

Supporting Information for:

Flame retardant concentrations are lower in university spaces meeting the new furniture flammability standard TB117-2013

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Analytical Methods

Materials and reagents

Solvents used during analysis were all of pesticide grade. *n*-hexane (Hex) was purchased from Acros Organics (Geel, Belgium). Acetone (Ac), dichloromethane (DCM), ethyl acetate (EA), *iso*-octane and methanol (MeOH) were purchased from Merck (Darmstadt, Germany).

Standards of BDE 28, 47, 85, 99, 100, 153, 154, 183, 196, 197, 203 and 209, α -HBCDD, β -HBCDD, γ -HBCDD, BTBPE, DBDPE, DBHCTD, EH-TBB, BEH-TEBP, HBB, TBBPA, DBE-DBCH isomers, TBP-AE, TBP-BAE, TBP-DBPE, OBTMPI, DP isomers, and labeled internal standards (IS) ¹³C-BDE 209, ¹³C- α -HBCDD, ¹³C- β -HBCDD, ¹³C- γ -HBCDD, and ¹³C-TBBPA were purchased from Wellington Laboratories (Guelph, ON, Canada). See Table SI1 for abbreviations and acronyms.

Standards of TPHP, TCEP, TEHP, EHDPP, TMP (mixture of 4 isomers), TDBPP and TDCIPP were purchased from Chiron AS (Trondheim, Norway). Triamyl phosphate (TAP; IS) was purchased from TCI Europe (Zwijndrecht, Belgium). Labeled TPP-d15 (IS) and TBOEP were purchased from Sigma Aldrich. TCPP (2 isomers) was purchased from Pfaltz & Bauer (Waterbury, CT, USA). Purity of analytical standards was >98%, except for TBOEP (>94%). Standard stock solutions were prepared in *iso*-octane, except for NBFRs which were prepared in a mixture of *iso*-octane:toluene (8:2, v/v).

Indoor dust SRM 2585 was purchased from the US National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA). Silica SPE cartridges (500 mg/3 mL, Bond Elut) were purchased from Agilent, while empty polypropylene tubes (3 mL) SPE cartridges and 500 mg/3 mL Supelclean ENVI- Florisil cartridges were purchased from Supelco (Bellefonte, PA, USA). Silica gel, anhydrous sodium sulfate (Na_2SO_4), and concentrated sulfuric acid (H_2SO_4 , 98%) were purchased from Merck. The preparation of acid impregnated silica (44%, w/w) was carried out as described elsewhere.¹ Glass test tubes were cleaned by soaking for at least 12 h in an alkali solution (diluted RBS 35, pH 11–12). After washing, the tubes were rinsed with water and dried at 100 °C for at least 12 h. The tubes were rinsed with Hex before use.

Sample Preparation

Due to the very comprehensive list of targeted flame retardants and the large differences in their physico-chemical properties, we used two separate sample preparation methods which led to three extracts per sample. These extracts were injected in various instruments, according to the expected presence of the FR groups.

Method I (Florisil fractionation)

The fractionation on Florisil was employed to measure the bulk of BFRs which elute in the first fraction (Fraction 1 – F1) and PFRs which elute in the 2nd fraction (Fraction 2 – F2). The method is largely based on the method described by Van den Eede et al.² In detail, a sample aliquot (around 30 mg) was accurately weighed and spiked with IS (^{13}C -BDE 209, BDE 77, BDE 128, CB 143, TCEP-d12, TBEP-d6, TDCPP-d15, TAP, and TPP-d15). Samples were extracted using 2.5 mL Hex:Ac (3:1 v/v) by a combination of vortexing and ultrasonic extraction (2 × 1 min vortex and 5 min ultrasonic extraction) which was repeated two times. After each extraction cycle, dust extracts were centrifuged at 3000 rpm for 3 min and supernatants were collected and transferred into clean glass tubes. The pooled supernatants were evaporated until dryness under a gentle nitrogen flow and redissolved in 1 mL Hex.

Prior to fractionation, Florisil® cartridges were prewashed with 6 mL of EA and 6 mL Hex. The extracts were quantitatively transferred and fractionation was achieved by eluting with 12 mL of Hex:DCM (1:1, v/v) (F1) and 10 mL of EA (F2). The 1st fraction (F1) was evaporated until 1 mL and quantitatively transferred onto acidified silica 5% cartridges (prewashed with 6 mL Hex) for a second clean-up. The target analytes were eluted with 10 mL of Hex:DCM (1:1 v/v), and afterwards evaporated until dryness under gentle nitrogen flow and reconstituted in 100 µL of *iso*-octane. The 2nd fraction (F2), containing PFRs, was evaporated until dryness under gentle nitrogen flow and resolubilized in 100 µL of *iso*-octane.

