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Authors

Allgood, Jaime M
Jimah, Tamara
McClaskey, Carolyn M
[et al.](#)

Publication Date

2017-02-01

DOI

10.1016/j.envres.2016.11.010

Peer reviewed



Potential human exposure to halogenated flame-retardants in elevated surface dust and floor dust in an academic environment



Jaime M. Allgood^a, Tamara Jimah^a, Carolyn M. McClaskey^{b,1}, Mark J. La Guardia^c,
Stephanie C. Hammel^{a,2}, Maryam M. Zeineddine^{a,3}, Ian W. Tang^a, Miryha G. Runnerstrom^a,
Oladele A. Ogunseitani^{a,*}

^a Department of Population Health & Disease Prevention, Program in Public Health, University of California, Irvine, CA 92697-3957, USA

^b Department of Cognitive Sciences, School of Social Sciences, University of California, Irvine, CA 92697-5100, USA

^c Department of Aquatic Health Sciences, Virginia Institute of Marine Science, College of William and Mary, Gloucester Point, VA 23062, USA

ARTICLE INFO

Keywords:

Chemicals
Dust
Electronics
Exposure
Flame-Retardants
Halogenated
Indoor
Pollutant
Toxic

ABSTRACT

Most households and workplaces all over the world possess furnishings and electronics, all of which contain potentially toxic flame retardant chemicals to prevent fire hazards. Indoor dust is a recognized repository of these types of chemicals including polybrominated diphenyl ethers (PBDEs) and non-polybrominated diphenyl ethers (non-PBDEs). However, no previous U.S. studies have differentiated concentrations from elevated surface dust (ESD) and floor dust (FD) within and across microenvironments. We address this information gap by measuring twenty-two flame-retardant chemicals in dust on elevated surfaces (ESD; n=10) and floors (FD; n=10) from rooms on a California campus that contain various concentrations of electronic products. We hypothesized a difference in chemical concentrations in ESD and FD. Secondly, we examined whether or not this difference persisted: (a) across the studied microenvironments and (b) in rooms with various concentrations of electronics. A Wilcoxon signed-rank test demonstrated that the ESD was statistically significantly higher than FD for BDE-47 ($p=0.01$), BDE-99 ($p=0.01$), BDE-100 ($p=0.01$), BDE-153 ($p=0.02$), BDE-154 ($p=0.02$), and 3 non-PBDEs including EH-TBB ($p=0.02$), BEH-TEBP ($p=0.05$), and TDCIPP ($p=0.03$). These results suggest different levels and kinds of exposures to flame-retardant chemicals for individuals spending time in the sampled locations depending on the position of accumulated dust. Therefore, further research is needed to estimate human exposure to flame retardant chemicals based on how much time and where in the room individuals spend their time. Such sub-location estimates will likely differ from assessments that assume continuous unidimensional exposure, with implications for improved understanding of potential health impacts of flame retardant chemicals.

1. Introduction

There is growing concern about possible health impacts due to human exposure to chemical flame retardants that are ubiquitous in consumer products. Several studies have established that flame retardant exposure through dust ingestion may increase the risk of adverse neurodevelopment in children (Eskenazi et al., 2011; Gascon et al., 2012; Herbstman et al., 2010; Roze et al., 2009), reduced thyroid functioning in children and adults (Chevrier et al., 2010), and infertility (Harley et al., 2010; Meeker and Stapleton, 2010). Therefore, the California's Safer Consumer Products Regulations Candidate

Chemicals list includes the following flame-retardants as hazardous to human health: polybrominated diphenyl ethers (PBDEs), 2-ethylhexyl 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 (HBCD), tris (2-chloroethyl) phosphate (TCEP), tris (1-chloro-2-propyl) phosphate (TCIPP), tris (1,3-dichloro-2-propyl) phosphate (TDCIPP), and tetrabromobisphenol-A (TBBPA) (State of California, 2014).

PBDEs are a flame retardant category most widely used in foam, plastic housings of electronics, and textiles until recent recognition of

* Corresponding author.

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

¹ Department of Otolaryngology – Head and Neck Surgery, Medical University of South Carolina, Charleston, South Carolina 29425-5500, USA.

² Nicholas School of the Environment, Duke University, Levine Science Research Center, Durham, NC 27708, USA.

³ Department of Interdisciplinary Studies, Program in Global Health and Medical Ethics, University of California, Berkeley, CA 94720-1960, USA.

their toxicity encouraged government regulations to stipulate voluntary phase-out or total ban in the United States and the European Union, respectively (Besis and Samara, 2012; Birnbaum and Staskal, 2004; Great Lakes Chemical Corp., 2005; European Court of Justice, 2008; State of California, 2003). PBDEs leach into human environments during normal usage of consumer products, and manufacturers have developed chemical alternatives to PBDEs (alt-PBDEs), although it is unclear if these are safer, including BTBPE, DBDPE, EH-TBB, and BEH-TEBP (Stapleton et al., 2008). Other flame retardants include bromine based chemicals such as the HBCDs used in polystyrene consumer products (Rani et al., 2014) and textiles (Kajiwara et al., 2009); TBBPA used in circuit boards (Zhou et al., 2014) and polymers (Sindik et al., 2015); and chlorinated organophosphates TCEP, TCIPP, and TDCIPP (Bergman et al., 2012) used in polyurethane foams (Van den Eede et al., 2011), textiles, and plastics (Van der Veen and de Boer, 2012).

