Exposure to PBDEs from Hand to Mouth Contact: Measurements of PBDEs on Hand Wipes from Individuals in the United States

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Introduction

The contamination of the environment with Polybrominated Diphenyl Ethers (PBDEs) is now considered ubiquitous as these chemicals have been detected in increasing concentrations in a variety of locations and samples (de Wit et al. 2006; Hale et al. 2006; Law et al. 2006). The prevalence of these compounds is most likely due to their increased use as flame retardant chemicals in a variety of consumer products such as furniture, TVs and electrical and electronic products (W.H.O. 1994; de Wit 2002). On a global scale, levels of PBDEs in human serum appear to be 20 fold higher in the North American population versus the European population (Hites 2004; Sjodin et al. 2004; Schecter et al. 2005). The reason for this difference is unclear, but may be related to the heavy use of pentaBDE, a commercial PBDE mixture used in polyurethane foam, in the United States. In fact, until 2005, the U.S. consumed 98% of the world market demand for pentaBDE, primarily as foam in furniture products (Hale et al. 2002; BSEF 2006).

The primary route of exposure to PBDEs among the general U.S. population is ambiguous at present but is most likely linked to dietary intake and/or indoor exposure (i.e. dust exposure or inhalation in the home). Recent studies have examined PBDE levels in market basket surveys (Huwe et al. 2002; Schecter et al. 2004; Schecter et al. 2006) and the results suggest that dietary intake may not be the dominating route of human exposure to PBDEs (McDonald 2005). Other studies have examined levels of PBDEs in indoor air and house dust (Wilford et al. 2004; Stapleton et al. 2005; Wilford 2005; Wu 2005) which suggests indoor exposure to dust may result in higher exposure than dietary exposure. However, it has proven quite difficult to determine the relative amount of exposure adults are receiving from indoor dust.

The present study was designed to further explore PBDE exposure routes in people by examining the adsorption of these chemicals to the hands from everyday routine behavior. Because PBDEs are added to numerous indoor products (e.g. TVs, computers, furniture, etc.), direct contact may lead to PBDE adsorption to the skin due to their lipophilic nature. Adsorption of these chemicals to the hands may lead to inadvertent ingestion from transfer to food that is eaten by the hands (e.g. French fries, sandwiches, etc.). In addition, hygiene (hand washing) and behavior (e.g. nail biting, cigarette smoking, finger licking) may influence the transfer of these chemicals from the hands to the mouth. To date, no studies have examined the levels of PBDE adsorbed to dermal surfaces. Results from this investigation will be useful for estimating potential exposure to PBDEs from skin adsorption and help elucidate additional exposure pathways.

Materials and Methods

Two different sampling campaigns have been conducted to date. In the first sampling study wipe samples from both forearms and hands were sampled from six individuals (5 adults and one 9 year old boy). Forearms and hands were sampled in the first phase to increase detection limits for the PBDE chemicals as it was believed the mass on just hands might be below detection limits. Since the results of this first sampling study revealed sufficient levels to measure PBDEs, the second sampling was adjusted to focus strictly on levels of PBDEs adsorbed to hands. During the second sampling event, 10 adults volunteered to have both hands wiped and measured for PBDEs. In agreeing to participate, all individuals signed a consent form and filled out a short questionnaire with information on age, gender, race, height, weight, residence, and occupation. Also included in the questionnaire were questions on the number of electronic appliances and foam containing furniture pieces in their residence and workplace.

North brand sterile gauze pads (3" x 3") soaked in isopropyl alcohol were used to collect the surface oils and associated PBDEs from the skin. Each individual was asked to collect their own sample and use one gauze pad per hand. Two aluminum foil dishes containing exactly 3.0 mL of isopropyl alcohol were prepared for each individual. Each person was instructed to open their sterile gauze pad, soak the pad in the isopropyl alcohol to collect the entire 3.0 mL of the alcohol and use the pad to wipe the entire surface area of their skin on each hand (or from elbow to fingertips on one arm for individuals in the first phase of this study having their forearms wiped). Immediately after wiping down the skin, the gauze pad was placed into a pre-cleaned glass 50 mL centrifuge tube and sealed. A second sterile gauze pad was then used in the same fashion to wipe the remaining hand (and forearm for participants in the first phase). Therefore, each sample from each individual contained two gauze pads that had been used to collect surface oils and PBDEs from the surface area of the skin.

Laboratory and field blanks were prepared by soaking gauze pads in 3.0 mL of isopropyl alcohol and using clean forceps to transfer the gauze from the package into the centrifuge tubes. All samples and blanks were spiked with two internal standards prior to extraction. Forty mL of dichloromethane was added to each centrifuge tube and then sonicated for 20 minutes. The dichloromethane containing the oils and PBDEs were decanted into a separate flask. This process was repeated two more times and the decanted solvent extracts were combined. The extract was reduced in volume using a rapid evaporation system and then filtered through a 0.45 µm PTFE filter to remove any remaining suspended cotton fibers. Total lipids in the extract were determined using gravimetric analysis of a sub-sample. The extracts were then injected into an HPLC to remove lipids using gel permeation chromatography. The extract was again reduced in volume using the rapid evaporation system, transferred to hexane and reduced in volume to 0.5 mL. A recovery standard was added and samples were analyzed using gas chromatography mass spectrometry operated in negative chemical ionization mode (GC/ECNI-MS), similar to analysis of dust samples (Stapleton et al. 2005).

