dust from 16 cars, 12 homes, and 18 offices in Lagos, Nigeria.

2. Materials and methods

2.1. Sample collection

Dust samples were collected from 16 private cars, 18 offices, and the living areas of 12 houses in Lagos, Nigeria (the largest city in Nigeria) between September and October 2014. House and office dust samples were collected using a vacuum cleaner, according to a standardised method (Harrad et al., 2008b). Briefly, 1 m^2 of carpeted floor was vacuumed for 2 min, while for bare floors, 4 m^2 surface was vacuumed for 4 min. Dust was retained using 25 μm pore size nylon sample socks (Allied Filter Fabric Pty Ltd, Australia) mounted in the furniture attachment tube of the vacuum cleaner. In cars, dust was sampled from the dashboard, seats, and the floor in the passenger cabin, as well as in the boot. After sampling, socks were closed with a twist tie, sealed in a plastic bag and stored at −20 °C until transportation via courier to the University of Birmingham for sieving and analysis. Before sampling, the furniture attachment and the vacuum tubing were cleaned thoroughly using an isopropanol-impregnated disposable wipe. At the time of sample collection, information on potential influences on BFR contamination was recorded. In homes and offices, this comprised the number and type of putative sources like electronic devices, foam-filled furniture and floor material; while in cars, the vehicle manufacturer and age was recorded. Prior to analysis, all dust samples were passed through a pre-cleaned, n-hexane rinsed 500 μm mesh testing sieve (UKGE Limited, UK), covered with the lid and shaken for 2–4 min. Sieved samples were stored in clean, n-hexane rinsed glass jars and stored at 4 °C until analysis.

2.2. Sample extraction

Accurately weighted aliquots of dust (~0.15 g) were loaded into pre-cleaned 66 mL cells containing 1.5 g Florisil and Hydromatrix (Varian Inc., UK) to fill the void volume of the cells, and spiked with internal (surrogate) standards (15 ng of each of BDE 77, BDE 128 and 30 ng of $^{13}C_{12}$-BDE 209) prior to pressurised liquid extraction (ASE 350, Dionex, Hemel Hempstead, UK) using hexane:dichloromethane (1:9, *v/v*) at 90 °C and 1500 psi. The heating time was 5 min, static time 4 min, purge time 90 s, flush volume 50%, with three static cycles (Harrad and Abdallah, 2011).

2.3. Clean up

The crude extracts were concentrated to 0.5 mL using a Zymark Turbovap® II then purified by loading onto SPE cartridges filled with 8 g of pre-cleaned acidified silica (44% concentrated sulfuric acid, *w/w*). The analytes were eluted with 25 mL of hexane:dichloromethane (1:1, *v/v*). The eluate was evaporated to dryness under a gentle stream of nitrogen then reconstituted in 100 μL of isooctane containing 2.5 ng of $^{13}C_{12}$-BDE 100 used as recovery determination (syringe) standard for QA/QC purposes.

2.4. Instrumental analysis

Target PBDEs (BDEs 17, 28, 49, 66, 100, 99, 85, 154, 153, 183, 197, 203, 196, 208, 207, 206, and 209) were quantified using a TRACE™ 1310 Gas Chromatograph coupled to ISQ™ single quadrupole mass spectrometer (ThermoScientific, Austin, TX, USA) operated in negative chemical ionisation mode. Chromatographic resolution of PBDEs was achieved on a HP5-MS capillary column (15 m × 0.25 mm × 0.1 μm; Agilent, CA, USA) according to a previously reported method (Harrad et al., 2008b). PCB analysis was