



# Emissions and fate of brominated flame retardants in the indoor environment: A critical review of modelling approaches



Ioannis Liagkouridis<sup>a,b,\*</sup>, Ian T. Cousins<sup>b</sup>, Anna Palm Cousins<sup>a</sup>

<sup>a</sup> IVL Swedish Environmental Research Institute, P.O. Box 21060, SE 100 31 Stockholm, Sweden

<sup>b</sup> ITM Department of Applied Environmental Science, Stockholm University, SE 106 91 Stockholm, Sweden

## HIGHLIGHTS

- Current emission models likely underestimate the release of low volatile BFRs from products.
- Material abrasion and direct material–dust partitioning are important, yet understudied emission mechanisms.
- Indoor surfaces can be significant sinks, but the mechanism is poorly understood.
- Indoor fate of low volatile BFRs is strongly associated with particle sorption and transport.
- A mass-balance approach of particle dynamics indoors will help to describe particle-bound transport.

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## ABSTRACT

This review explores the existing understanding and the available approaches to estimating the emissions and fate of semi-volatile organic compounds (SVOCs) and in particular focuses on the brominated flame retardants (BFRs). Volatilisation, an important emission mechanism for the more volatile compounds can be well described using current emission models. More research is needed, however, to better characterise alternative release mechanisms such as direct material–particle partitioning and material abrasion. These two particle-mediated emissions are likely to result in an increased chemical release from the source than can be accounted for by volatilisation, especially for low volatile compounds, and emission models need to be updated in order to account for these. Air–surface partitioning is an important fate process for SVOCs such as BFRs however it is still not well characterised indoors. In addition, the assumption of an instantaneous air–particle equilibrium adopted by current indoor fate models might not be valid for high-molecular weight, strongly sorbing compounds. A better description of indoor particle dynamics is required to assess the effect of particle-associated transport as this will control the fate of low volatile BFRs. We suggest further research steps that will improve modelling precision and increase our understanding of the factors that govern the indoor fate of a wide range of SVOCs. It is also considered that the appropriateness of the selected model for a given study relies on the individual characteristics of the study environment and scope of the study.

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## 1. Introduction

Brominated flame retardants (BFRs) are a diverse class of semi-volatile organic compounds (SVOCs) which have been reported to be present in the outdoor (see e.g. Covaci et al. (2011), de Wit (2002), Law et al. (2006)) as well as the indoor environment (see e.g. Harrad et al., 2010). BFRs have been extensively used as additives or reactive constituents to increase fire resistance of a wide range of materials and commercial products. Globally, the BFRs that have been most widely produced (see Table 1) and studied are polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDs) and

tetrabromobisphenol A (TBBPA). The use of certain BFRs has been associated with adverse effects on wildlife and human health (Darnerud, 2003; Simonsen et al., 2000). Due to their ubiquitous presence in the environment and their potential harmful effects, BFRs have undergone extensive risk assessments (EC, 2008; ECB, 2002, 2006) and are subject to regional or international regulatory restrictions and controls (BSEF, 2012). Having been characterised as bioaccumulative, toxic and persistent with a potential for long-range transport, penta- and octa-BDE commercial formulations were listed as persistent organic pollutants (POPs) under the Stockholm Convention on POPs, while HBCD is currently proposed for listing under the Convention. Additionally, the use of deca-BDE in electronic and electrical equipment has been prohibited in the EU since 2008 under the EU Restriction of Hazardous Substances Directive (RoHS) while a voluntarily phase-out of deca-BDE by BSEF

\* Corresponding author. Tel.: +46 8 598 56 300; fax: +46 8 598 56 390.

E-mail address: [ioannis.liagkouridis@ivl.se](mailto:ioannis.liagkouridis@ivl.se) (I. Liagkouridis).

**Table 1**  
Brominated flame retardant (BFR) applications (Waaaijers et al., 2013).

Application	Main BFR
Printed circuit boards	TBBPA
Electronic components encapsulations	
Technical laminate	
Housings of electronic products	TBBPA
Wiring parts	deca-BDE
Housings for business machines, toys, telephones, and others consumer electronics	Other BFRs
Electrical and electronic equipment, connectors, switches etc.	Brominated polystyrenes and other BFRs
Encapsulated electronic components	
Wire and cables	deca-BDE and other BFRs
Textile coatings	HBCD

(Bromine Science and Environmental Forum) member companies is expected to occur by the end of 2013 in the US and Canada (BSEF, 2012).

The continuous demand for flame retardants in combination with the gradual enforcement of bans and restrictions on the use of the most widely used BFRs has given way to two groups of alternative BFRs known as ‘emerging’ and ‘novel’ brominated flame retardants. ‘Emerging’ BFRs refer to chemicals which are “documented regarding production and use as FRs that have been shown to occur/distribute to the environment and/or wildlife, humans or other biological matrices” and ‘novel’ BFRs include chemicals which are “documented as potential FRs that have been shown to be present in materials or products” (Bergman et al., 2012). Following this shift towards alternative BFRs the scientific community has raised the alarm over potential health risks arising from their use (EFSA, 2012). Moreover, steps towards substitution of BFRs with non-halogenated or halogen-free flame retardants (HFFRs) have been made and an evaluation of the hazardous properties of these HFFR replacements performed (Waaaijers et al., 2013).

