# CONCENTRATIONS AND SEASONAL VARIATION OF POLYBROMINATED DIPHENYL ETHERS (PBDEs) IN HOUSE DUST FROM KOREA

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## Introduction

Flame retardants, in particular, polybrominated diphenyl ethers (PBDEs) have been used for several decades in many products found in homes such as furniture, mattresses, carpet pads and electronics. PBDEs are a class of additive brominated flame retardants with 209 congeners. Three types of commercial products of PBDEs were available with three different levels of bromination, i.e., penta-, octa-, and deca-BDE products (LaGuardia et al., 2006). The major environmental sources of PBDEs include PBDE containing products, such as polyurethane, computers, and televisions (Jones-Otazo et al., 2005). Similar to polychlorinated diphenyls (PCBs), a class of traditional persistent organic pollutants (POPs), PBDEs can undergo long-range transport to remote regions, accumulation in animal tissues, and biomagnification through food chains (Wang et al., 2007). The objective of this study was to investigate the concentrations and seasonal variations of PBDEs in in-house dust collected in Goyang, Korea. The electronic equipements, which are established the home, also analyzed to evaluate the source relation between the in house dust and electronics containing PBDEs.

## Materials and methods

A total of 5 in-house dust samples were collected from my home of Goyang, Korea, during four seasons from the January to the December in 2011. The dust was collected from the floor in the living room and bedrooms by vacuuming the floor, sofa, and electric appliances with a small vacuum cleaner. Before sample collections, the vacuum cleaner was thoroughly cleaned. Usually, 3~6 g of dust was collected. After collection, all samples were stored in clean amber glass containers and taken back to the laboratory. All the dust samples were passed through a 500 μm sieve (dust finer than this size was assumed to be readily ingested) to remove coarse particles and other residues, lyophilized, and then stored at −18 °C until the time of use.

PBDEs concentrations in the dust samples were determined by Soxhlet extraction and gas chromatography/mass spectrometry (GC/MS) as reported previously with modifications. Thirteen PBDE congeners, including BDE 47, 66, 100, 99, 85, 154, 153, 183, 196, 197, 206, 207 and 209 were quantified. Typically, the dust samples were extracted with 500 mL of toluene for 24 h. The extracts were concentrated, added to 30 mL of n-hexane, and then treated with concentrated $H_2SO_4$ (10 mL) to remove colored matters. The organic phases were loaded onto a multilayer silica–alumina column. After the internal standard was added, the organic solutions (100 μL) obtained were stored at −18 °C until the time of analyses.

Table 1. Collected samples in the house

| *Sample No.* | *Sampling period* | *Sample amounts(g)* | *Comments* |
|---|---|---|---|
| Sample-1 | January~March | 2.9840 | Spring season |
| Sample-2 | April~July | 2.9446 | Summer season |
| Sample-3 | August~October | 12.9165 | Fall season |

| | | | |
|---|---|---|---|
| Sample-4 | November~December | 6.2848 | Winter season |

## Results and discussion

**Concentration levels of PBDEs:** Thirteen PBDE congeners BDE-47, BDE-66, BDE-100, BDE-99, BDE-85, BDE-154, BDE-153, BDE-183 and BDE-209 were detected in dust samples. The mean and median values and concentration ranges are listed in Table 1. As shown in Table 1, the mean and median values of total PBDEs ($\Sigma^{all}$PBDEs) were 23.4 and 1.6 ng/g, respectively, and ranged from 0.02 to 426.7 ng/g.

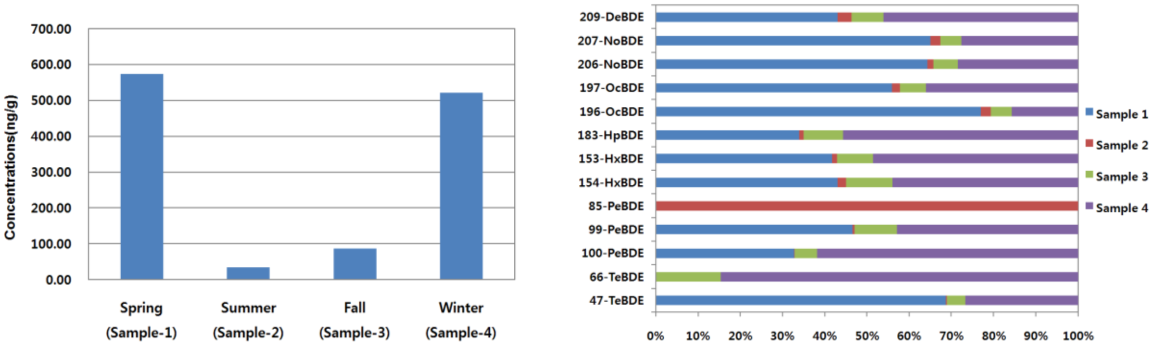
The presence of flame retardants in house dust is thought to originate mainly from household items containing PBDEs that breakdown to form dust particles and/or that off-gas to air followed by partitioning to dust. It is speculated that house dust behaves like a reservoir for PBDEs. The concentrations of 13 PBDE congeners in the samples were showed in Figure 1. The sum concentrations of the 13 congeners ranged from 35 to 573 ng/g with an average of 306 ng/g. The highest concentrations of PBDEs in house dust were observed for BDE 209, with a maximum concentration of 427 ng/g.

