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Polybrominated diphenyl ethers in residential dust: Sources of variability

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ABSTRACT

We characterized the sources of variability for polybrominated diphenyl ethers (PBDEs) in residential dust and provided guidance for investigators who plan to use residential dust to assess exposure to PBDEs. We collected repeat dust samples from 292 households in the Northern California Childhood Leukemia Study during two sampling rounds (from 2001 to 2007 and during 2010) using household vacuum cleaners and measured 22 PBDEs using high resolution gas chromatography–high resolution mass spectrometry. Median concentrations for individual PBDEs ranged from <0.1–2500 ng per g of dust. For each of eight representative PBDEs, we used a random-effects model to apportion total variance into regional variability (0–11%), intra-regional between-household variability (17–50%), within-household variability over time (38–74%), and within-sample variability (0–23%) and we used a mixed-effects model to identify determinants of PBDE levels. Regional differences in PBDE dust levels were associated with residential characteristics that differed by region, including the presence of furniture with exposed or crumbling foam and the recent installation of carpets in the residence. Intra-regional differences between households were associated with neighborhood urban density, racial and ethnic characteristics, and to a lesser extent, income. For some PBDEs, a decreasing time trend explained a modest fraction of the within-household variability; however, most of the within-household variability was unaccounted for by our mixed-effects models. Our findings indicate that it may be feasible to use residential dust for retrospective assessment of PBDE exposures in studies of children's health (e.g., the Northern California Childhood Leukemia Study).

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) have been used worldwide as chemical flame retardants to treat plastics and textiles in consumer products (U.S. Environmental Protection Agency, 2010). Three commercial PBDE mixtures, Penta-BDE (composed primarily of BDEs 99, 47, 100, 153, and 154), Octa-BDE (composed primarily of BDEs 183, 197, 207, 196, and 153), and Deca-BDE (composed primarily of BDE-209), have been manufactured (La Guardia et al., 2006). In the U.S., Penta- and Octa-BDEs are no longer used and Deca-BDE is being phased out (U.S. Environmental Protection Agency, 2012); however, consumer goods that have been treated with any of the three PBDE commercial mixtures can still be found in U.S. homes. Because PBDEs are not chemically bound to the polymers they treat, these additives can migrate into the environment. Indeed, PBDEs have been found in residential dust at high concentrations — with several studies of U.S. homes reporting median concentrations for major PBDE congeners of at least one part per million (Whitehead et al., 2011). Due to the State of California's unique flammability standards, dust samples from California homes have been reported to have exceptionally high levels of PBDEs (Dodson et al., 2012; Zota et al., 2008). Investigators have demonstrated that PBDE levels in paired samples of human serum and residential dust are significantly correlated, suggesting that dust ingestion is an important route of exposure to PBDEs in U.S. homes (Johnson et al., 2010; Stapleton et al., 2012a).

While many researchers have measured PBDEs in dust, most estimate human exposure to PBDEs using a single dust sample and only a few have sampled dust repeatedly in the same households and characterized the variability of dust measurements within households over time (Allen et al., 2008a; Batterman et al., 2009; Dodson et al., 2012; Harrad et al., 2008; Muenhor and Harrad, 2012; Vorkamp et al., 2011). The magnitude of temporal variability that exists in residential-dust measurements over years or decades has not been estimated and may be important for accurate assessments of long-term exposure. Moreover, previous investigations have not compared the

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magnitude of within-household temporal variability to the magnitude of between-household variability. When estimating the health effects related to a chemical exposure, it is this variance ratio that is related to the degree of exposure measurement error and predictive of the underestimation of risk estimates (Armstrong, 1998).

To characterize the long-term temporal variability of PBDE concentrations in residential dust, we analyzed 22 PBDEs, in dust samples collected in two rounds separated by 3–8 years. Because exposures to PBDEs have been associated with endocrine disruption (Chevrier et al., 2010; Meeker et al., 2009; Stapleton et al., 2011; Turyk et al., 2008), adverse birth outcomes (Chao et al., 2007; Harley et al., 2010, 2011), and adverse neurological development (Eskenazi et al., 2013; Herbstman et al., 2010); we also identified determinants of residential-dust PBDE levels and discuss strategies to limit human exposures to PBDEs. Finally, because Penta-BDE and Octa-BDE were banned for distribution in commerce on and after June 1, 2006 in California (Chan, 2004), we evaluate the long-term trends in residential-dust PBDE levels from 2001 to 2010.

2. Methods

2.1. Study population

Residential dust samples for our PBDE analysis were collected as part of the Northern California Childhood Leukemia Study, a case–control study conducted in the San Francisco Bay area and California Central Valley. Residential dust samples were originally collected from study homes as one strategy for identifying possible environmental risk factors for childhood leukemia and various persistent environmental contaminants including pesticides, polychlorinated biphenyls, and polycyclic aromatic hydrocarbons have been measured in the samples. Homes of children with leukemia and homes of healthy children were eligible for initial dust collection (from 2001 to 2007) if the children were 0–7 years-old at study enrollment. Subsequently, in 2010, a subset of the households that participated in the initial dust collection was eligible for repeated dust collection if the family was still living in the same home. Among 629 households who participated in the initial dust collection, 225 were eligible for a second dust collection and 203 households had two dust samples analyzed for PBDEs. For an additional 89 households who participated in the initial dust collection, but who were ineligible for repeated dust collection, we also analyzed their original dust sample for PBDEs. We obtained written informed consent from participating subjects in accordance with the institutional review boards' requirements at the University of California, Berkeley.

