BROMINATED FLAME RETARDANTS IN DUST FROM UNITED KINGDOM: WITHIN-ROOM SPATIAL AND TEMPORAL VARIABILITY

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Introduction

Polybrominated diphenyl ethers (PBDEs) are a group of widely used brominated (BFRs). Due to their presence in food, indoor air, household dust, and human tissues[1, 2, 3, 4], combined with evidence relating to their potential adverse effects on human health[5, 6], several jurisdictions have banned the marketing and use of Penta- and Octa-BDEs, while both the Deca-BDE flame retardant formulation and hexabromocyclododecane (HBCD) are the subjects of ongoing EU risk assessment[7, 8]. To date, estimates of human exposure to PBDEs (and other related contaminants) arising from the ingestion of indoor dust have relied on single "spot" measurements in a given microenvironment[3]. Some variability in contamination of dust with PBDEs between rooms in the same house has been suggested[9]. In contrast, nothing is known about: (a) within-room spatial variability and (b) within-room temporal variability. In both instances, substantial variation would have implications for estimates of human exposure. Given this, we studied in a small number of homes and offices, within-room spatial and temporal variability of PBDEs, along with two other BFRs, viz: decabromodiphenyl ethane (DBDPE) and 1,2-bis(2,4,6-tribromophenoxy)ethane (TBE).

Materials and methods

Indoor dust samples were collected in the West Midlands, UK. Samples were all taken under normal room use conditions, so as to reflect actual human exposures as far as possible. Within-room spatial variability in BFR concentrations in dust was studied in three homes and two offices. In each case, five samples were taken on the same day at different locations within the room under study as described above. Care was taken that there was no overlap between the area covered by each sample. Furthermore, in three homes, temporal variability in BFR concentrations in dust was studied by taking one sample per month over a period of either 9 or 10 months. In each room, care was taken to ensure that the same area of the room was sampled on each occasion. Concentrations of tri-hexa-BDEs were determined at the University of Birmingham, with those of higher brominated PBDEs, decabromodiphenyl ethane (DBDPE) and 1,2-bis(2,4,6-tribromophenoxy)ethane (TBE) measured at the University of Antwerp. The detailed sampling and analytical protocols are described elsewhere[3].

Results and discussion

Table 1 shows the concentrations and dust loadings (i.e. grams of dust per unit area of floor sampled) recorded in each sample, along with the relative standard deviation (RSD) of these parameters in each room. Figure 1 illustrates the RSD values for BDE-47, BDE-99, BDE-209, and dust loadings in each room studied. Comparison of these with the RSDs obtained from replicate analysis of indoor dusts SRM2585 and 2584, show that the spatial variability in concentrations of PBDEs in dust within the same room, exceeds substantially that attributable to analytical variability. While it is possible that our RSD value for analytical variability derived from analysis of SRMs may be an underestimate; it is equally possible that it may overestimate analytical variability for highly contaminated samples, for which reproducibility of measurement is generally better. While these findings could suggest that the entire surface of a room must be sampled in order to obtain a representative sample, one must also consider the possibility of appreciable variation in exposure depending on the location within the room, and that a more accurate estimate of exposure may be obtained via sampling only in the most-frequented parts of the room. It is also instructive to note that the comparable RSDs for both concentrations of PBDEs and dust loadings (Figure 1 and Table 1), suggest that at least some of the observed within-room spatial variability in concentrations of BFRs in dust is attributable to spatial variability in dust loadings.

Table 1: Spatial Variability in Concentrations (ng g^{-1}; ng m^{-2} in parentheses) of BFRs and Dust Loadings (g m^{-2}) within Same Room

