PBDEs , PCDD/Fs, AND PCBs IN INDOOR HOUSE DUST

Wenning RJ [1], Bock, M [2], Maier M [3], Luksemburg WJ [3]

[1]ENVIRON, Marketplace Tower, 6001 Shellmound Street, Suite 700, Emeryville, California, 94608, USA; [2]ENVIRON, 163 Commercial Street, Suite 402, Portland, Maine 04101, USA; [3]Vista Analytical Laboratory, Inc., 1104 Windfield Way, El Dorado Hills, California 95762, USA

Introduction

A growing number of studies have shown that certain chemicals and residues found in plastics, furniture, carpets, electronic equipment, and other household consumer products may be found in house dust, thereby providing another opportunity for human exposure.[1,2,3] Several recent studies have reported the presence of polybrominated diphenyl ethers (PBDEs) and polychlorinated dibenzo-p-dioxins (PCDD/Fs) in indoor house dust in the U.S. and several European countries at similar concentrations, though the distribution of congeners often vary widely.[4,5,6,7,8,9]

At present, concerns are increasingly focused on the occurrence of PBDEs in the environment and the potential for human exposure through foods and consumer products treated with flame retardants containing PBDEs.[10] These concerns are especially focused on children, because of the association shown in animal studies between exposure to certain PBDEs and effects on the thyroid and thyroid hormones, which are known to be critical to growth and development at young ages.[11] Materials used in consumer products that may contain PBDEs include high-impact polystyrene (e.g., television casings), latex (e.g., back coating of textiles), acrylonitrile-butadiene styrene (e.g., computer casings), and flexible polyurethane foam (e.g., upholstered furniture, mattresses, carpet padding). These products can contain anywhere from 2% to 18% by weight of PBDEs and may be a source of PBDEs in indoor dust.[12,13]

In this study, the concentrations of PBDEs, PCDD/Fs, and polychlorinated biphenyls (PCBs) were measured in house dust collected from air conditioner filters and vacuum cleaners used in residential houses in northern California, U.S, and Wellington, New Zealand. This is one of the few studies to also report PCBs in house dust. The results are summarized and compared to indoor dust data from other studies to identify similarities and differences in PCDD/F, PCB, PBDE levels, trends, and congener distributions. The significance in terms of overall human exposure was also considered using a screening-level human exposure model.

Materials and Methods

Indoor house dust was collected from two sources: air conditioner filters and vacuum cleaners. The dust from air conditioner filters was collected by scraping material from the filter directly into pre-cleaned amber glass jars with Teflon lids. Vacuum cleaner dust samples were collected using both standard household and specialized vacuum cleaning equipment. In 5 houses, indoor dust from household vacuum cleaners was collected by transferring the contents from the filter bag directly into pre-cleaned amber glass jars. In 4 houses, house dust was collected using a High Volume Small Surface Cyclone Vacuum Sampler (HVS3) as described in ASTM Method D-5438-00[14] using techniques as described in the literature.[15,16,17,18]

The HVS3 is a vacuum equipped with a cyclone and a fine-particle filter capable of capturing 99.95% of particles above 0.3 μm mean diameter. Using the HVS3, particulate matter was lifted by means of a flowing air stream passing through the sampling nozzle at a controlled velocity and flow rate and separated mechanically in a cyclone chamber. A Pell-Gelman glass fiber type A/E filter connected to the cyclone chamber served as a secondary capture device for fine particulates. The HVS3 allowed for height, airflow, and suction adjustments to reproduce systematically so that sampling conditions approximated consistent and repeatable conditions. Material collected from both the cyclone and glass fiber filter were combined prior to chemical analysis.

Chemical analysis was performed by Vista Analytical Laboratory (El Dorado Hills, CA, USA) by selective ion monitoring by high resolution gas chromatograph / mass spectrometer (HRGC/HRMS) using a modification of U.S. EPA Method 1614 for PBDEs, Method 1613 for PCDD/Fs and Method 1668 for PCBs. House dust samples were analyzed for 17 PBDEs (BDE-3, BDE-7, BDE-15, BDE-17, BDE-28/33, BDE-47, BDE-66, BDE-77, BDE-85, BDE-99, BDE-100, BDE-138, BDE-153, BDE-154, BDE-183, BDE-207 and BDE-209) and mono through nona homologues; 17 2,3,7,8-substituted PCDD/Fs and tetra through hepta homologues; and 209 PCB congeners. All results are reported in picograms per gram (pg/g) dry weight (dw) for PCDD/Fs and PCBs and nanograms per gram (ng/g) dw for PBDEs.

