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Authors

Allgood, Jaime M
Vahid, Kelsi Schechter
Jeeva, Keerthanaa
et al.

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Spatiotemporal analysis of human exposure to halogenated flame retardant chemicals

Jaime M. Allgood, Kelsi Schechter Vahid¹, Keerthanaa Jeeva², Ian W. Tang, Oladele A. Ogunseitan^{*}

Department of Population Health & Disease Prevention, University of California, Irvine, CA 92697-3957, United States

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 Σ PBDE ($p = 0.00$) and Σ 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.

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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

^{*} Corresponding author.

E-mail address: Oladele.Ogunseitan@uci.edu (O.A. Ogunseitan).

¹ Present affiliation: Department of Prevention and Community Health, Milken Institute School of Public Health, The George Washington University, Washington, DC 20052.

² Present affiliation: Industrial Hygiene, Environmental Health Division, School of Public Health, University of Minnesota, Minneapolis, Minnesota 55414.

(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-ethylhexyl)tetra-bromophthalate (BEH-TEBP), 1, 2-bis (2, 4, 6-tribromophenoxy) ethane (BTBPE), decabromodiphenyl ethane (DBDPE), α -, β -, & γ -hexabromocyclododecane (Σ 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 μ m). 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: α HBCD, β HBCD, γ HBCD reported as Σ 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,

1999; USEPA, 1992; Ott, 1990). Two external exposure estimates derived from Eq. (1a) FD and Eq. (1b) ESD were calculated for each FR chemical using the following two equations:

$$C_i = C_{FD} \times CR \quad (1a)$$

$$C_i = C_{ESD} \times CR \quad (1b)$$

For FD derived external exposure estimates, the particular FR concentration in each microenvironment (C_i) is the product of the concentration in the FD (C_{FD}) and the amount of dust that each participant comes into contact with per day (CR); and for ESD derived external exposure estimates, the particular FR concentration in each microenvironment (C_i) is the product of the concentration in the ESD (C_{ESD}) and the amount of dust that each participant comes into contact with per day (CR) (USEPA, 2016). We used the central tendency estimate for adults of 30 mg/day or 0.0208 mg/min as the amount of dust that each participant came into contact (USEPA, 2011).

Human exposure for each FR chemical was estimated using the following equation:

$$E = \sum_{i=1}^I C_i t_i \quad (2)$$

In Eq. (2), E = each participant's total integrated exposure estimate for each FR chemical, C_i = the dust concentration of the particular FR in each microenvironment from Eqs. (1a) or (1b), t_i = the amount of time the participant spent in the microenvironment based on time activity diaries, and I = the total number of microenvironments (Klepeis, 1999).

2.5. Statistical analyses

We used SPSS (Version 24.0) for statistical analysis. Sums of the exposure estimates for all the PBDEs ($\sum$ PBDEs) and non-PBDEs ($\sum$ non-PBDEs) were calculated based on ESD and FD chemical concentrations. Summary statistics were calculated for (a) the time spent in each microenvironment, (b) estimated FR exposure from ESD for each chemical, and (c) estimated flame retardant exposure from the FD for each chemical. We performed a paired t -test to determine whether the mean difference between each of pair of exposure estimates from ESD and FD was zero. The results were considered statistically significant when the p -value was 0.05 or less.

3. Results and discussion

3.1. Study participants and time activity patterns

Forty-three study participants completed time activity diaries for a 24-hour time period. Participants were 56% female and 79% between the ages of 18 and 25 years. Regarding ethnicity, 42% were Asian, 23% were Caucasian, 19% were Hispanic, 12% were African-American, and 5% were other or mixed. Table 1 shows the mean values of time spent by each study participant in each microenvironment.

