

Supplementary Material.

Brominated and organophosphorus flame retardants in South African indoor dust and cat hair

Martin Brits^{a,b,c*}, Sicco H. Brandsma^a, Egmont R. Rohwer^b, Jayne De Vos^c, Jana M. Weiss^d,
Jacob de Boer^a

^a Department of Environment and Health, Vrije Universiteit, Amsterdam, De Boelelaan 1085, 1081HV Amsterdam, The Netherlands

^b Department of Chemistry, Faculty of Natural and Agricultural Sciences, University of Pretoria, Lynnwood Road, Pretoria 0002, South Africa

^c National Metrology Institute of South Africa (NMISA), CSIR Campus, Meiring Naude Road, Pretoria 0040, South Africa

^d Department of Environmental Science and Analytical Chemistry, Stockholm University, Stockholm, SE-10691, Sweden,

*Corresponding author. +27 (0)12 841 2265; e-mail: mbrits@nmisa.org / m.brits@vu.nl

Table S1. Details on samples associated with vacuum cleaner bags (V-dust), freshly collected dust (F-dust) and cat hair (C-hair)

Table S2. List of OPFR target analytes with quantitation and qualification transition with associated collision energies and internal standards used.

Table S3. Calibration parameters listing the correlation coefficients (R^2), response factor (RF), response factor relative standard deviation (RSD) and limit of quantitation (ng)

Table S4. Results of triplicate spiking experiments for dust and cat hair samples, listing spiking concentration (ng), average percentage recoveries (%) and relative standard deviation (RSD)

Table S5. Recovery results for SRM 2585 with expanded relative uncertainty (%) values

Table S6. Comparison of concentrations (ng/g) of OPFRs in dust reference material (NIST SRM 2585) between different studies

Table S7. Expanded relative uncertainty (%) values for all target compounds in each matrix.
Detailed estimation of measurement uncertainty

Fig. S1. The uncertainty contribution of the uncertainty sources for FRs in dust

Fig. S2. The uncertainty contribution of the uncertainty sources for FRs in hair

Table S7. The concentration of BFRs in dust and cat hair samples (ng/g)

Table S8. The concentration of OPFRs in dust and cat hair samples (ng/g)

Fig. S3. Comparison of median concentrations of alkyl-OPFRs (sum of TNBP, TBOEP, TEHP), Cl-OPFRs (sum of TCEP, TCIPP, TDCPP) and aryl-OPFRs (sum of TPHP, EHDPP, TMPP, TIPPP) in indoor house dust ($\mu\text{g/g}$) from different studies

Table S1. Details on samples associated with vacuum cleaner bags (V-dust), freshly collected dust (F-dust) and cat hair (C-hair)

Houses	V-dust	F-dust	C-hair
House 1	V-dust 1	F-dust 1	C-Hair 1A C-Hair 1B
House 2	V-dust 2	F-dust 2	C-Hair 2A C-Hair 2B C-Hair 2C C-Hair 2D
House 3	V-dust 3	F-dust 3	C-Hair 3
House 4	V-dust 4	F-dust 4	C-Hair 4A C-Hair 4B
House 5	V-dust 5	F-dust 5	C-Hair 5
House 6	V-dust 6	F-dust 6	C-Hair 6
House 7	V-dust 7		
House 8	V-dust 8	F-dust 8	
House 9	V-dust 9	F-dust 9	
House 10	V-dust 10	F-dust 10	
House 11	V-dust 11		

Table S2. List of BFRs and OPFR target analytes with quantitation and qualification transition with associated collision energies and internal standards used.

Name	Abbreviation	CAS Number	Quantitation transition	Collision Energy	Qualification transition	Collision Energy	Internal standard
2,4,4'-Tribromodiphenyl ether	BDE28 ^a	41318-75-6	79		81		BDE58
2,2',4,4'-Tetrabromodiphenyl ether	BDE47 ^a	5436-43-1	79		81		BDE58
2,2',4,4',5-Pentabromodiphenyl ether	BDE99 ^a	60348-60-9	79		81		BDE58
2,2',4,4',6-Pentabromodiphenyl ether	BDE100 ^a	189084-64-8	79		81		BDE58
2,2',4,4',5,5'-Hexabromodiphenyl ether	BDE153 ^a	68531-49-2	79		81		BDE58
2,2',4,4',5,6'-Hexabromodiphenyl ether	BDE154 ^a	207122-15-4	79		81		BDE58
2,2',3,4,4',5',6-Heptabromodiphenyl ether	BDE183 ^a	207122-16-5	79		81		BDE58
2,2',3,3',4,4',5,5',6,6'-Decabromodiphenyl ether	BDE209 ^a	1163-19-5	486.4		484.4		¹³ C-BDE209
Bis(2-ethylhexyl) tetrabromophthalate	BEH-TEBP ^a	26040-51-7	463.6		461.6		¹³ C-BDE209
2-Ethylhexyl-2,3,4,5-tetrabromobenzoate	EH-TBB ^a	183658-27-7	356.7		358.7		¹³ C-BDE209
2,3,3',5'-Tetrabromodiphenyl ether	BDE58 ^a		79		81		
¹³ C ₁₂ -decabromodiphenyl ether	¹³ C-BDE209 ^a		494.4		496.4		
Tri-n-butyl phosphate	TNBP ^b	126-73-8	155.0 > 99.0	4	211.0 > 99.0	13	TNBP-d27
Tris (2-chloroethyl) phosphate	TCEP ^b	115-96-8	204.9 > 142.9	3	248.9 > 125.0	12	TCEP-d12
Tris (1-chloro-2-propyl) phosphate (multiple isomers)	TCIPP ^b	13674-84-5	200.9 > 99.1	30	276.9 > 125.1	10	TCEP-d12
Tris (1,3-dichloro-2-propyl) phosphate	TDCIPP ^b	13674-87-8	209.0 > 99.0	7	190.9 > 155.0	5	TDCIPP-d15
Tris (2-butoxyethyl) phosphate	TBOEP ^b	78-51-3	125.0 > 99.0	15	153.0 > 125.0	7	TPHP-d15
Triphenyl phosphate	TPHP ^b	115-86-6	215.0 > 168.0	25	169.0 > 115.0	35	TPHP-d15
2-Ethylhexyl diphenyl phosphate	EHDPP ^b	1241-94-7	251.1 > 77.0	35	251.1 > 152.0	32	TPHP-d15
Tris (2-ethylhexyl) phosphate	TEHP ^b	78-42-2	112.0 > 81.9	10	113.0 > 57.0	5	TPHP-d15
Tricresyl phosphate (mixture of 3 isomers)	TMPP ^b	1330-78-5	368.0 > 165.0	35	277.0 > 179.0	15	TPHP-d15
Tris (2-isopropylphenyl) phosphate	TIPPP ^b	64532-95-2	335.1 > 251.0	15	452.2 > 118.1	15	TPHP-d15
Tri-n-butyl phosphate-d27	d27-TNBP ^b		167.0 > 103.0	5	231.0 > 103.0	12	
Tris(2-chloroethyl) phosphate-d12	d12-TCEP ^b		261.0 > 131.0	13	213.0 > 148.0	4	
Tris(1,3-dichloro-2-propyl) phosphate-d15	d15-TDCIPP ^b		217.0 > 103.0	8	197.0 > 160.0	7	
Triphenylphosphate-d15	d15-TPHP ^b		243.0 > 176.0	22	223.0 > 176.0	35	