Results and Discussion

In the first sampling event, levels of PBDEs measured on the forearms and hands of the six individuals tested ranged from 7 to 540 ng ∑PBDEs. Using gender, height, and weight, surface area estimates for hands and forearms were calculated for each participant from the EPA Exposure Factors Handbook

(1997). Surface area estimates for the one 9 year old participant were calculated from the EPA's Child-Specific Exposure Factors Handbook. The concentration of PBDEs adsorbed to the skin was calculated on a surface area basis (ng/cm^2). The concentrations ranged from 0.003 to 0.4 ng/cm^2 and the highest concentration was observed in the 9 year old boy (See Figure 1) due to the reduced surface area in a smaller individual.

During the second sampling event, PBDEs were measured on wipes collected from hands of 10 adults. Levels of total PBDEs measured on just the two hands of the 10 individuals ranged from 3 to 1980 ng, and normalized to surface area, from 0.003 to 2 ng/cm^2 (See Figure 2). A comparison of the values measured between the first and second sampling events reveals no significant difference ($p<0.05$), suggesting the majority of PBDEs adsorbed to the skin surface area are present on the hands directly. Additionally, the range in total mass measured from the hand wipes ranges almost 4 orders of magnitude.

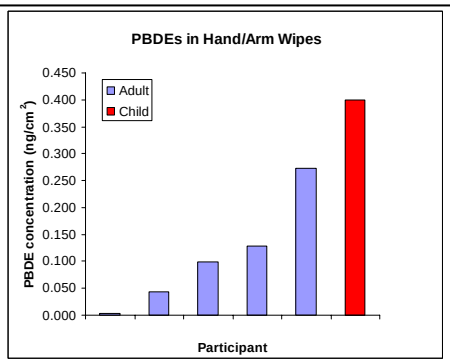

Figure 1. Surface area normalized concentrations of PBDEs adsorbed to hands and forearms of five adults and one 9 year old boy.

The mass of lipid measured gravimetrically from the hand wipe samples ranged from 5 to 30 mg. Normalization to lipid did reduce some of the variability among individual ∑PBDE levels. Analysis of the PBDE congener patterns was similar to PBDE congener patterns observed in dust. The contribution of BDE 209 to the ∑PBDE ranged from 0 to 94% among the ten individuals who had their hands wiped and ranged from <DL to 54 ng of BDE 209. BDE 99 and BDE 47 were generally found in greatest abundance and contributed 32 ± 5% and 29 ± 4% of the total PBDE mass. In some individuals tested, the BDE congener composition was very similar to the composition observed in the pentaBDE commercial mixture.

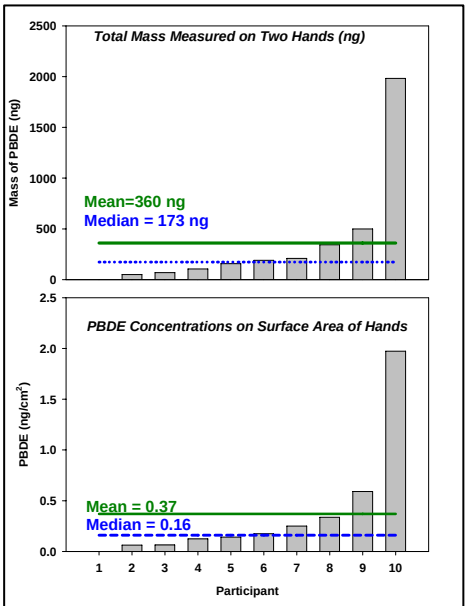

Figure 2. Total mass and concentration of PBDEs measured from wipe samples collected from the hands alone of 10 individuals.

Previous studies examining PBDEs in human milk and serum have found that approximately 5 to 10 % of the population possesses elevated levels of PBDEs that are an order of magnitude greater than the general median values (Schecter et al. 2003; Schecter et al. 2005). For example, analysis of 47 human breast milk samples by Schecter et al., 2003, revealed that the median concentration of ∑PBDEs was 34 ng/g lipid; however, 8 individuals had levels ranging from 103 to 418 ng/g lipid. The reason for the elevated levels in some individuals is currently unknown. Dietary estimates for adults in the United States range from 1 to 1.4 ng/kg/day (Schecter et al. 2006). From the results presented here, it seems likely that hand to mouth transfer may provide additional exposure at rates comparable to dietary sources. These routes of exposure may include transfer to finger food, particularly greasy fast food such as French fries, pizza,

burgers and sandwiches. Additionally, further exposure may be facilitated by individual behavior that would increase hand to mouth transfer such as nail biting, cigarette smoking and finger licking. Absorption through the skin surface may also be of importance. In a dermal exposure study using mice, Staskal et al. (Staskal et al. 2005) observed significant absorption of BDE 47 that was comparable to the absorption rate from dietary exposure. Therefore, adsorption of PBDEs to the hands may lead to accumulation from both hand to mouth contact and absorption through the skin.

This study demonstrates the importance of understanding exposure routes from adsorption of PBDEs to the hands. Given the variety of products known to contain PBDE chemicals that people come into contact with on a daily basis (e.g. furniture, remote controls, cell phones, etc.), it is probable that this contact leads to adsorption to skin oils. Further studies are needed to assess regional and temporal differences and to compare levels in children vs. adults. As part of a continuation of this study, the authors hope to recruit more children and monitor the same individuals over time.

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