Characterization of PBDEs in Household Dust: Comparison of Collection Methods and Relationship to Indoor Air

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Introduction. Polybrominated diphenyl ethers (PBDEs) are a class of brominated fire retardants that are commonly used in consumer products such as electronic appliances and furniture containing polyurethane foam. In our earlier work we found associations between PBDE concentrations in breast milk and house dust as well as dietary habits (Wu et al. 2006). However, this association could have been due to air concentrations, if dust and air were highly correlated. In this earlier study, a combined dust sample was collected by researchers from several areas of the house (Wu et al. 2006). It is not known if PBDE concentrations in dust vary within the home or how concentrations depend on the method of collection. For example, Colt et al (1998) found that the personal home-use vacuum cleaner can be used as a surrogate for more labor and time intensive researcher-collected dust methods.

The objectives of the current study were to sample dust in microenvironments within each home to determine if dust concentrations of PBDEs were homogeneously distributed between rooms. We also collected dust from the participants' vacuum cleaner bag along with researcher-collected dust to determine the comparability between dust collection methods. Finally, we compared concentrations in dust and air sampled from the same rooms.

Materials and Methods. Three household dust samples were collected from each of 20 participant's residences in the Boston, Massachusetts (USA) area from January through March, 2006. Two dust samples were researcher collected and the third was collected from the participant's vacuum cleaner. The study population was a convenience sample, with the simple inclusion criteria of owning a vacuum cleaner. To ensure the household vacuum bag was representative of the current home, participants were excluded if they had not changed their vacuum bag since moving into the current residence.

Researchers simultaneously collected dust in the participant's bedroom and main living area. Participant's were asked not to vacuum, sweep or clean for seven days prior to the visit. Dust samples were collected in accordance with a standardized protocol, using a Eureka Mighty-Mite vacuum cleaner (Model 3670) and crevice tool attachment (Rudel et al. 2003; Wu et al. 2006). Dust was captured by a cellulose extraction thimble (Whatman International) that was inserted between the crevice tool and the vacuum tube extender and secured using a rubber o-ring. After dust collection, the thimbles were wrapped in aluminum foil and sealed in polyethylene zip bags. Field sampling equipment was cleaned between each visit to prevent cross-contamination. Dust blanks were prepared using sodium sulfate powder as a surrogate for dust.

The entire floor-surface area equivalent of each room was vacuumed by gently drawing the crevice tool across the top of all surfaces. Emphasis was placed on not collecting dust from under furniture or in crevices between cushions. After vacuuming, the room surface area was measured. This sampling

strategy allows for calculating the two most common dust metrics: concentration (mass of contaminant per mass of dust) and dust loading (mass of contaminant per surface area sampled). The participant's vacuum bag was collected by removing the entire vacuum bag, wrapping the bag in aluminum foil and sealing the bag in a polyethylene zip bag. For participant's with a canister vacuum, the canister was emptied into a pre-cleaned amber jar and sealed in the same fashion. Dust samples were first sieved to remove large debris using ASTM certified metal sieves to collect dust particles <250 microns in size. Laboratory analysis of the dust samples was done by GC/MS ECNI, using the same methods described in a previous publication (Stapleton et al. 2005).

Results and Discussion. Table 1 presents the dust results for PBDE congeners with >50% detection on a concentration basis (ng/g). Also included is bistribromophenoxyethane (BTBPE). For researcher-collected dust, penta-product BDEs and deca-product BDEs were significantly higher in the main living area compared to the bedroom (p=0.01). The highest recorded value of total PBDEs in household dust, 269,300 ng/g, was found in one participant's vacuum bag. By percent of total PBDEs, the dust samples were generally dominated by BDE 209, followed by BDEs 99, 100 and 47; however, the relative contributions of each congener vary substantially by sample. For example, in dust samples collected from home vacuum bags, the percent contributions of penta-product BDEs to total PBDEs ranged from 0.4% to 98.5%, octa-product BDEs ranged from 0.02% to 11.1%, and deca-product BDEs ranged from 1.3% to 99.5%.