Due to the ubiquity of chemical flame retardants, we sought to investigate indoor dust as a component of models for estimating human exposure (Johnson-Restrepo and Kannan, 2009; Lorber, 2008). In the U.S., studies of indoor dust chemical concentrations have been limited to homes and offices (Allen et al., 2008; Batterman et al., 2009, 2010; Dodson et al., 2012; Harrad et al., 2008; Hwang et al., 2008; Imm et al., 2009; Johnson et al., 2010; Meeker et al., 2009; Quiros-Alcala, et al., 2011; Stapleton et al., 2005; Ward et al., 2014; Zota et al., 2008; Watkins et al., 2011). These studies suggest that flame retardants vary widely within microenvironments across the locations. In Swedish and Iraqi homes, indoor dust concentrations differed between the floor and elevated surfaces (Björklund et al., 2012; Al-Omran and Harrad, 2015). The presence of electronics may also affect flame retardant concentrations (Brandma et al., 2013; de Wit et al., 2012; Fulong and Espino, 2013; Harrad et al., 2004; He et al., 2015). For example, a prior study found that flame retardant concentrations decreased with increasing distance from a television set (Harrad et al., 2009). Therefore, electronic products at a dust sample collection site may be influential.

Accurate spatial location of dust sample collection is important because of the way people interact with the environment. In particular, small children spend time in contact with the floor, making floor dust a significant exposure point for this population (Johnson-Restrepo and Kannan, 2009; Lorber, 2008). Older children and adults, on the other hand, may spend time at the elevated surfaces in a room when sitting on a sofa or using a computer, making elevated surface dust a significant exposure point for this population. Thus, spatial location of the dust sample collection may impact human exposure estimates, and it is important that the collection location is congruent with how the chemical exposure occurs.

However, U.S. studies either combine floor and elevated surface dust in the same sample (Dodson et al., 2012; Watkins et al., 2011; Zota et al., 2008), sample the floor only (Quiros-Alcala et al., 2011; Stapleton et al., 2008), or sample from household vacuum bag dust (Imm et al., 2009; Meeker et al., 2009), thereby compromising the interpretation of data which may be relevant for direct exposure assessments. Consequently, it is imperative for us to collect information on how chemical flame retardant concentrations vary in elevated surface and floor dust samples, high versus low electronic presence areas, and across microenvironments. The information will allow exposure estimation and identification of populations that are vulnerable to excessive chemical exposure. Preventive measures may then be taken to reduce exposure to toxic flame retardants.

For the present study, we collected dust samples from elevated surfaces and floors at various locations on the campus of the University of California, Irvine. The microenvironments sampled included a bus, scientific laboratory, computer laboratory, gymnasium, and two each of domestic apartments, classrooms, and offices. The dust samples were collected to investigate a specific primary hypothesis: elevated surface dust flame retardant concentrations differ from floor dust flame

retardant concentrations. Additionally, we compared concentrations in dust from elevated surfaces and floors to examine whether or not flame retardant levels from these two sampling sites (a) vary across microenvironments and (b) vary based on number of electronic products in the sampled areas. In this study, we included two categories of flame retardants chemicals: polybrominated diphenyl ethers (PBDEs) congeners and other flame retardants referred to as non-polybrominated diphenyl ethers (non-PBDEs). The PBDEs congeners included in this analysis were BDE-28, BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, BDE-206, BDE-209 and the non-PBDEs were EH-TBB, BEH-TEBP, BTBPE, DBDPE, α HBCD, β HBCD, γ HBCD, TCEP, TCIPP, TDCIPP, and TBBPA.

2. Methods

2.1. Purposeful sampling of microenvironments

Two previous studies employed a strategy based on sampling for heterogeneity of microenvironments (de Wit et al., 2012; Thuresson et al., 2012). We adapted a similar strategy, known as maximum heterogeneity sampling, whereby locations are sampled using purposeful sampling techniques. Maximum heterogeneity sampling is typically used when sampling people and is conducted in a way that maximizes a key factor, but this study applies it to sampling microenvironments (Patton, 2002). The key factor we maximized was electronic presence which is described below. In considering this factor, we sampled from both high electronic presence areas and low electronic presence areas. Any similarities or differences in flame retardant concentrations between the elevated surface dust and floor dust are of value in understanding whether or not these two sites may impact flame retardant exposure estimates, because they emerge from areas of maximum variation (Patton, 2002). The specific locations sampled are listed in Table S1.

2.2. Electronic products

Microenvironments were sampled for maximum heterogeneity (Patton, 2002) based on low or high numbers of stationary electronic products. The type and count of electronic products in each sampled area are listed in Table S1. An electronic density score was calculated for each place by dividing the total number of electronic products by the square footage of the sampled room. Microenvironments with an electronic density score of 0.01 or greater ($n=6$) were categorized as high electronic presence areas (HEPA) and those with an electronic density score of 0.00 ($n=4$) were categorized as low electronic presence areas (LEPA).

2.3. Dust sampling

All indoor dust samples were collected between June 2013 and September 2013. Dust samples were collected following the methods of previous studies using an Eureka Mighty-Mite vacuum cleaner (Allen et al., 2008; Watkins et al., 2011; Zota et al., 2008). The crevice tool used for dust collection was welded by General Mechanical Inc. (Anchorage, Alaska) and contained a cellulose thimble (19×90 mm) held in place by a rubber o-ring. Dust samples were collected by slowly moving the crevice tool over surfaces in each of the two sampling areas—elevated surfaces and floors – for 15 min each whereby approximately 1 g of dust was collected per sample. The elevated surfaces sampling area included surfaces above the floor such as sofas, book cases, desks, tables, chairs, and counter tops that were approximately 2 feet or higher from the floor while floor dust samples were taken strictly from the floor. After collection, each dust sample was placed in foil and a polyethylene zip bag and then stored in our UC Irvine laboratory at -4 °C until they were shipped on dry ice to the College of William and Mary, Virginia, U.S. in September of 2013.