Study participants spent an average of 870 min per day at home, which is higher than at any other location. This is slightly lower than reported by other investigators, for example, Odeh and Hussein (2016) found that, on average, adult students in Jordan spent about 900 min at home (63%). Importantly, our finding of time spent at home is 70 min less than the 940 min reported by the U.S. National Human Activity Pattern Survey (NHAPS) as the average time spent by Americans indoors at home (Leech et al., 2002); 80 min less than the 950 min reported by the Canadian Human Activity Pattern Survey (CHAPS) that Canadians spend at home (Leech et al., 2002); and the average 940 min that Germans spend at home (Brasche and Bischof, 2005). Following time spent at home, other locations and classrooms also claim participant times at an average of 260 min and 160 min, respectively.

Table 1

Mean time period spent (minutes) per microenvironment for study participants.

	N	Mean time spent in minutes						
		Home	Transit	Class	Office	Lab	Gym	Other
All	43(100%)	870	53	160	83	11	12	260
Sex								
Male	19(44%)	850	59	140	94	16	12	270
Female	24(56%)	880	48	170	74	6.3	12	250
Age								
18–25	34(79%)	880	56	150	67	11	13	260
>25	9(21%)	830	40	170	140	10	6.7	250
Race								
Asian	18(42%)	890	63	120	66	10	2.5	280
Black	5(12%)	800	18	220	75	0.0	0.0	320
Hispanic	8(19%)	870	58	130	110	11	7.5	250
White	10(23%)	840	48	200	110	0.0	41	205
Other	2(5%)	960	53	190	0.0	90	0.0	150

The least average time period was spent in transit, laboratories, and gymnasiums at 53 min, 11 min, and 12 min, respectively. Time activity patterns are expected to vary according to age-group, geographic location, seasons, and cultural factors. In general, the pattern of time activities recorded in this study do not show extreme deviation from the norm given the demographic and occupational attributes of the population studied.

3.2. ESD compared to FD flame retardant human exposure estimates assuming 24 hours spent across microenvironments

Data on the mean 24-hour exposure to 10 PBDE and 8 non-PBDE FR chemicals for our sampled population are shown in Table 2. The information includes estimates based on chemical concentrations in ESD and FD, and the mean difference between the exposure estimates from the two sampling sites. A positive difference indicates a higher mean exposure estimate for ESD relative to FD; and a negative difference indicates a higher mean exposure estimate for FD relative to ESD.

Table 2

Comparison of external exposure estimates to flame retardant chemicals derived from elevated surface dust (ESD) and floor dust (FD) and based on 24 h spent across microenvironments.

Flame retardant chemicals	ESD		FD		n	Mean difference	95% CI for mean difference	p^a
	Mean	SD	Mean	SD				
PBDEs								
BDE-28	1.0	0.67	0.052	0.04	43	0.98	(0.78, 1.2)	0.00*
BDE-47	190	61	61	11	43	120	(1100, 140)	0.00*
BDE-85	12	4.1	5.9	1.9	43	5.7	(4.8, 6.5)	0.00*
BDE-99	210	52	97	12	43	110	(100, 1300)	0.00*
BDE-100	48	15	22	4.3	43	27	(23, 30)	0.00*
BDE-153	22	5.2	12	2.7	43	10	(9, 12)	0.00*
BDE-154	18	3.9	8.8	1.1	43	9.1	(8.0, 10)	0.00*
BDE-183	1.4	0.20	1.1	0.15	43	0.27	(0.19, 0.35)	0.00*
BDE-206	8.9	0.94	11	1.7	43	−2.3	(−2.5, −2.1)	0.00*
BDE-209	450	50	540	79	43	−88	(−98, −77)	0.00*
$\sum$ PBDE	960	160	750	97	43	200	(160, 240)	0.00*
Non-PBDEs								
EH-TBB	81	36	68	32	43	14	(3.7, 24)	0.00*
BEH-TEPH	190	85	41	9.4	43	150	(130, 170)	0.00*
BTBPE	0.02	0.06	0.22	0.05	43	−0.20	(−0.22, −0.18)	0.00*
DBDPE	17	7.4	18	2.4	43	−1.4	(−4.1, 1.2)	0.29
$\sum$ HBCD	13	0.60	16	1.0	43	−2.8	(−3.0, −2.6)	0.00*
TCEP	220	57	120	57	43	36	(32, 40)	0.00*
TCIPP	78	18	41	8.7	43	100	(88, 120)	0.00*
TDCIPP	950	970	370	27	43	580	(280, 880)	0.00*
TBBPA	15	1.4	20	2.9	43	−5.6	(−6.5, −4.6)	0.00*
$\sum$ non-PBDEs	1600	950	700	87	43	870	(580, 1200)	0.00*

^a Paired t -test.

