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Concentrations and loadings of organophosphate and replacement brominated flame retardants in house dust from the HOME Study during the PBDE phase-out

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Abstract

Polybrominated diphenyl ethers, a class of flame retardants and endocrine disruptors, have been substituted in new products by organophosphate (OPFR) and replacement brominated flame retardants (RBFR). OPFRs and RBFRs readily migrate from consumer products into dust where humans are exposed via incidental ingestion and inhalation. We quantified concentrations and loadings of OPFRs and RBFRs in house dust samples (n=317) collected from the homes of Cincinnati women between 2003 and 2006 and examined their associations with demographic and house characteristics. Tris-(1-chloro-2-propyl)-phosphate (TCIPP, geometric mean [GM]: 2,140 ng g⁻¹, range: 70.1-166,000 ng g⁻¹), tris-(1,3-dichloro-2-propyl)-phosphate (TDCIPP, GM: 1,840 ng g⁻¹, range: 55.2-228,000 ng g⁻¹), triphenyl phosphate (TPHP, GM: 1,070 ng g⁻¹, range: 34.1-62,100 ng g⁻¹), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB, GM: 59.5 ng g⁻¹, range: 2.82-7,800 ng g⁻¹), and bis-(2-ethylhexyl)-tetrabromophthalate (BEH-TEBP, GM: 121 ng g⁻¹, range 2.17-13,600 ng g⁻¹) were all detected in >90% of dust samples; tris-(2-chloroethyl)-phosphate (TCEP, GM: 669 ng g⁻¹, range: 56.8-160,000 ng g⁻¹) was detected in 80.1% of

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samples. Concentrations of EH-TBB and BEH-TEBP increased in house dust from 2003-2006. The number of people living in the home, race, education, floor type, and year of sample collection were associated with some OPFR and RBFR concentrations and loadings. This study suggests that OPFRs and RBFRs were ubiquitous in house dust during the PBDE phase-out and justifies more research on the consequences of exposure to these environmental chemicals.

Graphical Abstract

Introduction

Polybrominated diphenyl ether (PBDE) flame retardant chemicals were phased out of use in consumer products such as furniture foam and electronics due to human toxicity concerns beginning in 2004 (United States Environmental Protection Agency, n.d.). They were largely replaced with organophosphate flame retardants (OPFRs), which have been in use for other consumer and industrial applications for decades (Wei et al., 2015), and replacement brominated flame retardants (RBFRs), which are found in the flame retardant mixture Firemaster 550, along with OPFRs, and are also referred to as “novel brominated flame retardants” and “alternative brominated flame retardants” in the literature (Barr et al., 2010).

Tris (1,3-dichloro-2-propyl) phosphate (TDCIPP or TDCPP) is an OPFR that was removed from children’s sleepwear in the late 1970s because of evidence of carcinogenicity, but whose production has increased in recent decades for other consumer flame retardant uses. (Gold et al., 1978; van der Veen and de Boer, 2012) Tris (1-chloro-2-propyl) phosphate (TCIPP or TCPP) is also a suspected carcinogen, while tris (2-chloroethyl) phosphate (TCEP) and triphenyl phosphate (TPHP or TPP) are both neurotoxic in animal studies (van der Veen and de Boer, 2012). Firemaster 550 is a mixture of several flame retardant chemicals that have endocrine disrupting properties (Patisaul et al., 2013; Saunders et al., 2013). Two of its main brominated constituents, 2-ethylhexyl 2,3,4,5-tetrabromobenzoate (EH-TBB or TBB) and bis (2-ethylhexyl) tetrabromophthalate (BEH-TEBP or TBPH), are structurally similar to bis (2-ethylhexyl) phthalate (DEHP) and mono (2-ethylhexyl) phthalate (MEHP), a widely used plasticizer and its metabolite both with endocrine disrupting properties (Benjamin et al., 2017). Additionally, TPHP is used as a phthalate substitute in some personal care products such as nail polish, giving another potential route of exposure to this suspected toxicant (Young et al., 2018).

Similar to PBDEs, most OPFRs and RBFRs are not chemically bound to the products in which they are applied, and thus they migrate into house dust where humans can be exposed to these compounds via dust ingestion, the largest contributor to human exposure. Dermal sorption and inhalation are additional exposure routes for OPFRs and RBFRs (Kim et al.,

2019; Schreder et al., 2016). The human health consequences of exposure to OPFRs and RBFRs are largely unknown at this time, but increasing worldwide production volumes and the persistent nature of these chemicals in the environment have caused concern among consensus groups (Abbasi, 2016; Bennett et al., 2016).

