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Personal Exposure to Polybrominated Diphenyl Ethers (PBDEs) in Residential Indoor Air

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We used personal air samplers to measure indoor air exposure to polybrominated diphenyl ethers (PBDEs) for 20 residents of the Greater Boston Area (Massachusetts). Area air measures were simultaneously collected from two rooms in each participant's home. Total personal air concentrations (particulate + vapor) were 469 pg/m^3 for non-209 BDEs and 174 pg/m^3 for BDE 209, significantly higher than bedroom and main living room concentrations ($p = 0.01$). The ratio of personal air to room air increased from 1 for vapor-phase congeners to 4 for fully particulate-bound congeners, indicating a personal cloud effect. Bedroom and main living area air samples were moderately correlated for non-209 BDEs ($r = 0.45$, $p = 0.045$) and BDE 209 ($r = 0.58$, $p = 0.008$). Use of personal air concentrations increased estimates of inhalation exposure over those previously reported. Inhalation may account for up to 22% of the total BDE 209 exposure in U.S. adults.

Introduction

Polybrominated diphenyl ethers (PBDEs) are a class of compounds commonly used as fire retardants in consumer products. PBDEs are manufactured in three commercial products: Penta-, Octa- and Deca-bromo diphenyl ether. The Penta commercial product is generally used in products containing polyurethane foam and is composed mainly of tetra- to hexa-brominated congeners, dominated by BDE 47 (∼40%) and BDE 99 (∼46%) (*1*). The Octa and Deca commercial products are mainly used in electronics, with the Octa product composed primarily of hepta- to nona-brominated congeners and the Deca product comprised primarily of the fully brominated congener, BDE 209 (*1*).

PBDEs are persistent in the environment, highly hydrophobic, and bioaccumulate in biota and humans. Previous studies documented PBDEs in diverse biota and environmental media (*2*). In humans, PBDE body burdens in the general population increased for several decades and vary geographically; one influential study showed that body burdens in Sweden increased exponentially over a 25 year period (*3*). The highest PBDE concentrations in humans and the environment have been reported in the United States (*2*).

While animal studies indicate that PBDEs have endocrine disrupting and developmental neurotoxic effects (*4*−*10*), the effects on human health remain unknown. The European Union banned the Penta and Octa products; several U.S. States have implemented similar bans, including one banning Deca. However, vast reservoirs of PBDEs remain in existing consumer products, potentially contributing to environmental and human burdens of PBDEs for decades (*11*).

Given the widespread use of PBDEs in consumer products and the large percentage of time Americans spend in their homes (approximately 69% on average; *12*), there is considerable potential for exposure to PBDEs in the indoor residential environment. We previously showed that penta-like PBDEs in dust collected from the homes of first-time mothers were associated with PBDE concentrations in breast milk (*13*). Exposure to PBDEs in dust may occur via ingestion or dermal absorption. Alternatively, PBDEs in dust may be acting as a surrogate for PBDEs in indoor air.

Existing estimates of inhalation exposure to PBDEs are limited and may not be representative of personal exposure to PBDEs in the United States. Previous air measurements (1) relied on passive sampling methods that undersample particulates (*14*−*18*), (2) collected area samples (*14*−*21*) that underestimate exposure compared with personal air sampling (*22*−*26*), (3) were conducted in countries other than the United States (*14*−*21*), and/or (4) did not report air concentrations for BDE 209 in homes (*15*−*19*, *21*). The primary objectives of this study were to address these limitations by measuring personal exposure to PBDEs in residential indoor air.

Materials and Methods

Study Design. Indoor air samples were collected from 20 urban residences in the Greater Boston area (Massachusetts) from January to March 2006. The field investigation included both single- and multi-family residences and was conducted during the winter months to minimize variation due to ventilation. The study population was selected based on willingness to participate and therefore does not represent a random sample of area residents. Informed consent was obtained from all participants and the study was approved by the Institutional Review Board of Boston University Medical Center.