The potential for exposure to PCDDs, PCBs, and PBDEs in indoor house dust by children and adults was evaluated using a screening-level equation and exposure parameters from U.S. EPA.[19,20] Similarities and differences in congener distributions in this and other house dust studies were evaluated using principal components analysis to evaluate potential sources.

Results and Discussion

In this study, PBDEs, PCDD/Fs, and PCBs were found in house dust collected in Wellington, New Zealand, and northern California. Results are summarized in Figures 1 and 2.

In house dust, total PBDEs ranged widely, spanning nearly 2-orders of magnitude (1,100 to 96,300 ng/g). In the air conditioner filters, total PBDEs were 20-fold higher in the sample from a commercial office building as compared to samples collected from residential homes. Indoor dust collected from 11 vacuum cleaner filter bags ranged widely, from 1,100 to 17,700 ng/g, with a median of 11,900 ng/g. The vacuum dust sample collected from a rural California location was at the lower end of the range (3,000 ng/g); the single sample from New Zealand contained 10,200 ng/g. Consistent with the results of Stapleton[8], the predominant PBDE congeners in the dust samples are, in order, BDE-209 > BDE-99 > BDE-47 > BDE-100. PBDE congener distributions are presented in Table 1 and Figure 3. Indoor dust collected from air conditioner filters was dominated by DecaBDE. Indoor dust samples collected from vacuum cleaner filter bags was dominated by both DecaBDE and PentaBDEs, and to a lesser degree by TetraBDEs and HexaBDEs, with one sample exception where the dust contained predominantly Hexa- and HeptaBDEs.

As shown in Table 1, the TEQ_{WHO97} for PCDD/Fs ranged from 1.8 to 53 pg/g. The TEQ_{WHO97} for dioxin-like PCBs ranged from 0.099 to 1.3 pg/g. The range for total TEQ_{WHO97}, including both PCDD/Fs and dioxin-like PCBs was 1.9 to 55 pg/g. The lowest levels were found in the vacuum filter bag from New Zealand. The relative distributions of PCDD/Fs were fairly consistent among all samples, and the relative distributions of dioxin-like PCBs differ only for the sample from New Zealand, which has proportionally more PCB-123 than the U.S. samples.

The screening-level child exposure results and comparisons to noncancer health effect benchmark for PBDEs are summarized in Table 2, the results are similar for adults. For non-cancer health effects, theoretical exposures to PBDEs, PCDD/Fs (not shown), and PCBs (not shown) in house dust were well below levels of health concern – there were 2 to 3 orders of magnitude differences between screening non-cancer health effect benchmark values and predicted exposure concentrations. Using the total TEQ for PCDD/Fs and dioxin like PCBs for the composite (n=15) U.S. sample, the theoretical cancer risk was five in one million (5 in 1,000,000 or 5×10^{-6} risk). This is within the “target risk range” established by the USEPA of one in a million (1×10^{-6}) to one hundred in a million (100×10^{-6}). Principal components analysis (not shown) reveals differences in PBDE composition among the available studies published to date and between geographic areas, suggesting differences in flame retardant formulations and/or usage.

Conclusions

The results of this study indicate that house dust may contribute to human exposure to PBDEs, PCDD/Fs, and PCBs. Additional work, however, is clearly needed to better understand the significance of the house dust ingestion exposure pathway relative to intake from foods and other potential environmental sources. For example, questions exists whether current estimates and assumptions regarding the amount of house dust ingested by adults and children on a daily basis are correct.[21] It is generally assumed for screening purposes that individuals ingest, on average, approximately one-half of the amount of outdoor soil assumed to ingested every day (200 g/day for children and 50 g/day for adults[20]), though increasing evidence suggests current estimates may be exaggerated.[22]

Based on the results of this study, higher brominated PBDEs (decaBDE), as well as lower brominated PBDE congeners (e.g., tetraBDEs and pentaBDEs), are likely ubiquitous at low levels in residential houses and businesses. The results from this study, with the exception of the single high result for dust from the business air conditioner filter, compare well to the range found in 17 house dust samples from Stapleton[8], which was 780 to 29,700 ng/g.