Sugeng et al. examined electronics and furniture characteristics associated with dust OPFR concentrations and loadings in the Netherlands, but had a small sample size and no analysis of sociodemographic factors (Sugeng et al., 2018). Other studies of dust replacement flame retardants from birth cohort studies have only measured OPFRs in participants' homes (Castorina et al., 2017; Phillips et al., 2018), omitting RBFRs. Larger studies of factors associated with environmental levels of OPFRs and RBFRs in various populations are needed to fully understand the distributions and risk factors for these chemicals.

The Health Outcomes and Measures of the Environment (HOME) Study is a multiracial pregnancy and birth cohort with environmental exposures historically similar to US national averages (Kalloo et al., 2018; Woodruff et al., 2011). Women in this study were pregnant during the PBDE phase-out, so their exposures are historically relevant as they represent the beginning of the increase in use of OPFRs and RBFRs. The present study is part of the exposure assessment phase of a comprehensive examination of the developmental neurotoxicity of replacement flame retardant chemicals in this cohort. The specific goals of this article are to characterize both OPFR and RBFR dust concentrations and loadings in the homes of pregnant HOME Study women, and to examine factors associated with their variation.

Materials and methods

Participants

Between March 2003 and February 2006, 468 healthy pregnant women were enrolled in the HOME Study in Cincinnati, Ohio, USA. Enrollment criteria have been previously described (Braun et al., 2017). Briefly, women were eligible for the HOME Study if they were 1) 18 years of age, 2) at 16 $\pm$ 3 weeks' gestation, and 3) living in a home built before 1978 (related to the original randomized trial to reduce residential lead and injury hazards) (Braun et al., 2018). We restricted our final sample to women who remained in the study until delivery and who had a house dust sample collected during pregnancy (n=317). The study protocol was approved by the Institutional Review Board at Cincinnati Children's Hospital Medical Center, and all participants provided informed consent.

Data collection

The HOME Study staff used a High Volume Surface Sampler to collect house vacuum dust samples during a home visit scheduled at ~20 weeks pregnancy (ASTM D5438-17 Standard Practice for Collection of Floor Dust for Chemical Analysis, 2017; Colt et al., 1998). Samples were collected from the main activity room of the home from a 1 meter by 1 meter square area of floor for median sampling time of 275 seconds (range: 145-759 seconds). At the time of sample collection, trained study staff recorded the type of flooring (carpet; wood; tile; vinyl or linoleum) and visible cleanliness by observation (appears clean, some evidence

of housecleaning, or no evidence of housecleaning) in the room from which the sample was taken. Whole dust was stored at -20°C until it was shipped to the Virginia Institute of Marine Sciences for analysis.

We collected sociodemographic information via standardized questionnaires and interviews.

Analytical methods

The analysis of dust for OPFRs (TCEP, TCIPP, TDCIPP, and TPHP) and RBFRs (EH-TBB and BEH-TEBP) were conducted in accordance with La Guardia and Hale (La Guardia and Hale, 2015). Briefly, all samples were prepared for flame retardants analysis by passing them through a $300\text{ }\mu\text{m}$ sieve using a mechanical sieve shaker (Performer III SS-3, Gilson Company, Inc.). The shaker sieved each sample for 90 minutes at 60% 3600 rpm, retaining all material $< 300\text{ }\mu\text{m}$ for analysis ($\sim 20\%$ by weight of the original sample). These samples were stored at $< 0^{\circ}\text{C}$ until analyzed.