2.4. Chemical analyses

Dust samples were sieved (300 μm) and ~100 mg was subjected to accelerated solvent extraction (ASE 200, Dionex, Sunnyvale, CA, USA) with dichloromethane (DCM). Extracts were purified by size exclusion chromatography (SEC, Envirosep-ABC[®], 350 \times 21.1 mm. column; Phenomenex, Torrance, CA, USA). Each post-SEC extract was then reduced in volume (~1 mL) by placing in a water bath under a stream of nitrogen and solvent exchanged to hexane, reduced in volume (~1 mL) and added to the top of a solid phase 2-g silica glass extraction column (Isolute, International Sorbent Tech.; Hengoe Mid Glamorgan, UK). Each column was eluted with 3.5-mL hexane (fraction one), followed by 6.5 mL of 60:40 hexane/DCM and 8 mL DCM (fraction two) and 5 mL 50:50 acetone/DCM (fraction three). The second fraction, containing brominated flame retardants, was reduced in volume and solvent exchanged to methanol, then 400 ng of deca-chlorodiphenyl ether (DCDE) (Ultra Scientific, North Kingstown, RI, USA) was added as an internal standard. Fraction three containing chlorinated organophosphate flame retardants and TBBPA was reduced, solvent exchanged to methanol, and DCDE (400 ng; Ultra Scientific) added as an internal standard. Analytes in these purified extracts were chromatographically separated by UPLC (Acquity UPLC, Waters Corporation, Milford, MA, USA) operated in the gradient mode (100% methanol (A1) and 100% water (B1), equipped with a C18 UPLC analytical column (Acquity UPLC BEH C18, 1.7 μm , 2.1 \times 150 mm, Waters Corp.). Analytes were ionized by APPI, the dopant (acetone) was introduced (150 $\mu\text{l}/\text{min}$.) by a liquid chromatography pump (LC-20AD, Shimadzu Corporation, Kyoto, Japan) and product ions were detected by triple quadrupole mass spectrometer (3200 QTrap, AB Sciex, Framingham, MA, USA). Additional information on sample preparation and UPLC-APPI-MS/MS operation can be found in Schreder and La Guardia (2014).

2.5. Quality control and assurance

The dust samples were analyzed using a method previously published (Schreder and La Guardia, 2014) and qualified by the analysis of a National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) #2585 (house dust). Previously, SRM #2585 certified mean % recovery for Σ PBDEs was established at 65%; individual PBDEs recoveries range 38–114%. Referenced mean % recovery values for each additional class of flame-retardants (i.e. Σ alt-PBDEs, Σ HBCDs, Σ CIOPs and TBBPA) range 52–91%; individual recoveries range 51–142% (Table S2). This method was validated during this study using a performance-based QA/QC approach which included laboratory blanks, duplicate, surrogate and matrix spike recovery analysis. Of the 22 targeted flame-retardants the laboratory blanks only contained trace amounts of BDE-99 (9.08 and 20.7 ng/g) and BDE-209 (14.4 and 17.2 ng/g). The Relative Percent Difference (RPD) between individual FRs from the duplicate analysis ranged from 1% to 16%. The surrogate recoveries for BDE-166 and dTDCIPP ranged from 61% to 108% and 56% to 113% respectively and recoveries for all matrix spiked analytes were >54%, except for BDE-28 at 33% recovery (Table S3).

2.6. Statistical analyses

Summary statistics were calculated with non-detectable concentrations left at zero for all analytes. Sums of the PBDEs associated with the technical products (PentaBDE: BDE-28, BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-153 and BDE-154; and DecaBDE: BDE-206 and BDE-209) as well as sums of the total PBDEs and HBCDs (α -HBCD, β -HBCD, γ -HBCD) were calculated (La Guardia et al., 2006). Ratios (elevated surfaces/floor concentrations) were calculated to evaluate differences in flame retardant chemical concentrations based on dust collection site for which non-detectable concentrations were

replaced with a value of $1/2$ LOQ. Since most data were found to lack normal distribution, we conducted our analyses using the two following non-parametric tests: (i) a Wilcoxon signed rank test was used to further investigate the hypothesis of a significant difference in flame retardant concentrations comparing elevated surfaces and floor dust samples pairs each collected from the same place; and (ii) a Mann-Whitney rank sum test was conducted to test the hypothesis that the elevated surface and floor dust flame retardant concentrations from high versus low electronic presence areas are significantly different. The conventional p -value of 0.05 or less was used to assess the statistical significance of the results. Data analysis was performed in SPSS (Version 23).

3. Results and discussion

To investigate the relationship between flame retardant chemicals in elevated surface dust (ESD) and floor dust (FD), indoor dust samples were collected from microenvironments with either high or low numbers of electronic products in locations on the University of California, Irvine campus. The concentrations of 22 flame retardant chemicals were determined in dust samples collected from elevated surfaces ($n=10$) and floors ($n=10$) in a bus, scientific laboratory, computer laboratory, gymnasium, apartments ($n=2$), classrooms ($n=2$), and offices ($n=2$). The detection limit and frequency, median, and range of each flame retardant chemical are shown in Table 1. We detected 21 flame retardants, including 10 PBDE congeners, EH-TBB, BEH-TEBP, BTBPE, DBDPE, TBBPA, α -HBCD, β -HBCD, γ -HBCD, TCEP, TCIPP, and TDCIPP. Among these, 19 chemicals were found in at least 12 of the 20 dust samples. Overall, median concentrations for all detected flame retardants were greater in ESD than FD (see Table 1) and varied across microenvironments (see Fig. 1 and Table S4). This general trend persisted for most chemicals in HEPA and LEPA though the difference was magnified in HEPA (see Fig. 2 and Table S7). However, not all sampled locations had greater concentrations in ESD (see Table S5), suggesting that sample site location may be an important consideration in exposure estimation for some microenvironments, but not all microenvironments. In the following sections, we describe the ratio of chemical concentrations in ESD and FD, and then we compare these concentrations across sampled microenvironments, and between HEPA and LEPA.