^aAnalysed using GC-ECNI-MS

^bAnalysed using GC-EI-MS/MS

Table S3. Calibration parameters (using an eight-point calibration curve) listing the correlation coefficients (R^2), response factor (RF), response factor relative standard deviation (RSD) and limit of quantitation (LOQ)

Compound	Correlation coefficient (R^2)	Response factor (RF)	Response factor RSD (%)	LOQ dust (ng/g) ^a	LOQ hair (ng/g) ^b
BDE28	0.9999	1.49	2.6	1.3	0.13
BDE47	0.9996	0.97	11.1	5.2	0.52
BDE100	0.9999	1.56	2.2	0.9	0.09
BDE99	0.9998	1.52	3.8	4.6	0.46
BDE154	0.9999	1.60	3.7	4.0	0.40
BDE153	0.9999	1.48	3.2	5.5	0.55
BDE183	0.9997	1.11	5.1	1.9	0.19
BDE209	0.9998	0.64	8.4	12	1.2
EH-TBB	0.9998	0.14	9.6	23	2.3
BEH-TEBP	0.9995	0.22	5.7	24	2.4
TNBP	0.9998	1.10	2.4	157	15.7
TCEP	0.9998	1.16	4.1	58	5.8
TCIPP	0.9999	1.51	4.4	187	18.7
TDCIPP	0.9999	1.22	6.0	78	7.8
TBOEP	0.9987	0.18	14.2	131	13.1
TPHP	0.9999	1.11	6.8	16	1.6
EHDPP	0.9997	1.77	10.5	80	8.0
TEHP	0.9998	0.31	14.8	43	4.3
TMPP	0.9997	0.33	4.9	36	3.6
TIPPP	0.9994	0.48	11.5	49	4.9

^aBased on maximum dust sample intake of 50 mg

^bBased on maximum hair sample intake of 500 mg

Bold LOQ value represent analytes which were present in blanks

Table S4. Results of triplicate spiking experiments for dust and cat hair samples, listing spiking concentration (ng), average percentage recoveries (%) and relative standard deviation (RSD)

Compounds	Dust						Cat hair					
	Low spike (ng)	Low spike recovery (%)	RSD (%)	High spike (ng)	High spike recovery (%)	RSD (%)	Low spike (ng)	Low spike recovery (%)	RSD (%)	High spike (ng)	High spike recovery (%)	RSD (%)
BDE28	1.7	96	8.0	19	98	4.6	1.7	99	7.5	19	99	6.6
BDE47	1.7	92	5.7	19	99	4.1	1.7	94	8.4	19	97	4.4
BDE100	1.7	102	8.4	19	99	5.8	1.7	97	6.7	19	99	4.2
BDE99	1.7	93	5.4	19	101	4.7	1.7	97	11.5	19	103	5.0
BDE154	3.4	93	8.9	39	100	4.4	3.4	95	8.7	39	96	5.8
BDE153	3.4	94	6.7	39	98	5.6	3.4	95	11.5	39	99	6.3
BDE183	3.4	94	10.8	39	98	5.8	3.4	95	9.4	39	97	4.5
BDE209	8.5	95	1.9	97	96	7.5	8.6	99	9.9	97	99	5.8
EH-TBB	49	92	10.2	100	96	5.7	49	90	9.0	100	93	3.0
BEH-TEBP	48	94	4.5	97	96	5.0	48	95	3.5	97	97	3.7
TNBP	50	95	5.2	101	98	3.6	50	88	6.7	101	97	3.4
TCEP	48	102	2.5	98	105	2.7	48	102	4.2	101	99	3.6
TCIPP	50	97	6.3	101	94	15.2	50	95	19.6	101	99	19.3
TDCIPP	48	100	2.0	98	99	6.3	48	98	9.9	98	100	8.5
TBOEP	48	84	12.1	98	99	9.3	48	91	7.9	98	96	11.5
TPHP	50	97	9.3	100	103	8.0	50	96	12.3	100	99	9.1
EHDPP	50	100	7.5	101	102	3.1	50	99	11.5	100	105	7.6
TEHP	49	90	5.7	99	101	3.1	49	86	7.5	99	102	1.9
TMPP	49	94	17.2	99	105	10.9	49	94	14.7	99	105	12.7
TIPPP	48	87	4.7	98	99	3.0	48	85	1.7	98	101	0.7

Table S5. Results (mean concentration and standard deviation) of FR analysis of SRM 2585 dust samples, compared to certified and reference values, and with expanded relative uncertainty (U) values.