For OPFR and RBFR assays, dust ($\sim 100\text{ mg}$) was subjected to accelerated solvent extraction (ASE 200, Dionex, Sunnyvale, CA, USA) with dichloromethane (DCM) (Schreder et al., 2014). Sodium sulfate (NaSO_4) (baked at 400°C for >12 hours) was combined with the sample and added to each ASE extraction cell to reduce dead space volume within the cell. Surrogate standards (2, 3, 4, 4', 5, 6-hexabromodiphenyl ether (BDE-166), deuterated tris (1, 3-dichloro-2-propyl) phosphate (d15-TDCIPP) and, deuterated triphenyl phosphate (d15-TPP)) were added to each sample prior to extraction. Extracts were then purified by size exclusion chromatography (SEC, Envirosep-ABC®, $350 \times 21.1\text{ mm}$. column; Phenomenex, Torrance, CA, USA) using DCM as the mobile phase at 5 mL min^{-1} . Each post-SEC extract was solvent exchanged to hexane, reduced in volume and added to the top of a solid phase 2-g silica glass extraction column (Isolute, International Sorbent Tech.; Hengoed 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). Both fractions (fraction two containing RBFRs and fraction three containing OPFRs) were reduced, solvent exchanged to methanol and transferred to 2 mL vials. Decachlorodiphenyl ether (DCDE) was added to each as an internal standard. Analytes in these purified extracts were separated by ultra-high performance liquid chromatography (UPLC, Acquity UPLC, Waters Corporation, Milford, MA, USA) in the gradient mode (100% methanol (A1) and 100% water (B1)). The UPLC was equipped with a C18 UPLC analytical column (Acquity UPLC BEH C18, $1.7\text{ }\mu\text{m}$, $2.1 \times 150\text{ mm}$, Waters Corp.). Analytes were determined on a triple quadrupole mass spectrometer (3200 QTrap, AB Sciex, Framingham, MA, USA). Ionization was by atmospheric pressure photoionization (APPI). The APPI dopant (acetone) was introduced ($150\text{ }\mu\text{L min}^{-1}$) by a liquid chromatography pump (LC-20AD, Shimadzu Corporation, Kyoto, Japan). Negative product ions were detected in the Multiple Reaction Monitoring (MRM) mode. Quantitation ions for EH-TBB, BEH-TEBP and BDE-166 were: $m/z\ 79\ ([79\text{Br}]^-)$ and $m/z\ 81\ ([81\text{Br}]^-)$; and for TCEP, TCIPP, TDCIPP, dTDCPP and DCDE were: $m/z\ 35\ ([35\text{Cl}]^-)$ and $m/z\ 37\ ([37\text{Cl}]^-)$. For the non-chlorinated OPFRs, the quadrupole was operated in the Q1 positive ionization scan mode. Quantitation ions for TPHP were: $m/z\ 327$

([M+H]⁺) and m/z 328 ([M+2H]⁺); and for d15-TPP were: m/z 342 ([M+H]⁺) and m/z 343 ([M+H]⁺).

Quality Control

Analysis was performed in 16 batches with 18 to 20 samples per batch, for a total of 317 dust samples. Recovery warning limits were calculated for each surrogate at the levels of 100% $\pm$ 2 SD of the recovery rate distribution; compound measurements with recovery rates outside of the warning limits were excluded (n=85 or 4.2% of all values). Recovery warning limits for BDE-166, d15-TDCIPP, and d15-TPP were 62-138%, 61-139%, and 56-144%, respectively.

Analyte measurements were then corrected by dividing by their recovery rates. Mean recovery rates for BDE-166, d15-TDCIPP, and d15-TPP were 102%, 109%, and 86.7% respectively. Batches (n=3) were adjusted by subtracting the blank value from each uncorrected compound measurement if the blank value was greater than 100 ng g⁻¹ for a certain chemical. Each batch contained a duplicate measurement of one sample. When recording these values, we calculated a relative percent difference between the duplicates for each analyte. If one duplicate analyte measurement was more than two times higher than the other (considering the approximately log-normal distribution of the concentrations), we excluded all measurements for that compound from the batch (n=90 or 5.0% of all values).

Data Analysis

For OPFRs, samples < the limit of detection (LOD) (100 ng g⁻¹ for all chemicals) were replaced with LOD / $\sqrt{2}$ (n=70 or 3.9% of all values were replaced) before recovery rate adjustment and blank subtraction (Hornung and Reed, 1990), which resulted in some final corrected values below the LOD. For RBFRs, we used instrument reading values as low as 2 ng g⁻¹, to decrease the amount of missing data as concentrations of those chemicals were much lower than the OPFRs. All concentration values were log₁₀-transformed for data analysis due to right-skewed distributions. We calculated univariate statistics for each compound and created *OPFR* and *RBFR* concentrations from the equally-weighted respective individual analytes.

Floor dust loading (ng/m²) was also calculated for each compound by taking the analyte concentration (ng g⁻¹) for each sample multiplied by the sieved dust weight and then divided by the sampling area from which the dust was taken. For a small number of samples, the sieved dust weight was not recorded, which resulted in slightly different sample sizes between the dust concentration and loading data.