Fraction F1, containing PBDEs and NBFRs, was subjected to analysis by GC-ECNI/MS. The 2nd fraction (F2), containing PFRs, was subjected to analysis by GC-EI/MS.

Method II (Silica fractionation)

In detail, a sample aliquot (typically 30 mg) was accurately weighed and spiked with a mixture containing IS (^{13}C - α -, β -, γ -HBCDD, and ^{13}C -TBBPA). Samples were extracted using 2.5 mL Hex-Ac (3:1 v/v) by a combination of vortexing and ultrasonic extraction (2 × 1 min vortex and 5 min ultrasonic extraction) which was repeated three times. After each extraction cycle, dust extracts were centrifuged

at 3000 rpm for 3 min and supernatants were collected and transferred into clean glass tubes. The pooled supernatants were evaporated until dryness under a gentle nitrogen flow and redissolved in 1 mL Hex.

Prior to fractionation, silica cartridges were topped with 100 mg acid silica (44%) and prewashed with 6 mL of Hex. The extracts were quantitatively transferred and fractionation was achieved by eluting with 6 mL of Hex (discarded) and 12 mL of DCM (F3). This fraction contained HBCDDs and TBBPA and was resolubilized in 100 μ L of methanol and further subjected to LC-MS/MS analysis.

Chemical Analysis

GC/ECNI-MS Analysis

The analysis of F1, containing PBDEs and NBFRs was performed with an Agilent 6890 GC coupled to an Agilent 5973 MS operated in electrochemical negative ionization (ECNI) mode. The GC system was equipped with electronic pressure control and a programmable-temperature vaporizer (PTV). A volume of 2 μ L of cleaned extract was injected on a DB-5 column (15 m \times 0.25 mm \times 0.10 μ m) using solvent vent injection. The injection temperature was set at 90 $^{\circ}$ C, hold 0.04 min, ramp 700 $^{\circ}$ C/min to 295 $^{\circ}$ C. Vent time was 0.02 min and vent flow 75 mL/min. Injection was performed under a pressure of 10 psi until 1.25 min and purge flow to split vent of 50 mL/min after 1.25 min. The GC temperature program was 90 $^{\circ}$ C, hold 1.50 min, ramp 10 $^{\circ}$ C/min to 300 $^{\circ}$ C, hold 3 min, ramp 40 $^{\circ}$ C/min to 310 $^{\circ}$ C, hold 5 min. Helium was used as a carrier gas with a ramped flow rate of 1.0 mL/min until 20 min and then raised to 2.0 mL/min. The mass spectrometer was employed in selected ion monitoring (SIM) mode, with ions m/z 79 and 81 monitored the whole run time. For BDE 209, ions m/z 487 and 485 were used, while 13 C-BDE 209 was monitored using ions m/z 495 and 497. Dwell times were set on 35 ms. The ion source, quadrupole and interface temperatures were set at 250, 150 and 300 $^{\circ}$ C, respectively and the electron multiplier voltage was at 2200 V. Methane was used as moderating gas.

GC/EI-MS Analysis

Analysis of PFRs in fraction F2 was performed with an Agilent 6890 GC coupled to an Agilent 5973 MS operated in electron impact ionization (EI) mode. The GC system was equipped with electronic pressure control and a programmable-temperature vaporizer (PTV). One μ L of purified extract was injected on a HT-8 column (25 m \times 0.22 mm \times 0.25 μ m) using cold splitless injection. The injection temperature was set at 90 $^{\circ}$ C, hold 0.03 min, ramp 700 $^{\circ}$ C/min to 290 $^{\circ}$ C. Injection was performed using a pressure of 1 bar until 1.25 min and purge flow to split vent of 50 mL/min after 1.25 min. The GC temperature program was 90 $^{\circ}$ C, hold 1.25 min, ramp 10 $^{\circ}$ C/min to 240 $^{\circ}$ C, ramp 20 $^{\circ}$ C/min to 310 $^{\circ}$ C, hold 16 min. Helium was used as a carrier gas with a flow rate of 1.0 mL/min. The mass spectrometer was run in SIM mode. Dwell times ranged between 20 and 30 ms in different acquisition windows. The ion source, quadrupole and interface temperatures were set at 230, 150 and 300 $^{\circ}$ C, respectively, and the electron multiplier voltage was at 2200 V.

LC-MS/MS

The determination of individual HBCD isomers and TBBPA in the Fraction 3 (silica fractionation) was achieved using a dual pump Agilent 1100 Series liquid chromatograph equipped with autosampler and vacuum degasser. A Luna C18(2) reversed phase (RP) analytical column (150 mm \times 2 mm i.d., 3 μ m particle size, Phenomenex) was used for the separation of α -, β -, and γ -HBCDD. A mobile phase of (A) ammonium acetate 2mM in water/methanol (1:1 v/v) and (B) methanol at a flow rate of 0.250 mL/min was applied for elution of HBCDD isomers; starting at 75% (B) held for 2 min, then increased linearly to 100% (b) until 9 min; held until 12 min followed by a linear decrease to 70% (B) over 0.5 min and held for 7.5 min.