Compound	Assigned value (ng/g)		Batch 1 (V-dust) (n=3)		Batch 2 (F-dust) (n=3)		Batch 3 (C-hair) (n=3)		U%
			Mean (ng/g)		Mean (ng/g)		Mean (ng/g)		
BDE28	46.9	± 4.4 ^a	44.2	± 5.3	46.2	± 5.5	47.3	± 5.7	12
BDE47	497	± 46 ^a	488	± 64	502	± 66	507	± 67	13
BDE99	892	± 53 ^a	878	± 124	857	± 121	868	± 123	14
BDE100	145	± 11 ^a	143	± 19	144	± 19	152	± 20	13
BDE153	119	± 1 ^a	117	± 18	118	± 18	122	± 18	15
BDE154	83.5	± 2.0 ^a	82.4	± 11.6	83.8	± 11.8	82.4	± 11.6	14
BDE183	43.0	± 3.5 ^a	44.4	± 7.0	45.0	± 7.0	42.6	± 6.7	16
BDE209	2510	± 190 ^a	2410	± 376	2362	± 368	2417	± 377	16
TNBP	276	± 14 ^b	276	± 30	265	± 29	280	± 31	11
TCEP	925	± 149 ^b	927	± 100	948	± 103	947	± 103	11
TCIPP	1220	± 350 ^b	1502	± 258	1298	± 223	1395	± 240	17
TPHP	1190	± 130 ^b	1199	± 195	1274	± 208	1183	± 193	16

^a Certified value

^b Reference value

Table S5. Comparison of concentrations (ng/g) and standard deviation (SD) of OPFRs in dust reference material (NIST SRM 2585) between different studies

Compound	This study (n=9)		Brandsma et al. (2013)*		Cristale et al. (2018) (n=3)		Persson et al. (2018) (n=13)		Bergh et al. (2012) (n=7)		Van Den Eede et al. (2012) (n=6)	
	Mean (ng/g)	U	Mean (ng/g)	SD	Mean (ng/g)	SD	Mean (ng/g)	SD	Mean (ng/g)	SD	Mean (ng/g)	SD
EH-TBB	71	15			49	8					26	2
BEH-TEBP	950	170			1,018	14					574	49
TNBP	270	29	269	19	234	2	185	22	190	20	190	10
TCEP	940	102	792	127	714	8	550	99	840	60	680	60
TCIPP	1,204	360	944	264	778	6	603	110	880	140	860	70
TDCIPP	1840	276	1,556	529	1,808	2	1,230	240	2,300	280	3,180	70
TBOEP	76,800	15,900	73,464	32,324	54,848	2	60,000	7,010	82,000	6,500	63,000	2,000
TPHP	1,220	199	1,104	99	1,139	9	700	130	1,100	100	1160	140
EHDPP	1,430	230	963	202	978	2	741	100	1,300	120		
TEHP	334	38	265	111	331	2			370	40		
TMPP	1,030	225							740	110	1,140	30

* calculated from RSD/ CV%

Table S7. Expanded relative uncertainty (U) values for all target compounds in dust and cat hair matrix.

	Dust	Cat hair
	U (%)	U (%)
BDE28	15	20
BDE47	17	22
BDE100	17	17
BDE99	19	22
BDE154	19	21
BDE153	20	22
BDE183	21	24
BDE209	21	22
EH-TBB	14	15
BEH-TEBP	14	13
TNBP	14	15
TCEP	14	13
TCIPP	23	34
TDCIPP	20	23
TBOEP	28	27
TPHP	22	23
EHDPP	22	22
TEHP	14	16
TMPP	30	29
TIPPP	13	11

ESTIMATION OF UNCERTAINTY

Identification of uncertainty sources. The main parameters estimated to affect the measurand concentration were estimated from preparation of the standard and internal standard solutions, calibration curve, recovery and repeatability.

Estimation of the uncertainty derived from standard and internal solution preparation.

The combined standard uncertainty associated with the gravimetric preparation of the native and labelled standard stock solutions was calculated using Eq. (1)

$$u(C_{Std}) = C_{Std} \sqrt{\left(\frac{u(P)}{P}\right)^2 + (u_{dil})^2 + \left(\frac{u(W)}{W}\right)^2} \quad (1)$$

were,

$U(C_{Std})$	Combined standard measurement uncertainty of standard stock solution
C_{Std}	Concentration of the standard stock solution
$u(P)$	Standard measurement uncertainty from purity of standard solution
P	Purity of standard stock solution (obtained from manufacturers certificate)
u_{dil}	Combined standard measurement uncertainty for the dilution of standard stock solution
(W)	Weight of the standard stock solution
$u(W)$	Combined measurement uncertainty for weighing, obtained through Eq. (2)

$$u(W) = \sqrt{(u_{Std})^2 + (u_{solvent})^2 + (u_{Sample})^2} \quad (2)$$

where,

U_{Std}	Standard measurement uncertainty of balance from weighing standard stock solution
$U_{Solvent}$	Standard measurement uncertainty from weighing solvent
U_{Sample}	Standard measurement uncertainty from weighing sample

Estimation of the uncertainty from linear calibration graph. The combined uncertainty associated with the linear calibration was calculated using Eq. (3)

$$u(c_0) = \frac{s_{y/x}}{b} \sqrt{\frac{1}{m} + \frac{1}{n} + \frac{(x_0 - \bar{x})^2}{S_{xx}}} \quad S_{xx} = \sum_{i=1}^n (x_i - \bar{x})^2 \quad (3)$$

where,

$u(c_0)$	Calibration uncertainty
$s_{y/x}$	Residual standard deviation of the data points from the regression line
b	slope of linear calibration graph
m	Number of repeat measurements of sample to obtain the value for x_0
n	Number of calibration points
x_0	Calculated analyte concentration of sample using calibration curve
\bar{x}	Mean value for x in the calibration summated over n number of calibration points
x_i	Individual calibration concentrations obtained from the calibration curve

