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## Product screening for sources of halogenated flame retardants in Canadian house and office dust



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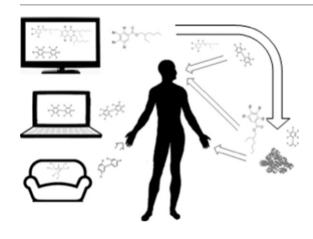
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#### GRAPHICAL ABSTRACT

- Concentrations of flame retardants in dust correlated with product surface wipes
- Most abundant FRs in electronics were PBDEs, TDCPP, DBDPE, EH-TBB and BEHTBP.
- Descending order of FRs in CRTs, TVs, PCs, A-V devices, and small household appliances
- Product wipe testing, but not XRF, useful for non-destructive screening of BFRs
- Removal of FR retardants from product surfaces raises concerns for human exposure.



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#### ABSTRACT

Human exposure to halogenated flame retardants (HFRs) such as polybrominated diphenyl ethers (PBDEs) and their replacements, can be related to exposure to indoor dust and direct contact with HFR-containing products. This study aimed to identify electronic products that contributed to HFRs measured in indoor dust and to develop a screening method for identifying HFRs in hard polymer products. Concentrations of 10 PBDEs and 12 halogenated replacements in dust and surface wipe samples of hard polymer casings of electronic products plus Br in the surfaces of those casing measured using X-ray fluorescence (XRF) were analyzed from 35 homes and 10 offices in Toronto (ON, Canada). HFR concentrations in dust and product wipes were positively correlated. Thus, we hypothesize that electronic products with the highest HFR concentrations contribute the most to concentrations in dust, regardless of the volatility of the HFR. Abundant HFRs in dust and product wipes were PBDEs (BDE-47, 99, 100, 153, 154, 183, 209), TDCPP, DBDPE, EH-TBB and BEHTBP. Older CRT TVs had the highest concentration of BDE-209 of all products tested. This was followed by higher concentrations of HFRs in PCs, Audio/Video (A/V) devices, small household appliances (HHAs) and flat screen TVs. The removal of HFRs from polymer surfaces using wipes supports concerns that HFRs could be transferred from these surfaces to hands as a result of direct contact with HFR-containing products. Surface wipe testing shows promise for screening additive HFRs. In

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comparison, the Br-content obtained using a handheld XRF analyzer did not correspond to concentrations obtained from surface wipe testing.

#### 1. Introduction

PBDEs, as three main commercial mixtures of c-pentaBDE, coctaBDE and c-decaBDE,<sup>2</sup> were widely used as flame retardants (FRs) in various consumer products beginning in the 1970s (Prevedouros et al., 2004; Abbasi et al., 2015). Elevated concentrations of PBDEs are well documented in indoor (e.g., Allen et al., 2008; Harrad et al., 2008, 2010; Zhang et al., 2011; Shoeib et al., 2012) and outdoor environments (e.g., de Wit, 2002; Hites, 2004; Melymuk et al., 2012), and have resulted in widespread human and ecosystem exposure (e.g., Sjödin et al., 2008; Siddique et al., 2012; Crimmins et al., 2012; Buttke et al., 2013). Exposure to PBDEs continues to raise concerns due to increasing evidence of their endocrine modulation effects (Bellanger et al., 2015 inter alia, Lyche et al., 2015 inter alia) such as alteration of thyroid and estrogen and androgen hormones (Ernest et al., 2012), delayed time to pregnancy (Harley et al., 2011), and developmental neurotoxicity (Herbstman et al., 2010; Eskenazi et al., 2013; Roth and Wilks, 2014).

As a result of health concerns and persistence, the congeners of cpenta- and c-octaBDE were added in 2009 to the list of chemicals for elimination under the Stockholm Convention (UNEP, 2010). The production of c-penta- and c-octaBDE was voluntarily phased out by chemical producers in the U.S. in 2004. Canada banned the production and new use of c-penta- and c-octaBDE in 2008 (Environment Canada, 2013). DecaBDE was listed for authorization under REACH<sup>3</sup> in 2010, meaning that decaBDE will be progressively replaced by alternative flame retardants in new products. In 2013, Norway nominated decaBDE for inclusion as a POP (persistent organic pollutant) under the Stockholm Convention (UNEP, 2013). As of 2010, three main manufacturers of decaBDE began to voluntarily phase out the export and sale of decaBDE for certain applications in Canada (Environment Canada, 2013). In Canada, there are no specific controls on PBDEs in products although this is currently under consideration (Environment Canada, 2015). The production, importation and sales of decaBDE were expected to be discontinued in the U.S. following 2013 (U.S. EPA, 2015). Despite the cessation in production and new uses in North America and Europe, the stock of PBDEs in in-use products, which was estimated to be ~120,000 tonnes in the U.S. and Canada in 2014 (Abbasi et al., 2015), remains a source of PBDEs to the indoor and ultimately outdoor environment.