The target analytes were baseline separated on the RP column with retention times of 4.0, 6.0, 6.8 and 7.4 min for TBBPA, α -, β - and γ -HBCDD, respectively. MS analysis was performed using an Agilent 6410 triple quadrupole MS system operated in the electrospray negative ionization mode. N₂ was used as drying gas at a flow of 10 L/min and heated to 300 °C. Nebulizer pressure was 35 psi and capillary voltage 4000 V. HBCDD isomers were quantified by isotope dilution. MS/MS detection operated in the MRM mode was used for quantitative determination of the HBCDD isomers based on m/z 640.6 to 81 and m/z 652.6 to 81 transitions for the native and ¹³C-labeled diastereomers, respectively. Fragmentor voltage and collision energy were set as 80 and 15 V, respectively. For quantitative determination of TBBPA, the following MRMs were used: m/z 542.6 to 81 and m/z 554.6 to 81 for the native and ¹³C-TBBPA, respectively.

Quality Control

In the 2015 batch, three field blanks and four procedural blanks were analyzed in the same batches as the samples and results are blank corrected. This implies subtraction of mean blank values (in pg) from the raw FR values (in pg) in the samples. Blank values, when detected, were <0.5% of minimum sample values in the batch. In the 2016 batch, three field blanks were analyzed; they were all non-detect. Field blanks were collected by field staff vacuuming 2.8 grams of sodium sulfate from a stainless steel tray. In the 2018 batch, two procedural blanks were analyzed; they were all non-detect.

Method limits of quantification (LOQs) were calculated as three times the standard deviation of blank values and divided by the amount of dust used for analysis (typically 30 mg). For compounds not detected in the blanks, the LOQ was calculated based on the signal to noise ratio 10/1, taking into account also the chromatogram's characteristics for the respective retention time (co-elution, noisy baseline, etc). LOQs are compound-specific variables and therefore spanned a large range of concentrations. We replaced non-detect values with the limit of detection (LOD)*detection frequency for the chemical. For chemicals with multiple isomers, such as TCIPP, TCP, DP, and HBCDD, we replaced non-detect values using this method for individual isomers before summing them.

The method has been validated as described by Van den Eede et al.² A series of optimization and spiking experiments were performed for BFRs and PFRs at two concentration levels, Q_{low} and Q_{high} , and three replicates for each level. Precision between different days were assessed using the same concentration levels spiked on a low contaminated dust sample, using three replicates per level and executed on three different days. Precision was within 12% for each set of triplicates and all analytes. The recovery was calculated by subtracting the blank concentrations and divided by the calculated concentration of a mixed solution of standards (having the same concentrations). Further details can be found in Van den Eede et al.²

SRM 2585 (Organic Contaminants in House Dust), which has certified values for PBDEs and indicative values for EH-TBB, BEH-TEBP, HBCDDs, chlorinated PFRs and TBOEP, was used to test the accuracy (Figure SI1). Concentrations of PBDEs, EH-TBB, BEH-TEBP, chlorinated PFRs and HBCDDs ranged between -9 and 30% relative difference from the certified values. Despite a few discrepancies, there does not appear to be a systematic bias to the samples and values were not adjusted.

The Toxicological Centre participated in 2015 in the 2nd Interlaboratory test on novel flame retardants (INTERFLAB2) in standard solutions and house dust and in 2018 to the 3rd Interlaboratory test on novel flame retardants (INTERFLAB3) in standard solutions and dust from e-waste facilities. Obtained values were between 5 and 25% from the consensus values for most investigated analytes. The full reports can be provided upon request.

All sample preparation, chemical analysis, and quality control measures are also described in Dodson et al. 2017³.

Table SI1. Measured flame retardants concentrations (ng/g dust; n=86, except TBBPA n=84)