Estimation of uncertainties associated with recovery. The combined uncertainty for selected analytes associated with the recovery from the CRM for dust was obtained from E.q (4)

$$u(R_m) = R_m \times \sqrt{\frac{s_{obs}^2}{n \times C_{obs}^2} + \left(\frac{u(C_{CRM})}{C_{CRM}}\right)^2} \quad (4)$$

where,

R_m Mean recovery, calculated from E.q (5)

$$R_m = \frac{C_{obs}}{C_{CRM}} \quad (5)$$

C_{obs}	Mean concentration of the results from the replicate analysis of the CRM
C_{CRM}	The certified value for the CRM, obtained from the certificate
S_{obs}	Standard deviation of the results from the replicate analyses of the CRM
n	Number of replicate analysis of the CRM
$u(C_{CRM})$	Standard uncertainty in the certified value for the CRM, obtained from the certificate

Spiking experiments at a low ($n=3$) and high ($n=3$) concentration were performed to assess for recovery. A Student's t-test was used to determine whether the mean recovery was significantly different from 1.0. The combined uncertainty associated with each spiking experiment was calculated using E.q (6) and included in the uncertainty estimation.

$$u(R_m) = R_m \times \sqrt{\frac{s_{obs}^2/n + s_{native}^2}{(C_{obs} - C_{native})^2} + \left(\frac{u(C_{spike})}{C_{spike}}\right)^2} \quad (6)$$

where,

R_m Mean recovery, calculated from E.q (7)

$$R_m = \frac{C_{obs} - C_{native}}{C_{spike}} \quad (7)$$

S_{obs} Standard deviation of the results from the replicate analyses of the spiked sample

n Number of replicate analysis of spiked sample

S_{native} Standard deviation of the mean of the results of repeat analyses of the unspiked matrix

C_{obs} Mean concentration of the results from the replicate analysis of the spiked sample

C_{native} Mean concentration of the analyte in the unspiked sample.

$u(C_{spike})$ Standard uncertainty in the concentration of the spiked sample

C_{spike} Prepared concentration of the analyte in the spiked sample

Estimation of uncertainty of repeatability. The standard uncertainty associated with the repeatability of the method was obtained from E.q (8)

$$u(r) = \frac{RSD}{\sqrt{n}} \quad (8)$$

where,

$u(r)$ Standard uncertainty of repeatability

RSD Relevant standard deviation of the repeat measurements

n Number of repeat measurements

The combined standard measurement uncertainty for the analytes in the matrices was calculated using Eq. (9) and multiplied by 2 (coverage factor) at 95% confidence level. The expanded relative uncertainty for the analytes in the two matrices are listed in Table S1

$$\frac{u_c(A)}{C_A} = \sqrt{\left(\frac{u(C_{Std})}{C_{Std}}\right)^2 + \left(\frac{u(C_{Istd})}{C_{Istd}}\right)^2 + \left(\frac{u(c_0)}{c_0}\right)^2 + \left(\frac{u(R_m)}{R_m}\right)^2 + u(r)^2} \quad (9)$$

where,

- $u_c(A)$ Combined standard measurement uncertainty of the analyte
- C_A Concentration of the analyte
- $u(C_{Std})$ Combined standard measurement uncertainty of standard solution
- C_{Std} Concentration of standard solution
- $u(C_{Istd})$ Combined standard measurement uncertainty of internal standard solution
- C_{Istd} Concentration of internal standard solution
- $u(C_0)$ Combined standard measurement uncertainty of calibration curve
- C_0 Calculated concentration of the analyte in the sample using calibration curve
- $u(R_m)$ Combined standard measurement uncertainty of recovery
- R_m Calculated recovery
- $u(r)$ Combined standard measurement uncertainty of repeatability

