Flame Retardant Exposures in California Early Childhood Education Environments

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Key words: child care; children; exposure; flame retardant; PBDEs

Abbreviations

BEH-TEBP: bis(2-ethylhexyl) tetrabromophthalate

ECE: early childhood education

EH-TBB: 2-ethylhexyl tetrabromobenzoate

PBDE: polybrominated diphenyl ether

TCEP: tris (2-chloroethyl) phosphate

TDCIPP: tris (1,3-dichloroisopropyl) phosphate

Abstract

Infants and young children spend as much as 50 hours per week in child care and preschool. Although approximately 13 million children, or 65% of all U.S. children, spend some time each day in early childhood education (ECE) facilities, little information is available about environmental exposures in these environments. We measured flame retardants in air and dust collected from 40 California ECE facilities between May 2010 and May 2011. Low levels of six polybrominated diphenyl ether (PBDE) congeners and four non-PBDE flame retardants were present in air, including two constituents of Firemaster 550 and two tris phosphate compounds [tris (2-chloroethyl) phosphate (TCEP) and tris (1,3-dichloroisopropyl) phosphate (TDCIPP)]. Tris phosphate, Firemaster 550 and PBDE compounds were detected in 100% of the dust samples. BDE47, BDE99, and BDE209 comprised the majority of the PBDE mass measured in dust. The median concentrations of TCEP (319 ng/g) and TDCIPP (2,265 ng/g) were similar to or higher than any PBDE congener. Levels of TCEP and TDCIPP in dust were significantly higher in facilities with napping equipment made out of foam (Mann-Whitney p-values<0.05). Child BDE99 dose estimates exceeded the RfD in one facility for children <3 years old. In 51% of facilities, TDCIPP dose estimates for children <6 years old exceeded age-specific "No Significant Risk Levels (NSRLs)" based on California Proposition 65 guidelines for carcinogens. Given the overriding interest in providing safe and healthy environments for young children, additional research is needed to identify strategies to reduce indoor sources of flame retardant chemicals.

1. Introduction

Many infants and young children spend as much as ten hours per day, five days per week, in child care and preschool centers. Nationally, 13 million children, or 65% of all U.S. children, spend some time each day in child care or preschool (Tulve et al., 2006). Early childhood education (ECE) facilities are varied and include home-based child care providers, centers operated like private schools, and programs run by government agencies (e.g., preschool in school districts or Head Start) or religious institutions. These facilities are located in a variety of building types, including homes, schools, commercial buildings, and portable classrooms.

Polybrominated diphenyl ethers (PBDEs) are flame retardant compounds that have been used in consumer products for decades, including polyurethane foam in furniture, child car seats, and related products. These compounds persist in the environment, and are commonly detected in house dust and human tissue (Sjödin et al., 2008a; Sjödin et al., 2008b). Several studies have reported higher PBDE serum levels among children compared to adults (Toms et al., 2009; Lunder et al., 2010; Eskenazi et al., 2011). Higher levels in children are likely attributable to increased exposure via non-dietary ingestion due to frequent hand-to-mouth contact (Wu et al., 2009; Johnson et al., 2010; Stapleton et al., 2012a).

A growing body of research has raised concern about the health effects of PBDE flame retardant exposure in children (Roze et al., 2009; Herbstman et al., 2010; Gascon et al., 2011; Eskenazi et al., 2013). Several studies have documented disruption of thyroid homeostasis, important for normal brain development, in pre- and postnatally exposed animals (Zhou et al., 2002; Dingemans et al., 2011) and humans (Turyk et al., 2008; Chevrier et al., 2010). Consistent with these studies, epidemiological evidence suggest adverse neurodevelopmental effects associated with early childhood PBDE exposure (Gascon et al., 2011; Eskenazi et al., 2013).

Restrictions on the use of PBDE flame retardants in the U.S. have resulted in increased use of replacement fire retardants in furniture, including Firemaster 550 and tris chloroalkyl phosphate (tris phosphate) flame retardants (Stapleton et al., 2012b). Prior to 1977, chlorinated tris [(1,3-dichloroisopropyl) phosphate (TDCIPP)] was used in children's sleepwear as a fire retardant; however, manufacturers voluntarily stopped using it in these products after it was found to be mutagenic (Blum and Ames, 1977; Gold et al., 1978). Recently, TDCIPP was listed as a carcinogen by the state of California (State of California Environmental Protection Agency). Today TDCIPP is widely used and commonly detected in furniture foam as well as infant products (Blum, 2007; Stapleton et al., 2011). To date, no studies have examined flame retardants in ECE facilities.

In the current study, we measured 14 PBDE flame retardants and four non-PBDE flame retardants, including two constituents of Firemaster 550 [2-ethylhexyl tetrabromobenzoate (EH-TBB) and bis(2-ethylhexyl) tetrabromophthalate (BEH-TEBP)] and two tris phosphate flame retardants [tris (2-chloroethyl) phosphate (TCEP) and TDCIPP], in 40 child care facilities in California. We also measured six PBDE congeners (i.e., BDE47, 99, 100, 153, 154, and 209) and four non-PBDE flame retardants (EH-TBB, BEH-TEBP, TCEP and TDCIPP) indoor and outdoor air. This study is the first to examine flame retardant compounds in ECE facilities in California and the United States.