3.1. Comparison of elevated surface dust (ESD) and floor dust (FD) flame retardant concentrations

To compare ESD and FD flame retardant concentrations, we measured 11 PBDE congeners in dust samples from both locations, nine of which were detected in at least 70% of the samples. BDE-66 was not detected in any location and BDE-28 was only found in 30% and 10% of ESD and FD samples, respectively. We measured 11 non-PBDEs, of which EH-TBB, BEH-TEBP, DBDPE, α -HBCD, β -HBCD, γ -HBCD, TCEP, TCIPP, and TDCIPP were detected in at least 70% of samples, while TBBPA was in 60% and BTBPE was in 20% of the samples tested. Median concentrations for each detected PBDE congener and non-PBDEs were greater in ESD than FD, and the median ratio of ESD to FD was two or more for most, but not all, chemicals.

The results shown in Table 1 indicate that median ESD to FD ratios were near 2.0 for BDE-85, BDE-153 and TBPH and at least two or more for BDE-47, BDE-99, BDE-100, BDE-154, Σ PentaBDE, BDE-206, BDE-209, Σ BDE, EH-TBB, DBDPE, TCEP, and TDCIPP, suggesting higher chemical concentrations in ESD than FD. A Wilcoxon signed-rank test showed that the difference between ESD and FD was statistically significant for BDE-47 ($Z=-2.60$, $p=0.01$), BDE-99 ($Z=-2.50$, $p=0.01$), BDE-100 ($Z=-2.50$, $p=0.01$), BDE-153 ($Z=-2.40$, $p=0.02$), BDE-154 ($Z=-2.29$, $p=0.02$), Σ PentaBDE ($Z=-2.50$, $p=0.01$), Σ BDE ($Z=-2.29$, $p=0.02$), EH-TBB ($Z=-2.29$, $p=0.02$), BEH-TEBP ($Z=-1.99$, $p=0.05$), and TDCIPP ($Z=-2.19$, $p=0.03$). Our findings

Table 1
Flame Retardant Concentrations (ng/g Dust) in Elevated Surface Dust (ESD) and Floor Dust (FD) Samples from the University of California, Irvine Campus.

| Chemical Name | Abbreviation | Elevated surface dust samples (n=10) | | | | Floor dust samples (n=10) | | | | Comparison of ESD and FD | |
|--|--------------|--------------------------------------|------|--------|---------|---------------------------|------|--------|-------|--------------------------|----------------|
| | | % > DL | min. | median | max. | % > DL | min. | median | max. | Ratio (median) | p ^a |
| polybrominated diphenyl ethers | ∑PBDEs | 100 | 3650 | 23508 | 141172 | 100 | 2435 | 6582 | 36377 | 3.37 | 0.02* |
| 2,4, 4'-tri BDE | BDE 28 | 30 | 0 | 0 | 789 | 10 | 0 | 0 | 47 | – | 0.11 |
| 2,2',4,4'-tetra BDE | BDE 47 | 100 | 794 | 3260 | 60350 | 100 | 292 | 1151 | 11000 | 4.71 | 0.01* |
| 2,3',4,4'-tetra BDE | BDE 66 | 0 | – | – | – | 0 | – | – | – | – | – |
| 2,2',3,4,4'-penta BDE | BDE 85 | 90 | 0 | 117 | 3730 | 80 | 0 | 110 | 2300 | 1.85 | 0.09 |
| 2,2',4,4',5-penta BDE | BDE 99 | 100 | 1310 | 5945 | 45200 | 100 | 763 | 2060 | 12700 | 3.00 | 0.01* |
| 2,2',4,4',6-penta BDE | BDE 100 | 100 | 206 | 1041 | 15900 | 90 | 0 | 353 | 4730 | 2.67 | 0.01* |
| 2,2',4,4',5,5'-hexa BDE | BDE 153 | 100 | 115 | 632 | 6140 | 80 | 0 | 210 | 3100 | 1.99 | 0.02* |
| 2,2',4,4',5,6'-hexa BDE | BDE 154 | 100 | 121 | 620 | 5710 | 80 | 0 | 190 | 903 | 2.08 | 0.02* |
| | ∑pentaBDE | 100 | 268 | 11694 | 133904 | 100 | 1055 | 4618 | 34780 | 3.37 | 0.01* |
| 2,2',3,4,4',5',6'-hepta BDE | BDE 183 | 70 | 0 | 75 | 242 | 60 | 0 | 15 | 280 | 1.01 | 0.21 |
| 2,2',3,3',4,4',5,5',6-nona BDE | BDE 206 | 100 | 28 | 127 | 412 | 90 | 0 | 25 | 605 | 2.21 | 0.09 |
| decabromodiphenyl ether | BDE 209 | 100 | 935 | 5545 | 21400 | 100 | 215 | 1300 | 28900 | 2.74 | 0.20 |
| | ∑decaBDE | 100 | 963 | 5672 | 21812 | 100 | 222 | 1312 | 29505 | 3.08 | 0.20 |
| Non-polybrominated diphenyl ethers | ∑non-PBDEs | 100 | 9080 | 103000 | 1390000 | 100 | 5760 | 22300 | 88600 | 2.77 | 0.07 |
| 2-ethylhexyl 2,3,4,5-tetrabromobenzoate | EH-TBB | 100 | 143 | 604 | 89200 | 100 | 113 | 325 | 15500 | 2.05 | 0.02* |
| 2-ethylhexyl 2,3,4,5-tetrabromophthalate | BEH-TEBP | 100 | 255 | 1441 | 44550 | 100 | 223 | 1246 | 4690 | 1.76 | 0.05* |
| 1, 2-bis (2,4,6-tribromophenoxy) ethane | BTBPE | 20 | 0 | 0 | 90 | 20 | 0 | 0 | 290 | 2.30 | 0.59 |
| decabromodiphenyl ethane | DBDPE | 100 | 144 | 344 | 2780 | 90 | 0 | 154 | 929 | 2.02 | 0.07 |
| tetrabromobisphenol-A | TBBPA | 60 | 0 | 476 | 1530 | 60 | 0 | 187 | 7260 | 1.28 | 0.89 |
| α-1, 2,5,6,9,10-HBCD | α-HBCD | 100 | 56 | 204 | 439 | 100 | 198 | 52 | 475 | 1.11 | 0.65 |
| β-1,2,5,6,9,10-HBCD | β-HBCD | 100 | 14 | 41 | 79 | 100 | 10 | 39 | 94 | 1.09 | 0.80 |
| γ-1,2,5,6,9,10-HBCD | γ-HBCD | 100 | 21 | 118 | 296 | 90 | 0 | 74 | 157 | 1.57 | 0.07 |
| hexbromocyclododecanes | ∑HBCD | 100 | 89 | 393 | 799 | 100 | 104 | 326 | 636 | 1.02 | 0.39 |
| tris (2-chloroethyl) phosphate | TCEP | 90 | 0 | 1315 | 12500 | 70 | 0 | 918 | 35300 | 2.23 | 0.31 |
| tris (1-chloro-2-propyl) phosphate | TCIPP | 100 | 1660 | 5975 | 65950 | 80 | 0 | 4265 | 49400 | 0.12 | 0.33 |
| tris (1,3-dichloro-2-propyl) phosphate | TDCIPP | 100 | 3680 | 34075 | 1380000 | 100 | 2980 | 12450 | 54200 | 2.60 | 0.03* |