Sample/ Congener #	Σtri-hexa-BDEs[a]	209	ΣBDEs[b]	DBDPE	TBE	Dust Loading
Home1a	24 (160)	13000 (86000)	13000 (87000)	<dl	<dl	6.8
Home1b	5.0 (28)	6600 (40000)	6600 (40000)	<dl	<dl	6.1
Home1c	9.6 (25)	18000 (48000)	18000 (48000)	<dl	<dl	2.6
Home1d	28 (130)	7900 (36000)	7900 (37000)	<dl	6.5 (30)	4.6
Home1e	3.4 (12)	6300 (27000)	6300 (27000)	<dl	<dl	4.3
RSD[c]	80	49	50	-	-	33
RSD[d]	97	48	49	-	-	
Home2a	250 (48)	-	250 (48)	-	-	0.19
Home2b	180 (37)	-	180 (37)	-	-	0.20
Home2c	200 (64)	-	200 (64)	-	-	0.31
Home2d	300 (53)	-	300 (53)	-	-	0.17
Home2e	180 (29)	-	180 (29)	-	-	0.16
RSD[c]	24	-	24	-	-	29
RSD[d]	29	-	29	-	-	-
Home3a	1900 (1800)	-	1900 (1800)	-	-	0.96
Home3b	1400 (1100)	-	1400 (1100)	-	-	0.77
Home3c	1100 (2300)	-	1100 (2300)	-	-	2.2
Home3d	1300 (1300)	-	1300 (1300)	-	-	1.1
Home3e	2600 (2300)	-	2600 (2300)	-	-	0.88
RSD[c]	37	-	37	-	-	49
RSD[d]	31	-	31	-	-	-
Office1a	160 (190)	-	160 (190)	-	-	1.1
Office1b	250 (190)	-	250 (190)	-	-	0.73
Office1c	270 (230)	-	270 (230)	-	-	0.76
Office1d	250 (200)	-	250 (200)	-	-	0.76
Office1e	130 (200)	-	130 (200)	-	-	1.5
RSD[c]	28	-	28	-	-	34
RSD[d]	4	-	4	-	-	-
Office2a	150 (140)	2800 (2600)	3000 (2700)	210 (190)	99 (90)	0.91
Office2b	470 (320)	4100 (2700)	4600 (3200)	200 (130)	11 (7.4)	0.67
Office2c	200 (130)	3000 (2000)	3200 (2200)	420 (280)	19 (13)	0.67
Office2d	170 (72)	2000 (860)	2300 (1000)	70 (29)	44 (19)	0.42
Office2e	100 (39)	4200 (1600)	4300 (1600)	34 (13)	78 (29)	0.37
RSD[c]	67	28	28	82	75	36
RSD[d]	77	39	40	87	106	-

[a] sum of PBDEs 28, 47, 49, 66, 99, 100, 153, and 154

[b] sum of PBDEs 28, 47, 49, 66, 99, 100, 153, 154, 183, 196, 197, 203, and 209

[c]relative standard deviation of concentrations expressed as ng g^{-1} for BFRs and g m^{-2} for dust loading

[d]relative standard deviation of concentrations expressed as ng m^{-2}

- denotes not measured or not calculated for samples from this room

We also studied within-room temporal variability in concentrations of all target PBDEs in three rooms (all homes). The concentrations and dust loadings recorded for these samples are in Table 2, with the RSD values for BDE-47, BDE-99, BDE-209, and dust loadings shown in Figure 1. In general, RSDs of concentrations of PBDEs and dust loadings in these samples exceed those observed in the spatial variability study. Together with our data on within-room spatial variability, these data provide the first empirically-derived indication of the uncertainty associated with an exposure assessment based on a single measurement of dust contamination from a given room at a given point in time. While the variability in the entire dataset of 9-10 monthly samples within each room does not appear excessive, caution is advised given the range of concentrations within each room. In the three rooms studied, the maximum Σtri-hexa-BDE concentration (ng g^{-1}) exceeds the minimum by a factor of ~50, 3.5, and 5.5 in home A, B, and C respectively, while for BDE-209, the corresponding figures are 7.5, ~400, and ~35. Clearly, substantial variation in estimates of exposure is possible, depending on when a given room is sampled.

Figure 1: Relative standard deviations (RSD) of dust loading, and concentrations of BDEs-47, 99, and 209 in SRMs 2584 and 2585[3] and microenvironments where within-room spatial (Spatial 1-5) and temporal (temporal 1-3) variation were studied.

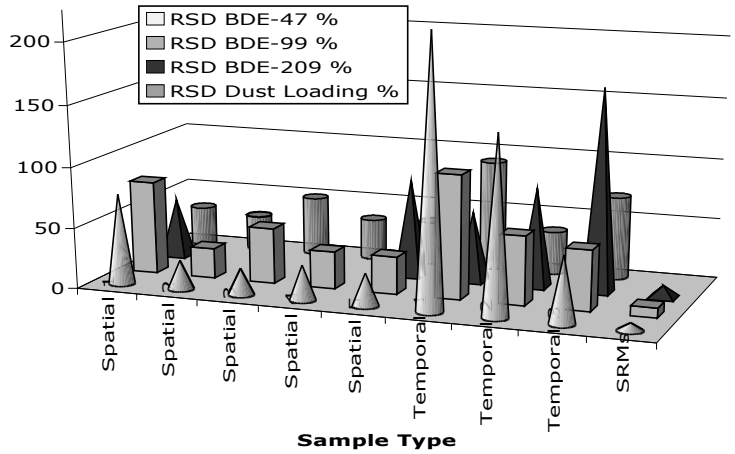

Table 2: Temporal Variability in Concentrations (ng g^{-1}; ng m^{-2} in parentheses) of BFRs[a] and Dust Loadings (g m^{-2}) within Same Room.