2. Materials and methods

Forty ECE facilities located in two northern California counties [Monterey (n=20) and Alameda (n=20)] participated in this study. Monterey County, CA is largely rural and agricultural, while Alameda County, CA, is predominantly urban or suburban. To recruit a diverse sample, we geographically coded center and large home-based licensed (>8 children) ECE facilities by zip code using publicly available databases (Community Care Licensing Division, 2010). The center-licensed facilities were divided into 12 geographical units with approximately equal population in each county while the home-based facilities were divided into 8 geographical units. For center-licensed facilities, a recruitment flyer was mailed to 15 randomly selected child care centers per geographical group in Alameda County (n=160). Recruitment flyers were sent to every child care center in our database in Monterey County (n=130). Participating ECE facilities were given a small gift certificate to a school-supplies retailer. We ultimately completed assessments at 28 child care centers and 12 home-based facilities between May 2010 and May 2011, including all four seasons.

2.1. Questionnaires and site visits

We administered questionnaires and performed facility inspections to characterize environmental quality in the ECE facilities. Information obtained included building type (home, school or office), building age, ECE type (home versus center), building materials, neighborhood type (residential, commercial or agricultural), and the presence of foam napping equipment, upholstered furniture and electronics.

2.2. Flame retardant measurement in dust

The dust sampling methods followed procedures described in the American Society for Testing Materials (ASTM) Standard Practice D 5438-05. Dust samples were collected using the High Volume Small Surface Sampler (HVS3) (Roberts et al., 1991). With the exception of one facility where no carpets or floor dust was present, dust samples were collected from carpets centrally located in the primary child care room where air sampling would take place (n=39). Dust samples were collected from at least 1 meter squared into cleaned, 250 ml amber glass bottles (I-CHEM, item# 341-0250). Bulk dust was sieved to 150 µm using a stainless steel sieve and aliquotted. Dust samples were analyzed by U.S. EPA's National Exposure Research Laboratory (Research Triangle Park, North Carolina) for 14 PBDEs (Clifton et al., 2013), and for two tris phosphates and two Firemaster 550 compounds by Battelle Memorial Institute (Columbus, Ohio). Both dust concentrations (i.e., ng/g) and dust loading (i.e., ng/m²) were determined. Detailed dust sampling, laboratory, and analytical QA/QC results are presented in the Supplementary Material (SM), Tables S1-S4.

2.3. Flame retardant measurement in air

Indoor air samples were collected over 6-10 hours when children were present at the ECEs. The indoor air sampling system used a single rotary vane pump installed in a stainless steel box, lined with foil-faced fiberglass sound insulation to reduce noise, to pull air through a manifold equipped with taper-tube flow meters (Key Instruments #10710). Air was pulled at 4 liters per minute onto two identical pre-cleaned polyurethane foam (PUF) plug cartridges in parallel. One cartridge was analyzed by Battelle Memorial Institute for the tris phosphate and Firemaster 550 compounds (and other compounds not reported here); the second cartridge was analyzed for selected PBDEs. Sampling methods did not include filters to collect particles upstream of the PUF plug, therefore, reported levels of less volatile PBDEs may be underestimated because fine particles with adsorbed PBDEs could pass through the PUF. Outdoor air samples were collected from a random subset of facilities (n=16). Outdoor air was pulled using SKC Universal XR Pumps checked before and after sampling with a Gilibrator® air flow calibrator. Detailed air sampling, laboratory, and analytical QA/QC results are presented in the SM, Tables S5-S6.

2.4. Data analysis

Statistical analysis included computation of descriptive statistics for non-PBDE compounds and individual and summed PBDE congeners measured in air and dust. Flame retardant levels below the method detection limit (MDL) were imputed to MDL/√2 (Hornung and Reed, 1991). We computed Spearman's rank correlation coefficients to compare the indoor air concentrations with dust concentrations and dust loading levels. The Mann-Whitney rank sum test was used to assess bivariate associations between PBDE and non-PBDE dust levels and potential predictors of pentaBDE and tris phosphate flame retardants in air and dust, including the presence (yes/no) of foam napping equipment and upholstered furniture. The association between levels of decaBDE (i.e., BDE209) and the presence of computers (yes/no) in the facilities was also examined. The sum of pentaBDEs was calculated by summing BDE47, 99, 100, 153, and 154 by weight (La Guardia et al., 2006). Stata software, version 11 (StataCorp LP, College Station, TX) was used for descriptive statistics and test of association, while Figures 1 and 2 were produced in R Version 2.14.1.