2.2. Collection of residential dust

We collected dust samples from subjects' household vacuum cleaners during two sampling rounds; from 2001 to 2007 and again during 2010. The median interval between repeated sample collections was 4.8 years (range of 2.6–8.6 years). During the first round of dust sampling, we collected vacuum cleaner dust and administered a questionnaire during an in-home visit. For the second round of dust sampling, we interviewed subjects via telephone and instructed them to mail the contents of their vacuum cleaners to the study center in prepaid parcels. We stored dust samples away from heat (4 °C or colder) and light prior to chemical analysis.

2.3. Laboratory analysis of PBDEs

The analytical protocol for PBDE analysis has been previously described (Whitehead, 2011). Briefly, we homogenized and fractionated the dust samples using a mechanical sieve shaker equipped with a 100-mesh sieve to obtain dust particles smaller than 150 μm. Portions of fine dust (0.2 g) were spiked with nine 13C-labeled internal standards, extracted via accelerated solvent extraction, purified by silica-gel column chromatography and gel permeation chromatography, concentrated to 250 μL, solvent exchanged into tetradecane, and spiked with two 13C-labeled recovery standards. Finally, we analyzed 22 PBDEs [BDEs 28, 32, 47, 66, 71, 99, 100, 153, 154, 155, 179, 183, 190, 196, 197, 201, 202, 203, 206, 207, 208, and 209] using isotope dilution/high resolution gas chromatography–high resolution mass spectrometry (HRGC–HRMS, DFS, Thermo-Finnigan, Bremen, Germany) equipped with a DB-5 column (15 m × 0.25 mm i.d., 0.1 μm film thickness, J&W Scientific, USA) and operated in electron impact ionization-selective ion monitoring (EI-SIM) mode (for a list of ions used to quantify PBDE concentrations refer to the Appendix, Table A1; for details regarding the thermal stability of BDE-209 during analysis refer to the Appendix, Table A2). Because our findings were similar for PBDE congeners within each homologue group, we present results for eight representative PBDEs, one from each homologue group from Tri- to Deca-BDE.

2.4. Household characteristics

Parents initially participated in structured in-home interviews designed to ascertain information relevant to childhood leukemia. Among other things, this questionnaire established general demographic information such as the annual household income, the educational attainment of the parents, and the ethnicity and race of the parents (mothers were categorized into three groups: Hispanic; non-Hispanic, White or Asian; and non-Hispanic, non-White, non-Asian, i.e., non-Hispanic mothers of other races). Subsequently, households participating in the repeated dust collection ($N = 203$) completed an additional telephonic questionnaire designed to ascertain information about sources of residential chemical exposures. The latter questionnaire covered topics related to sources of PBDEs (see Appendix, Table A3 for details), including the quantity and daily use of televisions and computers; the presence of upholstered furniture (quantity as well as quality, i.e., the presence of crumbling or exposed foam, and age, i.e., purchased before or after 2006); and the quantity and installation history (since move-in date) of carpeting. Moreover, this questionnaire obtained information about residential characteristics including, the square footage, construction date, construction material, and type of residence (e.g., single family home, apartment) as well as resident activities including, occupations, window and air conditioner use, and shoe removal habits. Finally, we gathered information about the characteristics of the vacuum cleaner including the type of vacuum and the frequency of its use.

We used a global positioning device to determine the latitude and longitude for each residence and classified each residence as belonging to one of six geographic regions as shown in Fig. 1. We linked each location to the corresponding U.S. Census block and identified each residence as urban, suburban, or rural based on the Census Bureau's delineations (U.S. Census Bureau, 2000).

2.5. Differences between duplicate, replicate, and repeat samples

We analyzed samples in batches of 12, with each batch consisting of 8 samples, 1 method blank, 1 duplicate sample pair (i.e., two 200-mg portions of fine dust taken from the same vacuum cleaner), and 1 inter-batch quality control sample (i.e., a 200-mg portion of fine dust taken from the quality control vacuum cleaner dust). Because we prepared and analyzed an inter-batch quality control replicate alongside each successive sample batch, the inter-batch quality control results illustrate the reproducibility of the dust preparation and analytical methods over the course of the study. Likewise, the duplicate samples illustrate the reproducibility of the dust preparation and analytical methods within each sample batch. For some batches, we replaced the inter-batch quality control sample with a National Institute of Standards and Technology Standard Reference Material 2585 (NIST SRM 2585) dust sample, which contained certified concentrations of eleven of the 22 PBDEs analyzed in this study. NIST has homogenized the SRM 2585 dust with a rigorous protocol, so results obtained from

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