We used multiple linear regression to estimate the difference in OPFR and RBFR concentrations or loadings using predictors chosen *a priori*: age, household income, race (non-Hispanic white vs. all others), education, year of sample collection, smoking status during pregnancy, number of people living in the home, floor surface type (hard floor vs. carpet), and visible cleanliness of floor. We considered maternal race and education to be socioeconomic proxies for the household. We did not have more detailed information on the furniture, carpet, mattress, electronics and other products that may shed flame retardants

over time. We also did not have details of other household members, including activities at home that may increase flame retardant deposit on the floor.

Models were not constructed to explore a specific hypothesis, but rather to examine distributions of flame retardant chemicals in relation to sociodemographic and housing characteristics. We defined hard floors as either wood, tile, or vinyl/linoleum flooring; wood floors were the most common of hard floors (83%). Visible cleanliness was assessed at two levels—clean (“appears clean”) or not clean (“some evidence of housecleaning” or “no evidence of housecleaning”). Income was assessed for potential non-linear associations using generalized additive models and was finally retained as a linear term due to the lack of non-linearity.

We used R for all analyses, (R Core Team, 2018) including packages *EnvStats* (Millard, 2013) and *mgcv* (Wood et al., 2016), with statistical significance at $p < 0.05$.

Results and discussion

Participant characteristics

A total of 317 pregnant women in this study were on average 29.4 years old, 62% were non-Hispanic white, 52% possessed a Bachelor’s degree or higher, 85% were non-smokers, and had an average of two other people living in the home with them. In the main living area where the dust sample was collected, 89% had carpet and 35% were visibly unclean (Table 1). Compared to other pregnant women in the study region, study participants are slightly older, more likely to be black (due to oversampling), more likely to have a college education or greater, but had similar rates of smoking during pregnancy and marriage (Braun et al., 2017).

Flame retardant levels

Concentrations of each analyte were log-normally distributed, and all OPFRs were detected at approximately 10 times higher concentrations than RBFRs (Figure 1), perhaps because OPFRs have been in use longer as additives to consumer products. We observed a high detection rate across analytes; the analyte with the lowest detection rate was TCEP at 80.1% detection, and the highest was TPHP with 100% detection (Table 2).

Geometric mean concentrations of OPFRs in descending order were TCIPP: 2,140 ng g⁻¹ (range: 70.1-166,000 ng g⁻¹), TDCIPP: 1,840 ng g⁻¹ (range: 55.2-228,000 ng g⁻¹), TPHP: 1,070 ng g⁻¹ (range: 34.1-62,100 ng g⁻¹), and TCEP: 669 ng g⁻¹ (range: 56.8-160,000 ng g⁻¹). TCIPP, TDCIPP, and TCEP levels are similar to those reported in the CHAMACOS cohort in California (Castorina et al., 2017) and the TESIE cohort in North Carolina (Phillips et al., 2018). TPHP levels are similar to CHAMACOS (Castorina et al., 2017), but our levels are higher than those from the Netherlands (Sugeng et al., 2018) and lower than those in TESIE (Phillips et al., 2018). Levels of TDCIPP in the house dust of a group of men in Massachusetts were very similar to ours, and that study found an interquartile range increase in TDCIPP concentration was associated with a 3% decrease in free thyroxine and a 17% increase in prolactin in the men (Meeker and Stapleton, 2010). These results could

indicate that the levels of TDCIPP in our cohort are in the range to display endocrine disruption impact.

EH-TBB had the lowest geometric mean dust concentrations of all measured analytes at 59.5 ng g⁻¹ (range: 2.82-7,800 ng g⁻¹), and BEH-TEBP had the second lowest concentration geometric mean at 121 ng g⁻¹ (range: 2.17-13,600 ng g⁻¹). These levels are similar to those reported from Belgian (Ali et al., 2011) and New Zealand (Ali et al., 2012) homes in 2008. EH-TBB is primarily used as a component of Firemaster 550 (Hoffman et al., 2014), which was introduced in 2003 to replace PBDE-based flame retardant products (Stapleton et al., 2008), thus explaining the relatively low concentrations of EH-TBB in these participants. Despite the low concentrations of RBFRs in these dust samples, the very high detection frequencies (>80%) for both RBFRs and OPFRs indicates that these compounds were nearly ubiquitous contaminants at the time of sample collection (between March 2003 and February 2006). We expect OPFR and RBFR use increased after PBDE-containing products reached their end of useful life, were discarded, and were replaced with products containing other flame retardants (Covaci et al., 2010; van der Veen and de Boer, 2012).