Table 1. Summary Statistics for PBDEs and BTBPE in dust (ng/g) by location

		Main Living Area (n = 20)	Bedroom (n = 20)	Home Vacuum Bag (n = 20)
BFR	**Congener**	**GM (GSD)**	**GM (GSD)**	**GM (GSD)**
Penta-product				
	BDE 17	0.6 (33.9)	0.2 (23.4)	0.2 (20.9)
	BDE 28/33	15.9 (3.6)	12.1 (2.3)	6.0 (3.0)
	BDE 47	1,885.8 (2.8)	850.6 (3.3)	332.4 (4.4)
	BDE 49	21.9 (10.7)	23.6 (2.6)	12.4 (2.9)
	BDE 66	12.7 (10.2)	15.3 (2.9)	6.0 (5.7)
	BDE 75	6.2 (6.1)	3.5 (5.5)	2.8 (4.2)
	BDE 85/155	124.0 (3.1)	51.8 (4.1)	16.6 (7.3)
	BDE 99	2,474.4 (2.9)	1,178.9 (4.0)	538.7 (3.6)
	BDE 100	440.0 (2.9)	207.0 (3.8)	74.3 (4.4)
	BDE 138	15.9 (11.5)	10.9 (7.2)	4.5 (6.1)
	BDE 153	234.4 (2.9)	124.2 (4.5)	47.0 (4.2)
	BDE 154	182.8 (2.9)	94.4 (4.3)	30.3 (7.8)
	∑ Penta-product BDEs	**5,502.3 (2.9)**	**2,640.7 (3.7)**	**1,187.2 (3.5)**
Octa-product				
	BDE 183	22.9 (6.1)	30.9 (7.4)	15.1 (3.7)
	BDE 196	1.2 (28.9)	0.9 (31.8)	3.3 (8.6)
	BDE 197	1.0 (37.0)	1.7 (41.7)	5.3 (5.8)
	BDE 203	1.3 (20.1)	1.6 (25.3)	4.4 (6.2)
	∑ Octa-product BDEs	**40.2 (6.0)**	**51.1 (7.1)**	**35.1 (3.4)**
Deca-product				
	BDE 206	87.4 (3.9)	43.6 (5.0)	41.7 (5.5)
	BDE 207	40.2 (7.3)	19.2 (13.7)	25.2 (7.5)
	BDE 208	27.9 (9.3)	9.2 (16.6)	26.3 (9.1)
	BDE 209	4,601.6 (4.4)	1,667.0 (6.8)	1,845.6 (5.5)
	∑ Deca-product BDEs	**4,812.2 (4.4)**	**1,858.9 (6.0)**	**1,974.6 (5.5)**
BTBPE				
	BTBPE	10.0 (16.8)	5.0 (27.5)	11.3 (3.9)

Factor analysis was used as an aid in guiding the analysis, using the *a priori* hypothesis that the congeners are related to the three commercial products. The results of this analysis yielded three distinct factor patterns for each sample type (bedroom, main living area, household vacuum bag), with congeners aligning with their respective commercial product, Penta, Octa and Deca [results not shown]. There were limited exceptions to the neat alignment of congeners along commercial product, consistent with observations that the congener distributions observed in the environment differ from the distributions in their source commercial product. A possible explanation for this is the breakdown of higher brominated congeners into lower brominated congeners, as described in Stapleton et al. (2006). In that study, BDE201 was observed to be a breakdown product of BDE209. When BDE201, a minor component(<0.8%) of the Octa but not Deca product (LaGuardia et al. 2006), was included in the factor analysis (originally excluded because of the sample percent detection was <50%),

we found that BDE201 loaded onto the Octa factor equally with the Deca factor, suggesting an equal contribution from source and breakdown product in the environment for this congener.

BTBPE was detected in 87% of dust samples. Similar to PBDEs, the results for BTBPE were log-normally distributed across participants. The ranges for BTBPE by location were 1.3 – 85.5 ng/g in the home vacuum bag, <LOD – 380.8 ng/g in the bedroom, and <LOD – 333.6 ng/g in the main living area.

For penta- and deca-product BDEs, the bedroom and main living area dust were moderately correlated (r = 0.45, p=0.047; r = .55, p=0.012, respectively) (Figure 1). For the octa-product BDEs, the correlation was less strong and not statistically significant (r =0.34, p=0.14). The lack of stronger correlations between the two rooms, along with the significantly higher concentrations found in main living area dust, suggests that microenvironments are playing an important role in the distribution of PBDEs in household dust. Sources in one room are creating concentrations that are not uniformly distributed across the home. The lack of a strong association further supports the idea that the sources are localized and not likely to be from a general home characteristic (ie. insulation).

Figure 1. Concentrations of PBDEs in researcher-collected household dust (ng/g), by room .

