Assessment of Decabromodiphenyl Ether (BDE-209) in the Indoor Environment: Implications on Environmental Exposure Assessment

Yu-Li Huang, Yu-Ping Wang, Dai-Rong Yang, Sheng-Yi Chen, Jenq-Ren Chen
Department of Safety, Health and Environmental Engineering, National Kaohsiung First University of Science and Technology, 2 Juoyue Road, Nantz District, Kaohsiung, 811, Taiwan.

Introduction

Polybrominated diphenyl ethers (PBDEs) have been an important type of flame retardant. PBDEs are chemically stable, and break down under high temperature to release incombustible gas that will further slow down the combustion process. On the other hand, because of their chemical stability, PBDEs are likely to accumulate upon release into the environment. Furthermore, PBDEs are hydrophobic and tend to accumulate within the biota and eventually in the human body. Various studies have shown substantial bioaccumulation and bioconcentration within the natural environment. For example, there are abundant measurement data on PBDEs in the atmosphere (Su *et al.*, 2007; Moon *et al*., 2007), soil (Hale *et al*., 2006), sediment (Oros *et al*., 2005), wastewater sludge (Oberg *et al*., 2002; Kupper *et al*., 2008), as well as measurement from wildlife specimens (She *et al*., 2002; Tuerk *et al*., 2005) and even food products (Schecter *et al*., 2010). Meanwhile, human biomonitoring studies also showed an increasing trend in body burden of PBDEs through measurement of adipose tissue (Fernandez *et al*., 2007), blood (Jakobsson *et al.*, 2002; [xii]Thuresson et al., 2006) or breastmilk (Meironyte *et al*., 1999), and the results were comparable across specimens on a lipid-based level (Hites et al., 2004). People with occupational exposures to PBDEs were found to have elevated body burden (Jakobsson *et al.*, 2002); on the other hand, within the general population, the total body burden were found to be on an increasing trend. In a recent study, food ingestion was deemed the most dominant exposure pathway for tetra- to hepta-PBDEs (Fromme *et al*., 2009), but exposure contributions for other congeners were not examined.

During the highest application period, PBDEs were widely used in commercial products such as upholstered furniture, curtain, consumer electronics, etc. PBDEs are typically added to the main product materials, and since there is no chemical binding with the main material, PBDEs may leach out readily throughout the product life. PBDEs are often used in electric and electronic products as well as in furniture, and these products are often used indoors. As a result, people are likely to come into direct contact with PBDEs upon its release into indoor environment. Among the 209 possible congeners, decabromodiphenyl ether (BDE-209) is the only congener still in production and use, and

its usage is expected to end within a few years. Nonetheless, because of the large amount of application in the past, issues related to environmental pollution of PBDEs (especially BDE-209) are expected to last for yeas to come.

There were limited data on pollutant levels for BDE-209 in earlier studies, partly because scientists did not consider it hazardous due to its extreme stability, but also because of difficulties in the analytical techniques. With the advancement of instrumental analysis techniques, data on BDE-209 measurement have increased almost on an explosive speed. Recent studies have shown that BDE-209 is abundant in the general living areas, e.g., cars, offices and homes. The published data were primarily from North America and Europe, with some from Singapore, Japan and Korea, but not a lot from Taiwan. Hence, the purpose of this study is to examine indoor levels of BDE-209 in the indoor environment in southern Taiwan.

Materials and Methods

This study was performed in Kaohsiung City, the largest metropolitan area in southern Taiwan. Interior dust samples were collected from various homes, offices and private cars. Each dust sample was collected from the floors using an electric vacuum cleaner with a thimble filter fitted to the vacuum inlet. After sampling, the collected dust samples were kept in the thimble filter and kept in a zipper bag. Before analysis, each sample was emptied from the filter and dried in the oven at 105˚C until total weight stabilizes. The dried dust sample was then passed through a 40-mesh sieve to remove excess debris. For chemical analysis, each pre-weighed dust sample and extracted with 20 mL of HPLC-grade acetone using a microwave extraction device (CEM MARS-5, CEM corporation, Matthews, North Carolina, USA). The extraction procedure included heating to 120 ˚C in 15 minutes at 800 W, followed by a holding time of 10 minutes. Afterwards, the cooled aliquot was filtered through a 0.45μm pore size PTFE filter.

The samples were analyzed with a high-performance liquid chromatograph equipped with a DAD (Agilent Model 1200 HPLC/UV/DAD, Agilent Technologies, Inc., Santa Clara, CA, USA). A C18 column (Zorbax Eclipse XDB-C18, 4.6×150 mm, 3.5 μm) was used for analysis, and the mobile phase was 100% methanol at a flow rate of 1 ml/min. The sample injection volume was 10 μl, and each sample run was set to 12 minutes. Instrumental calibration was performed over 0.6 – 60 μg/mL using the BDE-209 standard (purity >98%, Sigma-Aldrich, Inc., St. Louis, MO, USA), and the instrumental accuracy was measured with the purchased standard solution (50 mg/L, Sigma-Aldrich, Inc., St. Louis, MO, USA). The UV detector was set at 290 nm for detection and quantification of BDE-209. With the

above instrumental settings, BDE-209 could be detected at 8.9 minutes after sample injection, as shown in Figure 1. In some occasions when BDE-209 concentrations were below the calibration range, then the extracts were concentrated under nitrogen with an evaporation device (Tokyo Rikakikai Model MG-2200, Tokyo Rikakikai Co. Ltd., Tokyo, Japan). The final solution volume was measured before instrumental analysis.