FR Name	Abbrev.	%>LOQ	Median	Mean	GM	95th%tile	Max.
<i>Polybrominated Diphenyl Ethers</i>							
2,4,4'-tribromodiphenyl ether	BDE 28	76	5.6	30	6	140	480
2,2',4,4'-tetrabromodiphenyl ether	BDE 47	100	390	2800	470	14000	43000
2,2',3,4,4'-pentabromodiphenyl ether	BDE 85	99	26	200	36	870	3100
2,2',4,4',5-pentabromodiphenyl ether	BDE 99	100	520	3300	670	17000	52000
2,2',4,4',6-pentabromodiphenyl ether	BDE 100	100	84	710	110	3500	11000
2,2',4,4',5,5'-hexabromodiphenyl ether	BDE 153	99	47	390	65	1600	6900
2,2',4,4',5,6'-hexabromodiphenyl ether	BDE 154	100	44	330	62	1700	4500
2,2',3,4,4',5',6-heptabromodiphenyl ether	BDE 183	92	12	58	17	260	540
2,2',3,3',4,4',5,6'-octabromodiphenyl ether	BDE 196	91	18	150	22	630	5600
2,2',3,3',4,4',6,6'-octabromodiphenyl ether	BDE 197	63	5.3	30	6.9	170	430
2,2',3,4,4',5,5',6-octabromodiphenyl ether	BDE 203	71	5.6	42	7.6	150	1300
decabromodiphenyl ether	BDE 209	100	2400	88000	3500	350000	3500000
<i>Brominated Flame Retardants</i>							
2-ethylhexyl-2,3,4,5-tetrabromobenzoate	EH-TBB	100	690	9000	1100	24000	180000
bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate	BEH-TEBP	99	1300	3600	1500	15000	37000
tetrabromobisphenol A	TBBPA	94	95	1400	110	4200	66000
tetrabromochlorotoluene	TBCT	14	NA	NA	NA	14	670
para-tetra-bromoxylene	pTBX	8	NA	NA	NA	5.3	46
pentabromotoluene	PBT	34	NA	NA	NA	12	370
pentabromoethylbenzene	PBEB	30	NA	NA	NA	16	230
pentabromobenzene	PBBz	51	0.8	7.3	1.6	44	120
pentabromobenzylacrylate	PBBA	19	NA	NA	NA	65	670
hexabromobenzene	HBB	85	5	45	6.4	170	1100
hexachlorocyclopentadienyl-dibromocyclooctane	DBHCTD	19	NA	NA	NA	21	50
1,2-bis(2,4,6-tribromophenoxy)ethane	BTBPE	56	8.1	98	9.8	320	4300
decabromodiphenylethane	DBDPE	90	270	3100	260	8600	120000
alpha-1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane	a-DBE-DBCH	37	NA	NA	NA	44	260
beta-1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane	b-DBE-DBCH	40	NA	NA	NA	37	560

2-bromoallyl-2,4,6-tribromophenyl ether	TBP-BAE	9	NA	NA	NA	17	100
2,4,6-tribromophenyl allyl ether	TBP-AE	23	NA	NA	NA	6	140
2,4,6-tribromophenyl 2,3-dibromopropyl ether	TBP-DBPE	37	NA	NA	NA	28	160
octabromo-1,3,3-trimethyl-1-phenylindan	OBTMPI	29	NA	NA	NA	72	2800
<i>Dechlorane Plus</i>							
dechlorane plus	DPs	100	23	170	31	510	5100
<i>Hexabromocyclododecane</i>							
Hexabromocyclododecane	HBCDD	92	320	7700	340	5600	300000
<i>Organophosphate Flame Retardants</i>							
tris(2-chloroethyl) phosphate	TCEP	99	360	25000	500	24000	1300000
tris(1-chloroisopropyl) phosphate	TCIPP	100	2900	8800	3600	34000	150000
tris(1,3-dichloro-isopropyl) phosphate	TDCIPP	100	3800	41000	5500	250000	990000
tris(2,3-dibromopropyl) phosphate	TDBPP	37	NA	NA	NA	210	1400
triphenyl phosphate	TPHP	100	5700	11000	5800	46000	76000
tri-(2-butoxyethyl) phosphate	TBOEP	100	59000	180000	75000	800000	1500000
tri-(2-ethylhexyl) phosphate	TEHP	70	180	310	160	1100	1900
tris(4-butylphenyl) phosphate	TBuPhP	63	28	75	35	380	620
ethylhexyl diphenyl phosphate	EHDPP	100	930	2500	1000	8800	42000
tri-cresyl phosphate	TMPP	87	420	3000	450	7100	120000

Triplicate relative standard deviation for triplicates run in 2018 batch: anti-DP: 96.6%, syn-DP: 78.3, DBDPE: 93%, BDE 209: 68.8%, TBBPA: 53.2%, BDE 100: 40.8%, BDE 154: 37.5%, BDE 47: 34%, TDCIPP: 32.9%, BDE 153: 30.9%.

Table SI2. Comparisons of median and maximum values for 14 FRs with values reported in North American dust since 2006.