Acknowledgements
This work was supported entirely by Vista Analytical Laboratory and ENVIRON. The authors wish to thank private homeowners, whom remain anonymous, for permission to conduct indoor house sampling and use of results.

References

01. Roberts, JW, Dickey, P. 1995. Rev Environ Contam Toxicol. 143:59-78.
02. Marklund, A, Andersson, B, Haglund, P. 2003. Chemosphere. 53(9):1137-46.
03. Ferro, AR, Kopperud, RJ, Hildemann, LM. 2004. Environ Sci Technol. 38(6):1759-64.
04. Wittsiepe J, Ewers U, Mergner HJ, Lahm B, Hansen D, Volland G, Schrey P. 1997. Zentralbl Hyg Umweltmed. 199(6):537-50.
05. Rudel, RA, Camann, DE, Spengler, JD, Korn, LR, Brody, JG. 2003. Environ. Sci. Technol. 37:4543-4553.
06. Santillo, D, Labunska, I, Davidson, H, Johnston, P, Strutt, M, Knowles, O. 2003. Consuming Chemicals: Hazardous Chemicals in house dust as an indicator of chemical exposure in the home: Part I - UK. Greenpeace Research Laboratories Technical Note 01/2003.
07. Santillo, D, Labunska, I, Fairley, M, Johnston, P. 2003. Hazardous chemicals in house dusts as indicators of chemical exposure in the home: Part II : Germany, Spain, Slovakia, Italy and France. Greenpeace Research Laboratories Technical Note 02/2003.
08. Stapleton, H, Dodder, N, Schantz, M, Wise, S. 2004. Organohalogen Compounds. 66:3740-3744.
09. Sjodin, A, Papke, O, McGahee, E, Jones, R, Focant, J-F, Pless-Mulloli, T, Toms, L-M, Wang, R, Zhang, Y, Needham, L, Herrmann, T, Patterson, D. 2004. Organohalogen Compounds. 66:3817-3822.
10. Wenning, R.J. 2002. Chemosphere. 46(5):779-96.
11. McDonald, TA. 2002. Chemosphere. 46(5):745-55.
12. Alaee, M., Wenning, R.J. 2002. Chemosphere. 46(5):579-82.
13. ENVIRON. 2004. Children's Health Assessment of Pentabrominated diphenyl ethers. USEPA, Cincinnati.
14. ASTM Method D 5438-00, Standard Practice for Collection of Floor Dust for Chemical Analysis.
15. Roberts, JW, Budd, WT, Ruby, MG, Stamper, VR, Camann, DE, Fortman, RC, Sheldon, LS, Lewis, RG. 1991. Paper No. 91-150.2, 84th Annual Meeting, Air & Waste Management Association, Vancouver, British Columbia, June 16–21.
16. Roberts, JW, Han, W, Ruby, MG. 1993. Evaluation of Dust Samplers for Bare Floors and Upholstery. Battelle Subcontract No. 46534(g21733808)-00 03EQ, EPA Prime Contract No. 68-00-0007.
17. Roberts, JW, Ruby, MG. 1989. Development of a High Volume Surface Sampler for Pesticides," U.S. Environmental Protection Agency Report No. EPA 600/4-88/036.
18. Stamper, VR, Roberts, JW, Ruby, MG. 1990. Development of a High Volume Small Surface Sampler for Pesticide and Toxics in House Dust. Research Triangle Institute Report No. RTI/171-01/02F, Research Triangle Park, NC,
19. U.S. EPA. 1997. Risk Assessment Guidance for Superfund. Vol I. Human Health Evaluation manual (Part A) EPA/540/1-89/002.
20. U.S. EPA. 1997. Exposure Factors Handbook. EPA/600/P-95/002Fa.
21. Hays, SM, Cushing, CA, Leung, HW, Pyatt, DW, Holicky, KC, Paustenbach, DJ. 2003. Journal of Children's Health. 1(4):449–475.
22. Ruby, M.V., Fehling, K.A., Paustenbach, D.J., Landenberger, B.D., Holsapple, M.P. 2002. Environ Sci Technol. 36(22):4905-11.