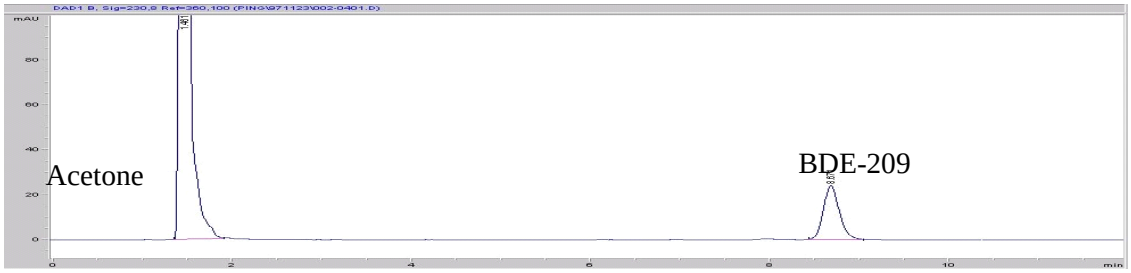

Figure 1. HPLC chromatogram of BDE-209

Results and Discussion

In all, 34 dust samples were collected, including 13 from cars, 7 from private homes, 4 from offices, and 10 from research laboratories. The results are summarized in Table 1. As is shown in the table, concentrations from different types of interior environment varied substantially, with samples from homes, offices and laboratory approximately at the same magnitude level (several thousand ng/g). in contrast, dust samples from cars showed substantially higher concentrations both in terms of arithmetic and geometric mean concentrations. Only four dust samples were collected from different offices, and three resulted in similar concentrations (2,000, 2,100 and 3,800 ng/g); the fourth sample was substantially higher than others, but because of the limited sample size, it could not be determined whether the results adequately represent a "normal" (or typical) range. In addition, the amount of electric and electronic appliances differ in offices, so might this affect the indoor levels for BDE-209, but this could not be confirmed in this study since the amount of appliances were not recorded with each indoor environment. Further study is warranted to estimate BDE-209 levels in an office environment.

Table 1. Summary of sampling outcomes

Sample source	Private home	Car	Office	Laboratory
Total samples measured	7	13	4	10
Concentration range(ng/g)	1,100 – 12,000	2,700 – 38,000	2000 – 16000	2000 - 7900
Mean concentration (ng/g)*	5,600 (3,700)	14,000 (12,000)	6,000 (6,900)	3,800 (1,800)
Geometric mean (ng/g)+	4,400 (2.2)	10,000 (2.3)	4000 (2.7)	3,400 (1.5)

*+Numbers in parentheses denote arithmetic (geometric) standard deviations.

The measured BDE-209 concentrations from homes, cars and laboratories were within the same level of magnitude. Within the same type of indoor environment, the concentrations appeared to follow a lognormal distribution, with geometric standard deviation between 2.2 to 2.7 (Table 1). The results from this study were within the range reported in literature comparable to those found in the literature; nonetheless, variations in BDE-209 levels among these studies suggest that there might be other factors affecting the indoor levels. Also, the large variation in indoor levels may lead to varying exposures due to differing duration spent within each environment. Further studies are warranted on the indoor levels of BDE-209 and potential exposure resulting from the indoor environment.

Acknowledgement

This study was funded by National Science Council of Taiwan under project no. 97-2221-E-327-010.

References

Su Y, Hung H, Sverko E, Fellin P, Li H, 2007. Atmos Environ 41:8725-35.

Moon HB, Kannan K, Lee SJ, Choi M, 2007. Chemosphere 66:585-93.

Hale RC, La GM, Harvey E, Gaylor MO, Mainor TM, 2006. Chemosphere 64: 181-6.

Oros DR, Hoover D, Rodigari F, Crane D, Sericano J. 2005. Environ Sci Technol 39:33–41.

Öberg K, Warman K, Öberg T, 2002. Chemosphere, 48: 805-9.

Kupper T, de Alencastro LF, Gatsigazi R, Furrer R, Grandjean D, Tarradellas J, 2008. Chemosphere 71(6):1173-80

She JW, Petreas M, Winkler J, Visita P, McKinney M, Kopec D, 2002. Chemosphere 46:697–707.

Tuerk KJS, Kucklick JR, Becker PR, Stapleton HM, Baker JE. 2005. Environ Sci Technol, 39:692–8.

Schecter A, Colacino J, Patel K, Kannan K, Yun SH, Haffner D, Harris TR, Birnbaum L. 2010, Toxicol Appl Pharmacol, doi:10.1016/j.taap.2009.10.004

Fernandez MF, Araque P, Kiviranta H, Molina-Molina H, Rantakokko P, Laine O, Vartiainen T, Olea N, 2007. Chemosphere, 66(2):377-83.

Jakobsson K, Thuresson K, Rylander L, et al. Chemosphere, 2002, 709-16.

Thuresson K, Höglund P, Hagmar L, et al. Environ Health Perspect, 2006, 114:176-81.

Meironyte D, Noren K, Bergman A. J Toxicol Environ Health A, 1999, 58(6):329 - 341

Hites RA. Environ Sci Technol, 2004, 38:945–56.

Fromme H, Körner W, Shahin N, Wanner A, Albrecht M, Boehmer S, Parlar H, Mayer R, Liebl B, Bolte G. 2009. Environ Int, Volume 35(8):1125-35.