Air Sample Collection. At each residence, one personal air sample and two area samples (bedroom, main living area) were collected simultaneously during a one-week period using identical air sampling equipment and media. Air sampling pumps (Casella Apex) were connected downstream of sampling media via $^1/_4$ in. Tygon tubing and calibrated to a flow rate of 2 L/min. Flow rates were monitored at the beginning and end of the 7-day sampling period, exhibiting negligible change in flow (<0.2%) during the study period (average total volume sampled = 9.1 m^3). The sampling media consisted of a glass fiber filter (GFF) followed by a polyurethane foam (PUF) plug (76 mm). The GFF had a nominal pore size of 0.06 μm and was used to capture particulate-bound PBDEs while the PUF plug was used to capture vapor-phase PBDEs.

The bedroom and main living area were chosen as the rooms most likely to contain PBDE sources. The main living area was defined as the room other than the bedroom where the participant spent the most time, typically a living room or family room. Area air sampling media were mounted on tripods approximately 1.2 m (4 ft) from the ground. The third

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pump was worn by the participant in a hip-pouch and the sampling media was clipped to the shirt collar, within 30 cm of the breathing zone. Participants were asked to turn on all pumps in the evening when they came home from work and turn them off in the morning. All pumps were stopped if the participant left their residence at any time in the evening and turned on upon their return. At night, the personal pumps were placed near the bed with the sampling media as close to the participant's breathing zone as possible. Room temperature and humidity were recorded at the start and end of the study period.

Prior to sampling, the GFFs were baked at 450 °C for 4 h and the PUFs were Soxhlet extracted to remove any residual PBDEs prior to sampling. After sampling, media were wrapped in clean foil to prevent ultraviolet exposure, sealed in pre-cleaned glass jars, and placed on ice. All sampling equipment was cleaned before re-deployment. Air sampling included 8 field blank samples (13% of total) and 7 duplicate area samples (12% of total). Duplicate personal samples were not collected to minimize burden on participants.

Laboratory Analysis. The GFFs and PUFs were extracted using an automatic pressurized fluid extractor. Air sampling units were disassembled and individual GFFs and PUFs were placed into pre-cleaned 34 mL stainless steel extraction cells. Each GFF and PUF was spiked with two quantification standards, 4′-fluoro-2,3,3′,4,5,6-hexabromodiphenyl ether (FBDE 160) and ^{13}C labeled BDE 209. The void volume of all cells was filled with hydromatrix (Dionex, Sunnyvale, CA). Cells containing PUFs were extracted with HPLC-grade petroleum ether; cells containing GFFs were extracted using HPLC-grade dichloromethane. Laboratory blanks, consisting of hydromatrix filled cells, were extracted alongside the air samples. Samples were extracted by heating and pressurizing the cells to 100 °C and 1500 psi for 5 min with the appropriate solvent. Each sample was extracted over three cycles and collected into amber collection vials. Each extract was reduced in volume to 200 μL using a gentle stream of purified nitrogen gas, and then filtered through glass wool plugged disposable pipettes.

Extracts were analyzed for PBDEs using a gas chromatograph (GC, Agilent 5890) coupled to a mass spectrometer (Agilent 5975) operated in electron capture negative ionization (GC/ECNI-MS) mode. A 0.25 mm (i.d.) × 15 m fused silica capillary column coated with a 5% phenyl methylpolysiloxane (0.25 μm film thickness) was used for the separation of PBDE congeners. On-column injection was employed in the GC, and the injection port was set to track the oven temperature. The oven temperature program was held at 80 °C for 2 min followed by a temperature ramp of 12 °C/min to 140 °C, followed by a temperature ramp of 5 °C/min to a final temperature of 280 °C, which was held for an additional 20 min. The transfer line temperature was maintained at 280 °C and the ion source was held at 200 °C. A suite of 27 individual BDE congeners was measured in all samples. Tri- through octa-BDE congeners, and FBDE 160, were quantified by monitoring bromide ions (m/z 79 and 81). All three nona-BDE congeners and BDE 209 were quantified by monitoring molecular ion fragments (m/z 484.6 and 486.6), while ^{13}C BDE209 was monitored through m/z 494.6 and 496.6. The relative standard deviation (RSD) was 5.97%, calculated from the duplicate samples.

Questionnaire. Home characteristics were recorded via questionnaire and during a walk-through visit conducted by the research team. Surface area, volume, and carpet floor coverage were measured. A researcher-administered questionnaire was used to collect information about residence (e.g., age of dwelling, dwelling type, area, heating, window use), household cleaning habits (e.g., frequency, equipment used), and a detailed inventory of electronics and furniture in the bedroom and main living area (e.g., number of electronics, patterns of usage, age of products).