Sample/ Congener #	Σtri-hexa-BDEs[b]	209	ΣBDEs[c]	DBDPE	Dust Loading
Home A1	2.7 (26)	7200 (96000)	7200 (97000)	<dl	13.5
Home A2	3.8 (24)	16000 (110000)	16000 (120000)	<dl	7.3
Home A3	9.6 (25)	18000 (48000)	18000 (48000)	<dl	2.6
Home A4	12 (51)	5300 (23000)	5300 (24000)	<dl	4.4
Home A5	13 (46)	10000 (35000)	10000 (35000)	<dl	3.4
Home A6	14 (37)	16000 (44000)	16000 (45000)	<dl	2.8
Home A7	19 (42)	14000 (32000)	14000 (32000)	<dl	2.2
Home A8	150 (200)	22000 (28000)	22000 (29000)	<dl	1.3
Home A9	13 (12)	17000 (17000)	17000 (17000)	<dl	0.96
Home A10	29 (38)	42000 (56000)	43000 (57000)	<dl	1.3
RSD[d]	156	58	58	-	91
RSD[e]	99	61	61	-	-
Home B1	38 (1.9)	3600 (180)	3600 (190)	<dl	0.05
Home B2	38 (20)	27 (14)	65 (34)	<dl	0.53
Home B3	53 (28)	3000 (1600)	3000 (1600)	<dl	0.53
Home B4	45 (28)	2500 (1600)	2600 (1600)	<dl	0.63
Home B5	39 (24)	1600 (940)	1600 (970)	<dl	0.60
Home B6	51 (36)	8700 (6200)	8700 (6200)	58 (41)	0.71
Home B7	51 (42)	3600 (2900)	3800 (3000)	35 (28)	0.81
Home B8	40 (30)	3700 (2800)	4500 (3400)	29 (22)	0.75
Home B9	130 (83)	12000 (7700)	12000 (7800)	57 (36)	0.63
RSD[d]	52	82	80	120	36
RSD[e]	64	94	92	117	-
Home C1	51 (7.1)	3100 (440)	3200 (450)	<dl	0.14
Home C2	140 (210)	2400 (3600)	2500 (3800)	290 (430)	1.5
Home C3	74 (37)	1700 (850)	1800 (900)	1400 (690)	<dl
Home C4	120 (53)	1100 (480)	1200 (530)	270 (130)	0.47
Home C5	270 (190)	1600 (1100)	1900 (1400)	1100 (750)	0.71
Home C6	150 (70)	3800 (1800)	4000 (1900)	1500 (680)	0.47
Home C7	380 (160)	6500 (2700)	6900 (2900)	110 (45)	0.41

Home C8	250 (140)	1300 (690)	1600 (850)	1500 (780)	0.54
Home C9	210 (38)	36000 (6400)	36000 (6500)	70 (13)	0.18
RSD[d]	54	166	161	90	69
RSD[e]	70	93	89	83	-

[a]BDE-28 and TBE were detected in less than two samples in each room and are excluded
[b] sum of PBDEs 28, 47, 49, 66, 99, 100, 153, and 154
[c] sum of PBDEs 28, 47, 49, 66, 99, 100, 153, 154, 183, 196, 197, 203, and 209
[d]relative standard deviation of concentrations expressed as ng g^{-1}
[e]relative standard deviation of concentrations expressed as ng m^{-2}
- denotes not calculated for samples from this room

Until now, estimates of human exposure to PBDEs and related contaminants via the ingestion of indoor dust have been based on a "default" dust ingestion rate regardless of the dust loading of the room. However, this may not always be appropriate. To illustrate, consider two identically-dimensioned and ventilated rooms, containing identical PBDE emission strengths, but different dust loadings. In which will exposure be greater? One may hypothesise there to be a lower mass-based concentration of PBDE due to dilution in a dustier room, but will the reduced exposure arising from the lower mass-based PBDE concentration be mitigated to some degree by a greater dust ingestion rate in such a microenvironment? While there are no data addressing the influence of dust loading on dust ingestion rates (acquisition of such data is recommended), our data on the temporal variations in PBDE concentrations and dust loadings in Home A, offer an opportunity to evaluate the extent to which higher dust loadings dilute concentrations of PBDEs. A plot of dust loading versus PBDE concentration should be linear with a negative slope, provided: (a) PBDE emissions in the room remain essentially constant throughout the monitoring period, and (b) the sources of the dust and of PBDEs are independent. Considering the entire dataset for Home A for those congeners detected in all samples (Table 2), only BDE-153 displays a significant ($p<0.05$) negative correlation. However, inclusion of samples A8 and A10 violate condition (a) as they were taken during periods when significant changes occurred. If these samples are excluded, then BDE-47, BDE-99, and BDE-153, all display significant ($p<0.05$) negative correlation. Interestingly, no correlation was observed for BDE-209, suggesting the source of this congener may be related to the source of the dust (e.g. abraded fabric fibers), thus violating condition (b). While more data are required to evaluate fully the hypothesis that higher dust loadings dilute PBDE concentrations, unless the source of PBDEs and of dust are the same, our observations in this room suggest it may have some validity, and more detailed study is recommended.

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