2.5. Non-cancer risk estimation

A screening-level risk assessment was conducted to evaluate flame retardant exposures to children in the ECE facilities. There are currently no health-based reference concentrations to evaluate flame retardants in air. Thus, we calculated non-dietary ingestion child exposure-dose estimates based on the measured dust concentrations for children in four distinct age groups (birth to <1 year; 1 to <2 years; 2 to <3 years; and 3 to <6 years). Assuming a soil and dust intake rate of 60 mg per day for children <1 year and 100 mg per day for children ≥1 year old (US EPA, 2011), we used standard equations relating concentration, intake rate, and an exposure factor to standard body weights for each age category (ATSDR, 2005). We assumed that gastrointestinal absorption of these compounds was 100%. Since children are not present in ECE facilities every day, we calculated the exposure factor assuming a child spends five days per week and 48 weeks per year (which accounts for four weeks away from child care for holidays and vacation). We also assumed exposure over one year (ATSDR, 2005). Detailed information on the calculations is presented in the SM.

Child dose estimates were compared to U.S. EPA chronic oral reference doses (RfDs) for BDE47, 99, 153 and 209 (U.S. EPA, 2008a, b, c, d). The hazard quotient, defined as the ratio of an observed dose to an RfD, was used to express risk relative to the RfDs. If the hazard quotient is greater than 1, the exposure dose estimate exceeded health-based exposure limits. Because the health-based reference values include safety factors, however, exposures exceeding these levels are not necessarily likely to result in adverse health effects (ATSDR, 2005).

2.6. No Significant Risk Levels (NSRLs) for Cancer

Under California's Proposition 65, the Office of Environmental Health Hazard Assessment (OEHHA) has set "Safe Harbor Levels" called No Significant Risk Levels (NSRLs) for carcinogenic substances, defined as the daily intake level posing a one in 100,000 (10^{-5}) excess risk of cancer over a lifetime (OEHHA, 2001); the NSRL for TDCIPP is 5.4 µg/day (OEHHA, 2011).

Using this benchmark, we computed a child-specific NSRL for TDCIPP based on age-specific child body weights (US EPA, 2011) and OEHHA's guidelines to define Safe Harbor Levels that account for the increased sensitivity of very young children, which incorporates an age sensitivity factor (ASF) of 10 for children below the age of two years and an ASF of 3 for children between the ages of two and six years (OEHHA, 2001). Age-adjusted NSRLs were calculated for four age groups (i.e., birth to <1 year; 1 to <2 years; 2 to <3 years; and 3 to <6 years):

$$NSRL_{\ child}\left(\frac{\mu g}{day}\right) = \frac{\dfrac{NSRL_{adult}\left(\frac{\mu g}{day}\right)}{BW_{\ adult}(70\ kg)} \times BW_{child}\ (Varies\ by\ Age\ Group, kg)}{ASF\ (Varies\ by\ Age\ Group)}$$

It should be noted that an age-specific NSRL, such as the $\text{NSRL}^{\text{child (0 to <1 year)}}$, is the estimated daily intake for that age range, which contributes $1/70^{\text{th}}$ (assuming a 70 year lifetime) of the target lifetime cancer risk in that particular year of life. If the ratio of a child's TDCIPP oral dose estimate (µg/day) to age-adjusted NSRL (µg/day) is greater than 1, the exposure dose estimate exceeded the 10^{-5} threshold.

3. Results and discussion

3.1. ECE facility and child characteristics

The 40 ECE facilities served a total of 1,764 children. Building types included single family detached homes (38%), traditional school buildings (28%), portable school buildings (23%), office buildings (8%), and churches (5%). Half the facilities were in buildings constructed after 1970, with the oldest structure built in 1903 and the most recent built in 2008. Twenty-six (65%) of the facilities were in residential neighborhoods, 8 (20%) were in commercial areas, five (13%) were adjacent to agricultural fields, and one (3%) was in a rural/ranch area.

The average attendance per facility was 44 children (range=4-200). Seventy-six percent of the children were 3+ years old, 19% were 2-3 years, and 5% were less than 2 years of age; 95% of the children spent at least 1-2 hours outside each day, with some spending up to 6 hours outside, depending on the weather. Thirty-seven percent of children spent >8 hours per day in child care, 41% spent 5-8 hours, and 22% spent less than 5 hours.

3.2. Flame retardant levels in dust

Table 1 summarizes flame retardant dust concentrations in the ECE facilities. Thirty-nine dust samples were analyzed for 14 PBDE congeners, EH-TBB, BEH-TEBP, TCEP and TDCIPP. Where duplicate samples were measured, the average was used.

We detected PBDEs in 100% of the dust samples collected. The median levels of PBDE flame retardants in dust were somewhat lower than levels reported in other studies focusing on residential environments in California, possibly due to the frequent cleaning that occurs in ECE facilities (Zota et al., 2008; Quiros-Alcala et al., 2011; Dodson et al., 2012). For example, the median BDE99 level in the ECE facilities (1,031 ng/g) was very similar to a study of other California homes (1,100 ng/g) (Dodson et al., 2012) and more than 4 times lower than a study of low-income California homes (4,965 ng/g)(Quiros-Alcala et al., 2011). Maximum flame retardant levels in dust from ECE facilities were similar to the upper-bound levels measured in other California studies.

