# PBDEs AND PCBs IN INDOOR AIR: LEVELS AND FACTORS INFLUENCING THEIR CONCENTRATIONS

Hazrati S and Harrad S

Division of Environmental Health and Risk Management, University of Birmingham, Edgbaston, Birmingham, B15 2TT, UK

## Introduction

Polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) are two classes of persistent organic compounds widely detected in all environmental compartments as a result of their intensive use in industrial and commercial applications. PCBs were applied in public building constructions for various purposes, mainly as an additive to concrete, caulking, grout, paints, permanent elastics, Thikol rubber sealants, and flame retardant coatings of acoustic ceiling tiles. Owing to their thermal stability, high bromine content (good flame retardant properties), and relatively low costs, PBDEs are used as flame-retardant additives (i.e. they are only dissolved in the material) in different resins, polymers, and substrates at levels ranging from 5 up to 30% [1]. The end use for these flame-retarded polymers is generally in electronic and electrical equipment such as computers, TV sets, and household appliances. They are also added to wires and cables, textiles, polyurethane foam, carpets and upholstery used in household and business furnishings, cars, buses, trucks, and aircraft. Substantial amounts of PCBs and PBDEs may be released through evaporation from a variety of sources present in indoor microenvironments and result in elevated concentrations of these compounds in indoor air. Relatively little is known about PCB and PBDE concentrations in indoor environments and the factors influencing their levels. In this paper we report the PCB and PBDE concentrations in a wide range of indoor microenvironments.

## Material and methods

Fully sheltered polyurethane foam disks (140 mm diameter, 12 mm thickness, 360.6 $cm^2$ surface area, and 0.01685 g $cm^{-3}$ density) were deployed in a number of indoor microenvironments including 29 homes, 29 offices, 3 public indoor environments, and 25 cars for a period of approximately one month in the West Midlands of the UK. PUF disks were treated with a known amount of sampling efficiency standards prior to deployment in the field. The extraction, purification and analysis methods are described elsewhere[2]. Sampling and analytical methods were evaluated by analysing 5 replicate passive samples and 6 replicates of SRM 1944. Recoveries of internal/surrogate standards and sampling efficiency standards were also calculated for all samples. To estimate concentrations of PCBs and PBDEs in indoor air previously reported passive sampling rates were applied[2].

## Results and discussion:

### *PCB and PBDE concentrations in indoor air microenvironments*

ΣPCB in this study constitutes the sum of the 57 quantified PCB chromatographic peaks in most of the samples, while ΣPBDE refers to the sum of 10 BDE congeners including BDEs 17, 28, 47, 49, 66, 85, 99, 100, 153, and 154. Table 1 summarises the concentrations found in offices, homes, public indoor environments, and cars. Average concentrations of ΣPCB and ΣPBDE for all indoor microenvironments studied were 8.8 and 0.269 ng $m^{-3}$, respectively. Excluding cars, average indoor air concentrations of ΣPCB and ΣPBDE for all other indoor microenvironments are 11.6 ng $m^{-3}$ (SD=22; median = 3.55; from 0.49 to 101.8 ng $m^{-3}$) and 0.109 ng $m^{-3}$ (SD=0.2; median = 0.047; from 0.004 to 1.4 ng $m^{-3}$), respectively. With respect to PCB, the most contaminated indoor environments were found to be public indoor microenvironments followed by offices, homes, and cars. Mann-Whitney analysis revealed ΣPCB concentrations in offices to be statistically significantly higher than those in homes ($p<0.001$), which is consistent with an earlier study in the West Midlands[3] where ΣPCB concentrations in workplaces were reported to be higher than domestic indoor microenvironments but contrasts with the work of Macleod[4] who reported PCB levels in 11 homes (Mean = 278 ng $m^{-3}$) to be higher than those measured in 3 office buildings (Mean = 78 ng $m^{-3}$).

A few previous studies have reported elevated concentrations of PCBs in a range of indoor microenvironments at various locations around the world. Such studies have generally focused on buildings with known PCB sources, like PCB containing sealants, caulking, and transformers where PCB concentrations as high as 1000-7500 ng $m^{-3}$ have been reported in buildings with PCB-containing permanently elastic sealants[5, 6]. In the most recent study, a number of homes with PCB containing sealants have been monitored and ∑PCB concentrations ranging from <100 to >6000 ng $m^{-3}$ (mean = 790 ng $m^{-3}$) have been reported[7]. In some cases the concentrations significantly exceeded occupational exposure limits recommended by OSHA, NIOSH, and ACGIH. While ∑PCB concentrations found in this study are much lower than the values reported for such buildings with known PCB sources, they are comparable with the geometric mean of 18 ng $m^{-3}$ reported by Vorhees et al.[8] and the most spatially relevant study for comparison[3], in which PCB levels were reported to be between 1.1 and 69 ng $m^{-3}$ (mean = 9 and median =3.9 ng $m^{-3}$) in 14 different indoor microenvironments from the West Midlands. Importantly, Mann-Whitney test analysis revealed no statistically significant differences in ∑PCB concentrations between our data and those of the latter study[3] (P>0.50) supporting our previous finding that no decline in the contamination of indoor air with PCBs in the West Midlands over the last ca. 8 years is evident[2].