bold indicates an ESD to FD median ratio above 2, DL=detection limit (which is 1 ng/g for each chemical)

^a Wilcoxon sign rank test.

* p < 0.05.

suggest that several flame retardants are found in higher concentrations in ESD relative to FD.

The observed differences in chemical concentrations between ESD

and FD can be explained by two main factors. Firstly, as previously suggested, dust from elevated surfaces may represent smaller particles that accumulate congeners that have been volatilized (Rauert et al.,

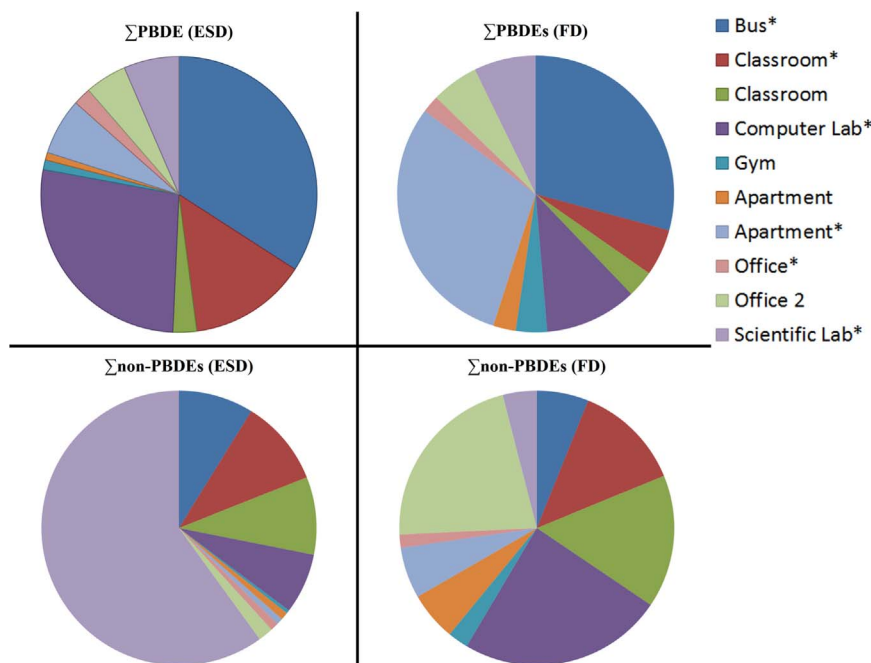


Fig. 1. Patterns of ΣPBDEs and Σnon-PBDEs by microenvironment for elevated surface dust (ESD) and floor dust (FD).