We observed that BEH-TEBP and EH-TBB concentrations increased by year of sample collection (Figure 2). Geometric mean concentrations for samples collected in 2005 and 2006 were significantly higher than those collected in 2003, although 2004 sample concentrations did not differ significantly from 2003 samples. This evidence of increasing RBFR levels in dust likely reflects the growing usage of these chemicals in consumer products during this period and justifies further research at later time points to determine whether environmental contamination by RBFRs has plateaued, or if it is still increasing.

We also calculated chemical dust loading for each participant's home. This is a measure of the quantity of an analyte per square meter of sampled floor space and is summarized for each compound in Table 2. Average loading values ranged from 14.5 ng/m² (EH-TBB) to 519.8 ng/m² (TCIPP). All of our OPFR dust concentrations and loadings were higher than those found by Sugeng et al. in the Netherlands from 2013-2015, who did not measure RBFRs (Sugeng et al., 2018). Chemical dust loading values can give an additional sense of scale when interpreting exposure data for substances found in house dust—it may be difficult to conceptualize a gram of dust, but a square meter of floor space is easily imagined. Additionally, chemical dust loading may be a better predictor of chemical body burden. Lanphear et al. studied lead exposure in children and found that dust lead loading was more strongly predictive of blood lead levels than dust lead concentration (Lanphear et al., 2002).

Associations with personal and housing characteristics

Univariate associations between dust OPFR and RBFR dust concentrations are presented in Supplemental Table 1. We used two linear regression models for each chemical and for their class summaries to assess factors associated with dust concentration (Table 3) and dust loading (Table 4). Women's age, income, smoking status, and their home's visible cleanliness were not significantly associated with levels of the OPFRs or RBFRs in any of the models. For both model types, one additional person living in the home was associated with a 17-23% increase in the dust concentrations or loadings of some individual chemicals.

More people living in the home could mean more furniture in the main living area and more personal electronic equipment, both of which may contribute to the flame retardant burden. Also, additional people in the home would produce more wear-and-tear and faster breakdown of products containing flame retardants, releasing them into dust more readily. Abbasi et al. analyzed product wipes from household electronics for the presence of flame retardants, and found that personal computers, small household appliances, and audio/visual equipment in particular contained large quantities of BEH-TEBP relative to other flame retardant chemicals (Abbasi et al., 2016).

Concentrations of TCEP, TCIPP, BEH-TEBP, and OPFRs were 69%, 113%, 79%, and 54% higher respectively among non-Hispanic white homes compared to homes of all other race women. Additionally, TCEP concentrations were 271% as high in homes of women with a Bachelor's degree compared to those with a high school education or less, and 222% as high in homes of women with some graduate school. BEH-TEBP concentrations were 74% higher in homes of women with some college education. These findings of higher exposure in white, more highly educated women's households could be reflective of the age of furniture and electronics present in their homes and the transition from PBDEs to replacement flame retardant chemicals. At the time that these samples were taken, newer furniture and electronic devices would have contained higher concentrations of OPFRs and RBFRs, and older furniture and electronics would have contained higher concentrations of PBDEs (Covaci et al., 2010; van der Veen and de Boer, 2012). Additionally, Vuong et al. found white, highly educated HOME Study women had lower serum PBDE concentrations compared to non-white women with lower education (Vuong et al., 2015).

We did not find women's income to be associated with dust OPFRs or RBFRs, despite the associations with their race and education. While all three variables are used as socioeconomic status proxies, none completely captures this metric. We speculate that the poor prediction power of income is related to the high variability in this factor among the study women, which also depends on other members of the household. Race and education are more stable variables, which may make them better socioeconomic status substitutes in this circumstance.

Point estimates for dust concentration and dust loading models agreed in directionality for all factors except floor type (Figure 3). Having hard floors was associated with higher dust OPFR concentration but lower dust OPFR loading. One possible explanation is that carpeting contains materials, such as fibers, which undergo breakdown and could create a larger dust volume, thus "diluting" the concentration of OPFRs in the samples. In fact, we observed approximately three times greater dust sample volumes from homes with carpeting versus homes with hard floors (data not shown). These results indicate that homes with hard floors had smaller amounts of dust, but higher OPFR chemical concentrations in their dust. Floor type was associated with RBFR loading, but not concentration (Tables 3 and 4).

