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Spatiotemporal analysis of human exposure to halogenated flame retardant chemicals

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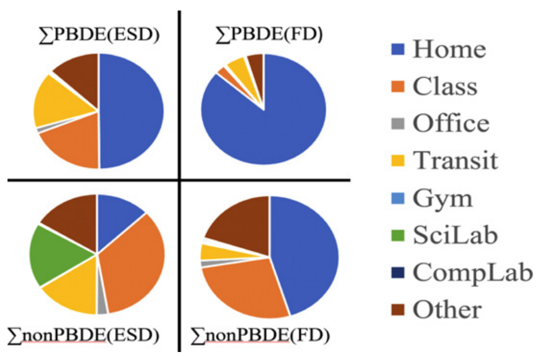
HIGHLIGHTS

- Study participants recorded time activity patterns in diaries.
- Estimation of participant exposure to 18 flame retardant chemicals in different microenvironments
- Comparison of mean exposure estimates through elevated surface dust (ESD) and floor dust (FD).
- Exposure to most flame retardant chemicals was statistically significantly higher in ESD than FD.

GRAPHICAL ABSTRACT



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ABSTRACT

Human exposure to flame retardants occurs in microenvironments due to their ubiquitous presence in consumer products and building materials. Recent research suggests higher levels of exposure through elevated surface dust (ESD) compared to floor dust (FD). However, it is unclear whether this pattern is consistent in different microenvironments beyond the home. We hypothesized that time spent in various microenvironments will significantly modify the pattern of human exposure to flame retardant chemicals in ESD and FD. We tested this hypothesis by collecting time activity diaries from 43 participants; and by estimating human exposure to 10 polybrominated diphenyl ether and 8 non-polybrominated diphenyl ether flame retardant chemicals, based on chemical concentrations measured in different microenvironments visited by the participants. The results of paired *t*-tests show that, with some notable exceptions, estimates of human exposure to most chemicals through ESD are statistically significantly higher for ∑PBDE ($p = 0.00$) and ∑non-PBDEs ($p = 0.00$) than through FD. This study reinforces the need to integrate temporal, locational, and elevation dimensions in assessing human exposure to potentially toxic flame retardant chemicals.

1. Introduction

Assessments of human exposure to toxic chemicals consider the concentration of chemicals that individuals encounter in each specific location where they spend time over a given period. There has been particular interest in estimating human exposure to flame retardant (FR) chemicals due to possible health implications (Kim et al., 2014). In the U.S., indoor dust is considered the primary source of FR exposure

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(Johnson-Restrepo and Kannan, 2009; Lorber, 2008). Many U.S. FR studies have concentrated on sampling the home (Dodson et al., 2012; Johnson et al., 2010; Quiros-Alcala et al., 2011; Ward et al., 2014). However, FR chemical concentrations have been shown to vary across sampled locations (Allgood et al., 2017; Brommer and Harrad, 2015; Cequier et al., 2014; La Guardia and Hale, 2015; Mizouchi et al., 2015). Thus, accounting for where a person spends time is important for refining the precision of estimates of human exposure to FR chemicals.

Additionally, there is evidence that many FR chemical concentrations differ between elevated surface dust (ESD) and floor dust (FD) (Allgood et al., 2017; Al-Omran and Harrod, 2016; Björklund et al., 2012; Cequier et al., 2014; Xu et al., 2016). Yet many U.S. studies do not account for this difference (Dodson et al., 2012; Quiros-Alcala et al., 2011; Watkins et al., 2011). Studies which consider sample elevation mainly focus on the home, ignoring exposure from other locations (Al-Omran and Harrod, 2016; Björklund et al., 2012; Cequier et al., 2014; Xu et al., 2016). It is unknown whether the pattern of higher FR exposure through ESD compared with FD persists when chemical exposure in microenvironments other than the home are considered.

The knowledge gap is wider in cases that estimate human exposure to FR chemicals based on uncorroborated assumptions about time spent in locations. Previous studies assume exposures over 24-h at home or school (Ali et al., 2012; Quiros-Alcala et al., 2011; Wikoff et al., 2015). Other studies rely on a pre-existing Flemish time survey, and adopted a 'typical' time pattern assuming proportion of time spent per day is 72% at home, 23.8% at the office, 4.2% in transport (Ali et al., 2011; Harrad et al., 2008a, 2008b; Roosens et al., 2010). Accounting for actual time spent in microenvironments over 24 h may lead to different exposure estimates. Additionally, estimates of human exposure may differ between ESD and FD with comparable FR concentrations when a temporal dimension is considered.

Time activity diaries account for time spent in different spaces, and have been informative for investigations regarding human exposure to black carbon (Dons et al., 2011), pesticides (Tulve et al., 2008), and ultrafine particles (Buonanno et al., 2014). However, to our knowledge, no studies have estimated FR chemical exposure based on time activity diaries for people in various microenvironments with known chemical concentrations.