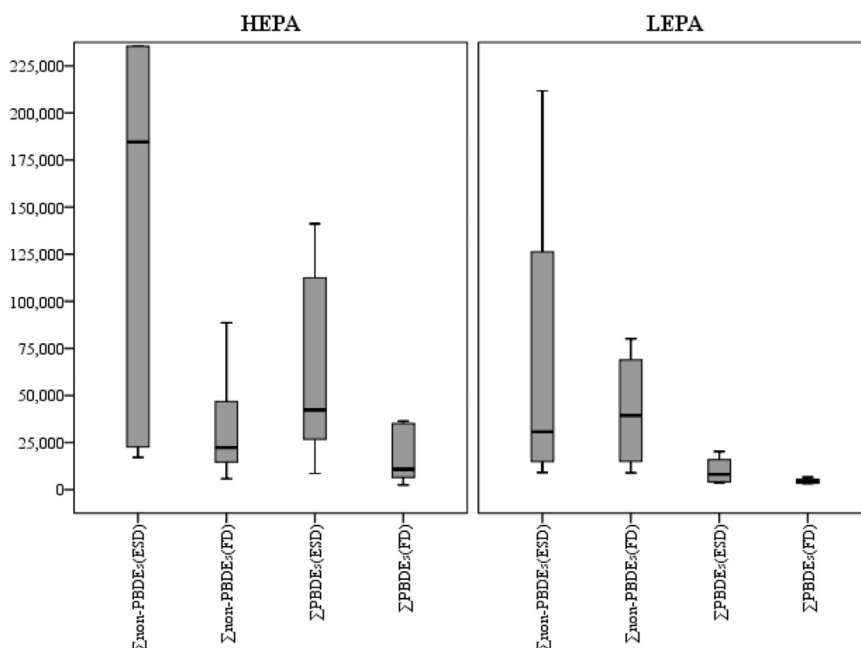


Fig. 2. The median and range of Σ non-PBDEs and Σ PBDEs in elevated surface dust (ESD) and floor dust (FD) for High Electronic Presence Areas (HEPA) and Low Electronic Presence Areas (LEPA).

2015); while dust from the floor most likely contains larger particles with chemicals leached from consumer products into the ambient environment. Dirt, sand, and soil brought in from the outside may also dilute the concentration of target chemicals in indoor dust (Allen et al., 2013; Cao et al., 2014). Secondly, flame retardant concentrations may be locally high in the ESD collected directly from the surface of consumer products in which they are embedded (Meeker et al., 2009). Previous studies have found a greater correlation between hand wipes and body burden of chemicals than between body burden and floor dust from the home or office (Stapleton et al., 2012; Watkins et al., 2011). It is possible that hand wipes approximate exposure through contact with elevated surfaces rather than through floor dust.

Whereas most flame retardant chemicals were found at higher median concentrations in ESD than FD, notable exceptions were BDE-183, α HBCD, β HBCD, γ HBCD, Σ HBCD, and TBBPA, which had median ratios near 1.0 indicating similar ESD and FD concentrations. Further, TCIPP had a median ratio less than 1.0 suggesting that concentrations are higher in FD than ESD. This indicates that some flame retardant chemicals may not occur in higher concentrations in ESD than in FD. We cannot rule out the possibility that some products in the sampled locations may have been manufactured with smaller amounts of the aforementioned flame retardants.

Our results should be interpreted cautiously because of the small sample size, but previous reports support the validity of our findings. For example, Allen et al. (2013) calculated ratios of vent to floor dust in airplanes and found median ratios of two or higher for most detected chemicals, though contrary to our findings median BDE-99 and BDE-209 ratios were near 1.0 while BDE-183 was below 1.0. Björklund and Sellström (2012) also compared ESD and FD PBDE and HBCD concentrations in Swedish homes, observing a similar general trend of greater chemical concentrations in elevated surface compared to floor dust except HBCD was lower in ESD. Further, Al-Omran and Harrad (2015) found median ESD BDEs at higher concentrations than median FD levels. Overall, the general observation across studies of median ratios near or above 2.0 underscores the finding of higher chemical concentrations in dust samples collected at elevated surfaces compared to the floor.

These results have important implications for chemical exposure estimation. Exposure is possibly underestimated for most flame

retardants when relying on FD samples or combinations of ESD and FD samples, particularly for populations spending time at elevated surfaces. Additional studies that use probability sampling methods are needed to confirm our findings.

3.2. Comparison of elevated surface dust (ESD) and floor dust (FD) concentrations across microenvironments

With the exception of apartments, this is the first U.S. study that directly compares ESD and FD in a gymnasium, offices ($n=2$), classrooms ($n=2$), laboratories ($n=2$), and a campus transportation bus. In Fig. 1, the Σ PBDEs and Σ non-PBDEs patterns from indoor ESD and FD are compared across these microenvironments on the UC Irvine campus. Also, the ratio of ESD to FD for each chemical and location sampled is shown in Table S5. Our results show that, in most but not all microenvironments, the concentrations differ across ESD and FD.

3.2.1. Apartments

Unlike most of the other microenvironments, apartments had similar ESD and FD chemical concentrations. These ratios were mostly below 2.0 and similar to the median ratios reported from Iraqi homes (Al-Omran and Harrad, 2015) and homes in Norway (Cequier et al., 2014). Also, in both apartments, PBDE congeners' concentrations were similar except there was one anomalous pattern in the BDE-209 levels of the HEPA apartment. The concentrations of BDE-209 from the two sampled apartments were approximately 18 to 68 times higher than the median concentrations reported by other investigators (Dodson et al., 2012; Björklund and Sellström, 2012; Al-Omran and Harrad, 2015). This discrepancy may be due to the possibility that a piece of material with BDE-209 was included in our dust sample, indicating that high BDE-209 exposure is potentially a concern if people are exposed this way. However, we only sampled two apartments and these concentrations may not be indicative of the levels of BDE-209 to which the general population is exposed. Further, the extremely high concentrations that we detected produced the unusual trends in BDE-209 that we observed when comparing BDE-209 and Σ PBDEs concentrations in the apartment to other microenvironments. The trends of the non-PBDEs were similar to the other BDE congeners. When comparing TDCIPP, our ESD levels (10,900 ng/g and 13,200 ng/g) and our FD levels

(13,300 ng/g and 11,600) were approximately 5 to 43 times higher than the median or mean concentrations reported by other investigators who also sampled homes but did not differentiate between dust from two sampling sites (Meeker and Stapleton, 2010; Dodson et al., 2012; Brandsma et al., 2014), which may reflect an increase in the use of TDCIPP over time.

