

Are we exposed to halogenated flame retardants from both primary and secondary sources?

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Table S1: List of NFRs and PBDEs for analysis, along with CAS numbers, molecular formula, molecular weight, and LogK_{ow} and LogK_{OA}^{1,2}

	Full Name	CAS, Molecular Formula	Molecular weight (g/mol)	LogK _{ow}	LogK _{OA}
Novel flame-retardants (NFRs)					
TBP-AE	Allyl 2,4,6-tribromophenyl ether Also known as: 1,3,5-Tribromo-2-(prop-2-en-1-yloxy)benzene	3278-89-5, C ₉ H ₇ Br ₃ O	371	5.6	8.6
PBBz	Pentabromobenzene	608-90-2, C ₆ HBr ₅	473	6.4	9.1
PBT	Pentabromotoluene	87-83-2, C ₇ H ₃ Br ₅	487	7.0	9.6
PBEB	Pentabromoethyl benzene	85-22-3, C ₈ H ₅ Br ₅	501	7.5	10
HBB	Hexabromobenzene	87-82-1, C ₆ Br ₆	552	7.3	10.4
EH-TBB	Ethylhexyl-tetrabromobenzoate	26040-51-7, C ₁₅ H ₁₉ Br ₄ O ₂	550	8.8	12.3
BEH-TEBP	Bis(2-ethylhexyl) tetrabromophthalate	26040-51-7, C ₂₄ H ₃₄ Br ₄ O ₄	706	12	16.9
s-DDC-CO	Dechlorane Plus (syn and anti isomers)	13560-89-9, C ₁₈ H ₁₂ Cl ₁₂	654	11.3	14.8
a-DDC-CO	Dechlorane Plus (syn and anti isomers)	13560-89-9, C ₁₈ H ₁₂ Cl ₁₂	654	11.3	14.8
OBTMPI	Octabromotrimethylphenyl indane	1084889-51-9, C ₁₈ H ₁₂ Br ₈	868	13	17.8
DBDPE	Decabromodiphenylethane	84852-53-9, C ₁₄ H ₄ Br ₁₀	971	13.6	19.2
Polybrominated diphenyl ethers (PBDEs)					
BDE-17	2,2',4-Tribromodiphenyl ether	147217-75-2, C ₁₃ H ₉ Br ₃ O ₂	437	5.88	9.3
BDE-28	2,4,4'-Tribromodiphenyl ether	41318-75-6, C ₁₂ H ₇ Br ₃ O	407	5.88	9.5
BDE-47	2,2',4,4'-Tetrabromodiphenyl ether	5436-43-1, C ₁₂ H ₆ Br ₄ O	486	6.77	10.5
BDE-49	2,2',4,5'-Tetrabromodiphenyl ether	243982-82-3, C ₁₂ H ₆ Br ₄ O	486	6.37*	-
BDE-66	2,3',4,4'-Tetrabromodiphenyl ether	189084-61-5, C ₁₂ H ₆ Br ₄ O	486	6.77	10.8
BDE-71	2,3',4',6-Tetrabromodiphenyl ether	189084-62-6, C ₁₂ H ₆ Br ₄ O	486	6.77	10.4
BDE-85	2,2',3,4,4'-Pentabromodiphenyl ether	182346-21-0, C ₁₂ H ₅ Br ₅ O	565	7.66	11.7
BDE-99	2,2',4,4',5-Pentabromodiphenyl ether	60348-60-9, C ₁₂ H ₅ Br ₅ O	565	7.66	11.3
BDE-100	2,2',4,4',6-Pentabromodiphenyl ether	189084-64-8, C ₁₂ H ₅ Br ₅ O	565	7.66	11.1
BDE-138	2,2',3,4,4',5'-Hexabromodiphenyl ether	182677-30-1, C ₁₂ H ₄ Br ₆ O	644	8.55	-
BDE-153	2,2',4,4',5,5'-Hexabromodiphenyl ether	68631-49-2, C ₁₂ H ₄ Br ₆ O	644	8.55	11.8
BDE-154	2,2',4,4',5,6'-Hexabromodiphenyl ether	207122-15-4, C ₁₂ H ₄ Br ₆ O	644	8.55	11.9
BDE-183	2,2',3,4,4',5',6-Heptabromodiphenyl ether	207122-16-5, C ₁₂ H ₃ Br ₇ O	722	9.44	12
BDE-190	2,2',3',4,4',5',6-Heptabromodiphenyl ether	189084-68-2, C ₁₂ H ₃ Br ₇ O	722	9.44	-
BDE-209	Decabromodiphenyl ether	1163-19-5, C ₁₂ Br ₁₀ O	959	12.1	-