Chemical	Highest median ^a	References for comparison	Highest maximum ^b	References for comparison
BDE 47				
BDE 99				
BDE 183				
BDE 209			✓	[3-22]
EH-TBB			✓	[3, 6, 7, 9-13, 15-17, 23-29]
BEH-TEBP				
DBDPE	✓	[3, 9, 12, 13, 15, 16, 25, 27]	✓	[3, 9, 12, 13, 15, 16, 25, 27]
HBB				
TBBPA				
HBCDD				
TCEP			✓	[3, 4, 6, 9, 11-13, 15, 16, 28-31]
TCIPP				
TDCIPP				
TPHP				

^ageometric means used for comparison if median values not reported

^b95% values reported if maxima not reported

Table SI3. Estimated marginal means (95% confidence intervals) from baseline, adjusted, and post-2004 adjusted models

FR		TB133	TB117	TB117-2013
BDE 47	baseline	540 (310, 930)*	870 (440, 1700)*	110 (46, 280)
	adjusted	690 (400, 1200)*	1600 (740, 3600)*	110 (53, 250)
	adjusted (furnished after 2004)	300 (110, 780)	1400 (460, 4400)*	98 (44, 220)
BDE 99	baseline	750 (460, 1200)*	1300 (690, 2400)*	170 (73, 380)
	adjusted	1100 (700, 1700)*	2400 (1200, 4600)*	190 (99, 350)
	adjusted (furnished after 2004)	660 (300, 1400)*	2100 (840, 5200)*	170 (89, 320)
BDE 183	baseline	15 (10, 23)*	38 (23, 63)*	5.1 (2.6, 9.7)
	adjusted	18 (12, 29)*	28 (15, 54)*	5 (2.7, 9.4)
	adjusted (furnished after 2004)	19 (8.7, 43)*	19 (7.4, 48)*	5 (2.6, 9.7)
BDE 209	baseline	11000 (6200, 18000)*	1500 (760, 2800)	760 (320, 1800)
	adjusted	11000 (5700, 22000)*	2100 (790, 5600)	820 (320, 2100)
	adjusted (furnished after 2004)	4700 (1700, 13000)*	3100 (950, 10000)*	590 (250, 1400)
EH-TBB	baseline	860 (530, 1400)	2200 (1200, 4000)*	540 (240, 1200)
	adjusted	920 (500, 1700)	2200 (920, 5300)*	520 (220, 1200)
	adjusted (furnished after 2004)	1700 (530, 5200)	4100 (1100, 16000)*	470 (180, 1200)
BEH-TEBP	baseline	2100 (1400, 3100)*	1200 (730, 2000)	990 (520, 1900)
	adjusted	2000 (1300, 3100)*	1400 (730, 2600)	810 (440, 1500)
	adjusted (furnished after 2004)	1700 (790, 3600)	2100 (870, 5100)	790 (420, 1500)
DBDPE	baseline	760 (420, 1400)*	80 (38, 170)	110 (41, 280)
	adjusted	850 (430, 1600)*	170 (65, 440)	120 (49, 310)
	adjusted (furnished after 2004)	440 (120, 1600)	220 (51, 990)	98 (34, 280)
HBB	baseline	15 (9.7, 23)*	2.3 (1.3, 3.9)	3.7 (1.8, 7.5)
	adjusted	15 (8.5, 25)*	2.5 (1.1, 5.5)	3.5 (1.7, 7.6)
	adjusted (furnished after 2004)	17 (5.7, 50)*	3.6 (1, 13)	3.4 (1.4, 8.3)
TBBPA	baseline	180 (110, 300)*	110 (57, 200)	40 (17, 91)
	adjusted	170 (93, 320)*	63 (26, 150)	35 (15, 82)
	adjusted (furnished after 2004)	440 (120, 1600)*	53 (11, 250)	31 (11, 92)
HBCDD	baseline	670 (420, 1100)*	300 (160, 540)*	72 (33, 160)
	adjusted	960 (530, 1700)*	300 (130, 700)*	71 (31, 160)
	adjusted (furnished after 2004)	3100 (880, 11000)*	360 (82, 1600)	85 (30, 240)
TCEP	baseline	1000 (580, 1800)*	330 (160, 680)	140 (54, 370)
	adjusted	1800 (870, 3600)*	1100 (390, 3000)*	170 (63, 450)
	adjusted (furnished after 2004)	310 (160, 600)*	380 (180, 810)*	130 (74, 210)
TCIPP	baseline	4200 (3000, 5900)	2700 (1800, 4100)	4000 (2300, 7000)
	adjusted	3900 (2700, 5800)	2700 (1500, 4700)	4200 (2400, 7100)
	adjusted (furnished after 2004)	3200 (1700, 6300)	3500 (1600, 7600)	3900 (2300, 6800)
TDCIPP	baseline	6600 (4000, 11000)	5700 (3000, 11000)	3000 (1300, 6900)
	adjusted	7600 (3900, 15000)	5300 (2100, 14000)	2700 (1100, 6600)
	adjusted (furnished after 2004)	2400 (1200, 4900)	4300 (1900, 9800)	2300 (1300, 4000)
TPHP	baseline	5000 (3600, 6900)	6000 (4000, 9000)	8400 (4900, 14000)
	adjusted	5700 (3800, 8700)	7200 (4000, 13000)	8400 (4700, 15000)
	adjusted (furnished after 2004)	4600 (1900, 11000)	9600 (3500, 27000)	7400 (3600, 15000)

* $p < 0.05$, and ** $p < 0.005$.

