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

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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. Secondarily, 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-ehtylhexyl)tetrabromophthalate (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

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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 (Sindiku 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 (Brandsma 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 dipheyl 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, αHBCD, βHBCD, γHBCD, 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.