Figure 1 shows the relative proportion of each PBDE congener's mass to total PBDE mass in the ECE facilities. BDE47, BDE99, and BDE209 comprised the bulk of the PBDE mass measured in the dust samples. Although BDE47 and BDE99 are no longer used, furniture and other long-lasting products containing these materials are still found in many buildings (Great Lakes Chemical Corporation, 2005). BDE209 is currently used in plastic electronic casings and textiles, and in many dust samples (~40%), it was the dominant congener measured.

Use of tris phosphate flame retardants (TCEP and TDCIPP) is increasing as a replacement for PBDEs. These tris phosphate compounds were detected in 100% of the dust samples (Table 1). The median TDCIPP concentrations (2,265 ng/g) and TCEP (319 ng/g) were similar to or higher than any median of individual PBDE congeners. The median TDCIPP level measured in the ECE facilities was also slightly higher than levels reported in a previous study of homes in California (2,100 ng/g) and Massachusetts (1,752 ng/g) (Stapleton et al., 2009; Dodson et al., 2012). Components of the Firemaster 550 flame retardant mixture (EH-TBB and BEH-TEBP) were also detected in 100% of the dust samples, with median levels of 362 and 133 ng/g, respectively. These levels were within an order of magnitude of those reported in other California homes [i.e., EH-TBB (100 ng/g) and BEH-TEBP (260 ng/g)] (Dodson et al., 2012). Firemaster 550 has been used as a replacement for penta-PBDEs (Dodson et al., 2012).

The compounds TDCIPP (median= 6,046 ng/m²) and BDE209 (median = 2,924 ng/m²) had the highest loading values across the flame retardants measured. Flame retardant loading values are presented in the SM, Table S7.

3.3 Predictors of flame retardant concentrations in dust

Twenty-nine facilities (74%) had upholstered furniture present in the child care room where dust sampling occurred and 17 (43%) had napping equipment made out of foam. Dust concentrations of all of the individual pentaBDE congeners (BDE47, 99, 100, 153 and 154) and the sum of pentaBDE congeners were significantly higher in the 29 facilities with any upholstered furniture or foam napping equipment present (e.g., Σ pentaBDE = 2,642 versus 1,362 ng/g; Mann-Whitney p-values<0.05) (see SM, Table S8 and Figure S1). Concentrations of the tris phosphate flame retardants TCEP and TDCIPP were significantly higher in facilities with foam napping equipment present compared to facilities without foam napping equipment (medians= 643 versus 261 ng/g and 2,837 versus 1,539 ng/g, respectively; Mann-Whitney p-values<0.05) (Figure 2 and SM Table S9). Similarly, concentrations of the pentaBDE flame retardants were higher in facilities where foam napping equipment was present compared to locations without foam napping equipment (median BDE-99 levels=1,119 ng/g versus 740 ng/g, respectively), albeit the differences were not statistically significant (see SM, Table S9). While consumer electronics have been associated with decaBDE (BDE209) levels in dust (Webster et al., 2009), we did not find significantly higher BDE209 dust concentrations in rooms with a computer or television.

3.4. Flame retardant levels in air

Table 2 summarizes flame retardant concentrations in indoor air. Outdoor air flame retardant concentrations are presented in the SM (Table S10). Forty indoor and 16 outdoor air samples were analyzed for 5 PBDE congeners, EH-TBB, BEH-TEBP, TCEP and TDCIPP. Due to laboratory problems with low recovery of the labeled BDE209 in some sample surrogates, BDE209 was quantified in just 7 indoor air samples, but was detectable in all of these samples (median=1.4 ng/m^3). The pentaBDE congeners, BDE47 and BDE99, were commonly detected indoors (>MDL = 90% and 95%, respectively) and outdoors (>MDL = 56% and 75%, respectively). Levels of BDE47 and BDE99 were notably higher indoors compared to outdoors (mean indoor to outdoor [I/O] ratio = 17.6 and 10.0, respectively)(see SM, Table S11). Levels of TDCIPP and TCEP were also higher indoors compared to outdoors (I/O ratio = 2.6 and 6.0, respectively). In general, flame retardant levels in air were low, often below detection at the median or less than 1 ng/m^3. As would be expected, indoor air levels were higher than outdoor levels for several flame retardants, likely associated with volatilization or re-suspension of contaminated dust particles indoors. Flame retardant levels in indoor air were not associated with the presence of upholstered furniture, foam napping equipment, or computers (data not shown).

Although some correlations attained statistical significance, overall, flame retardant levels in air were weakly correlated to levels in dust on a concentration or loading basis (see SM, Table S12), likely due to the relatively low volatility of these compounds. Indoor air and dust concentrations of TDCIPP were significantly correlated (Spearman rho = 0.34; p-value < 0.05), while BEH-TEBP indoor air concentrations and dust loading levels were significantly correlated (rho = 0.37; p-value < 0.05).