ΣPBDE concentrations in indoor air were on average more than 2 orders of magnitude lower than those of PCBs. Unlike PCBs, cars were identified to be the most contaminated indoor microenvironments within the locations studied, followed by public indoor environments, offices, and homes (see Table 1) implying that cars may be a potentially important source of human exposure to PBDEs. As with the PCBs, Mann-Whitney analysis revealed ΣPBDE concentrations in offices to be statistically significantly (p<0.05) higher than those found for homes, which is in agreement with our earlier study that reported workplaces to be generally more contaminated than domestic indoor microenvironments[9]. The reason for these differences in PCB and PBDE concentrations observed is unknown but might be related to differences in either the number and strength of sources present in each indoor microenvironment or room usage pattern and other factors influencing indoor air concentrations (e.g. room temperature and ventilation rates)[2].

ΣPBDE concentrations in this study are lower than those reported previously for indoor environments using high volume air samplers[9, 10], perhaps due to differences in sampling method or simply because of the inter/intra building and seasonal variations observed in different monitoring campaigns[2]. The most relevant data for comparison are two recent studies in which PUF disk passive samplers were employed to monitor indoor air concentrations of PBDEs in residential indoor environments [11, 12]. Our findings are comparable but still lower than the results reported by Wilford et al.,[11] who found ∑PBDE concentrations (sum of 10 congeners) in 74 Canadian residential homes to be on average 0.26 ng $m^{-3}$. However, indoor air concentrations of ΣPBDE in Birmingham are higher than recently reported values for Kuwait indoor microenvironments in which average concentrations of 0.015 and 0.03 ng $m^{-3}$ have been reported for homes and general workplaces, respectively. The differences between these three studies may be a reflection of the PBDE usage profile in North America, Europe, and Asia.

***Factors influencing PCB and PBDE concentrations in indoor environments***

The time slot covering the 1950s and 1970s is the most likely period in which PCBs may have been applied in buildings construction materials or used in sealants, plasticizers, and paints. PCBs remained in use in closed systems e.g. in capacitors and light ballasts even after they were banned from all new uses in the late 1970s. As a result, the age of buildings may greatly influence PCB levels of indoor environments in industrialised countries. To study any variations in ∑PCB concentrations of building with different age groups, sampling sites were classified into three different age groups. Specifically: buildings constructed prior to 1950 (n = 9, average ∑PCB =5.8, and SD = 4 ng $m^{-3}$); between 1950 and 1979 (n = 33, average ∑PCB = 19.9, and SD = 5 ng $m^{-3}$); and between 1979 and 2005 (n = 17, average ∑PCB = 2, and SD = 1.8 ng $m^{-3}$). As expected, the highest ∑PCB concentrations were found in buildings constructed between 1950 and 1979 followed by those constructed prior to 1950 (Figure 1). However, Mann-Whitney U test analysis revealed no statistically significant differences in ∑PCB concentration between buildings constructed prior to 1950 and 1950-1979 (p=0.36). The reason for this observation might be related to refurbishments performed in some of the old buildings during the PCB usage period or the presence of other PCB containing electrical appliances. However, ∑PCB concentrations in buildings constructed between 1950 and 1979 were statistically significantly higher than those built post 1979

(Mann-Whitney U test, $p<0.001$) suggesting that restrictions on PCB usage have effectively reduced PCB concentrations in indoor environments. Despite this, it is important to note that average ∑PCB concentrations found in buildings constructed well after the introduction of usage restrictions are much higher than those reported for outdoor air in Birmingham [13]. For instance, ∑PCB concentrations in three offices located in a building constructed in 1998 and located approximately 1.5 km distant from the EROS monitoring site within the University of Birmingham, were respectively on average 5, 6, and 7 times higher than those values recorded for outdoor air at EROS over a similar period. Mann-Whitney test analysis revealed average ∑PCB concentrations in buildings constructed post 1980 (n = 17, average ∑PCB = 2 ng $m^{-3}$) to be statistically significantly higher than outdoor air concentrations (n = 26, average ∑PCB = 0.255 ng $m^{-3}$) recorded at the EROS site in 2001-2 ($p < 0.0001$). These significant differences imply that there are still some minor PCB sources that affect the indoor air quality in quite new buildings, which clearly require further research.