3.2.2. Exercise gymnasium

In general, ESD to FD ratios were near 1.0 and similar to what we discovered in apartments. The two exceptions were TCEP and DBDPE (see Table S5). This may be due to the presence of these chemicals in gymnasium equipment. Relative to the apartments, chemical concentrations were generally lower in the gymnasium. Although to our knowledge, this is the first direct comparison of ESD and FD concentrations in an exercise gymnasium, one study measured Σ PBDEs and Σ non-PBDEs in gymnasiums relative to homes (La Guardia and Hale, 2015), but our results differ. The gymnasiums sampled by the previous investigators contained foam pits, whereas the gymnasium we sampled did not. This discrepancy suggests that the types of equipment used in public gymnasiums should be considered as an important factor in estimating risks of chemical exposures that may differ from the home.

3.2.3. Offices

Unlike the apartments and the gymnasium, offices did not have ESD to FD ratios of 1.0. The ratios were greater than 3.0 for most BDE congeners. The exceptions were BDE-209 and BDE-206 (see Table S5). However, the non-BDEs ESD to FD ratios were below 2.0. The exception to this was in the HEPA office (see Table S5). These results show that patterns of chemical concentrations varied between offices and apartments, in agreement with other studies (de Wit et al., 2012; He et al., 2015; Brommer and Harrad, 2015). However, one study found a similar Σ PBDEs median in sampled apartments and offices (de Wit et al., 2012). Our results differed, possibly because those investigators did not differentiate between ESD and FD as we did; and potentially because of our small sample size, particularly if our findings are exceptional compared to concentrations in apartments and offices that were not included.

3.2.4. Classrooms

ESD to FD ratios were mostly ≥ 2.0 , indicating that classrooms have higher concentrations of chemicals in ESD than FD. The exceptions were the HBCDs and TCIPP in both classrooms, and others noted in Table S5. Chemical concentrations in classrooms were generally at least twice as high as concentrations measured in the apartments. The higher concentrations may reflect the impact of regulatory policies regarding flammability standards, particularly if the furniture in the classrooms complies with State policy TB 133 (State of California, 1991). The main exception was the FD Σ PBDEs, which was 6–9 times lower in the classrooms than in the HEPA apartment due to an unusual level of BDE-209 in the HEPA apartment (discussed in the *Apartments* section). Compared to a previous study of a Philippines university, the ESD BDE-209 concentrations in our study were similar to Classroom 1, and higher than Classroom 2 (Fulong and Espino, 2013). Differences may be because we sampled directly from the surfaces of the room contents while those investigators did not.

3.2.5. Laboratories

The laboratories that we investigated had some of the highest chemical concentrations detected relative to the other microenvironments, especially in ESD. Chemical concentrations in ESD may be due to office chairs with exposed foam, which may be why we observed high Σ non-PBDEs. Consistent with the presence of exposed foam, the TDCIPP concentrations here are higher than those reported in other studies, though they did not differentiate between ESD and FD (Van den Eede et al., 2011; He et al., 2015). With respect to PBDEs,

generally Σ PBDEs were higher in the laboratories than the apartments, however, the difference was not as pronounced as with the Σ non-PBDEs (Fig. 1). Also, the ratios that we observed in the laboratories were ≥ 2.0 for most chemicals, in contrast to the ratios of near 1.0 for the apartments. The few exceptions are listed in Table S5. The high chemical concentrations that we measured in this study suggest that there is a potential risk for underestimation of flame retardant exposure for occupations and populations that spend time in laboratories.

3.2.6. Campus transportation bus

Most ESD to FD ratios were 3.0–3.2. The exceptions are noted in Table S5. This indicates that, on the transportation bus, most chemical concentrations were at least thrice as high in ESD than FD. To our knowledge, this is the first study to report concentrations of flame retardants in a transportation bus. However, these results are similar to an airplane study that found median ratios of vent to floor dust above 2.0 for most flame retardant chemicals (Allen et al., 2013). Also, based on the findings of our study, there were higher concentrations of Σ PBDEs on the bus compared to the apartments (see Fig. 2a-b). High chemical concentrations may be attributable to the U.S. National Highway Traffic Safety Administration's Standard 302 on flammability (U.S. Dept. of Transportation, 1972). These results are consistent with previous studies that sampled cars and found significantly higher flame retardant concentrations in cars than homes (Brommer and Harrad, 2015; Hassan and Shoeib, 2015). Our observations suggest that exposure to PBDEs may be underestimated for populations traveling on the bus, whereas previous studies assumed constant exposures at home for an entire day and night. Future studies should include probability sampling of transportation buses in addition to analysis of time activity patterns for bus passengers.

3.3. High Electronic Presence Areas (HEPA) and Low Electronic Presence Areas (LEPA)

To compare chemical concentrations in ESD and FD based on number of electronic products, we sampled both HEPA and LEPA. A description of the type of electronics, the count of electronics, and the square footage of each sampled area is included in Table S1. Table S7 shows the HEPA and LEPA median and range for each detected PBDE congener and AFR chemical in ESD and FD. Fig. 2 shows the comparison of ESD and FD Σ PBDEs and Σ non-PBDEs, in HEPA and LEPA. Overall, both HEPA and LEPA had ESD with higher median flame retardant concentrations than FD except for Σ non-PBDEs in LEPA.

3.3.1. Elevated surface dust (ESD)

The data presented in Fig. 2 shows that the median Σ PBDEs was approximately 5 times and Σ non-BDEs was approximately 3 times greater in HEPA than in LEPA, indicating higher concentrations in HEPA than in LEPA. A similar pattern was observed for the median of most non-PBDEs chemicals except DBDPE, HBCDs, and TBBPA.