* calculated in Virtual Computational Chemistry Laboratory (<http://www.vcclab.org>)

-: not available

Table S2: Method detection limits (MDL) for sample types. MDL for cell phones (wiped area = 50 cm^2) is 2 times of that for other electronic products (wiped area = 100 cm^2).

		Air (pg/m^3)	Dust (ng/g)	Product Wipes (pg/cm^2)	Hand Wipes (pg/cm^2)
NFRs	TBP-AE	0.14	0.10	0.08	0.03
	PBBz	0.03	0.03	0.02	0.01
	PBT	0.07	0.05	0.05	0.02
	PBEB	0.07	0.05	0.04	0.02
	HBB	0.03	0.02	0.02	0.01
	EH-TBB	1.4	1.0	0.8	0.30
	BEH-TEBP	3.5	2.5	2.0	0.80
	s-DDC-CO	0.34	0.25	0.2	0.08
	a-DDC-CO	0.34	0.25	0.2	0.08
	OBTMPI	7.0	5.0	4.0	1.6
PBDEs	DBDPE	8.4	6.0	4.8	2.0
	BDE-17	0.22	0.15	0.12	0.05
	BDE-28	0.22	0.15	0.12	0.05
	BDE-47	0.22	0.15	0.12	0.05
	BDE-49	0.22	0.15	0.12	0.05
	BDE-66	0.22	0.15	0.12	0.05
	BDE-71	0.22	0.15	0.12	0.05
	BDE-85	0.22	0.15	0.12	0.05
	BDE-99	0.22	0.15	0.12	0.05
	BDE-100	0.22	0.15	0.12	0.05
	BDE-138	0.29	0.20	0.16	0.06
	BDE-153	0.29	0.15	0.16	0.06
	BDE-154	0.22	0.15	0.12	0.05
	BDE-183	0.29	0.20	0.16	0.06
	BDE-190	0.14	0.10	0.08	0.03
	BDE-209	18	13	10	4.5

Table S3: Analysis of SRM 2585 house dust samples (ng/g; n=5) for NFRs and PBDEs.

		SRM measurements (n=5)	Certified or reference values	Average percentage of certified or indicative values (%)
NFRs	PBT	nd	nd ^a	-
	PBEB	4.3	9.0 ^a	48
	HBB	4.8	4.5 ^a	110
	EH-TBB	90	26-54 ^{a-d}	350-170
	BEH-TEBP	2300	500-1300 ^{a-d}	450-170
	DBDPE	nd	nd ^{a-d}	-
PBDEs	BDE-17	12	12 ^e	100
	BDE-28	47	47 ^e	100
	BDE-47	500	500 ^e	100
	BDE-85	53	44 ^e	110
	BDE-99	1000	890 ^e	110
	BDE-100	150	150 ^e	99
	BDE-138	16	15 ^e	100
	BDE-153	140	120 ^e	110
	BDE-154	110	84 ^e	120
	BDE-183	49	43 ^e	110
	BDE-209	2100	2500 ^e	94

^a Brown et al., 2014 (n=7)

^b Van den Eede et al., 2012 (n=6)

^c Sahlström et al., 2012 (n=5)

^d Ali et al., 2011 (n=2)

^e SRM 2585, NIST

Table S4: Recoveries (%) of NFRs and PBDEs and surrogate standards in spiked wipe blanks (n=3).