Unlike PCBs, concentrations of three major PBDE congeners (i.e. PBDE 47, 99, and 100) as well as the ∑PBDE concentrations were significantly positively correlated with the construction year of the buildings at the 95% confidence interval (i.e. R=0.3 and $p<0.05$) (Figure 2). This is expected, since the potential sources and usage pattern of PBDEs in indoor environments are totally different from those of PCBs. PBDE 47 (r = 0.27 and p = 0.03), PBDE 99 (r = 0.31 and p = 0.01), and ∑PBDE (r=0.29 and p = 0.02) also demonstrated significant positive correlation with the number of personal computers present in the indoor environments studied. However, no significant correlation between the number of PUF containing items of furniture and concentrations of PBDE congeners were observed. Surprisingly, concentrations of most PBDE congeners in carpeted rooms were lower than in non-carpeted rooms (Mann-Whitney U test, $p<0.05$). The reason for this observation is not clear but might be related to the fact that most of the non-carpeted rooms were offices and public indoor microenvironments (containing more personal computers), in which PBDE concentrations were significantly higher than homes. When a similar statistical analysis was carried out for naturally ventilated homes, no significant differences were observed between PBDE concentrations of carpeted and non-carpeted rooms. These observations suggest that the number of items of electrical and electronic equipment (especially personal computers) exerts a bigger influence on PBDE concentrations in indoor air than the number of PUF containing furniture or carpets.

**Acknowledgments**
The authors gratefully acknowledge the provision of scholarship to Mr. Sadegh Hazrati by the Iranian Ministry of Health and Medical Education.

**References:**

1. WHO. 1994, IPCS, *Environmental Health Criteria 162*, Geneva: World Health Organization.
2. Hazrati S. and Harrad S. *Organohalogen Comp* 2005; 67: 1033.
3. Currado G. M. and Harrad S. *Environ Sci Technol* 1998; 32: 3043.
4. Macleod K. E. *Environ Sci Technol* 1981; 15: 926.
5. Balfanz E., Fuchs J. and Kieper H. *Chemosphere* 1993; 26: 871.
6. Burkhardt U., Bork M., Balfanz E. and Leidel J. *Das Offentliche Gesundheitswesen* 1990; 52: 567.
7. Kohler M., Tremp J., Zennegg M., Seiler C., Minder-Kohler S., Beck M., Lienemann P., Wegmann L. and Schmidt P. *Environ Sci Technol* 2005; 39: 1967.
8. Vorhees D. J., Cullen A. C. and Altshul L. M. *Environ Sci Technol* 1997; 31: 3612.
9. Harrad S., Wijesekera R., Hunter S., Halliwell C. and Baker R. *Environ. Sci Technol.* 2004; 38: 2345.
10. Shoeib M., Harner T., Ikonomou M. and Kannan K. *Environ Sci Technol* 2004; 38: 1313.
11. Wilford B. H., Harner T., Zhu J. P., Shoeib M. and Jones K. C. *Environ Sci Technol* 2004; 38: 5312.
12. Gevao B., Al-Bahloul M., Al-Ghadban A. N., Ali L., Al-Omair A., Helaleh M., Al-Matrouk K. and Zafar J. *Atmos. Environ.* 2006; 40: 1419-26
13. Robson M. PhD Thesis, Division of Environmental Health and Risk Management, University of Birmingham, UK, 2004.