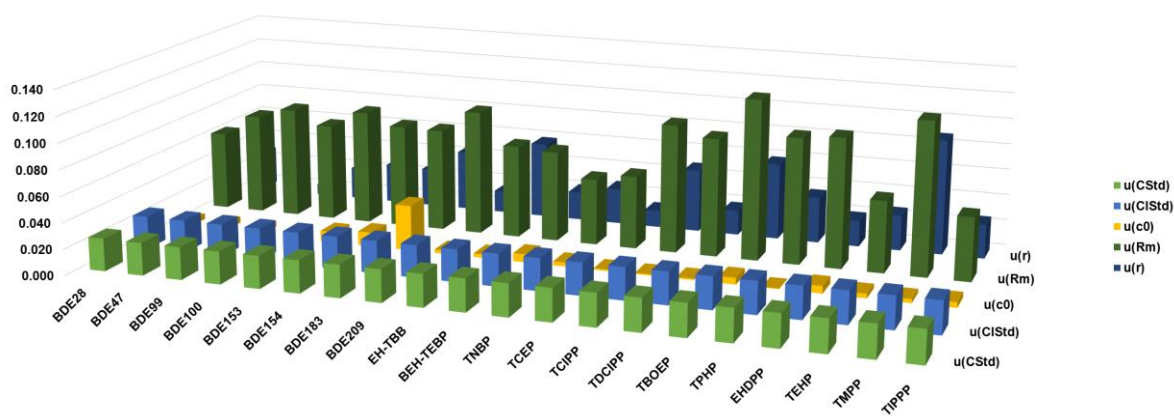


Fig. S1. The uncertainty contribution of the uncertainty sources for FRs in dust

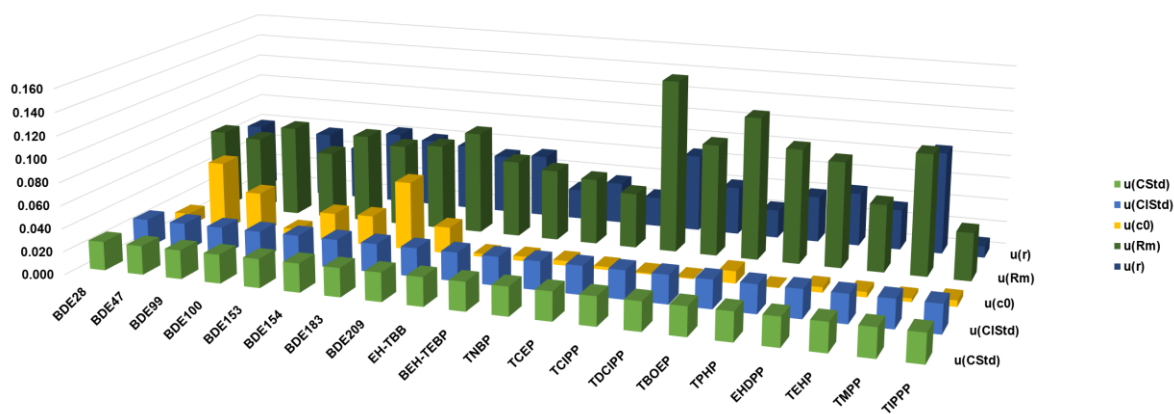


Fig. S2. The uncertainty contribution of the uncertainty sources for FRs in hair

Table S8. The concentration of BFRs in dust and cat hair samples (ng/g)

	BDE28	BDE47	BDE100	BDE99	BDE154	BDE153	BDE183	BDE209	EH-TBB	BEH-TEBP
V-Dust 1	<LOQ	6.12	1.11	11.1	<LOQ	9.42	2.34	635	28.6	44.3
V-Dust 2	<LOQ	6.04	<LOQ	13.0	<LOQ	<LOQ	<LOQ	135	<LOQ	43.8
V-Dust 3	<LOQ	44.8	6.92	29.6	<LOQ	<LOQ	15.5	204	<LOQ	156
V-Dust 4	<LOQ	7.31	<LOQ	13.5	<LOQ	10.4	3.63	272	27.9	154
V-Dust 5	<LOQ	18.6	3.13	21.2	4.24	<LOQ	<LOQ	161	<LOQ	95.4
V-Dust 6	<LOQ	6.94	<LOQ	10.5	<LOQ	<LOQ	2.92	76.6	<LOQ	29.9
V-Dust 7	<LOQ	6.06	<LOQ	10.4	<LOQ	<LOQ	4.52	857	28.1	38.6
V-Dust 8	<LOQ	6.17	<LOQ	10.4	<LOQ	7.69	5.29	567	<LOQ	38.1
V-Dust 9	<LOQ	14.3	2.93	20.6	4.15	7.90	4.15	198	<LOQ	44.1
V-Dust 10	2.85	35.3	7.87	29.0	4.81	10.4	4.07	329	39.3	58.7
V-Dust 11	2.58	99.5	6.83	142	13.4	18.4	6.91	565	36.4	246
F-Dust 1	<LOQ	6.80	1.14	17.0	4.81	<LOQ	3.61	4590	30.0	68.1
F-Dust 2	<LOQ	6.46	1.45	12.7	<LOQ	6.67	5.10	1430	28.3	117
F-Dust 3	5.01	21.4	4.04	18.3	<LOQ	23.5	67.1	570	<LOQ	85.4
F-Dust 4	<LOQ	8.24	1.08	14.0	4.09	8.79	18.9	2510	298	12400
F-Dust 5	<LOQ	11.5	1.43	12.7	<LOQ	<LOQ	3.20	618	221	80.2
F-Dust 6	<LOQ	8.32	<LOQ	13.1	<LOQ	<LOQ	4.73	877	31.2	76.9
F-Dust 8	<LOQ	7.50	1.25	11.2	<LOQ	<LOQ	5.81	868	28.7	64.7
F-Dust 9	<LOQ	15.0	2.87	12.7	4.60	7.53	6.53	882	30.0	74.5
F-Dust 10	3.00	36.9	7.80	32.2	4.83	8.68	4.75	653	24800	1080
C-Hair 1A	<LOQ	0.608	<LOQ	1.06	0.429	<LOQ	0.336	14.1	<LOQ	3.73
C-Hair 1B	0.204	0.824	0.124	1.46	0.525	0.877	0.336	13.8	3.09	5.04
C-Hair 2A	<LOQ	0.687	0.096	0.892	<LOQ	0.713	0.541	10.1	3.55	13.7
C-Hair 2B	<LOQ	0.677	<LOQ	0.797	<LOQ	<LOQ	<LOQ	9.13	3.38	19.0
C-Hair 2C	<LOQ	0.695	<LOQ	0.868	<LOQ	<LOQ	0.342	10.6	3.00	13.0
C-Hair 2D	<LOQ	0.666	<LOQ	0.634	<LOQ	<LOQ	<LOQ	8.80	3.05	15.9
C-Hair 3	0.310	1.19	0.149	0.636	<LOQ	<LOQ	<LOQ	5.45	<LOQ	8.27
C-Hair 4A	<LOQ	0.736	0.116	1.07	<LOQ	1.00	0.390	10.6	3.60	8.98
C-Hair 4B	0.149	0.650	0.121	0.937	<LOQ	0.916	0.314	8.01	3.29	6.66
C-Hair 5	0.163	0.870	0.143	1.10	<LOQ	1.03	0.251	4.33	3.54	5.96
C-Hair 6	0.502	1.78	0.261	1.18	<LOQ	<LOQ	<LOQ	6.64	<LOQ	6.54

Table S9. The concentration of OPFRs in dust and cat hair samples (ng/g)