Figure 1. Total PBDE Concentrations Measured in Dust Samples.

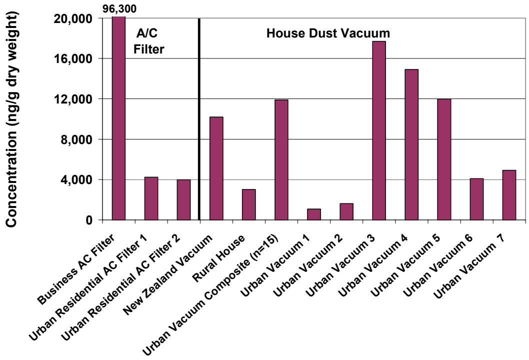

Figure 3. Relative Distribution of PBDEs in Dust.

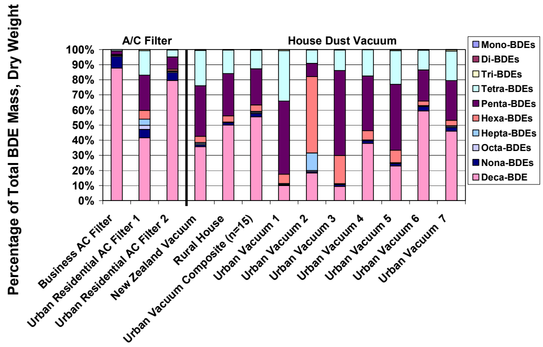

Figure 2. PCDD/F and Dioxin-like PCB TEQ_{WHO97} Concentrations Measured in Dust Samples

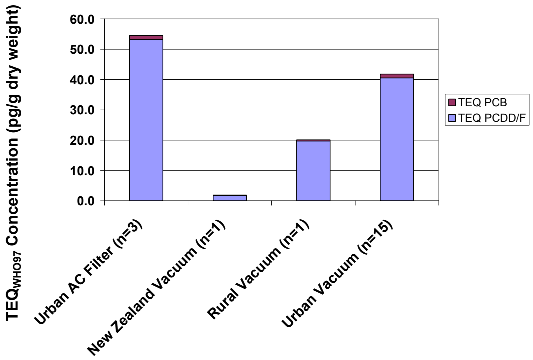

Table 1. Summary of Select PBDE Congener and Dioxin Toxicity Equivalent PCDD/F and PCB Concentrations in Dust Samples.

Parameter	Units	Mean	Min	-	Max
BDE-28/33	ng/g	21	0.9	-	59
BDE-47	ng/g	1,120	44	-	2,460
BDE-66	ng/g	53	20	-	113
BDE-85	ng/g	296	34	-	7,450
BDE-99	ng/g	2,140	207	-	7,660
BDE-100	ng/g	373	32	-	1,580
BDE-153	ng/g	276	21	-	1,440
BDE-154	ng/g	200	17	-	1,110
BDE-183	ng/g	39	4	-	157
BDE-209	ng/g	9,052	107	-	84,500
PCDD/F Total Dioxin TEQs	pg/g	28	1.8	-	53
PCB Total Dioxin TEQs	pg/g	0.8	0.099	-	1.3
Combined TEQs	pg/g	30	1.9	-	55

Table 2. Results of the screening-level calculation of daily intake of PBDEs from house dust collected from residential houses.

Chemical and Receptor	ADD (mg/kg-day)	Alternative Benchmarks		U.S. EPA Benchmarks	
		Benchmark	Hazard Index	RfD	Hazard Index
DecaBDE, Child	5×10^{-5}	4	0.00001	0.01	0.005
Mono-NonaBDEs, Child	4×10^{-5}	0.04	0.001	0.002	0.02