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Flame retardant exposures in California early childhood education environments

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ABSTRACT

Infants and young children spend as much as 50 h 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^{-1}) and TDCIPP (2265 ng g^{-1}) 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,b). 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

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.

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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 m 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^{-1}) and dust loading (i.e., ng m^{-2}) were determined. Detailed dust sampling, laboratory, and analytical QA/QC results are presented in Supplementary Material (SM), Tables S1–S4.

2.3. Flame retardant measurement in air

Indoor air samples were collected over 6–10 h 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 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 $MDL/\sqrt{2}$ (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 penta-BDE 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

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