	TNBP	TBOEP	TEHP	TCEP	TCIPP	TDCIPP	TPHP	EHDPP	TMPP	TIPPP
V-Dust 1	275	960	130	4350	3590	790	617	212	93.1	58.7
V-Dust 2	284	2170	91.3	310	1470	338	484	783	147	<LOQ
V-Dust 3	1490	6700	<LOQ	458	8610	668	619	313	334	636
V-Dust 4	274	6430	179	1270	1700	208	1130	422	75.5	57.4
V-Dust 5	<LOQ	3510	210	628	1880	343	448	371	418	59.9
V-Dust 6	<LOQ	2910	90.6	8630	1990	2140	277	187	92.8	<LOQ
V-Dust 7	245	1640	695	1780	139000	610	4970	6580	92.9	75.4
V-Dust 8	324	36000	221	2210	81100	790	2470	480	76.7	<LOQ
V-Dust 9	4610	5910	172	4180	2490	1390	911	379	635	55.6
V-Dust 10	307	1680	83.4	647	21900	289	415	329	140	61.9
V-Dust 11	294	11900	446	553	12800	458	1390	802	474	82.2
F-Dust 1	203	8670	142	1550	4410	558	2140	162	77.2	<LOQ
F-Dust 2	958	3140	109	7610	3160	1530	565	763	127	74.6
F-Dust 3	1730	13000	319	402	8970	28300	2600	748	314	933
F-Dust 4	396	21600	225	10000	10700	57700	2130	4380	141	116
F-Dust 5	<LOQ	586	234	1040	7010	322	3480	354	88.2	91.3
F-Dust 6	194	21500	125	16900	2090	2050	470	334	1130	<LOQ
F-Dust 8	212	485	110	919	178000	372	2660	165	52.2	55.6
F-Dust 9	<LOQ	885	141	1730	2310	1340	391	733	147	57.3
F-Dust 10	210	1240	272	5220	18700	11700	6500	862	647	1440
C-Hair 1A	<LOQ	56.2	10.3	26.3	285	19.2	53.2	18.0	15.6	<LOQ
C-Hair 1B	<LOQ	65.6	12.8	25.3	264	26.9	62.1	29.9	14.3	6.58
C-Hair 2A	23.1	387	22.6	22.8	159	91.1	130	78.0	35.5	7.64
C-Hair 2B	22.2	396	21.2	22.5	160	99.3	123	77.4	37.5	<LOQ
C-Hair 2C	22.5	414	20.9	21.6	152	93.2	128	74.3	29.0	5.55
C-Hair 2D	20.6	364	20.3	20.6	149	85.9	111	63.3	30.7	<LOQ
C-Hair 3	<LOQ	488	10.0	23.1	245	13.2	18.0	16.2	19.9	24.6
C-Hair 4A	<LOQ	485	54.2	53.9	372	16.5	51.6	184	8.81	5.81
C-Hair 4B	<LOQ	418	50.6	53.3	338	19.1	51.9	188	7.64	5.49
C-Hair 5	<LOQ	177	42.9	32.6	344	32.5	31.0	39.5	113	6.26
C-Hair 6	31.0	283	13.8	47.3	297	24.4	12.0	53.7	12.2	<LOQ