Table 1. **Concentration (ng/g) of PBDE in house dust from Korea**

| PBDE Compounds | Mean ± SD | Median | Range |
|---|---|---|---|
| 47-TeBDE | 2.54±3.19 | 1.59 | 0.02-6.98 |
| 66-TeBDE | 0.03±0.05 | 0.01 | <LOD-0.11 |
| 100-PeBDE | 6.94±7.90 | 5.27 | 0.03-17.17 |
| 99-PeBDE | 2.13±1.98 | 2.26 | 0.04-3.97 |
| 85-PeBDE | 0.003±0.01 | 0.00 | <LOD-0.01 |
| 154-HxBDE | 0.25±0.22 | 0.27 | 0.02-0.44 |
| 153-HxBDE | 0.62±0.58 | 0.62 | 0.03-1.20 |
| 183-HpBDE | 1.92±1.90 | 1.66 | 0.09-4.28 |
| 196-OcBDE | 1.13±1.58 | 0.47 | 0.11-3.46 |
| 197-OcBDE | 1.93±1.98 | 1.63 | 0.15-4.32 |
| 206-NoBDE | 26.34±30.28 | 18.05 | 1.52-67.72 |
| 207-NoBDE | 29.38±34.03 | 19.16 | 2.87-76.32 |
| 209-DeBDE | 231.16±210.05 | 233.87 | 30.23-426.68 |

**Seasonal Variation and Congener Pattern of PBDEs:** The seasonal variation showed in Figure 1. As show in the Figure 1, the concentration level of PBDEs in this study decreasing in the following order: Sample-1(spring)>Sample-4(winter)>Sample-3(fall)>Sample-2(summer). The highest concentration observed in spring season. This phenomena may be estimated the ventilation of the in-house because there are limited ventilation in winter and spring season.

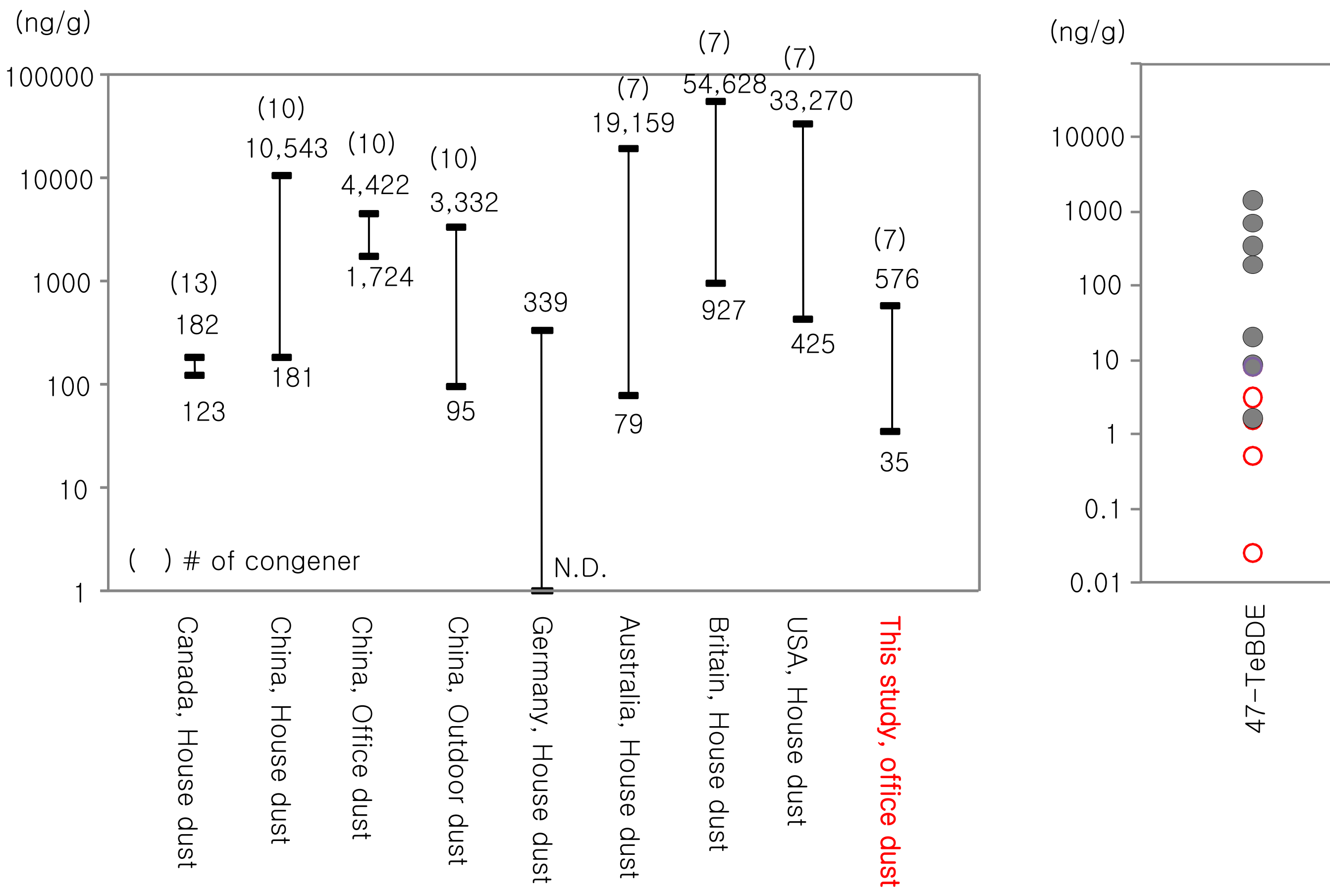
The average contribution of PBDEs congeners in this study decreasing in the following order: BDE-209>BDE-207 and BDE-206>BDE-197>BDE-196>BDE-99>BDE-100>BDE-47. The specific pattern in this study exhibited two significantly different trends. Firstly, higher brominated BDEs from octa- to deca-BDEs were the dominant congeners, and accounted for 75~99% of ΣPBDEs. BDE-209 (average 94%) was the dominant deca-BDE in this study. The electronic equipements, which are established the home, also analyzed to evaluate the source relation between the in house dust and electronics containing PBDEs. The relation will be discussed.