3.5. Flame retardants health risk characterization

Brominated flame retardants are known neurotoxicants (Eskenazi et al., 2013) and evidence indicates that some are also endocrine disrupters (Legler and Brouwer, 2003; Hamers et al., 2006; Johnson et al., 2013). The RfDs for several PBDE congeners were based on adverse neurobehavioral effects in animals (U.S. EPA, 2008a, b, c, d). We compared child dose estimates for the measured flame retardants to health-based non-cancer RfDs when available. For this assessment, we grouped children into four age groups (birth to <1 year; 1 year to <2 years; 2 years to <3 years and 3 years to <6 years). Child intake estimates of BDE99 for the birth to <3 year old age groups, based on U.S. EPA non-dietary ingestion assumptions, exceeded the oral RfD (0.0001 mg/kg-day) in one facility (3%) (US EPA, 2011). The hazard quotients calculated for the birth to <3 year old age groups used the 95th percentile and maximum oral dose estimates, and ranged from 0.6 to 0.7 and 0.7 to 0.9, respectively, for BDE47, and ranged from 0.6 to 0.8 and 0.2 to 1.5, respectively, for BDE99. A potential limitation of this screening risk assessment is that we used an intake rate of 60 mg-dust/day for children <1 year and 100 mg-dust/day for children ≥ 1 year (US EPA, 2011) to represent exposure while attending an ECE facility, which may overestimate intake because children spend less than a full day in childcare. In addition, the screening risk assessment did not consider mixed exposures. The age-specific

PBDE dose estimates and hazard quotients for the four PBDE congeners with oral RfDs (BDE47, -99, -153, -209) are presented in the SM, Tables S14 (a-d).

The 50th and 95th percentile oral exposure dose estimates (µg/day) for TDCIPP exceeded the age-specific NSRL in all four age groups assessed (See SM, Table S15). The TDCIPP 50th and 95th percentile NSRL ratios for the four age groups were 1.7 and 28.1; 1.7 and 27.7; 1.4 and 22.9; and 1.0 and 17.0, respectively. Child TDCIPP exposure estimates exceeded age-adjusted NSRL benchmarks based on carcinogenicity in 51% of facilities for children <6 years old.

4. Conclusions

This is the first study to report air and dust levels of PBDE and non-PBDE fire retardants in child care environments. A total of 40 indoor and 16 outdoor air samples, and 39 dust samples were collected and analyzed for flame retardant compounds. Airborne levels were generally low, but flame retardants were always present in dust. Flame retardant concentrations in dust were higher in facilities where upholstered furniture and foam napping equipment was present, consistent with other studies showing tris chloroalkyl phosphate flame retardants in infant products (Stapleton et al., 2011). In some cases the individual differences were not statistically significant; however, the overall trend of higher levels, especially for the pentaBDE congeners, suggests that these furnishings were associated with increased pentaBDE contamination in dust. Screening-level child dose estimates of congener BDE-99, based on conservative non-dietary ingestion assumptions, exceeded the RfD in one facility for children <3 years old. The manufacture, distribution, and processing of products containing pentaBDEs (BDE47, BDE99, BDE100, BDE153 and BDE154) was banned in California as of June 1, 2006 (California Health and Safety Code); however, results of this study confirm the persistence of these chemicals in the indoor environment. Replacement furniture fire retardants such as chlorinated tris (TDCIPP), which is listed as a carcinogen on California's Proposition 65, and Firemaster 550, a suspected endocrine disruptor in animals (Patisaul et al., 2013), have come into wider use. Child TDCIPP exposure estimates in this study exceeded age-adjusted NSRL benchmarks based on carcinogenicity in 51% of facilities for children <6 years old. Our findings demonstrate that flame retardant exposures are occurring in ECE environments and suggest that more research is necessary to assess the potential health risks to children and adult staff, and, if warranted, to develop and implement policies to mitigate these exposures.

Acknowledgments

This research was funded by the California Air Resources Board (ARB Contract No. 08-305). This work does not necessarily reflect the opinion or official policy of the ARB. Although this work was reviewed by U.S. EPA and approved for publication, it does not necessarily reflect the opinion or official policy of the U.S. EPA. Co-author Walter Weathers has recently retired from the U.S. EPA.

References

ATSDR, 2005. Public Health Assessment Guidance Manual. Atlanta, GA: Center for Disease Control.

Blum, A., 2007. The fire retardant dilemma. Science 318, 194-195.

Blum, A., Ames, B.N., 1977. Flame-retardant additives as possible cancer hazards. Science 195, 17-23.

California Health and Safety Code, Division 104, sections §108920-108923.

Chevrier, J., Harley, K.G., Bradman, A., Gharbi, M., Sjödin, A., Eskenazi, B., 2010. Polybrominated diphenyl ether (PBDE) flame retardants and thyroid hormone during pregnancy. Environmental health perspectives 118, 1444-1449.