The replacement of PBDEs with "novel flame retardants" (NFRs) has resulted in a proliferation of other brominated flame retardants (BFRs) and halogenated and non-halogenated organophosphate flame retardants (OPFRs) (Ceresana, 2014). A growing literature is documenting the levels of NFRs in indoor dust (e.g., Ali et al., 2011; Cao et al., 2014; Cequier et al., 2014; de Wit et al., 2012; Newton et al., 2015; Shoeib et al., 2012; Stapleton et al., 2012a) and outdoor environments (e.g., Ma et al., 2012, 2013; Salamova and Hites, 2011, 2013).

Exposure to halogenated flame retardants (HFRs), particularly in North America, can occur through ingestion and inhalation of contaminated house dust (Jones-Otazo et al., 2005; Lorber, 2008; Trudel et al., 2011). Several studies have correlated concentrations of PBDEs in house dust with those in serum and breast milk (Björklund et al., 2012; Johnson et al., 2010; Wu et al., 2007; Watkins et al., 2011). However, given the stronger correlation between PBDEs in hand wipes or hand-to-mouth behavior and serum (rather than dust), PBDE transfer via hands may be the more likely route of exposure (Stapleton et al., 2008, 2012b; Watkins et al., 2011, Buttke et al., 2013). Similar evidence of exposure via hands is now emerging for NFRs (Stapleton et al., 2014). Thus, direct contact with FR-containing products and FR-contaminated dust are the two suggested sources of FR to hand transfer (Stapleton et al., 2008, 2014).

To minimize exposure from FR-contaminated dust or direct contact with FR-containing products, the sources need to be traced back to products with the highest FR release rate or mobility. Studies that have sampled dust from specific room locations have found that particular products, such as electronics, contribute to FR concentrations in dust (Muenhor and Harrad, 2012; Harrad et al., 2009). By assessing patterns among rooms, several studies have found correlations between FRs in dust and the prevalence of electronic equipment or furniture containing polyurethane foam (PUF) (Hazrati and Harrad, 2006; de Wit et al., 2012). Recently, Li et al. (2015) found a strong positive correlation between the power consumption of electronics and PBDE levels in a large room, which they attributed to heat generated from in-use electronics enabling the release of FRs. In contrast, several studies have failed to find a correlation between the PBDE concentrations in house dust and the number of electronic products or furniture in indoor environments (Kang et al., 2011; Kefeni and Okonkwo, 2012).

Upon failing to find a correlation between PBDEs in house dust and the number of products likely to have contained PBDEs, Allen et al. (2008) used X-ray fluorescence (XRF) to identify products containing bromine (Br) as an indicator of PBDEs. They found a correlation between Br levels measured by XRF (XRF-Br) and PBDE concentrations measured by means of GC-MS and an association between Br levels in products and PBDE concentrations in house dust. Stapleton et al. (2011) confirmed the results of Allen et al. (2008) for foam samples, but reported that XRF-Br over-predicted Br determined by GC-MS in those samples containing Firemaster 550. They also reported false positives of XRF-Br in foam products that vielded OPFRs upon GC-MS analysis. Kajiwara et al. (2011) also used XRF to screen for FRs in selected electronic products. Imm et al. (2009), using XRF to identify the sources of PBDEs in 38 U.S. households, found that XRF-Br from televisions (TVs) and upholstered living room chairs were correlated with total pentaBDE congeners in passive air samplers. They also reported that XRF-detected Br levels in sleeping pillows and vehicle seats were strongly correlated with PBDE concentrations in participants' lipid-adjusted blood serum. By means of XRF and forensic microscopy, Webster et al. (2009) explained the mechanisms of PBDEs migration from PBDE-containing products and their distribution in house dust. Three hypotheses have been proposed to account for the migration of additive FRs to dust from products or more specifically, the polymer to which they have been added: (1) volatilization from the polymer followed by air-dust partitioning, (2) abrasion of the polymer surface causing the release of FR-enriched particles or fibers, and (3) direct transfer of FRs from the FR-containing polymer to dust (Kemmlein et al., 2003; Takigami et al., 2008; Webster et al., 2009; Rauert et al., 2014a, 2014b inter alia). Volatilization is expected to be the main mechanism for the release of more volatile compounds whereas abrasion is considered more likely for less volatile compounds (Webster et al., 2009; Rauert et al., 2014a).

The main goal of this study was to understand which products act as a source of PBDEs and replacement HFRs to indoor dust in the context of human exposure. We hypothesized that higher concentrations of HFRs in products would be related to higher concentrations in associated dust. Second, we aimed to further develop the rapid and nondestructive technique of product wipe testing to identify selected HFRs in products.

<sup>&</sup>lt;sup>2</sup> Congeners of each PBDE commercial mixture considered in this study: c-PentaBDE: BDE-17, -28, -47, -71, -99, -100, -153, and -154; c-OctaBDE: BDE-153, -154, and -183; c-DecaBDE: BDE-209.

<sup>&</sup>lt;sup>3</sup> REACH is the regulation on Registration, Evaluation, Authorization and Restriction of Chemicals. It entered into force in 2007. It streamlines and improves the former legislative framework on chemicals in the European Union (EU).

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