* $p < 0.01$.

Table S2 compares our study median exposure estimates to median exposure estimates gleaned from prior studies based solely on home dust sampling. Accounting for 24 h spent across microenvironments results in higher mean human exposure estimates when using ESD samples compared to FD samples for most, but not all, FR chemicals.

3.2.1. ESD compared to FD flame retardant human exposure estimates for PBDEs

The $\sum$ PBDEs mean exposure estimates were observed as higher for estimates from ESD compared to those from FD as shown in Fig. 1. The $\sum$ PBDEs mean difference in estimated exposure between the two sampling sites was about 20% higher or ~210 ng/day greater for exposure estimates from ESD relative to FD, and the difference was statistically significant ($p = 0.00$). This trend persisted for all but BDE-206 and BDE-209; these were ~2.3 ng/day ($p = 0.00$) and ~88 ng/day ($p = 0.00$) lower, respectively, from ESD-based exposure estimates than FD-based exposure estimates. Our BDE-209 results diverged from Al-Omran and Harrod (2016) and Björklund et al. (2012) who demonstrated higher ESD-based exposure than FD-based exposure when only home-based exposure is considered (see Table S2); perhaps our sampled home had older BDE-209 containing products that were closer to the floor such as textiles (Kajiwara and Takigami, 2013) than those in the other two studies. Fig. S1a shows that for ESD-based exposure estimates, the home contributes about half and the other microenvironments contribute the other half of total estimated exposure to $\sum$ PBDEs; though for FD-based $\sum$ PBDE exposure estimates, the home is the major contributor as shown in Fig. S1b.

The highest mean PBDE difference between ESD and FD derived exposures estimates was from BDE-47 and the second highest was from BDE-99, representing 120 ng/day ($p = 0.00$) and 110 ng/day ($p = 0.00$), respectively. BDE-47 and BDE-99 exposure estimates may be highest in ESD based estimates compared to FD based estimates due to a lower molecular weight than higher brominated BDE congeners; they may partition into the air more so than heavier BDEs and later settle into ESD more so than FD, because they land there first (Rauert et al., 2015). These results are slightly different from those reported by Björklund et al. (2012) where BDE-47 and BDE-99 were the third and fifth highest in difference, between Swedish house ESD and FD exposure estimates. Al-Omran and Harrod (2016) found BDE-99 and BDE-47 were the PBDE congeners with the second and third highest difference, respectively, for ESD relative to FD exposure estimates from their sampled Iraqi houses. The differences between studies may be reflective

of the diversity of FR use between the U.S., Sweden, and Iraq and the distribution of FR chemicals in ESD and FD in the home compared to the distribution in various microenvironments, a unique feature of the present study. These results provide a cautionary note regarding potential underestimation of exposure in studies where only FD samples are used to estimate all measured PBDE FR chemicals, particularly for individuals spending time near elevated surfaces and outside of the home. Our results support the conclusion that for most PBDE congeners, higher levels of exposure may occur through ESD than from FD, particularly when spatiotemporal dimensions of exposure are included in the assessment.