BDE-99 and TDCIPP were the respectively dominant PBDE and non-BDE chemicals identified. The median BDE-99 concentration was approximately 8 times higher in HEPA than in LEPA (16200 ng/g compared to 2190 ng/g) and accounted for 38% and 27% of the HEPA and LEPA median Σ BDE, respectively. The median non-PBDEs chemical TDCIPP was approximately 3 times greater in HEPA versus LEPA (59200 ng/g v. 18700 ng/g) and accounted for 57% and 61% of the median HEPA and LEPA Σ non-PBDEs, respectively. The results demonstrating that BDE-99 and TDCIPP concentrations are higher in HEPA than LEPA suggest that electronic products leach BDE-99 and TDCIPP. Previous reports suggest that these chemicals are found mainly in furniture with foam (Stapleton et al., 2009) whereas only low concentrations have been detected in electronics (Brandsma et al., 2014; Kajiwara et al., 2011; Hibbert and Ogunseitan et al., 2014). In

the present study, we did not identify other products in the sample locations that could have contributed to the concentrations of BDE-99 and TDCIPP. Therefore, it is important that future investigations consider source apportionment of flame retardants chemicals, including foam products.

3.3.2. Floor dust (FD)

The observed pattern of flame retardant chemicals was similar to ESD, although with lower concentrations; with the exception that median Σ non-PBDEs were higher in LEPA than HEPA. The median Σ PBDEs was approximately 2.5 times greater in HEPA than LEPA while the median Σ non-PBDEs was ~1.7 times greater in LEPA than HEPA, as shown in Fig. 2. Higher PBDEs in HEPA than LEPA is expected given that a previous study found higher concentrations of PBDEs in living room dust than in bedroom dust, possibly due to differences in the number of electronics (Allen et al., 2008). Non-PBDEs may be higher in LEPA due to flame retardants used to manufacture materials for room floors (Van der Veen and de Boer, 2012).

The main PBDE congener in HEPA was BDE-99 and in LEPA was BDE-209 while TDCIPP was the main non-PBDEs chemical in both HEPA and LEPA. The median concentration of BDE-99 was approximately 4 times higher in HEPA (3840 ng/g) than in LEPA (956 ng/g), and accounted for 36% of the median Σ PBDEs. Conversely, the non-PBDEs median concentrations were greater in LEPA (see Table S7 for exceptions). The median concentration of TDCIPP was approximately 1.7 times higher in LEPA (18,800 ng/g) than in HEPA (11,100 ng/g), and accounted for 50% (HEPA) and 48% (LEPA) of the median Σ non-PBDEs, respectively. TDCIPP is not typically used to manufacture electronic products (Cao et al., 2014; Stapleton et al., 2009), but the specific reason for its lower concentration in HEPA is unclear and requires future studies that take into consideration foam and flooring materials.

The median concentration of BDE-209 was higher in LEPA (1410 ng/g) than HEPA (1110 ng/g) and accounted for 34% of the median Σ PBDEs in LEPA. BDE-209 is known for widespread use in electronic products, and we were surprised to find lower concentrations in HEPA (Birnbaum and Staskal, 2004). This result may reflect the policy-driven phase-out of BDE-209 from electronic products (Great Lakes Chemical Corp., 2005). A recent study found that dust from electronics has lower concentrations of BDE-209 than of resorcinol bis(diphenylphosphate) (RBDPP) and bisphenol A bis(diphenylphosphate) (BPA-BDPP), implying the use of these chemicals as newer alternatives to BDE-209 (Brandsma et al., 2013). We did not include these chemical alternatives in this study. Increasing use of RBDPP and BPA-BDPP in electronic products may explain why BDE-209 was detected at lower levels in HEPA than in LEPA and they will be included in future studies.

3.3.3. Implications for exposure estimation

The pattern of higher flame retardant concentrations in ESD relative to FD persisted in HEPA and LEPA. The median ESD to FD ratio, Σ PBDEs was approximately 4 for HEPA and approximately 2 for LEPA while Σ non-PBDEs was approximately 4 for HEPA (Table S6). The difference between ESD and FD concentrations from HEPA were found to be statistically significant for BDE-47 ($Z=-2.13$, $p=0.03$), BDE-99 ($Z=-2.13$, $p=0.03$), BDE-100 ($Z=-2.13$, $p=0.03$), BDE-153 ($Z=-2.13$, $p=0.03$), Σ BDE ($Z=-2.13$, $p=0.03$), and TCEP ($Z=-2.56$, $p=0.01$). These results support the hypothesis that human exposure to flame retardant chemicals is underestimated when only floor dust is analyzed in environments with a high number of electronic products containing PBDEs. However, underestimation of exposure to non-PBDEs may not be a problem for LEPA because the Σ non-PBDEs ratio was approximately 1.0. Additionally, a Mann-Whitney U test showed that in LEPA, only BDE-183 ($Z=-2.09$, $p=0.04$) was statistically significantly different in ESD and FD. This finding may be explained

by the fewer number of electronic products leaching flame retardants in LEPA, resulting in homogeneous flame retardant concentrations in elevated surfaces and on floors.

Our results suggest that a high electronic presence has more of an impact on flame retardant concentrations in ESD than FD. The implication is that estimates of human exposure based on FD or a combination of FD and ESD may underestimate true exposure, especially when there is a high number of electronic products in the sampled environment. Additional research should investigate the hypothetical association between ESD flame retardant concentrations and physiological markers of human exposure.

Acknowledgements

This research was supported in part by the University of California multi-campus research and education in green materials (UC-44157). We thank Daniel Nguyen of the UC Irvine Health Policy Research Institute for assistance with Fig. 1. J.M.A. acknowledges funding from the UC Irvine Program in Public Health Summer Research Stipend (2013–2016). This is Contribution No. 3600 of the Virginia Institute of Marine Science, College of William & Mary.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.envres.2016.11.010.

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