The indoor environment is of particular importance for human exposure to organic contaminants since many chemicals are released from consumer products used indoors resulting in high indoor concentrations and the majority of people spend on average 85–98% of their time indoors (Long et al., 2000; Schweizer et al., 2007; Wilford et al., 2004). Numerous studies that aimed to determine BFR levels in indoor air and dust have been conducted (e.g. Abdallah et al., 2008; Ali et al., 2011; Allen et al., 2007; Batterman et al., 2009; Saito et al., 2007; Shoeib et al., 2004, 2012; Sjodin et al., 2008; Stapleton et al., 2005; Stuart et al., 2008; Wilford et al., 2004, 2005). Many of these studies have reported elevated BFR concentrations in indoor air, ranging up to 3 orders of magnitude compared to outdoors. Studies that attempt to identify and quantify indoor exposure routes (Fromme et al., 2009; Harrad et al., 2004; Jones-Otazo et al., 2005; Lorber, 2007) and link indoor levels with levels in humans (Karlsson et al., 2007; Wu et al., 2007) and possible health implications (Johnson et al., 2013; Meeker et al., 2009) have also been published. In addition, the indoor environment as a potential source of BFRs to the outdoor environment has been advocated (Björklund et al., 2012; Cousins, 2012).

Chemical fate models have been developed in order to study indoor fate of organic chemicals (e.g. Bennett and Furtaw, 2004; Matoba et al., 1998; Zhang et al., 2009). However, the utilisation of modelling tools for such purposes can be challenging due to limitations in model parameterisation. This review aims to explore the existing understanding of the indoor fate of BFRs and current modelling approaches. We critically examine two of the key individual components needed to understand and model indoor fate, namely; emissions and indoor fate processes. We also critically review previous attempts to model the emissions and indoor fate of SVOCs. The key limitations in the current understanding of fate and modelling approaches are discussed and suggestions for further research made with the goal of providing a road

map for future experimental and modelling research focused on understanding the indoor fate and exposure of BFRs.

## 2. BFR sources and emissions indoors

### 2.1. Linking concentrations to indoor sources

It is believed that the frequent occurrence of BFRs in the indoor environment is a result of a large amount of BFR-containing products typically located indoors (Table 1). BFRs are incorporated in polymeric materials in quantities up to 33% (Alaee et al., 2003). A summary of the global annual production of polymers and the associated BFR content in 2001 is given in Table 2. Harju et al. (2009) presented an overview of the potential occurrence of alternative BFRs in polymeric and non-polymeric materials along with their commercial applications.

Only few studies have been successful in correlating BFR levels in indoor air and dust with BFR-containing products which can act as indoor sources (de Wit et al., 2012; Harrad et al., 2004), although several attempts have been made (Hazrati and Harrad, 2006; Stapleton et al., 2005; Wilford et al., 2004, 2005; Wu et al., 2007; Zhang et al., 2011). Researchers suggest that a number of product-related factors such as numbers, age, BFR content, usage pattern of products as well as environment-specific factors including temperature, humidity, ventilation rate etc. influence such complex relationships; the role of different microenvironments is often pointed out (Allen et al., 2008a; Hazrati and Harrad, 2006). The above hypothesis gained further support in a recent study by de Wit et al. (2012) who reported good correlations between PBDEs and HBCD in indoor air and dust and microenvironment characteristics.

Despite the fact that a clear, consistent relationship between indoor levels and possible sources is hard to obtain, some general trends have been observed. Harrad et al. (2004) measured the highest PBDE levels in rooms equipped with large numbers of computers and PUF (polyurethane foam) chairs, whereas the least contaminated samples came from indoor environments containing no PUF. Based on a sampling campaign in 74 houses, Wilford et al. (2004) reported an association between indoor air concentrations of PBDEs and practices such as building maintenance work and introduction of new possible sources such as personal computers and carpets. Hazrati and Harrad (2006) observed a sharp decrease (80%) of PBDEs in indoor air after a personal computer was replaced with a newer model. In a series of studies conducted in Japan (Hirai et al., 2006; Takigami et al., 2008; Tamade, 2002) elevated BFR concentrations were found in dust accumulated inside TV sets compared to typical house dust concentrations. The role of electronic equipment as a significant source of BFRs to the indoor environment was also advocated by Zhang et al. (2011) who showed that electronic equipment was the main source of PBDEs in areas with higher concentrations; PUF furniture and carpets were identified as likely sources in areas with lower PBDE concentrations. Characterisation of the bromine content of BFR-treated products using XRF technology has also been successful in linking indoor sources and concentrations (Allen et al., 2008b).

### 2.2. Indoor emission of BFRs: rates and mechanisms

It is evident that to sufficiently identify and quantify indoor presence and exposure to BFRs, apart from quantifying their presence in products, a solid understanding of the mechanisms through which these compounds migrate from treated products is required. Emissions are the key input parameter to drive a predictive indoor fate model. Information on emissions can be provided as experimentally derived rates/factors or by using modelling approaches. Volatilisation (gas-phase emission) has so far been considered as the main release mechanism of BFRs from treated products to indoor air, especially for the more volatile compounds. Once volatilised certain BFRs may subsequently partition to dust (Schripp et al., 2010). The elevated levels of very low

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