3.2.2. ESD compared to FD flame retardant human exposure estimates for non-PBDEs

The $\sum$ non-PBDEs mean exposure estimates were observed as higher for estimates from ESD compared to those from FD as shown in Fig. 1. The $\sum$ non-PBDE mean ESD human exposure estimate was over 2 times higher or ~900 ng/day higher than the FD human exposure estimate ($p = 0.00$). The greater difference between non-PBDEs relative to PBDEs may reflect the effect of PBDE phase-out policies (Besis and Samara, 2012; Birnbaum and Staskal, 2004; Great Lakes Chemical Corp., 2005; State of California, 2003) and the present study's accounting for time spent in the home and additional microenvironments where non-PBDEs are typically higher in ESD than FD (Allgood et al., 2017). Fig. S1c-d illustrates the contribution of microenvironments to total $\sum$ non-PBDE exposure estimates.

However, in Table 2, we note similar high mean BDE-47 and BDE-99 exposure estimates to exposure estimates for their replacement chemicals EH-TBB and BEH-TEPH (Covaci et al., 2011). This may be due to the academic environment sampled containing both older PBDE-containing products and newer products containing EH-TBB and BEH-TEPH as their replacements. Policy-driven trends in exposure to FRs were also reported by Xu et al. (2016) and Al-Omran and Harrod (2016) who sampled only the home, which may be more likely to contain newer products with non-PBDEs. Whereas Cequier et al. (2014) reported that most non-PBDEs were higher in ESD than FD, the differences were not statistically significant, and the authors assumed erroneously as we have shown in the present study, that all 24 h of daily exposure occurred in the home.

In the cases of BTBPE, DBDPE, $\sum$ HBCD, and TBBPA, our results buck the trend of higher exposures through ESD than through FD, with differences of ~0.20 ng/day ($p = 0.00$), ~1.4 ng/day ($p = 0.29$), ~2.8 ng/day ($p = 0.00$), and ~5.6 ng/day ($p = 0.00$) lower through ESD than through FD, respectively. While the differences between exposure derived from ESD compared to FD are statistically significant for BTBPE, $\sum$ HBCD, and TBBPA, the practical difference is a negligible matter of picograms per day. These low exposure levels may be background concentrations rather than from products in the sampled environments. TBBPA is an exception and may be present, particularly if it is used as a reactive FR (Covaci et al., 2011) and thus not as likely to leach out into the environment as when used as an additive FR. Other investigators suggest that this distinction is not persistent (Al-Omran and Harrod, 2016; Cequier et al., 2014).

Among the five other non-PBDE chemicals, the highest observed non-PBDE difference between estimated human exposure from ESD and FD was for TDCIPP (580 ng/day; $p = 0.00$), and the second highest was for BEH-TEPH (150 ng/day; $p = 0.00$). These findings suggest that the difference between human exposure estimates from ESD and FD persists for most non-PBDEs when spatiotemporal dimensions of chemical exposures are considered.

4. Conclusion and future research

This research supports the hypothesis that accounting for time spent across microenvironments significantly influences estimates of human exposure to various forms of FRs. The results should be cautiously

Fig. 1. Range of total human exposure estimates (ng/day). We accounted for time spent across microenvironments, and the estimates were derived from elevated surface dust (ESD) and floor dust (FD) for $\sum$ PBDEs and $\sum$ non-PBDEs, respectively.

interpreted because we extrapolated chemical concentration data acquired from specific spatial locations to categories of microenvironments in which our study participants spent time. However, the validity of our results is supported by previous studies that reported higher FR concentrations in home ESD when compared to FD (Al-Omran & Harrod, 2016; Björklund et al., 2012; Xu et al., 2016). Importantly, our conclusion contrasts with the 12-sample home-focused Cequier et al. (2014) conclusion that PBDE and non-PBDE FR chemicals in ESD and FD do not require differentiation. Future studies may include personal exposure samplers and automated time activity pattern recorders (e.g. those using Geographic Information System coordinates) to refine the collection of data on exposure history. Ultimately, policies to replace toxic chemicals in consumer products that shed FRs will be more effective in reducing or eliminating exposures. Such policies will be better informed with studies such as the present one that provides evidence for opportunity for exposure and hazard characterization in risk assessment.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2017.07.157>.

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