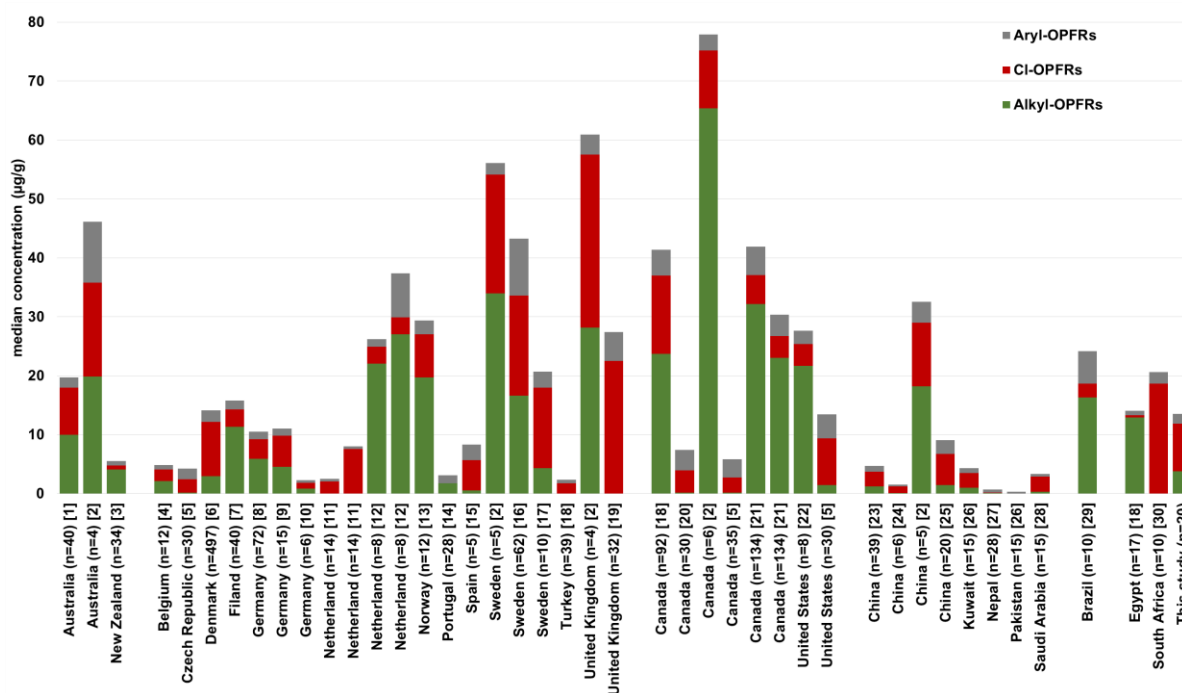


Fig. S3. Comparison of median concentrations of alkyl-OPFRs (sum of TNBP, TBOEP, TEHP), CI-OPFRs (sum of TCEP, TCIPP, TDCPP) and aryl-OPFRs (sum of TPHP, EHDPP, TMPP, TIPPP) in indoor house dust ($\mu\text{g/g}$) from different studies. [1] (C. He et al., 2018), [2] (Wong et al., 2017), [3] (Ali et al., 2012), [4] (Van den Eede et al., 2011), [5] (Vykoukalová et al., 2017), [6] (Langer et al., 2016), [7] (Rantakokko et al., 2019), [8] (Zhou and Püttmann, 2019), [9] (Zhou et al., 2017), [10] (Brommer et al., 2012), [11] (Sugeng et al., 2017), [12] (Brandsma et al., 2014), [13] (Cequier et al., 2014), [14] (Coelho et al., 2016), [15] (Cristale et al., 2016), [16] (Luongo and Östman, 2016), [17] (Bergh et al., 2011), [18] (Shoeib et al., 2019), [19] (Brommer and Harrad, 2015)(mean values), [20] (Liu and Mabury, 2018), [21] (Fan et al., 2014), [22] (Kim et al., 2019), [23] (Cao et al., 2019), [24] (M.-J. He et al., 2018), [25] (Tan et al., 2017), [26] (Ali et al., 2013), [27] (Yadav et al., 2017), [28] (Ali et al., 2016), [29] (Cristale et al., 2018), [30] Abafe and Martincigh, (2019).

REFERENCES

- Abafe, O.A., Martincigh, B.S., 2019. Concentrations, sources and human exposure implications of organophosphate esters in indoor dust from South Africa. *Chemosphere*. doi:10.1016/j.chemosphere.2019.04.175
- Ali, N., Dirtu, A.C., Eede, N. Van den, Goosey, E., Harrad, S., Neels, H., 't Mannetje, A., Coakley, J., Douwes, J., Covaci, A., 2012. Occurrence of alternative flame retardants in indoor dust from New Zealand: Indoor sources and human exposure assessment. *Chemosphere* 88, 1276–1282. doi:10.1016/j.chemosphere.2012.03.100
- Ali, N., Ali, L., Mehdi, T., Dirtu, A.C., Al-Shammari, F., Neels, H., Covaci, A., 2013. Levels and profiles of organochlorines and flame retardants in car and house dust from Kuwait and Pakistan: Implication for human exposure via dust ingestion. *Environ. Int.* 55, 62–70.

doi:10.1016/j.envint.2013.02.001

- Ali, N., Eqani, S.A.M.A.S., Ismail, I.M.I., Malarvannan, G., Kadi, M.W., Albar, H.M.S., Rehan, M., Covaci, A., 2016. Brominated and organophosphate flame retardants in indoor dust of Jeddah, Kingdom of Saudi Arabia: Implications for human exposure. *Sci. Total Environ.* 569–570, 269–277. doi:10.1016/j.scitotenv.2016.06.093
- Bergh, C., Torgrip, R., Emenius, G., Östman, C., 2011. Organophosphate and phthalate esters in air and settled dust - a multi-location indoor study. *Indoor Air* 21, 67–76. doi:10.1111/j.1600-0668.2010.00684.x
- Bergh, C., Luongo, G., Wise, S., Östman, C., 2012. Organophosphate and phthalate esters in standard reference material 2585 organic contaminants in house dust. *Anal. Bioanal. Chem.* 402, 51–59. doi:10.1007/s00216-011-5440-2
- Brandsma, S.H., De Boer, J., Leonards, P.E.G., Cofino, W.P., Covaci, A., Leonards, P.E.G., 2013. Organophosphorus flame-retardant and plasticizer analysis, including recommendations from the first worldwide interlaboratory study. *TrAC - Trends Anal. Chem.* 43, 217–228. doi:10.1016/j.trac.2012.12.004
- Brandsma, S.H., de Boer, J., van Velzen, M.J.M., Leonards, P.E.G., 2014. Organophosphorus flame retardants (PFRs) and plasticizers in house and car dust and the influence of electronic equipment. *Chemosphere* 116, 3–9. doi:10.1016/j.chemosphere.2014.02.036
- Brommer, S., Harrad, S., 2015. Sources and human exposure implications of concentrations of organophosphate flame retardants in dust from UK cars, classrooms, living rooms, and offices. *Environ. Int.* 83, 202–207. doi:10.1016/j.envint.2015.07.002
- Brommer, S., Harrad, S., Van Den Eede, N., Covaci, A., 2012. Concentrations of organophosphate esters and brominated flame retardants in German indoor dust samples. *J. Environ. Monit.* 14, 2482–2487. doi:10.1039/c2em30303e
- Cao, D., Lv, K., Gao, W., Fu, J., Wu, J., Fu, J., Wang, Y., Jiang, G., 2019. Presence and human exposure assessment of organophosphate flame retardants (OPEs) in indoor dust and air in Beijing, China. *Ecotoxicol. Environ. Saf.* 169, 383–391. doi:10.1016/j.ecoenv.2018.11.038
- Cequier, E., Ionas, A.C., Covaci, A., Marcé, R.M., Becher, G., Thomsen, C., 2014. Occurrence of a broad range of legacy and emerging flame retardants in indoor environments in Norway. *Environ. Sci. Technol.* 48, 6827–6835. doi:10.1021/es500516u
- Coelho, S.D., Sousa, A.C.A., Isobe, T., Kim, J.W., Kunisue, T., Nogueira, A.J.A., Tanabe, S., 2016. Brominated, chlorinated and phosphate organic contaminants in house dust from Portugal. *Sci. Total Environ.* 569–570, 442–449. doi:10.1016/j.scitotenv.2016.06.137
- Cristale, J., Aragão Belé, T.G., Lacorte, S., Rodrigues de Marchi, M.R., 2018. Occurrence and human exposure to brominated and organophosphorus flame retardants via indoor dust in a Brazilian city. *Environ. Pollut.* 237, 695–703. doi:10.1016/j.envpol.2017.10.110
- Cristale, J., Hurtado, A., Gómez-Canela, C., Lacorte, S., 2016. Occurrence and sources of brominated and organophosphorus flame retardants in dust from different indoor environments in Barcelona,