Compound	Sample 1	Sample 2	Sample 3	Mean
TBP-AE	95	110	94	98
PBBz	100	110	98	100
PBT	93	97	91	94
PBEB	94	97	90	94
HBB	110	110	110	110
EH-TBB	96	98	95	97
BEH-TEBP	91	89	100	94
s-DDC-CO	110	99	110	110
a-DDC-CO	110	100	110	110
OBTMPI	45	48	65	53
DBDPE	82	83	89	85
BDE-17	100	100	97	100
BDE-28	100	100	96	100
BDE-47	98	99	94	97
BDE-66	96	96	89	94
BDE-71	95	97	91	94
BDE-85	96	97	95	96
BDE-99	97	98	94	96
BDE-100	98	100	100	100
BDE-138	95	95	100	97
BDE-153	99	99	96	98
BDE-154	94	93	87	91
BDE-183	96	94	100	97
BDE-190	93	93	110	97
BDE-209	79	81	84	81
Surrogate Standards				
mPBBz	100	110	99	100
mHBB	110	110	100	110
F-BDE-100	100	110	99	100
F-BDE-154	92	95	90	92
F-BDE-208	62	68	85	72

Table S5: Spearman's rho correlation coefficients for NFRs and PBDEs in bedroom dust for compounds with >70% DF. Only significant correlations are indicated.

BR Dust	PBDEs												
	BDE-17	BDE-28	BDE-47	BDE-49	BDE-66	BDE-85	BDE-99	BDE-100	BDE-138	BDE-153	BDE-154	BDE-183	BDE-209
BDE_17	1												
BDE_28	.928	1											
BDE_47	.866	.909	1										
BDE_49	.751	.733	.716	1									
BDE_66	.837	.814	.931	.668	1								
BDE_85	.731	.793	.895	.610	.855	1							
BDE_99	.786	.848	.960	.661	.908	.950	1						
BDE_100	.739	.777	.887	.583	.847	.920	.944	1					
BDE_138	.676	.700	.852	.554	.845	.931	.932	.918	1				
BDE_153	.691	.676	.783	.607	.808	.842	.855	.833	.881	1			
BDE_154	.782	.828	.926	.653	.883	.964	.982	.959	.955	.893	1	.	
BDE_183					.355	.432	.343	.383	.504	.424	.415	1	
BDE_209													1

Table S6: p-values from Wilcoxon signed rank tests for differences of NFRs and PBDEs between paired samples in the air and dust of bedrooms (BRs) and most used rooms (MURs) and paired hand palms versus backs (in blue). p-values for Mann-Whitney U tests for differences between hand-held versus non-hand-held e-devices (in red) with median concentrations in parentheses. Hand-held devices were cell phones, home phones, and tablets, while non-hand-held devices were TVs, stereos, and desktop computers. We tested the difference between median concentrations of hand-held vs non-handheld e-devices when laptops were categorized as either non-hand-held¹ vs hand-held².

	BRs - MURs		hands	Hand-held vs non-hand-held (median concentrations in parentheses, pg/cm ²)		
	air	dust		palms vs backs	laptops taken as non-hand-held ¹	laptops taken as hand-held ²
TBP-AE	0.069	0.0010	*	*	*	*
PBBz	0.012	*	*	*	*	*
PBT	0.011	0.24	0.015	0.087 (0.060; 0.14)	0.40 (0.07; 0.19)	
EH-TBB	0.62	0.76	0.044	<0.001 (22; 3.5)	<0.001 (19.; 1.9)	
BEH-TEBP	*	0.80	0.075	0.0060 (17; 4.9)	<0.001 (15; 3.5)	
s-DDC-CO	*	0.84	0.13	0.0090 (0.51; 0.60)	0.10 (0.53; 0.61)	
a-DDC-CO	*	0.58	0.15	0.012 (0.68; 0.77)	0.090 (0.71; 0.77)	
OBTMPI	*	*	*	<0.001 (4.4; 4.9)	<0.001 (4.3; 5.3)	
DBDPE	*	0.18	0.0020	<0.001 (12; 82)	<0.001 (13; 160)	
BDE-17	0.91	0.47	0.16	*	*	*
BDE-28	0.99	0.29	0.33	*	*	*
BDE-47	0.73	0.10	0.25	0.0010 (8.6; 2.9)	<0.001 (9.2; 1.8)	
BDE-49	0.82	0.88	0.12	0.030 (0.31; 0.19)	0.91 (0.31; 0.16)	
BDE-66	*	0.030	0.017	0.0010 (0.21; 0.17)	0.18 (0.22; 0.15)	
BDE-71	*	0.065	*	*	*	*
BDE-85	*	0.051	0.43	0.0010 (0.24; 0.21)	0.59 (0.28; 0.17)	
BDE-99	0.27	0.10	0.29	0.0010 (8.1; 2.5)	<0.001 (8.7; 1.4)	
BDE-100	*	0.22	0.88	0.0080 (0.39; 0.25)	0.79 (0.43; 0.19)	
BDE-138	*	0.17	0.91	*	*	*
BDE-153	*	0.0070	0.12	<0.001 (0.27; 0.30)	<0.001 (0.26; 0.33)	
BDE-154	*	0.061	0.26	<0.001 (0.24; 0.20)	0.13 (0.26; 0.18)	
BDE-183	*	0.98	*	<0.001 (0.16; 0.24)	<0.001 (0.16; 0.28)	
BDE-209	*	0.88	0.0020	<0.001 (27; 79)	0.0010 (32; 99)	