Data Analysis. PBDE concentrations were blank-corrected by congener and matrix (GFF, PUF) using the mean of the field blanks. Limits of detection (LOD) were calculated by congener as three times the standard deviation of the field blanks. Values below the LOD were assigned a value of $^1/_2$ LOD. Since BDE 209 levels in field blanks varied by extraction batch, blank correction and LOD calculations for BDE 209 were conducted separately for each extraction batch.

Statistical analyses for air data were performed using SAS statistical software package, version 9.1. Analyses included univariate descriptive statistics, Spearman and Pearson correlations, scatterplots, simple linear regression, paired t-tests, and exploratory factor analysis. As congener-specific analyses indicated that the data were log-normally distributed, statistical analyses were performed on the natural log-transformed data with significance defined at the $\alpha = 0.05$ level.

Exploratory factor analysis was performed using congeners as the variables, with communality estimates set to the squared multiple correlation between the variables (SAS Proc Factor, Priors = SMC). The number of factors retained was determined by accepting all factors above the average eigenvalue and by examining scree plots. Rotated factor patterns were calculated using orthogonal rotations (rotate = varimax) as the factors were found to be uncorrelated when using oblique rotations (rotate = promax).

To examine relationships of congeners based on their physical chemistry, we estimated the fraction on particulate (ϕ) using a theoretical model (*21*) based on log K_{oa} (see Supporting Information). Although ϕ could also be estimated from our experimental data (comparing GFF to PUF), it may not reflect environmental conditions due to imperfect fitting of the GFF into the sampling tube or blow-off of PBDEs from particles on the filter.

Results and Discussion

Table 1 presents summary statistics for PBDEs in total air (GFF plus PUF) for all detected congeners, excluding BDE 71 which was only detected in 4 of 60 samples. Major congeners associated with the Octa commercial product (e.g., BDE 183) were not detected. The ΣBDE measure includes BDE 209, a congener that has not been widely reported in residential indoor air. To facilitate comparison to earlier studies, we include a summary measure for non-209 BDEs. The geometric mean (GM) concentration in personal air was 766 pg/m^3 for ΣBDEs and 469 pg/m^3 for non-209 BDEs. The dominant congeners were, by percent of total, BDEs 47, 209, and 99.

Personal Exposure. Geometric mean ΣBDE concentrations were significantly higher in personal air compared to bedroom (66% higher, $p = 0.001$) and main living area air (69% higher, $p = 0.002$). To examine these differences on a congener-specific basis, paired t-tests were performed between personal air and room air (Table 1). Statistically significant differences between personal air and room air were not found for lower brominated congeners; however, as the degree of bromination increased, the magnitude and significance of the difference between personal air and room air increased.

Personal inhalation exposure can be affected by a personal cloud that is influenced by individual activity patterns, including resuspension of dust (*27*). To further examine the personal cloud for PBDEs, we used the estimated particle−gas partition coefficient (see Supporting Information). For congeners detected in both personal and room air, Figure 1 graphs the ratio of personal air to room air versus the theoretical fraction on particulate (ϕ) for each congener. As ϕ increases, the ratio of personal to room air increases

TABLE 1. Summary Statistics for PBDE Concentrations in Personal and Room Air[a]