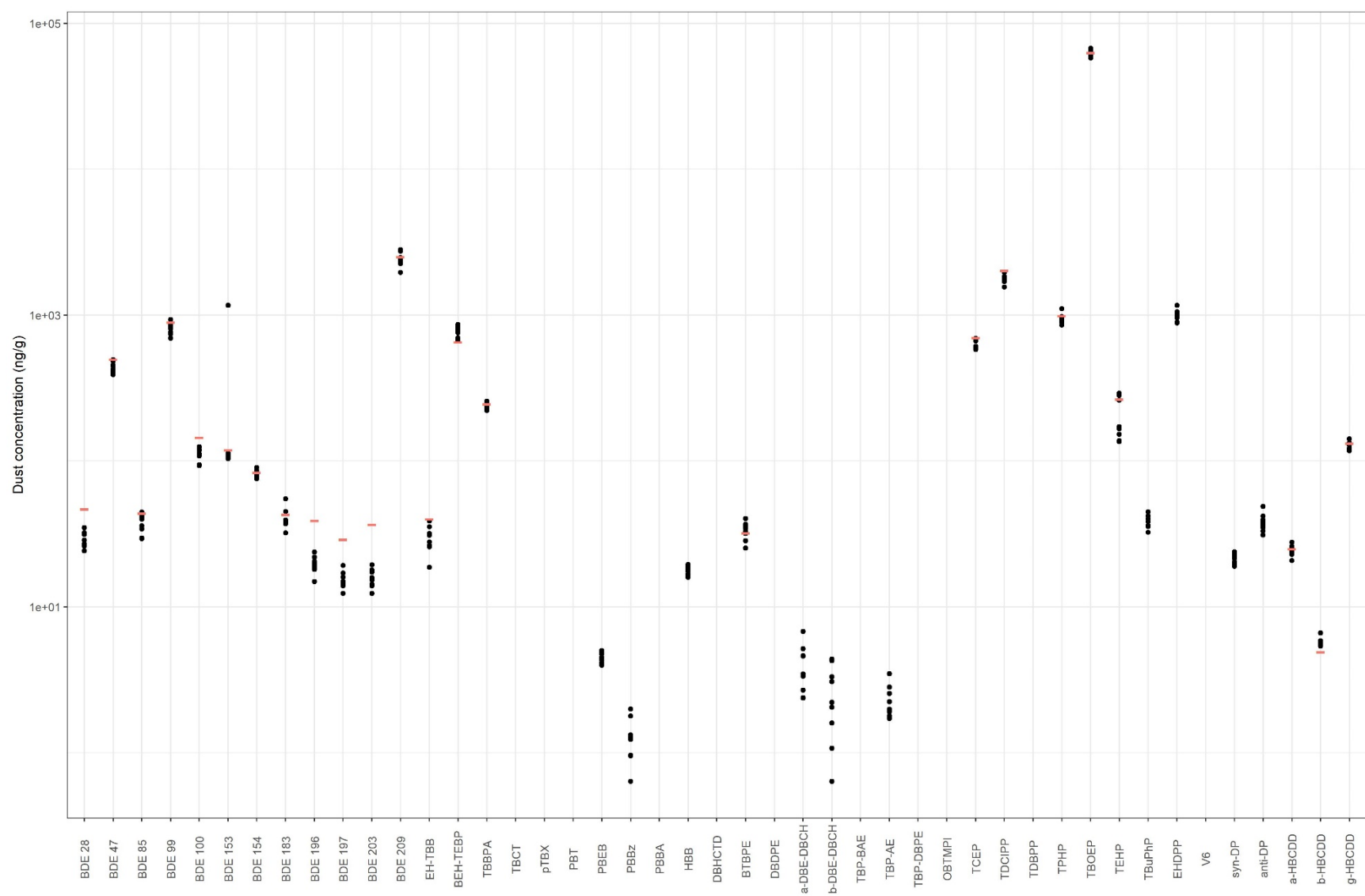


Figure S11. Comparison of Standard Reference Material (SRM) (n=9; black dots) with available certified/indicative values (red dashes).