**Table 1: Summary of ∑PCB and ∑PBDE concentrations (ng m$^{-3}$) in indoor microenvironments studied**

| | Office | | Home | | Public indoor | | Car | |
|---|---|---|---|---|---|---|---|---|
| Name | ΣPBDE | ΣPCB | ΣPBDE | ΣPCB | ΣPBDE | ΣPCB | ΣPBDE | ΣPCB |
| Min | 0.01 | 0.82 | 0.004 | 0.49 | 0.03 | 1.08 | 0.01 | 0.39 |
| Percentile 5 | 0.01 | 1.1 | 0.005 | 0.6 | 0.04 | 1.9 | 0.01 | 0.41 |
| Percentile 25 | 0.0 | 3.5 | 0.0 | 1.2 | 0.1 | 5.3 | 0.0 | 0.8 |
| GeoMean | 0.1 | 7.8 | 0.0 | 2.0 | 0.1 | 9.4 | 0.1 | 1.1 |
| Percentile 50 | 0.1 | 5.8 | 0.0 | 1.8 | 0.1 | 9.6 | 0.0 | 0.9 |
| Average | 0.2 | 18.2 | 0.1 | 2.8 | 0.1 | 30.7 | 0.7 | 1.4 |
| Standard deviation | 0.3 | 26.9 | 0.1 | 2.6 | 0.1 | 44.2 | 1.9 | 1.2 |
| Percentile 75 | 0.1 | 20.1 | 0.1 | 3.8 | 0.2 | 45.6 | 0.2 | 1.7 |
| Percentile 90 | 0.4 | 43.1 | 0.1 | 6.3 | 0.2 | 67.2 | 1.8 | 2.5 |
| Percentile 95 | 0.6 | 88.4 | 0.2 | 8.9 | 0.2 | 74.4 | 4.2 | 2.6 |
| Max | 1.4 | 101.8 | 0.2 | 9.8 | 0.2 | 81.5 | 8.2 | 6.1 |

**Figure 1: Concentrations of ∑PCB in building with different age groups**

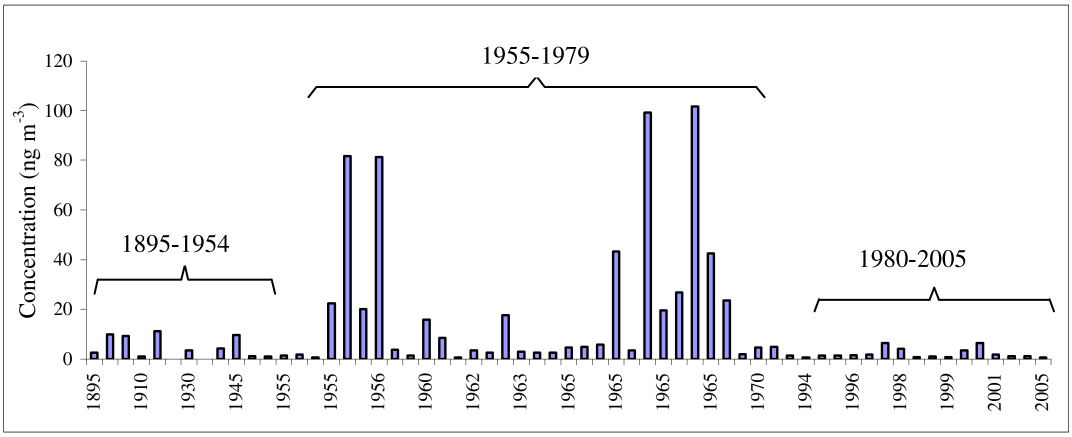
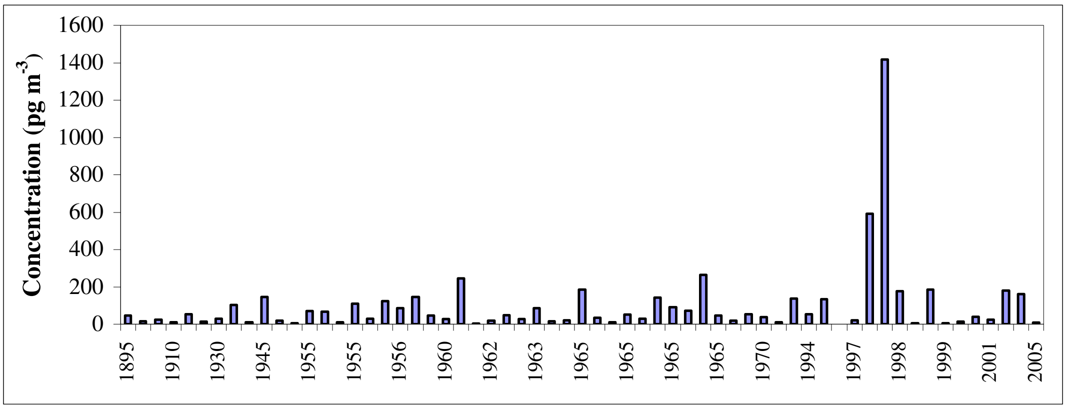



**Figure 2: Distribution of ∑PBDE concentrations in building with different ages**



Concentration (pg m-3)
1600
1400
1200
1000
800
600
400
200
0
1895
1910
1930
1945
1955
1955
1956
1960
1962
1963
1965
1965
1965
1965
1970
1994
1997
1998
1999
2001
2005