	personal air ($n = 20$)			bedroom ($n = 20$)				main living area ($n = 20$)			
congener	% detect	GM (GSD) (pg/m^3)	range (pg/m^3)	% detect	GM (GSD) (pg/m^3)	range (pg/m^3)	*p*-value[b]	% detect	GM (GSD) (pg/m^3)	range (pg/m^3)	*p*-value[c]
BDE 17	80	7.6 (2.3)	<4−58.1	75	8.1 (2.9)	<3.4−46.7	0.83	75	7 (3)	<4.0−81.5	0.79
BDE 28/33	100	29.6 (1.7)	<23.6−98.4	90	27.3 (2.3)	<11.4−102.3	0.51	90	25.4 (2.3)	<11.2−166.2	0.28
BDE 47	100	226.8 (2.3)	<147.6−1393	95	157.9 (2.7)	<62.4−784.3	0.02	90	145.1 (2.6)	<61.8−2371.4	0.02
BDE 49	75	9.1 (3.2)	<3.4−59.5	65	6 (2.9)	<3.4−35.7	0.05	75	7.2 (3.1)	<3.4−88.3	0.39
BDE 66	45	3.7 (2.1)	<3.4−18.9	45	3.5 (2.1)	<2.8−15.9	0.67	60	3.5 (2.3)	<3.4−59.6	0.57
BDE 85/155	35	3.8 (2.6)	<3.4−39.5	15	2.7 (1.7)	<3.4−16.7	0.01	40	2.5 (1.5)	<3.4−9.4	0.05
BDE 99	85	110.8 (2.8)	<41.6−879.4	90	66.9 (2.2)	<56.8−385.7	0.00	80	60.3 (2.1)	<49.2−552.6	0.00
BDE 100	85	22.2 (2.8)	<8.8−177.2	80	14.4 (2.4)	<8.8−101.2	0.01	75	12 (2.3)	<10.4−156.3	0.01
BDE 153	75	8.6 (3.2)	<3.2−73.7	60	4 (2.3)	<3.2−26.7	0.00	60	3.5 (1.8)	<3.2−10.9	0.00
BDE 154	75	9.1 (3.1)	<4.2−78.9	55	6.1 (3.1)	<4.2−138.7	0.03	60	5.2 (2.5)	<4.2−60.5	0.01
BDE 209	45	173.6 (3.2)	<52.4−1635.6	30	94.8 (2.0)	<51.0−268.6	0.01	45	94.2 (2.4)	<47.8−651.2	0.03
Σnon-209 BDEs[d]		469.1 (2.2)	151.2−2479.6		324.7 (2.3)	92.9−1342.6	0.01		288.6 (2.3)	82.1−3512.2	0.01
ΣBDE		765.7 (2.2)	230.2−2684.4		460.4 (2.0)	174.5−1532.6	0.00		452.8 (1.9)	224.1−3538.4	0.00

[a] Sum of PUF (vapor phase) and filter (particulate phase). GM = geometric mean. GSD = geometric standard deviation. Several congeners were not detected in any of 60 air samples (BDEs 25, 30, 75, 116, 119, 138, 156, 181, 183, 190, 191, 203, 205, 206). BDE 71 was only detected in 4 of 60 air samples. [b] Difference between GM of personal and GM of bedroom air using paired *t*-test. [c] Difference between GM of personal and GM of main living area air using paired *t*-test. [d] Sum of non-209 congeners.

FIGURE 1. Average ratio of personal to room PBDE air concentrations and theoretical fraction of PBDE on particulate (ϕ), by congener.

(Spearman correlation $r = 0.70$, $p < 0.001$). For BDE 17 and 28, lower brominated congeners that are mostly in the vapor phase at room temperature, there is no difference between personal air and room air (average personal/room ratio = 1). For BDE 99 and 153, congeners that are more likely to be found in the particulate phase, the ratio increases to 2. For BDE 209, almost entirely in particulate phase ($\phi > 0.9999$), the concentration in personal air is on average 4 times higher than area air. The ratio of 1 for lower-brominated congeners suggests that personal activities are not increasing volatilization or that the congeners are more evenly distributed in the indoor environment. Our findings of increased differences between personal air and room air as the degree of bromination increases are consistent with a personal cloud, as degree of bromination corresponds with a greater likelihood of partitioning to particulate, and resuspension by human activity.

The personal cloud effect we observed is consistent with previous studies (*28*). Ferro et al. (*29*) found that specific household activities (e.g., folding blankets, walking, dry dusting) increased personal exposure to particulate matter 1.4−1.6 times compared to a stationary area monitor. They also observed a trend of increased ratios of personal to room air with increased particle size, and attributed this to resuspension of dust. Similarly, elemental analysis of air sampling filters in an indoor air study estimated that bromine concentrations in personal air samples were 90% higher than area air samples (*23*).

Within-Home Variability. Personal air was strongly correlated with bedroom air for non-209 BDEs ($r = 0.80$, $p < 0.0001$) and BDE 209 ($r = 0.67$, $p = 0.001$), and moderately correlated with main living area air ($r = 0.59$, $p = 0.006$; $r = 0.37$, $p = 0.10$, respectively) (See Figure S4, Supporting Information). The strong correlations between personal air and bedroom air were expected due to the large fraction of sampling time that participants spent in the bedroom.