Figure SI2: Priority flame retardant uses and applications from EPA Chemical/Product Categories Database³²

References

1. Covaci, A.; Schepens, P., Simplified method for determination of organochlorine pollutants in human serum by solid-phase disk extraction and gas chromatography. *Chemosphere* **2001**, *43*, (4-7), 439-47.
2. Van den Eede, N.; Dirtu, A. C.; Ali, N.; Neels, H.; Covaci, A., Multi-residue method for the determination of brominated and organophosphate flame retardants in indoor dust. *Talanta* **2012**, *89*, 292-300.
3. Dodson, R. E.; Rodgers, K. M.; Carey, G.; Cedeno Laurent, J. G.; Covaci, A.; Poma, G.; Malarvannan, G.; Spengler, J. D.; Rudel, R. A.; Allen, J. G., Flame Retardant Chemicals in College Dormitories: Flammability Standards Influence Dust Concentrations. *Environ Sci Technol* **2017**, *51*, (9), 4860-4869.
4. Shen, B.; Whitehead, T. P.; Gill, R.; Dhaliwal, J.; Brown, F. R.; Petreas, M.; Patton, S.; Hammond, S. K., Organophosphate flame retardants in dust collected from United States fire stations. *Environ Int* **2018**, *112*, 41-48.
5. Watkins, D. J.; McClean, M. D.; Fraser, A. J.; Weinberg, J.; Stapleton, H. M.; Sjodin, A.; Webster, T. F., Exposure to PBDEs in the office environment: evaluating the relationships between dust, handwipes, and serum. *Environmental Health Perspectives* **2011**, *119*, (9), 1247-52.
6. Stapleton, H. M.; Misenheimer, J.; Hoffman, K.; Webster, T. F., Flame retardant associations between children's handwipes and house dust. *Chemosphere* **2014**, *116*, 54-60.
7. Hoffman, K.; Garantzotis, S.; Birnbaum, L. S.; Stapleton, H. M., Monitoring indoor exposure to organophosphate flame retardants: hand wipes and house dust. *Environ Health Perspect* **2015**, *123*, (2), 160-5.
8. Batterman, S.; Godwin, C.; Chernyak, S.; Jia, C.; Charles, S., Brominated flame retardants in offices in Michigan, USA. *Environ Int* **2010**, *36*, (6), 548-56.
9. Allgood, J. M.; Jimah, T.; McClaskey, C. M.; La Guardia, M. J.; Hammel, S. C.; Zeineddine, M. M.; Tang, I. W.; Runnerstrom, M. G.; Ogunseitan, O. A., Potential human exposure to halogenated flame-retardants in elevated surface dust and floor dust in an academic environment. *Environ Res* **2017**, *153*, 55-62.
10. Hammel, S. C.; Hoffman, K.; Lorenzo, A. M.; Chen, A.; Phillips, A. L.; Butt, C. M.; Sosa, J. A.; Webster, T. F.; Stapleton, H. M., Associations between flame retardant applications in furniture foam, house dust levels, and residents' serum levels. *Environ Int* **2017**, *107*, 181-189.
11. Bradman, A.; Castorina, R.; Gaspar, F.; Nishioka, M.; Colon, M.; Weathers, W.; Egeghy, P. P.; Maddalena, R.; Williams, J.; Jenkins, P. L.; McKone, T. E., Flame retardant exposures in California early childhood education environments. *Chemosphere* **2014**, *116*, 61-6.
12. Dodson, R. E.; Perovich, L. J.; Covaci, A.; Van den Eede, N.; Ionas, A. C.; Dirtu, A. C.; Brody, J. G.; Rudel, R. A., After the PBDE phase-out: a broad suite of flame retardants in repeat house dust samples from California. *Environ Sci Technol* **2012**, *46*, (24), 13056-66.
13. Schreder, E. D.; La Guardia, M. J., Flame retardant transfers from U.S. households (dust and laundry wastewater) to the aquatic environment. *Environmental Science & Technology* **2014**, *48*, (19), 11575-83.
14. Bennett, D. H.; Moran, R. E.; Wu, X. M.; Tulse, N. S.; Clifton, M. S.; Colon, M.; Weathers, W.; Sjodin, A.; Jones, R.; Hertz-Picciotto, I., Polybrominated diphenyl ether (PBDE) concentrations and resulting exposure in homes in California: relationships among passive air, surface wipe and dust concentrations, and temporal variability. *Indoor Air* **2015**, *25*, (2), 220-9.