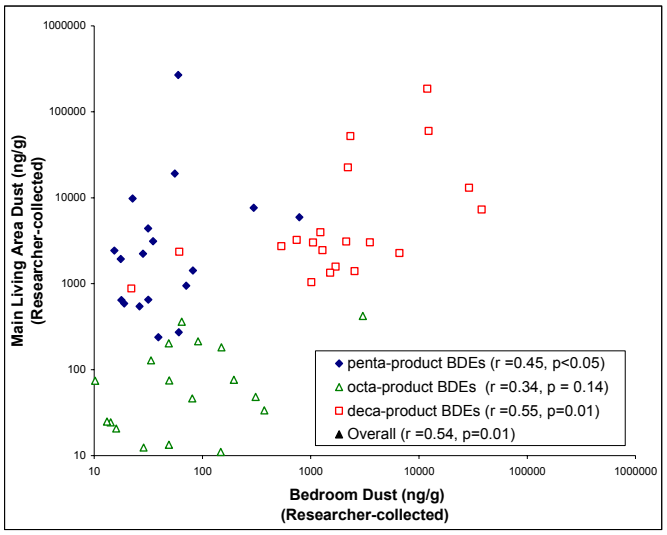
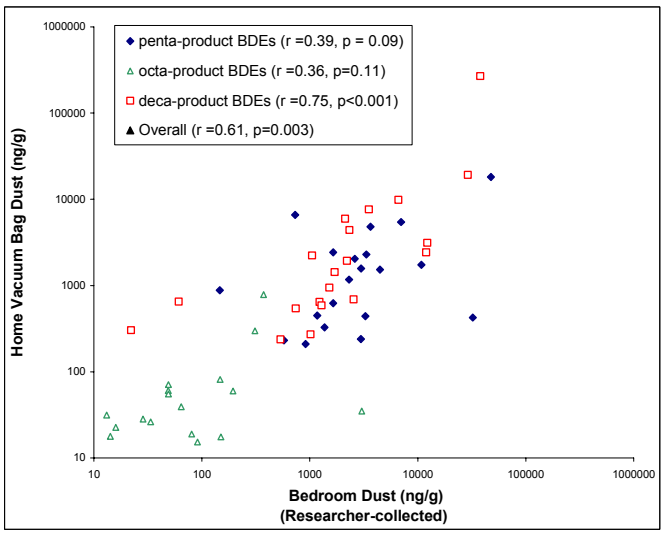


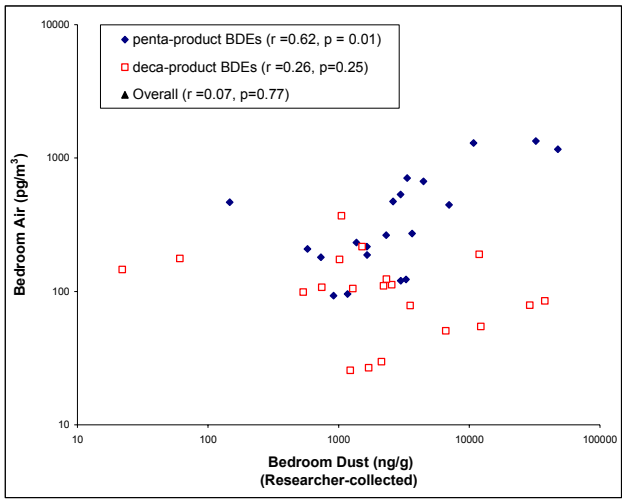
Figure 2. Comparison of researcher-collected bedroom dust and home vaccum bag dust.



The researcher collected dust for the main living area and participant's vacuum bag were not correlated for any of the three congener groupings. However, the dust collected in bedrooms and participant's vacuum bag were strongly correlated for deca-product BDEs (r = 0.75, p<0.001), and moderate to strongly correlated for total BDEs (r = 0.61, p=0.003) (Figure 2). Suggestive evidence of a moderate association was observed for the penta-product BDEs (r = 0.39, p=0.09). Possible explanations for the difference in correlations between rooms include greater dust loading in the bedroom, differential cleaning in the two rooms (ie. sweeping in living room), or more frequent use of the vacuum cleaner in the bedroom. Our results show that vacuum bag dust appears to be a suitable surrogate for researcher-collected dust, but only for the bedroom and only for deca-product BDEs.

PBDEs were measured in air for the same bedrooms and main living areas as the researcher collected dust, previously described in Allen et al. (2006). For the bedroom, there was a moderate correlation of penta-product BDEs in air and dust (r = 0.62, p=0.01), and no association found for deca-product BDEs (Figure 3). For the main living area, there were no associations found between air and dust for either penta- or deca-product BDEs (octa-product BDEs were not detected in the air samples). For total BDEs, there were no associations seen for either room. The lack of an association for deca-product BDEs and total BDEs, and penta-product BDEs in the main living area, indicates that dust cannot generally be used as a predictor of airborne concentrations.

Figure 3. Comparison of PBDEs in bedroom dust and air.by penta-[1] and deca-products[2].



[1] For air, sum of congeners 17, 28/33, 47, 49, 66, 85/155, 99, 100, 153, 154
[2] For air, only includes congener 209

These results demonstrate the importance of examining microenvironments when conducting exposure assessments in locations with a heterogeneous distribution of sources. The use of a participant's vacuum cleaner bag is a cost effective and simple means of estimating dust concentrations in the indoor environment. These benefits must be weighed against the evidence that this method may not be representative of room-specific and congener-specific exposures. Future studies may look at both dust collection methods related to the resident's body burdens of PBDEs to determine which is the most accurate predictor of human exposure to PBDEs.

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