There were no statistically significant differences between the bedroom and main living area for either non-209 BDEs or BDE 209 (paired *t*-test, $p = 0.56$, $p = 0.97$, respectively). However, room air samples were only moderately correlated for non-209 BDEs ($r = 0.45$ $p = 0.045$) and BDE 209 ($r = 0.58$, $p = 0.008$) (Figure 2). These results suggest that PBDE levels in indoor air may vary across rooms within a residence, possibly due to different PBDE sources in each microenvironment.

Congener Relationships. We used factor analysis to evaluate the relationships between PBDE congeners. PBDEs, a class of related compounds with both common characteristics (sources) and variable characteristics (physical chemistry), are ideally suited for this type of analysis. The

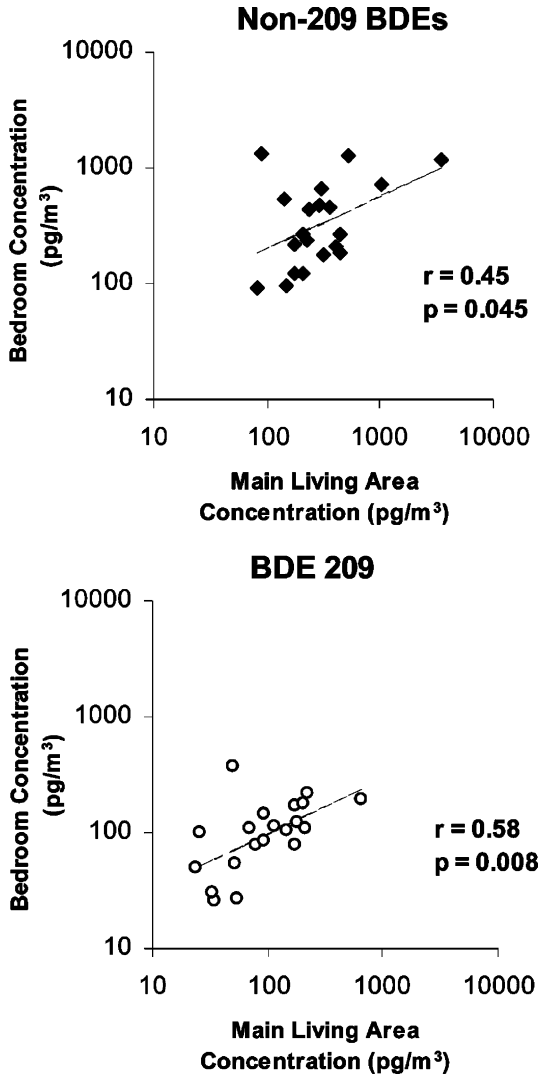

FIGURE 2. Pearson correlations of air concentrations (pg/m³) by sample location for non-209 BDEs and BDE 209.

factor analysis for personal air yielded three distinct factors, explaining 92% of the total variability in the original variables. Factor 1 loadings were dominated by congeners associated with the Penta product. All congeners except BDE 209 and BDE 17 had significant Factor 1 loadings (0.49−0.98), explaining 67% of the variability. Factor 2, accounting for 13% of the overall variability, was dominated by the exclusive loading of BDE 209 (0.88) and represented the Deca product.

Factor 3, accounting for the remaining 12% of explained variability, yielded rotated factor loadings related to the physical chemistry of the congeners. Figure 3 plots the rotated factor loadings for Factor 3 against the theoretical percent of chemical on particulate (ϕ, described earlier). A strong linear relationship is present; congeners with lower ϕ values are more heavily loaded onto Factor 3, while congeners expected to be found mostly on particulate have low, and even negative, factor scores. This indicates that in addition to their common sources, the physical chemistry of a congener (specifically the likelihood of being particulate-bound) partially determines the extent to which congeners are observed together in personal air samples. The results of the factor analysis support the separation of congeners into two groups: non-209 BDEs and BDE 209.