* Results not presented due to low detection frequencies in that sample matrix.

Table S7. Spearman's rho correlation coefficients between each pair of variables (only significant correlations presented) for NFRs and PBDEs.

		NFRs										PBDEs														
		TBP-AE	PBBz	PBT	PBEB	EH-TBB	BEH-TEBP	S-DDC-CO	a-DDC-CO	OBTMPI	DBDPE	BDE-17	BDE-28	BDE-47	BDE-49	BDE-66	BDE-71	BDE-85	BDE-99	BDE-100	BDE-138	BDE-153	BDE-154	BDE-183	BDE-190	BDE-209
Hand palms - Dust	BR Dust			.39#		.69#	.50#				.32*	.47#	.56#	.42#				.42#	.35*	.54#		.32*	.52#			
	MUR Dust			.39*		.63#	.64#						.57#	.47*					.43*	.43*		.45*	.42*			
Hand palms - Electronics	Cell Phones					.48#	.41*							.48#					.48#	.56#						
	Home Phones					.68*													.66*					.82#		
	Tablets																									
	Laptops					.48#	.39*							.74#	.41*				.59#	.72#				.37*	.49#	
	Desktops						.58*																			
	Televisions													.34#												
	Stereos							.82*	.82*																	
Televisions - dust	BR Dust						.25*																			
	MUR Dust						.47#																			

Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2-tailed).