Clifton, M.S., Wargo, J.P., Weathers, W.S., Colon, M., Bennett, D.H., Tulve, N.S., 2013. Quantitative analysis of organophosphate and pyrethroid insecticides, pyrethroid transformation products, polybrominated diphenyl ethers and bisphenol A in residential surface wipe samples. Journal of chromatography. A 1273, 1-11.

Community Care Licensing Division, 2010. In: California Department of Social Services, Accessed at http://www.ccld.ca.gov/docs/ccld_search/ccld_search.aspx on January 10th, 2010.

Dingemans, M.M., van den Berg, M., Westerink, R.H., 2011. Neurotoxicity of brominated flame retardants: (in)direct effects of parent and hydroxylated polybrominated diphenyl ethers on the (developing) nervous system. Environmental health perspectives 119, 900-907.

Dodson, R.E., Perovich, L.J., Covaci, A., Van den Eede, N., Ionas, A.C., Dirtu, A.C., Brody, J.G., Rudel, R.A., 2012. After the PBDE phase-out: a broad suite of flame retardants in repeat house dust samples from California. Environmental science & technology 46, 13056-13066.

Eskenazi, B., Chevrier, J., Rauch, S.A., Kogut, K., Harley, K.G., Johnson, C., Trujillo, C., Sjödin, A., Bradman, A., 2013. In utero and childhood polybrominated diphenyl ether (PBDE) exposures and neurodevelopment in the CHAMACOS study. Environmental health perspectives 121, 257-262.

Eskenazi, B., Fenster, L., Castorina, R., Marks, A.R., Sjödin, A., Rosas, L.G., Holland, N., Guerra, A.G., Lopez-Carillo, L., Bradman, A., 2011. A comparison of PBDE serum concentrations in Mexican and Mexican-American children living in California. Environmental health perspectives 119, 1442-1448.

Gascon, M., Vrijheid, M., Martinez, D., Forns, J., Grimalt, J.O., Torrent, M., Sunyer, J., 2011. Effects of pre and postnatal exposure to low levels of polybromodiphenyl ethers on neurodevelopment and thyroid hormone levels at 4 years of age. Environment international 37, 605-611.

Gold, M.D., Blum, A., Ames, B.N., 1978. Another flame retardant, tris-(1,3-dichloro-2-propyl)-phosphate, and its expected metabolites are mutagens. Science 200, 785-787.

Great Lakes Chemical Corporation, 2005. Great Lakes chemical corporation completed phase-out of two flame retardants. In PR Newswire; PR Newswire: Indianapolis, 2005.

Hamers, T., Kamstra, J.H., Sonneveld, E., Murk, A.J., Kester, M.H., Andersson, P.L., Legler, J., Brouwer, A., 2006. In vitro profiling of the endocrine-disrupting potency of brominated flame retardants. Toxicological sciences : an official journal of the Society of Toxicology 92, 157-173.

Herbstman, J.B., Sjödin, A., Kurzon, M., Lederman, S.A., Jones, R.S., Rauh, V., Needham, L.L., Tang, D., Niedzwiecki, M., Wang, R.Y., Perera, F., 2010. Prenatal exposure to PBDEs and neurodevelopment. Environmental health perspectives 118, 712-719.

Hornung, R., Reed, L., 1991. Estimation of average concentration in the presence of nondetectable values. Appl Occup Env Hyg 5, 46-51.

Johnson, P.I., Stapleton, H.M., Mukherjee, B., Hauser, R., Meeker, J.D., 2013. Associations between brominated flame retardants in house dust and hormone levels in men. The Science of the total environment 445-446, 177-184.

Johnson, P.I., Stapleton, H.M., Sjödin, A., Meeker, J.D., 2010. Relationships between polybrominated diphenyl ether concentrations in house dust and serum. Environmental science & technology 44, 5627-5632.

La Guardia, M.J., Hale, R.C., Harvey, E., 2006. Detailed polybrominated diphenyl ether (PBDE) congener composition of the widely used penta-, octa-, and deca-PBDE technical flame-retardant mixtures. Environmental science & technology 40, 6247-6254.

Legler, J., Brouwer, A., 2003. Are brominated flame retardants endocrine disruptors? Environment international 29, 879-885.

Lunder, S., Hovander, L., Athanassiadis, I., Bergman, A., 2010. Significantly higher polybrominated diphenyl ether levels in young U.S. children than in their mothers. Environmental science & technology 44, 5256-5262.

OEHHA, 2001. Office of Environmental Health Hazard Assessment (OEHHA). Proposition 65: Process for Developing Safe Harbor Numbers. California Environmental Protection Agency; 2001. http://oehha.ca.gov/prop65/policy_procedure/pdf_zip/SafeHarborProcess.pdf.

OEHHA, 2011. Office of Environmental Health Hazard Assessment (OEHHA). Evidence on the Carcinogenicity of Tris(1,3-Dichloro-2-Propyl) Phosphate. July 2011. http://oehha.ca.gov/prop65/hazard_ident/pdf_zip/TDCPP070811.pdf.