Strengths and Limitations

This study is the largest to date to estimate exposure to OPFRs and RBFRs via house dust in an established pregnancy and birth cohort. Serum PBDE concentrations in HOME Study women were similar to those of women in the NHANES (2003-2004) (Chen et al., 2014), a

nationally representative sample of the United States population, so we may expect exposure to OPFRs and RBFRs in the HOME Study to be similar to the NHANES during that period as well. One of the strengths of the HOME Study is the addition of individual-level data available for analysis and covariate adjustment. We leveraged this data set in our multiple regression analysis to explore sociodemographic and house characteristics associated with changes in dust OPFRs and RBFRs concentrations and loadings, some of which have not previously been reported.

However, we lacked information on cleaning frequency and types of furniture and electronics present in the room, which would likely affect concentrations of OPFRs and RBFRs. Sugeng et al. found that increased frequency of vacuuming and dusting was associated with lower dust levels of flame retardants, and that the presence and use patterns of various electronic devices also affected analyte levels (Sugeng et al., 2018). Also, many types of carpeting contain flame retardant chemicals, but we were unable to account for this in our analysis. We suggest that other cohort studies planning to analyze flame retardant exposures collect these covariates when household and biological specimens are collected to obtain a more complete picture of how the home environment contributes to dust levels of OPFRs and RBFRs.

Conclusions

In this cohort of women pregnant during the PBDE phase-out, concentration of RBFRs in house dust increased from 2003-2006. OPFRs were present at approximately 10 times higher concentrations than RBFRs, although RBFRs were detected in 99.4% of samples and OPFRs were detected in 100% of samples. Sociodemographic and housing characteristics were both associated with concentrations and loadings of these chemicals in house dust.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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Abbreviations:

OPFR	organophosphate flame retardant
RBFR	replacement brominated flame retardant
TCIPP	tris-(1-chloro-2-propyl)-phosphate
TDCIPP	tris-(1,3-dichloro-2-propyl)-phosphate
TPHP	triphenyl phosphate

EH-TBB	2-ethylhexyl-2,3,4,5-tetrabromobenzoate
BEH-TEBP	bis-(2-ethylhexyl)-tetrabromophthalate
PBDE	polybrominated diphenyl ether
DEHP	bis (2-ethylhexyl) phthalate
HOME	Health Outcomes and Measures of the Environment
ASE	accelerated solvent extraction
DCM	dichloromethane
BDE-166	(2,3,4,4',5,6-hexabromodiphenyl ether
d15-TDCIPP	deuterated tris (1,3-dichloro-2-propyl) phosphate
D15-TPP	deuterated triphenyl phosphate
SEC	size exclusion chromatography
DCDE	decachlorodiphenyl ether
UPLC	ultra-high performance liquid chromatography
APPI	atmospheric pressure photoionization
MMR	multiple reaction monitoring
LOD	limit of detection
CR	concentration ratio
CI	confidence interval
HS	high school
LR	loading ratio

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Highlights:

- We measured OPFRs and RBFRs in 317 women's house dust during the PBDE phase-out
- OPFR concentrations were about 10x higher than RBFR concentrations
- RBFR concentrations increased from 2003-2006
- Both sociodemographic and housing characteristics were related to dust levels

Figure 1: Distribution of house dust flame retardant concentrations. The Y-axis is on the log₁₀ scale. OPFRs: TCEP, TCIPP, TDCIPP, and TPHP. RBFRs: BEH-TEBP and EH-TBB.

Figure 2:

Dust concentrations of RBFRs by year from 2003-2006. Geometric mean of sample concentrations were significantly increased in 2005 and 2006 compared to 2003 levels. The Y-axes are on the log₁₀ scale. *p<0.05, **p<0.01

Figure 3:

Adjusted ratios and 95% confidence intervals for floor type variable (hard floor vs. carpet) in regression models of chemical dust concentrations and chemical dust loadings. Values <1 indicate higher chemical values for carpeted floors, and values >1 indicate higher chemical values for hard floors. Adjusted for age, income, smoking, race, education, year of dust collection, and visible cleanliness.

