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Direct contact between dust and HBCD-treated fabrics is an important pathway of source-to-dust transfer

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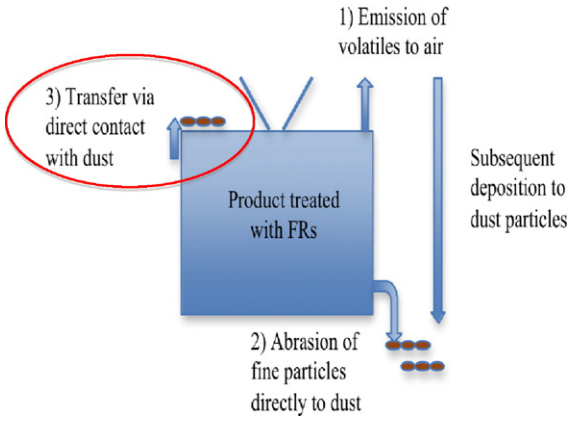
HIGHLIGHTS

- Transfer of HBCDs via direct contact between a curtain and dust was studied.
- Direct curtain:dust contact led to substantial transfer.
- Transfer is rapid yet source:dust equilibrium was not reached after 1 week of contact.
- Results imply regular cleaning of source surfaces may reduce contamination of dust.

GRAPHICAL ABSTRACT



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ABSTRACT

Hexabromocyclododecanes (HBCDs) are a class of brominated flame retardant that have found extensive application in consumer products used widely in indoor environments. Although uncertainty remains about the human health impacts of HBCDs, ingestion of HBCD-contaminated indoor dust has been shown to be a particularly significant exposure pathway for young children. Despite this, understanding of the mechanisms via which HBCD transfer from products to indoor dust remains incomplete. In this study, an in-house test chamber was used to investigate transfer of HBCDs from a treated textile sample to indoor dust via direct textile:dust contact. Results were compared with previous data using the same test chamber to examine other pathways via which HBCDs transfer from products to dust, and highlighted HBCD transfer via direct source:dust contact as being particularly important. This novel finding was corroborated by complementary experiments that examined HBCD transfer via direct contact, from other treated textiles to three major components of indoor dust: artificial indoor dust, soil particles, and cotton linters.

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1. Introduction

Hexabromocyclododecanes (HBCDs) are one of the most widely produced classes of brominated flame retardants (BFRs) used to flame retard polystyrene foams for building insulation, fabrics like furniture covers and curtains, and high impact polystyrene casings for electronic equipment (Harrad et al., 2010; Weil and Levchik, 2007). They are incorporated into products via an "additive" process where the HBCD formulation is physically rather than chemically bound to the product/polymer. Consequently, their release into the surrounding environment is relatively facile, leading to their ubiquitous presence in indoor air and dust (Harrad et al., 2010; Covaci et al., 2006). Concentrations in dust can vary by orders of magnitude (Abdallah et al., 2008b) up to 1.1 mg ΣHBCDs g^{-1} (Allen et al., 2013). Such elevated concentrations are of concern, given dust ingestion contributes an estimated 63% of the exposure of UK toddlers to ΣHBCDs (Abdallah et al., 2008a).

Currently hypothesized pathways of BFR transfer from products to dust include: (1) volatilization of BFRs from products with subsequent partitioning to dust; (2) abrasion via physical wear and tear of products, resulting in transfer of particles or fibers of the product directly to dust and (3) transfer via direct contact between product and dust (Suzuki et al., 2009; Takigami et al., 2008; Wagner et al., 2013; Webster et al., 2009). Test chambers constitute a potentially important tool for investigating source-to-dust transfer of BFRs. While test chamber studies to date have focused largely on contaminant emissions to air to determine specific emission rates (SERs) (Rauert et al., 2014a), the mass transfer to dust of phthalates from wall paint and vinyl flooring has been investigated in modified test chambers (Clausen et al., 2004; Schripp et al., 2010). These studies demonstrated phthalate transfer from source to dust occurred via volatilization with subsequent partitioning to dust, and via direct source:dust contact. More recently, a test chamber study by two of the current authors demonstrated rapid and substantial transfer of PBDEs from a TV casing sample to dust (Rauert and Harrad, 2015).

We have reported previously test chamber experiments simulating source-to-dust transfer of HBCD via partitioning post volatilization and via abrasion (Rauert et al., 2014b, 2015). However, to our knowledge, transfer of HBCD via direct source:dust contact has hitherto not been investigated.

This study is the first experimental investigation of HBCD transfer to dust through direct source:dust contact, using a test chamber and a HBCD-treated curtain as the source. Results are compared to previous data reporting HBCD transfer to dust via transfer pathways (1) and (2). Moreover, given the lack of previous data concerning transfer of HBCDs via direct source:dust contact; we report a series of complementary, more detailed follow-up experiments that examine HBCD transfer via direct contact, from four different treated textiles to three major components of indoor dust: artificial indoor dust, soil particles, and cotton linters.