Patisaul, H.B., Roberts, S.C., Mabrey, N., McCaffrey, K.A., Gear, R.B., Braun, J., Belcher, S.M., Stapleton, H.M. 2013. Accumulation and endocrine disrupting effects of the flame retardant mixture Firemaster® 550 in rats: an exploratory assessment. Journal of biochemical and molecular toxicology 27, 124-36.

Quirós-Alcalá, L., Bradman, A., Nishioka, M., Harnly, M.E., Hubbard, A., McKone, T.E., Eskenazi, B., 2011. Concentrations and loadings of polybrominated diphenyl ethers in dust from low-income households in California. Environment international 37, 592-596.

Roberts, J.W., Budd, W.T., Ruby, M.G., Bond, A.E., Lewis, R.G., Wiener, R.W., Camann, D.E., 1991. Development and field testing of a high volume sampler for pesticides and toxics in dust. Journal of exposure analysis and environmental epidemiology 1, 143-155.

Roze, E., Meijer, L., Bakker, A., Van Braeckel, K.N., Sauer, P.J., Bos, A.F., 2009. Prenatal exposure to organohalogens, including brominated flame retardants, influences motor, cognitive, and behavioral performance at school age. Environmental health perspectives 117, 1953-1958.

Sjödin, A., Papke, O., McGahee, E., Focant, J.F., Jones, R.S., Pless-Mulloli, T., Toms, L.M., Herrmann, T., Muller, J., Needham, L.L., Patterson, D.G., Jr., 2008a. Concentration of polybrominated diphenyl ethers (PBDEs) in household dust from various countries. Chemosphere 73, S131-136.

Sjödin, A., Wong, L.Y., Jones, R.S., Park, A., Zhang, Y., Hodge, C., Dipietro, E., McClure, C., Turner, W., Needham, L.L., Patterson, D.G., Jr., 2008b. Serum concentrations of polybrominated diphenyl ethers (PBDEs) and polybrominated biphenyl (PBB) in the United States population: 2003-2004. Environmental science & technology 42, 1377-1384.

Stapleton, H.M., Klosterhaus, S., Eagle, S., Fuh, J., Meeker, J.D., Blum, A., Webster, T.F., 2009. Detection of organophosphate flame retardants in furniture foam and U.S. house dust. Environmental science & technology 43, 7490-7495.

Stapleton, H.M., Klosterhaus, S., Keller, A., Ferguson, P.L., van Bergen, S., Cooper, E., Webster, T.F., Blum, A., 2011. Identification of flame retardants in polyurethane foam collected from baby products. Environmental science & technology 45, 5323-5331.

Stapleton, H.M., Eagle, S., Sjödin, A., Webster, T.F., 2012a. Serum PBDEs in a North Carolina toddler cohort: associations with handwipes, house dust, and socioeconomic variables. Environmental health perspectives 120, 1049-1054.

Stapleton, H.M., Sharma, S., Getzinger, G., Ferguson, P.L., Gabriel, M., Webster, T.F., Blum, A., 2012b. Novel and high volume use flame retardants in US couches reflective of the 2005 PentaBDE phase out. Environmental science & technology 46, 13432-13439.

State of California Environmental Protection Agency, Office of Environmental Health Hazard Assessment. Safe Drinking Water and Toxic Enforcement Act of 1986. Chemicals Known to the

State to Cause Cancer or Reproductive Toxicity. 2011. http://www.oehha.ca.gov/prop65/prop65_list/files/P65single102811.pdf.

Toms, L.M., Sjödin, A., Harden, F., Hobson, P., Jones, R., Edenfield, E., Mueller, J.F., 2009. Serum polybrominated diphenyl ether (PBDE) levels are higher in children (2-5 years of age) than in infants and adults. Environmental health perspectives 117, 1461-1465.

Tulve, N.S., Jones, P.A., Nishioka, M.G., Fortmann, R.C., Croghan, C.W., Zhou, J.Y., Fraser, A., Cavel, C., Friedman, W., 2006. Pesticide measurements from the first national environmental health survey of child care centers using a multi-residue GC/MS analysis method. Environmental science & technology 40, 6269-6274.

Turyk, M.E., Persky, V.W., Imm, P., Knobeloch, L., Chatterton, R., Anderson, H.A., 2008. Hormone disruption by PBDEs in adult male sport fish consumers. Environmental health perspectives 116, 1635-1641.

U.S. EPA, 2008a. 2,2',4,4',5,5'-Hexabromodiphenyl Ether (BDE-153) in Support of Summary Information on the Integrated Risk Information System (IRIS). National Center for Environmental Assessment. Washington, DC.

U.S. EPA, 2008b. Toxicological Review of 2,2',4,4'-Tetrabromodiphenyl Ether (BDE-47) in Support of Summary Information on the Integrated Risk Information System (IRIS). National Center for Environmental Assessment: Washington, DC.

U.S. EPA, 2008c. Toxicological Review of 2,2',4,4',5-Pentabromodiphenyl Ether (BDE-99) in Support of Summary Information on the Integrated Risk Information System (IRIS). National Center for Environmental Assessment: Washington, DC.