In this study of spatiotemporal exposure, we collected time activity diaries from a sample population present in academic microenvironments with known concentrations of FR chemicals. We investigated ten congeners of polybrominated diphenyl ethers (PBDEs) – BDE-28, BDE-47, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, BDE-206, BDE-209; and eleven congeners of non-polybrominated diphenyl ethers (non-PBDEs) – 2-ethyl-hexyl 2, 3, 4, 5-tetrabromobenzoate (EH-TBB), Bis(2-ehtylhexyl)tetra-bromophthalate (BEH-TEBP), 1, 2-bis (2, 4, 6-tribromophenoxy) ethane (BTBPE), decabromodiphenyl ethane (DBDPE), α-, β-, & γ-hexabromocyclododecane ($\sum$HBCD), tris (2-chloroethyl) phosphate (TCEP), tris (1-chloro-2-propyl) phosphate (TCIPP), tris (1,3-di-chloro-2-propyl) phosphate (TDCIPP), and tetrabromobisphenol-A (TBBPA). With these data, we tested the hypothesis that adding a refined temporal dimension will modify estimates of human exposure to FR chemicals across microenvironments with ESD and FD.

2. Methods

2.1. Time activity diaries

The research protocol for human participants was approved by UC Irvine Institutional Review Board. From March 2014 to March 2015, we recruited 43 participants to complete time activity diaries. Participants were included if they were at least 18 years of age, and lived within the academic environment. Each participant was asked to complete a time activity diary for a 24-hour time period during a weekday and a corresponding questionnaire. Participants that returned the time activity diary and questionnaire received a $5 gift card.

The procedure for recording time activity followed previously published method by Olds et al. (2009). We included 14 predefined categories to assess the type of microenvironment in which individuals spent each increment of recorded time, such as home (apartment, dormitory house), travel (car/taxi), travel (foot/bicycle), travel (bus), classroom (with computer), classroom (without computer), office (with computer), office (without computer), wet laboratory, retail store, restaurant, gymnasium, other outdoor space, and other indoor space. For time estimation, all time spent in travel (car/taxi & bus), classroom, office, and laboratory were consolidated into independent categories; and the category of "other" was created from a composite of retail store, restaurant, travel (foot/bicycle), other outdoor space, and other indoor space. We used previously reported concentrations (see Table S1) of each FR chemical measure in each microenvironment, except for the "other" category for which the median value of all sampled locations was used (Allgood et al., 2017). Each study participant responded to questions about demographic characteristics.

2.2. Dust sampling

Specific procedures for dust sample collection are chronicled in Allgood et al. (2017). Briefly, indoor ESD and FD samples were collected from microenvironments on the UC Irvine campus from June 2013–September 2013 using a Eureka Mighty-Mite vacuum cleaner with a crevice tool attached (Allen et al., 2008). The crevice tool was dragged across two sampling areas in each microenvironment for about 15 min each. The two sampling areas included elevated surfaces (surfaces approximately 2 ft above the floor or higher such as sofas and desks) and the floor.

2.3. Chemical analyses

Specific procedures for the dust sample preparation, extraction, chemical analyses methods, and quality control/quality assurance (QC/QA) are chronicled in Allgood et al. (2017). Briefly, accelerated solvent extraction (ASE) was applied to ~100 mg of each ESD and FD sample that had been sieved (300 μm). Then each extract was purified with size exclusion chromatography (SEC). Next, each post-SEC extract was reduced in volume and added to the top of an extraction column. Three fractions were then created with fraction two containing brominated FRs (PBDEs: BDE-28, BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, BDE-206, BDE-209; HBCDs: αHBCD, βHBCD, γHBCD reported as $\sum$HBCD, and brominated non-PBDEs: EH-TBB, BEH-TEBP, BTBPE, DBDPE) and fraction three containing TCEP, TCIPP, TDCIPP and TBBPA. The analytes were separated by ultra-performance liquid chromatography (UPLC), ionized by atmospheric pressure photoionization (APPI), and product ions were detected by triple quadrupole mass spectrometer (MS/MS). The analytical methods were validated using a QC and QA approach that used laboratory blanks, duplicate, surrogate and matrix spike recovery analysis. Additionally, Schreder and La Guardia (2014) describe in further detail the implemented dust sample preparation, chemicals used, extraction methods, UPLC-APPI-MS/MS, and QC/QA methods.

2.4. Flame retardant exposure estimation

We used scenario evaluation which is an indirect approach to estimate cumulative external exposure to FR chemicals (USEPA, 1992). Participants from UC Irvine were assumed to be exposed to previously measured FR chemicals measured in ESD and FD in UC Irvine microenvironments (Allgood et al., 2017). External exposure was estimated separately from FD and ESD by multiplying the indoor dust chemical concentration with time spent in each location and adding the exposure encountered in each location where time was spent over 24 h (Klepeis,