(a) Seasonal Variation (b) Congeners pattern

**Figure 1. Seasonal Variation and congeners pattern of PBDEs**

**Comparison the PBDEs concentration:** The PBDEs concentrations in dust can be compared to those found in another house dust study as shown in Figure 2. Generally, the concentrations of PBDEs detected in this study are lower than the concentrations reported from developed countries. The PBDE concentrations in in-house dust in the present study were much lower than those observed in China, UK (927~54,628 ng g−1) and USA (425~33,270 ng g−1). The average concentration of BDE-209 detected in this study is 2–3 orders less than the reported values from UK (Harrad et al., 2008), Japan (Suzuki et al., 2006) and USA (Batterman et al., 2010).



(a) Total concentration

**Figure 2. Comparison the PBDEs concentration**

**Source of the PBDEs:** In the environmental media, deca-BDEs commercial formulations, especially in indoor dust which was considered the major source of exposure to higher brominated BDEs in humans in recent studies. The detected congener except BDE-209 can be estimated the results of environmental decomposition. Recently, deca-BDE was mainly used so BDE-209 was decomposed as the usage process as shown in Figure 4. From this decomposition mechanism, the other congener's detected ratio except BDE-209 contents was not high ratio; therefore the result can be estimated as the result of environmental decomposition. The presence of flame retardants in house dust is thought to originate mainly from household items containing PBDEs that breakdown to form dust particles and/or that off-gas to air followed by partitioning to dust. It is speculated that house dust behaves like a reservoir for PBDEs. The relation of the source between electronic equipements and in house dust also discussed.

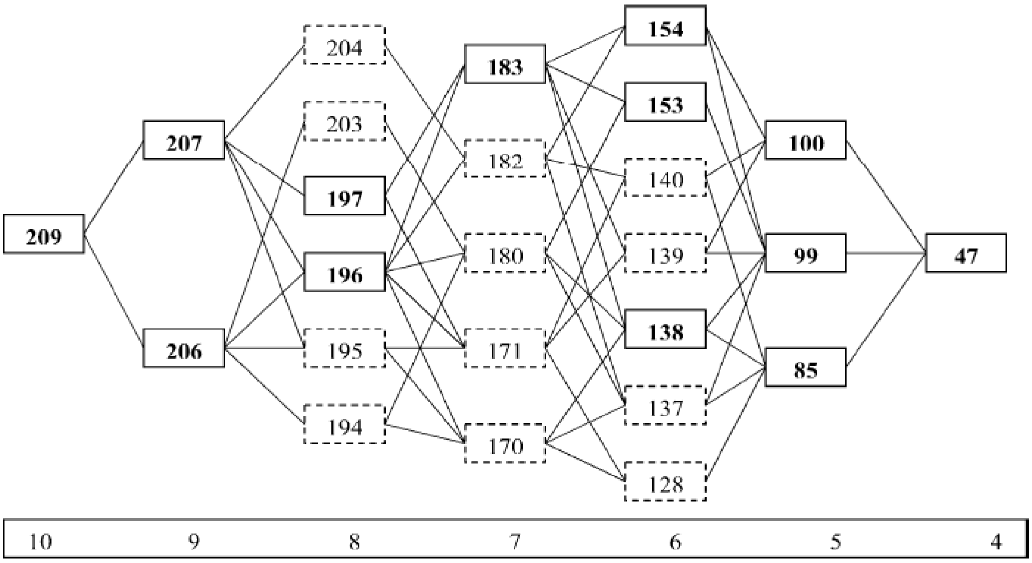


Figure 3. Debromination process of Deca-BDE by exposuing the sun

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