Table 1:**Cohort demographics and characteristics**

	N (mean)	% (range)
Total dust samples	317	100.0%
Household size (number of people)	(3.03)	(2-8)
Age at delivery (years)	(29.4)	(18-45)
Race		
White	196	61.8%
Non-white	121	38.2%
Ethnicity		
Non-Hispanic	309	97.5%
Hispanic	8	2.5%
Education		
High school or less	77	24.3%
Some college	76	24.0%
Bachelor's degree	92	29.3%
Any graduate school	72	22.7%
Smoking status during pregnancy		
Smoker	22	6.9%
Non-smoker	270	85.2%
Missing	25	7.9%
Household income		
\$30,000	101	31.9%
\$30,001-60,000	80	25.2%
>\$60,000	136	42.9%
Year		
2003	56	17.7%
2004	138	43.5%
2005	108	34.1%
2006	15	4.7%
Floor type		
Carpet	282	89.0%
Hard floor	30	9.5%
Missing	5	1.6%
Visible cleanliness		
Clean	206	65%
Less clean	97	30.6%
Missing	14	4.4%

Table 2:

Dust concentration and loading summary statistics for OPFRs and RBFRs

Dust Concentration (ng g⁻¹)						
	TCEP	TCIPP	TDCIPP	TPHP	BEH-TEBP	EH-TBB
Total samples	305	305	305	286	269	251
%>LOD ^a	80.1%	98.7%	99.1%	100.0%	99.4%	92.7%
Min	<LOD	<LOD	<LOD	<LOD	2.17	2.82
1st quartile	183	876	902	516	63.1	27.4
Geometric mean	669	2140	1840	1070	121	59.5
Median	759	1860	1860	995	115	45.8
3rd quartile	1810	4570	3720	2080	233	108
Max	160000	166000	228000	62100	13600	7800
Dust Loading (ng/m²)						
Total samples	300	300	300	283	265	247
Min	<LOD	<LOD	<LOD	<LOD	0.20	0.26
1st quartile	52.4	166	197	93.8	12.2	4.77
Geometric mean	161	520	449	246	30.5	14.5
Median	159	424	461	231	34.3	13.3
3rd quartile	525	1220	1030	646	71.3	38.3
Max	34900	47300	102000	10400	1230	2270

^aLOD = 100 ng g⁻¹ for TCEP, TCIPP, TDCIPP and TPHP. LOD = 2 ng g⁻¹ for BEH-TEBP and EH-TBB using instrument reading values.

Missing values were replaced with $LOD / \sqrt{2}$. Values were then recovery rate corrected and blank subtracted.

Table 3

Adjusted associations between factors and flame retardant chemical concentrations in house dust samples.

	<u>TCEP</u>		<u>TCIPP</u>		<u>TDCIPP</u>		<u>TPHP</u>		<u>BEH-TEBP</u>		<u>EH-TBB</u>		<u>OPFRs</u>		<u>RBFRs</u>	
	CR	95% CI	CR	95% CI	CR	95% CI	CR	95% CI	CR	95% CI	CR	95% CI	CR	95% CI	CR	95% CI
Household size	1.09	0.92–1.30	1.13	0.98–1.31	1.14	1.00–1.31	1.13	0.98–1.31	1.20	1.08–1.34	1.10	0.96–1.26	1.13	1.00–1.28	1.19	1.06–1.34
Race																
White	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Non-white	0.59	0.35–0.98	0.47	0.30–0.72	0.72	0.48–1.08	1.01	0.66–1.56	0.56	0.39–0.78	0.70	0.46–1.05	0.65	0.45–0.95	0.71	0.50–1.02
Education																
HS or less	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Some college	1.09	0.59–2.01	1.03	0.62–1.72	1.19	0.73–1.93	1.32	0.79–2.21	1.74	1.16–2.60	1.03	0.62–1.69	1.13	0.72–1.76	1.47	0.96–2.26
Bachelor's Degree	2.71	1.30–5.62	1.70	0.92–3.14	1.21	0.68–2.16	1.25	0.68–2.32	1.66	1.02–2.71	0.98	0.54–1.81	1.57	0.92–2.65	1.34	0.80–2.23
Some graduate school	2.22	1.03–4.82	1.36	0.71–2.61	1.47	0.80–2.72	1.61	0.85–3.06	1.82	1.10–3.03	1.14	0.60–2.15	1.35	0.77–2.34	1.54	0.90–2.65
Year																
2003	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
2004	0.79	0.35–1.77	0.76	0.38–1.51	1.30	0.68–2.48	1.10	0.57–2.14	1.58	0.90–2.77	1.52	0.72–3.21	0.95	0.53–1.72	1.63	0.89–3.00
2005	0.60	0.26–1.38	0.86	0.43–1.73	1.21	0.63–2.34	1.53	0.78–3.00	1.96	1.10–3.49	1.98	0.93–4.23	0.99	0.54–1.81	1.90	1.02–3.54
2006	0.60	0.24–1.48	0.75	0.35–1.60	1.46	0.71–2.98	1.27	0.61–2.65	2.74	1.48–5.05	3.01	1.36–6.67	0.94	0.49–1.80	2.92	1.51–5.64
Floor type																
Carpet	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Hard floor	1.72	0.88–3.36	1.12	0.64–1.96	1.99	1.17–3.38	2.11	1.21–3.67	0.82	0.53–1.26	1.00	0.59–1.68	1.97	1.23–3.18	0.85	0.53–1.35
Visible Cleanliness																
Clean	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Less clean	0.69	0.44–1.07	0.79	0.54–1.14	0.83	0.58–1.18	0.90	0.62–1.31	0.88	0.66–1.18	0.94	0.67–1.32	0.78	0.57–1.08	0.86	0.63–1.15