15. Stubbings, W. A.; Schreder, E. D.; Thomas, M. B.; Romanak, K.; Venier, M.; Salamova, A., Exposure to brominated and organophosphate ester flame retardants in U.S. childcare environments: Effect of removal of flame-retarded nap mats on indoor levels. *Environ Pollut* **2018**, *238*, 1056-1068.
16. La Guardia, M. J.; Hale, R. C., Halogenated flame-retardant concentrations in settled dust, respirable and inhalable particulates and polyurethane foam at gymnastic training facilities and residences. *Environ Int* **2015**, *79*, 106-14.
17. Cowell, W. J.; Stapleton, H. M.; Holmes, D.; Calero, L.; Tobon, C.; Perzanowski, M.; Herbstman, J. B., Prevalence of historical and replacement brominated flame retardant chemicals in New York City homes. *Emerg Contam* **2017**, *3*, (1), 32-39.
18. Allen, J. G.; McClean, M. D.; Stapleton, H. M.; Webster, T. F., Linking PBDEs in house dust to consumer products using X-ray fluorescence. *Environmental Science & Technology* **2008**, *42*, (11), 4222-8.
19. Stapleton, H. M.; Eagle, S.; Sjodin, A.; Webster, T. F., Serum PBDEs in a North Carolina toddler cohort: associations with handwipes, house dust, and socioeconomic variables. *Environmental Health Perspectives* **2012**, *120*, (7), 1049-54.
20. Sjodin, A.; Papke, O.; McGahee, E.; Focant, J. F.; Jones, R. S.; Pless-Mulloli, T.; Toms, L. M.; Herrmann, T.; Muller, J.; Needham, L. L.; Patterson, D. G., Jr., Concentration of polybrominated diphenyl ethers (PBDEs) in household dust from various countries. *Chemosphere* **2008**, *73*, (1 Suppl), S131-6.
21. Darrow, L. A.; Jacobson, M. H.; Preston, E. V.; Lee, G. E.; Panuwet, P.; Hunter, R. E., Jr.; Marder, M. E.; Marcus, M.; Barr, D. B., Predictors of Serum Polybrominated Diphenyl Ether (PBDE) Concentrations among Children Aged 1-5 Years. *Environ Sci Technol* **2017**, *51*, (1), 645-654.
22. Imm, P.; Knobeloch, L.; Buelow, C.; Anderson, H. A., Household exposures to polybrominated diphenyl ethers (PBDEs) in a Wisconsin Cohort. *Environ Health Perspect* **2009**, *117*, (12), 1890-5.
23. Stapleton, H. M.; Klosterhaus, S.; Eagle, S.; Fuh, J.; Meeker, J. D.; Blum, A.; Webster, T. F., Detection of Organophosphate Flame Retardants in Furniture Foam and US House Dust. *Environmental Science & Technology* **2009**, *43*, (19), 7490-7495.
24. Johnson, P. I.; Stapleton, H. M.; Mukherjee, B.; Hauser, R.; Meeker, J. D., Associations between brominated flame retardants in house dust and hormone levels in men. *Science of the Total Environment* **2013**, *445-446*, 177-84.
25. Brown, F. R.; Whitehead, T. P.; Park, J. S.; Metayer, C.; Petreas, M. X., Levels of non-polybrominated diphenyl ether brominated flame retardants in residential house dust samples and fire station dust samples in California. *Environ Res* **2014**, *135*, 9-14.
26. Fan, X.; Kubwabo, C.; Rasmussen, P. E.; Wu, F., Non-PBDE halogenated flame retardants in Canadian indoor house dust: sampling, analysis, and occurrence. *Environmental Science and Pollution Research International* **2016**, *23*, (8), 7998-8007.
27. Stapleton, H. M.; Allen, J. G.; Kelly, S. M.; Konstantinov, A.; Klosterhaus, S.; Watkins, D.; McClean, M. D.; Webster, T. F., Alternate and new brominated flame retardants detected in US house dust. *Environmental Science & Technology* **2008**, *42*, (18), 6910-6916.
28. Percy, Z.; La Guardia, M. J.; Xu, Y.; Hale, R. C.; Dietrich, K. N.; Lanphear, B. P.; Yoltan, K.; Vuong, A. M.; Cecil, K. M.; Braun, J. M.; Xie, C.; Chen, A., Concentrations and loadings of organophosphate and replacement brominated flame retardants in house dust from the home study during the PBDE phase-out. *Chemosphere* **2020**, *239*, 124701.
29. Castorina, R.; Bradman, A.; Stapleton, H. M.; Butt, C.; Avery, D.; Harley, K. G.; Gunier, R. B.; Holland, N.; Eskenazi, B., Current-use flame retardants: Maternal exposure and neurodevelopment in children of the CHAMACOS cohort. *Chemosphere* **2017**, *189*, 574-580.
30. Keimowitz, A. R.; Strunsky, N.; Wovkulich, K., Organophosphate flame retardants in household dust before and after introduction of new furniture. *Chemosphere* **2016**, *148*, 467-72.
31. Liu, X.; Cao, Z.; Yu, G.; Wu, M.; Li, X.; Zhang, Y.; Wang, B.; Huang, J., Estimation of Exposure to Organic Flame Retardants via Hand Wipe, Surface Wipe, and Dust: Comparability of Different Assessment Strategies. *Environ Sci Technol* **2018**, *52*, (17), 9946-9953.

32. U.S. Environmental Protection Agency, CPChem: Chemical and Product Categories.
<http://actor.epa.gov/cpcat> (July 23, 2020).