U.S. EPA, 2008d. Toxicological Review of Decabromodiphenyl Ether (BDE-209) in Support of Summary Information on the Integrated Risk Information System (IRIS). EPA/635/R-07/008F. National Center for Environmental Assessment: Washington, DC.

US EPA, 2011. U.S. EPA. Exposure Factors Handbook 2011 Edition (Final). U.S. Environmental Protection Agency, Washington, DC, EPA/600/R-09/052F, 2011.

Webster, T.F., Harrad, S., Millette, J.R., Holbrook, R.D., Davis, J.M., Stapleton, H.M., Allen, J.G., McClean, M.D., Ibarra, C., Abdallah, M.A., Covaci, A., 2009. Identifying transfer mechanisms and sources of decabromodiphenyl ether (BDE 209) in indoor environments using environmental forensic microscopy. Environmental science & technology 43, 3067-3072.

Wu, N., McClean, M.D., Brown, P., Aschengrau, A., Webster, T.F., 2009. Participant experiences in a breastmilk biomonitoring study: a qualitative assessment. Environmental health : a global access science source 8, 4.

Zhou, T., Taylor, M.M., DeVito, M.J., Crofton, K.M., 2002. Developmental exposure to brominated diphenyl ethers results in thyroid hormone disruption. Toxicological sciences : an official journal of the Society of Toxicology 66, 105-116.

Zota, A.R., Rudel, R.A., Morello-Frosch, R.A., Brody, J.G., 2008. Elevated house dust and serum concentrations of PBDEs in California: unintended consequences of furniture flammability standards? Environmental science & technology 42, 8158-8164.

Table 1. Flame retardant levels in dust (ng/g) from ECE facilities (n=39).

Analyte	>MDL[a] (%)	Mean	SD	25^{th} %	Median	75^{th} %	95^{th} %	Max
BDE47	100	1,717.0	3,085.7	263.8	768.9	1,326.5	11,699	15,116
BDE99	100	2,351.0	4,637.4	393.5	1,031.1	1,627.3	13,230	25,522
BDE100	100	471.2	945.0	86.8	211.5	330.9	2,010.6	5,525.0
BDE118	76.9	25.0	24.3	10.2	24.2	26.8	108.3	121.9
BDE153	100	297.1	633.1	63.8	125.1	177.8	1,285.8	3,783.3
BDE154	100	229.0	498.7	49.7	94.1	167.8	914.4	3,031.6
BDE183	87.2	26.0	27.7	12.4	17.3	27.1	113.2	139.2
BDE190	2.6	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	16.5
BDE197	89.7	24.0	20.0	16.1	17.3	20.9	33.7	70.8
BDE203	20.5	16.9	22.5	<MDL	<MDL	<MDL	36.8	69.2
BDE205	0.0	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL
BDE206	66.7	101.4	176.5	<MDL	48.3	73.3	330.7	1,085.5
BDE207	100	79.5	86.1	37.5	46.7	84.1	282.1	481.1
BDE209	100	2,588.4	3,363.1	882.5	1,442.5	2,635.8	11,369	16,792
∑ BDE	100	7,956.6	10,671.0	2,227.0	4,225.0	9,637.7	32,598	55,328
EH-TBB	100	1,062.3	2,510.1	216.2	362.4	712.3	6,557.9	14,812
BEH-TEBP	100	431.1	1,191.9	80.6	132.9	327.6	1,299.3	7,489.7
TCEP	100	935.9	1,580.2	203.1	319.1	663.5	6,750.7	6,834.9
TDCIPP	100	6,189.4	12,710.5	1,458.3	2,265.0	5,803.1	36,927	70,931

[a]MDL: Method detection limit

Table 2. Summary of flame retardant indoor air concentrations (ng/m^3).

Analyte	N	>MDL[a] (%)	Mean	SD	25th %	Median	75th %	95th %	Max
BDE47	40	90.0	0.52	0.67	0.07	0.26	0.62	2.16	2.67
BDE99	40	95.0	0.19	0.21	0.06	0.12	0.24	0.67	0.93
BDE100	40	37.5	0.01	0.02	<MDL	<MDL	0.01	0.05	0.08
BDE153	40	20.0	0.33	1.24	<MDL	<MDL	<MDL	1.43	7.62
BDE154	40	5.0	<MDL	<MDL	<MDL	<MDL	<MDL	0.12	4.60
BDE209 [b]	7	100	1.63	1.31	0.97	1.39	1.65	4.46	4.46
EH-TBB	40	15.0	0.58	2.60	<MDL	<MDL	<MDL	2.29	16.23
BEH-TEBP	40	17.5	0.23	0.87	<MDL	<MDL	<MDL	0.99	5.39
TCEP	40	65.0	2.69	3.89	<MDL	0.91	3.05	12.94	15.34
TDCIPP	40	90.0	0.59	0.36	0.40	0.53	0.72	1.25	1.99

[a]MDL: Method detection limit; [b] BDE209 was only analyzed from the first seven ECE facilities sampled.