Abbreviations: CR = concentration ratio; CI = confidence interval; HS = high school.

Models also adjusted for age, income, and smoking.

Bolded terms are statistically significant at p<0.05.

Table 4

Adjusted associations between factors and flame retardant chemical loadings in house dust samples.

	<u>TCEP</u>		<u>TCIPP</u>		<u>TDCIPP</u>		<u>TPHP</u>		<u>BEH-TEBP</u>		<u>EH-TBB</u>		<u>OPFRs</u>		<u>RBFRs</u>	
	LR	95% CI	LR	95% CI	LR	95% CI	LR	95% CI	LR	95% CI	LR	95% CI	LR	95% CI	LR	95% CI
Household size	1.13	0.93–1.38	1.17	0.99–1.40	1.18	1.01–1.39	1.17	1.00–1.38	1.23	1.06–1.43	1.17	0.98–1.39	1.16	1.00–1.35	1.24	1.06–1.43
Race																
White	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Non-white	0.77	0.42–1.40	0.61	0.37–1.03	0.95	0.59–1.52	1.23	0.76–1.97	0.88	0.56–1.39	1.04	0.62–1.75	0.84	0.53–1.32	1.09	0.70–1.71
Education																
High school or less	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Some college	0.95	0.46–1.94	0.90	0.48–1.67	1.03	0.59–1.81	1.19	0.67–2.10	1.51	0.88–2.58	0.85	0.45–1.61	1.01	0.59–1.75	1.31	0.77–2.24
Bachelor's Degree	2.15	0.91–5.06	1.35	0.64–2.84	0.96	0.49–1.88	1.09	0.55–2.15	1.45	0.76–2.78	0.80	0.37–1.74	1.29	0.68–2.46	1.15	0.60–2.20
Some graduate school	1.89	0.76–4.67	1.16	0.53–2.55	1.25	0.62–2.55	1.34	0.66–2.71	1.42	0.72–2.80	0.89	0.39–2.00	1.13	0.57–2.24	1.27	0.64–2.50
Year																
2003	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
2004	0.60	0.23–1.56	0.58	0.26–1.34	1.00	0.47–2.11	0.85	0.41–1.77	1.17	0.56–2.48	1.00	0.39–2.60	0.71	0.34–1.48	1.23	0.57–2.65
2005	0.30	0.11–0.80	0.43	0.19–1.01	0.61	0.28–1.31	0.77	0.36–1.61	1.00	0.46–2.16	0.95	0.36–2.50	0.50	0.24–1.06	0.97	0.45–2.12
2006	0.31	0.11–0.88	0.38	0.15–0.96	0.74	0.32–1.69	0.63	0.28–1.42	1.45	0.64–3.28	1.62	0.59–4.47	0.49	0.22–1.08	1.63	0.71–3.73
Floor type																
Carpet	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Hard floor	0.62	0.28–1.36	0.40	0.20–0.80	0.72	0.39–1.32	0.79	0.43–1.46	0.34	0.19–0.60	0.46	0.24–0.89	0.76	0.42–1.36	0.36	0.20–0.64
Visible Cleanliness																
Clean	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Not clean	1.04	0.62–1.74	1.18	0.75–1.86	1.25	0.83–1.87	1.37	0.90–2.07	1.18	0.81–1.72	1.42	0.92–2.19	1.15	0.78–1.71	1.18	0.81–1.72

Abbreviations: LR = loading ratio; CI = confidence interval.

Models also adjusted for age, income, and smoking.

Bolded terms are statistically significant at p<0.05.