Household Characteristics. With one exception (*19*), previous studies have not reported finding any statistically significant associations between PBDE concentrations in air

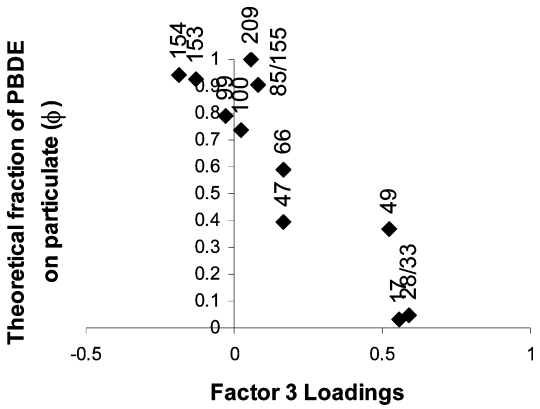

FIGURE 3. Associations between Factor 3 loadings from factor analysis and theoretical fraction on particulate (ϕ) by congener for personal air.

and characteristics of buildings or their contents (*14*−*18*, *20*, *21*). We looked for associations between furniture, electronics, and air concentration on a congener-specific and congener-group basis (non-209 BDEs and BDE 209). We plotted area air concentrations for each room against counts of furniture and electronics for that room, but observed no significant associations. We created dichotomized variables (high/low groups based on median) using both raw furniture counts, and a weighted furniture count where couches were assigned twice the value of chairs. For both dichotomized variables, the geometric mean main living area air concentration for non-209 BDEs was greater for participants in the higher furniture count groups (351 [3] v. 238 pg/m³; 397 v. 252 pg/m³, respectively), but the results were not statistically significant. Counts of electronics were analyzed in a similar manner using BDE 209 concentrations as the outcome variable, with no statistically significant or suggestive findings. We found no other significant associations between PBDE concentrations in indoor air and home characteristics (ie. dwelling type, heating, cleaning habits).

Our inability to detect statistically significant associations with room contents may in part be due to non-differential exposure misclassification (e.g., large differences in PBDE content between otherwise similar objects), producing a bias toward the null. Future studies attempting to examine the relationship between indoor PBDE levels and consumer products should consider methods that identify products containing PBDEs.

Significance of Inhalation Exposure. The geometric mean personal air concentrations of non-209 BDEs (469 pg/m³) were similar to residential indoor air concentrations measured in Toronto (median = 436 pg/m³, $n = 4$; BDE 17, 28/33, 47, 85, 99, 100, 153, 154, 183) using high-volume sampling (*21*), and higher than those measured in Ottawa (median = 100 pg/m³, $n = 74$; BDE 17, 28, 47, 66, 71, 85, 99, 100, 153, 154) using passive air sampling (*18*). Our results were also higher than concentrations (median = 129 pg/m³, $n = 7$; BDE 47, 99, 100, 153, 154) measured in Birmingham (UK) using high-volume samplers (*19*). Not surprisingly, the concentrations we measured in personal air in homes were considerably lower than those measured in occupational studies of electronics recyclers (*30*, *31*).

To evaluate the potential significance of personal exposure to PBDEs in indoor air, we estimated relative exposure contributions of air, food, and dust ingestion for adults and children using USEPA (*32*) exposure factors and median environmental concentration values (Table 2). Because of the large uncertainty for estimates of incidental dust ingestion, we used both mean and high dust ingestion rates (*32*, *33*). We assumed our median personal air concentrations;

TABLE 2. Estimated Relative Contributions of Three Exposure Routes to Percent Daily Intake of Non-209 BDEs and BDE 209

	adult[a]				child[a]			
	Non-209 BDEs		BDE 209		Non-209 BDEs		BDE 209	
	intake (ng/day)	% of intake	intake (ng/day)	% of intake	intake (ng/day)	% of intake	intake (ng/day)	% of intake
mean dust ingestion								
air[b]	5.6	11.0%	3.5	22.4%	3.2	2.0%	2.0	2.9%
food[c]	33.3	65.3%	6.5	41.7%	24.9	15.7%	6.0	8.7%
dust[d] (mean)	12.1	23.7%	5.6	35.9%	130.5	82.3%	60.8	88.4%
	51.0	100.0%	15.6	100.0%	158.6	100.0%	68.8	100.0%
high dust ingestion								
air	5.6	1.7%	3.5	2.4%	3.2	0.5%	2.0	0.7%
food	33.3	10.1%	6.5	4.5%	24.9	4.1%	6.0	2.2%
dust (high)	290.0	88.2%	135.0	93.1%	580.0	95.4%	270.0	97.1%
	328.9	100.0%	145.0	100.0%	608.1	100.0%	278.0	100.0%