2. Materials and methods

2.1. Experimental design

2.1.1. Initial test chamber experiments

The cylindrical stainless steel test chamber employed to investigate HBCD migration from the test curtain to dust is illustrated in Fig. 1a. With dimensions of 10 cm diameter and 20 cm height, volume of 1570 cm^3, and internal surface area of 785 cm^2, chamber internal temperature was monitored by a LogTag TRIX-8 temperature data logger. An aluminum mesh shelf was placed half way down the chamber. A HBCD treated curtain (7 cm × 3 cm rectangle) was placed on a clean glass fiber filter situated on the shelf, and a thin layer of previously characterized house dust (Rauert et al., 2015) placed evenly on top of the curtain (~500 mg). Concentrations of HBCD diastereomers in the curtains used in test chamber experiments were 18,000,000 ng g^{-1} for α-HBCD, 7,500,000 ng g^{-1} for β-HBCD, and 17,000,000 ng g^{-1} for γ-HBCD (Kajiwara et al., 2013). Dust utilized in test chamber experiments contained low concentrations of HBCDs and PBDEs (ΣHBCDs = 110 ng g^{-1} and ΣPBDEs = 280 ng g^{-1}). The percentage carbon and nitrogen content of this dust was determined, with results supplied as supplementary data (Table SD-1).

The chamber was sealed and left at room temperature (22 ± 1 °C) for either 24 h or 1 week. Post-experiment dust was gently removed from the source by gentle tapping, and homogenized via vortex mixing, ready for analysis. Each time period was studied in quadruplicate with duplicate dust subsamples (200 mg) from each experiment analyzed for HBCDs.

2.1.2. Follow-up experiments

Fig. 1b illustrates the experimental configuration employed in follow-up experiments. Four different kinds of HBCD-treated fabrics including 2 curtains (Curtain-1 and -2) and 2 vehicle seat fabrics (Seat fabric-1 and -2) were used. These materials were manufactured in Japan prior to HBCD's listing as a persistent organic pollutant (POP) under the Stockholm Convention (Annex A). HBCD concentrations in these fabrics ranged from 8,400,000 ng g^{-1} to 21,000,000 ng g^{-1} (Table 3). Each fabric material tested was fixed within a rectangular aluminum frame (outer dimensions 115 mm × 150 mm) with two windows each 53 mm × 83 mm. Prior to each experiment, the test fabric surface was vacuumed to remove small abraded fibers to minimize their inadvertent incorporation in the test dust. Three different types of test particles were used in follow-up experiments. These were: (1) artificial indoor dust (JIS Z 8901 Class 15, see below); (2) fine soil particle (calcined Kanto loam at 800 °C, JIS Z 8901 Class 8, median diameter 6.6–8.6 μm, density 2.9–3.1 g/cm^3, ignition loss 0–4%); and (3) cotton linters (<1.5 μm in diameter, <1 mm in length) (JIS, 2006). The artificial indoor dust was a mixture of test particles, containing 72% fine soil particles (JIS Z 8901 Class 8), 23% carbon black (JIS Z 8901 Class 12, 0.03–0.20 μm, 1.7–1.9 g/cm^3) and 5% cotton linters. The fine soil particles used are primarily inorganic, with negligible organic matter since they were prepared by calcination of loam soil at 800 °C. Initial concentrations of HBCDs in the three test dust types were all below detection limits.

Approximately 20–50 mg of test dust, soil or lint were placed within a 25 mm diameter circle on the surface of the HBCD-treated fabric material in the upper window (Fig. 1b) using a glass cylinder. Excessive dust was removed by gently tapping the reverse side of the aluminum frame. The exposed fabric surface in the lower window (Fig. 1b) was used as a control without dust. An additional blank using dust applied to aluminum foil, instead of a HBCD treated fabric, was also conducted. Each frame was then shielded from light with aluminum foil and maintained at 28 °C and 50% relative humidity in a thermohygrostat test chamber (IG400, Yamato Scientific, Tokyo, Japan). Three experimental durations were examined: 1, 4, and 7 days. Transfer of HBCDs to artificial indoor dust was examined for each of the 4 HBCD-treated fabric materials over three experimental durations. In contrast, transfer to the fine soil particle and cotton linter samples were examined over a single 4 day period for curtains only. Duplicate tests were conducted for Curtain-1 (all dust types) and Seat fabric-1 (artificial indoor dust only). Following each experiment, dust was collected from the surface of each fabric window (with or without dust) and from the aluminum foil by a low volume vacuum pump equipped with a stainless steel filter holder (KS-25, ADVANTEC, Tokyo, Japan) containing a glass fiber filter (GB-100R, 25 cm diameter, 0.6 μm pore size, ADVANTEC, Tokyo, Japan). Dust collected was weighed and analyzed for HBCDs.

2.2. Determination of concentrations of HBCDs

2.2.1. Test chamber experiment samples

Dust from test chamber experiments was analyzed using modified in-house methods (Rauert et al., 2015). A detailed description is provided as supplementary data. Method blanks run with each batch of samples were conducted by extracting a pre-cleaned 66 mL cell filled with