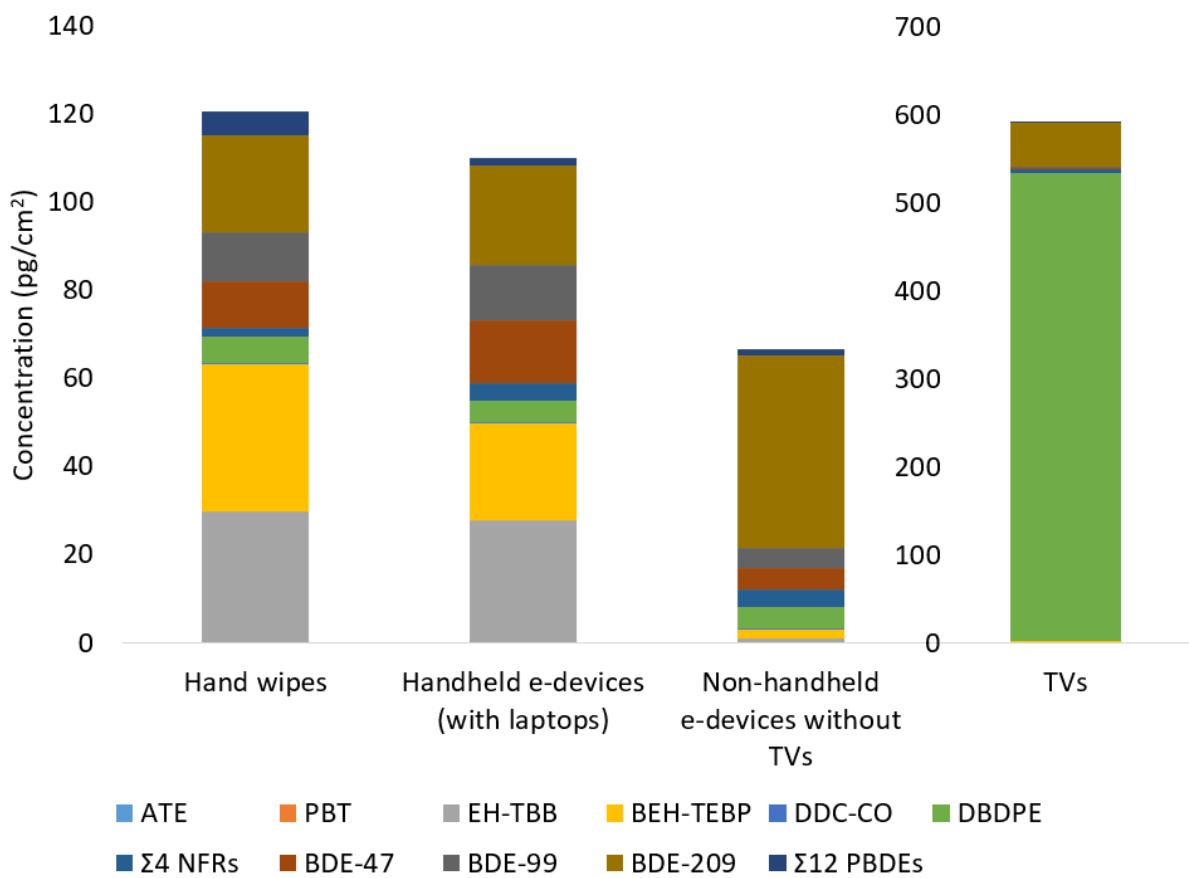


Fig. S1: Median concentrations of HFRs in hand (average of palm and back) and e-devices wipes.

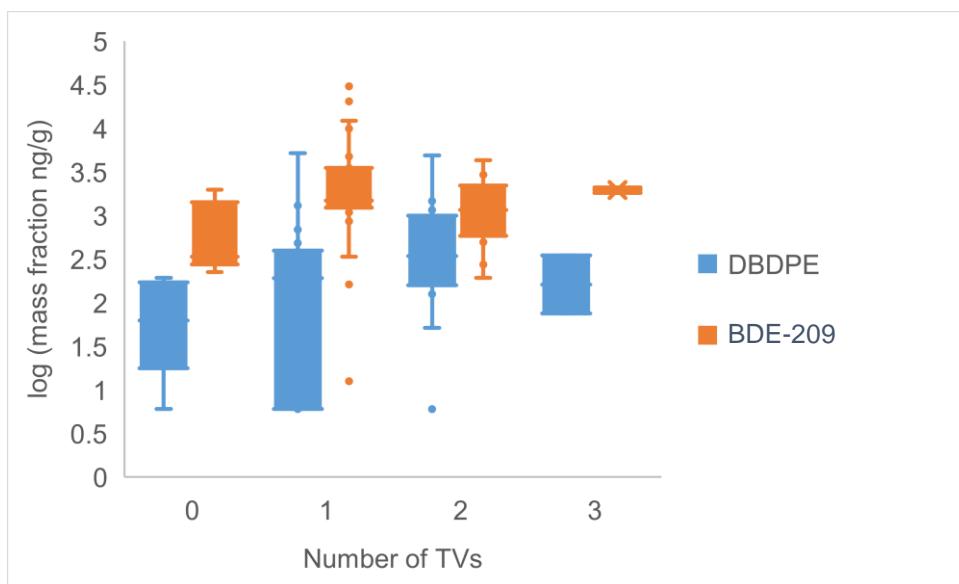


Fig. S2. DBDPE and BDE-209 mass fractions plotted against the number of TVs from the same homes. Boxes show first and third quartiles, whiskers define non-outlier range, and dots are outliers.

References

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