[a] Inhalation, food, and dust ingestion rates are from EPA Exposure Factors Handbook (*32*). Adult age was considered 20+ years. Child age was considered 1−5 years. For child fish consumption, EPA only reports intake estimates for ages 0−9 years. All other estimates based on 1−5 year definition. Adult weight was estimated as 65 kg and child weight was estimated as 16 kg. [b] Air concentrations from personal air data in current study. Intake estimates are based on inhalation rates of 13.25 m^3/day for adults, and 7.55 m^3/day for children, and assuming 90% of time spent indoors. [c] Food concentration data from Schecter et al. (*34*). Intake estimates based on median values of non-BDE209 and BDE209, by meat, dairy, and fish consumption. Fish intake estimates for children are for ages 0−9 years. [d] Dust concentration data from Stapleton et al. (*35*). Mean dust ingestion estimated as 4.16 mg/day for adults and 45 mg/day for children. High dust ingestion estimated as 100 mg/day for adults and 200 mg/day for children (*32, 33*).

the median concentrations in food (meat, dairy, and fish) and dust were drawn from previous U.S. studies (*34*, *35*).

For children, inhalation exposure accounts for <3% of total exposure regardless of the assumed incidental dust ingestion rate. For adults, the relative contributions of exposure route are particularly sensitive to uncertain rates of dust ingestion. For high estimates of incidental dust ingestion, inhalation exposure is minor (1.7% for non-209 BDEs and 2.4% for BDE 209). For mean estimates of incidental dust ingestion, inhalation accounts for a substantial fraction of total exposure: 11% of non-209 BDE and 22% of BDE 209. Use of low dust ingestion rates would further increase the importance of inhalation.

Several studies have estimated the relative importance of inhalation exposure to non-209 BDEs (*19*, *33*, *36*, *37*), finding that inhalation accounts for 3−7% of total inhalation exposure for adults, on average. When we used our personal air concentrations for non-209 BDEs, we estimated the inhalation contribution for adults to be 11%, assuming mean ingestion rates. When we examined BDE 209, a congener not included in previous inhalation estimates, the contribution of inhalation exposure significantly increased, accounting for 22% of BDE 209 exposure in adults.

Our estimates of inhalation exposure are likely to be low. We used personal, indoor air concentration from homes and estimated that people spend 90% of time indoors (*12*). The 10% of outdoor exposure was not included in our calculation. Additionally, air concentrations in European offices (*17*, *19*) and industrial occupational settings (*30*, *31*) were higher than those in residences. While our air sampling method more thoroughly captures particulate-bound PBDEs than passive methods, our samplers did not exclude particles too large to reach the alveoli where lung−blood gas exchange occurs; depending on the size fractionation of particulate, our results may somewhat overestimate inhalation exposure. However, overall exposure to airborne particulate may be more accurately represented: the larger particles may be directly ingested or transported via the mucociliary escalator, representing a secondary exposure pathway for airborne particulate.

Our estimates of the relative contribution of inhalation and dust ingestion to PBDE exposure are limited by the large uncertainty surrounding dust ingestion rates and the difficulty of directly measuring dust ingestion. Without knowledge of body burdens of PBDEs for participants in this study, we could not empirically determine which route was most important. Future work might measure personal air in offices or other microenvironments (automobiles) along with a biomarker of exposure.

To our knowledge, our results are the first measurements of personal air concentrations of PBDEs in a non-occupational setting and the first indoor air measures of PBDEs in the United States; they include BDE 209, a congener that has not been widely reported. We found that personal air concentrations of less volatile PBDEs exceeded area measurements, consistent with a personal cloud effect, and that inhalation of BDE 209 may account for a significant percent of overall PBDE exposure in U.S. adults.

Acknowledgments

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Supporting Information Available

Calculating fraction on particulate (ϕ); temperature adjustments for log K_{oa}; theoretical and observed ϕ; sensitivity analyses; correlations between personal and area air. This material is available free of charge via the Internet at http://pubs.acs.org.

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