- Spain. *Environ. Res.* 149, 66–76. doi:10.1016/j.envres.2016.05.001
- Fan, X., Kubwabo, C., Rasmussen, P.E., Wu, F., 2014. Simultaneous determination of thirteen organophosphate esters in settled indoor house dust and a comparison between two sampling techniques. *Sci. Total Environ.* 491–492, 80–86. doi:10.1016/j.scitotenv.2013.12.127
- He, C., Wang, X., Thai, P., Baduel, C., Gallen, C., Banks, A., Bainton, P., English, K., Mueller, J.F., 2018. Organophosphate and brominated flame retardants in Australian indoor environments: Levels, sources, and preliminary assessment of human exposure. *Environ. Pollut.* 235, 670–679. doi:10.1016/j.envpol.2017.12.017
- He, M.-J., Lu, J.F., Ma, J.Y., Wang, H., Du, X.F., 2018. Organophosphate esters and phthalate esters in human hair from rural and urban areas, Chongqing, China: Concentrations, composition profiles and sources in comparison to street dust. *Environ. Pollut.* 237, 143–153. doi:10.1016/j.envpol.2018.02.040
- Kim, U.J., Wang, Y., Li, W., Kannan, K., 2019. Occurrence of and human exposure to organophosphate flame retardants/plasticizers in indoor air and dust from various microenvironments in the United States. *Environ. Int.* 125, 342–349. doi:10.1016/j.envint.2019.01.065
- Langer, S., Fredricsson, M., Weschler, C.J., Bekö, G., Strandberg, B., Remberger, M., Toftum, J., Clausen, G., 2016. Organophosphate esters in dust samples collected from Danish homes and daycare centers. *Chemosphere* 154, 559–566. doi:10.1016/j.chemosphere.2016.04.016
- Liu, R., Mabury, S.A., 2018. Unexpectedly High Concentrations of a Newly Identified Organophosphate Ester, Tris(2,4-di-tert-butylphenyl) Phosphate, in Indoor Dust from Canada. *Environ. Sci. Technol.* 52, 9677–9683. doi:10.1021/acs.est.8b03061
- Luongo, G., Östman, C., 2016. Organophosphate and phthalate esters in settled dust from apartment buildings in Stockholm. *Indoor Air* 26, 414–425. doi:10.1111/ina.12217
- Persson, J., Wang, T., Hagberg, J., 2018. Organophosphate flame retardants and plasticizers in indoor dust, air and window wipes in newly built low-energy preschools. *Sci. Total Environ.* 628–629, 159–168. doi:10.1016/j.scitotenv.2018.02.053
- Rantakokko, P., Kumar, E., Braber, J., Huang, T., Kiviranta, H., Cequier, E., Thomsen, C., 2019. Concentrations of brominated and phosphorous flame retardants in Finnish house dust and insights into children's exposure. *Chemosphere* 223, 99–107. doi:10.1016/j.chemosphere.2019.02.027
- Shoeib, T., Webster, G.M., Hassan, Y., Tepe, S., Yalcin, M., Turgut, C., Kurt-Karakuş, P.B., Jantunen, L., 2019. Organophosphate esters in house dust: A comparative study between Canada, Turkey and Egypt. *Sci. Total Environ.* 650, 193–201.
- Sugeng, E.J., Leonards, P.E.G., van de Bor, M., 2017. Brominated and organophosphorus flame retardants in body wipes and house dust, and an estimation of house dust hand-loadings in Dutch toddlers. *Environ. Res.* 158, 789–797. doi:10.1016/j.envres.2017.07.035
- Tan, H., Peng, C., Guo, Y., Wang, X., Wu, Y., Chen, D., 2017. Organophosphate Flame Retardants in House Dust from South China and Related Human Exposure Risks. *Bull. Environ. Contam.*

Toxicol. 99, 344–349. doi:10.1007/s00128-017-2120-8

- Van den Eede, N., Dirtu, A.C., Neels, H., Covaci, A., 2011. Analytical developments and preliminary assessment of human exposure to organophosphate flame retardants from indoor dust. *Environ. Int.* 37, 454–461. doi:10.1016/j.envint.2010.11.010
- Van Den Eede, N., Dirtu, A.C., Ali, N., Neels, H., Covaci, A., 2012. Multi-residue method for the determination of brominated and organophosphate flame retardants in indoor dust. *Talanta* 89, 292–300. doi:10.1016/j.talanta.2011.12.031
- Vykoukalová, M., Venier, M., Vojta, Š., Melymuk, L., Bečanová, J., Romanak, K., Prokeš, R., Okeme, J.O., Saini, A., Diamond, M.L., Klánová, J., 2017. Organophosphate esters flame retardants in the indoor environment. *Environ. Int.* 106, 97–104. doi:10.1016/j.envint.2017.05.020
- Wong, F., Suzuki, G., Michinaka, C., Yuan, B., Takigami, H., de Wit, C.A., 2017. Dioxin-like activities, halogenated flame retardants, organophosphate esters and chlorinated paraffins in dust from Australia, the United Kingdom, Canada, Sweden and China. *Chemosphere* 168, 1248–1256. doi:10.1016/j.chemosphere.2016.10.074
- Yadav, I.C., Devi, N.L., Zhong, G., Li, J., Zhang, G., Covaci, A., 2017. Occurrence and fate of organophosphate ester flame retardants and plasticizers in indoor air and dust of Nepal: Implication for human exposure. *Environ. Pollut.* 229, 668–678. doi:10.1016/j.envpol.2017.06.089
- Zhou, L., Hiltcher, M., Püttmann, W., 2017. Occurrence and human exposure assessment of organophosphate flame retardants in indoor dust from various microenvironments of the Rhine/Main region, Germany. *Indoor Air* 27, 1113–1127. doi:10.1111/ina.12397
- Zhou, L., Püttmann, W., 2019. Distributions of organophosphate flame retardants (OPFRs) in three dust size fractions from homes and building material markets. *Environ. Pollut.* 245, 343–352. doi:10.1016/j.envpol.2018.11.023