

SUPPORTING INFORMATION

Has the phase-out of PBDEs affected their environmental levels?

Trends of PBDEs and their replacements in the Great Lakes atmosphere

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Table S1. Measurements (averages or ranges in pg/m³) of PBDEs and six selected alternative flame retardants in the atmosphere on a global scale. Studies near e-waste sites or landfills have not been included because those data do not represent ambient levels and are not comparable to the IADN data reported in this study. Atmospheric PBDE levels include all the congeners reported in each study.

Location	Year	Num.	PBDEs	PBEB	HBB	TBE	DBDPE	TBB	TBPH	Ref
Toronto, Canada	2007-2008	76	0.47-110							1
Southern Ontario	2002	~36	n.d.-105							2
Point Petre, Lake Ontario	2002-2004	32	8.8							3
Barcelona, Spain	2009-2010	13	6.9-48							4
Kuwait	2008-2010	48	32-57							5
Chile	2007	6	5-10							6
United Kingdom	2000-2010	71	~10							7
United Kingdom to Norway	2000-2004	40	~0.4-3.4							8
United Kingdom to Norway	2004-2008	44	~0.05-1.3							9
Northern Italy	2005	1	107							10
Tuscany, Italy	2008	19	n.d.-30							11
Switzerland	2007	38	24							12
Turkey	2005	60	6-150							13
Nuuk, Greenland	2004-2005	24	1.1							14
Eastern Mediterranean	2006	26	3.9							15
Brisbane, Australia	2008-2010	18	5.9-11							16
Antarctica	2009-2010	5	0.67-3.0							17
Canadian High Arctic	2002-2004	104	9.3							18
Arctic	2002-2005	480	1.1-8.6							19
European Arctic	2009	10	0.09-2.0		0.001-0.66	n.d.-0.06			n.d.-0.08	20
Bohai Sea and the Arctic	2003	49	2.2-200							21
East Asia to the Arctic	2010	17	0.07-8.1		0.1-5.9	n.d.-1.6		n.d.-8.9	n.d.-3.4	22
SE Asia to Antarctica	2010-2011	20	n.d.-6.6		0.12-26			0.21-0.4	n.d.-2.8	23
East and South China Seas	2008	18	11							24
Bengal Bay & Andaman Sea	2008	10	3.2							24
Indian Ocean	2008	13	5.1							24
Atlantic Ocean (off Africa)	2008	18	2.9							24
Indian Ocean	2004-2005	12	2.5							25
North Sea	2010	40	0.2-11		0.09-6.3					26
Atlantic & Southern Oceans	2008	17	0.3-5		0.04-11					27
Canadian High Arctic	2006-2008	72	1.2-55			0.16-1.9		0.74	0.8	28

Location	Year	Num.	PBDEs	PBEB	HBB	TBE	DBDPE	TBB	TBPH	Ref
Tibetan Plateau	2006-2008	17	0.83-5.2			0.57-20		0.54	0.38	28
Cape Grim, Australia	2004-2005	2	n.d.							29
China, Malaysia, Philippines	2004-2005	7	n.d.-17							29
Europe	2004-2005	9	n.d.-4.7							29
Botswana, S. Africa, Ghana	2004-2005	3	n.d.-6							29
Bermuda, Canada, USA	2004-2005	7	n.d.-24							29
Costa Rica, Cuba	2004-2005	2	1-6							29
Argentina, Bolivia, Chile,	2004-2005	7	n.d.-8							29
Antarctica, Alaska, Norway	2004-2005	4	n.d.-5.3							29
China	2004	32	<0.13-340							30
South Korea	2004	15	2.0-27							30
Japan	2004	20	5.0-71							30
Singapore	2004	10	10.0-29							30
South Korea	2006	38	3.8-24							31
Guangzhou, China	2003-2004	21	790-25,000							32
Hong Kong, China	2003-2004	23	9.5-1280							32
Urban South China	2004	128	350-7900							33
Remote SW China	2005-2006	59	1.6-58							34
Shanghai, China	2006	25	110-740							35
Guangzhou, China	2006-2007	49	610							36
Rural Southern China	2007-2008	60	37-950	0.1-4.8	0.42-14	n.d.-28	4-1400			37
Northeastern China	2008-2009	48	104							38
Yangtze River Delta, China	2009-2010	84	839-1250							39

*n.d. = not detectable

Table S2. The regression results for parameter $a_0 \pm$ standard errors for P $\leq 5\%$; otherwise, the cell is blank.

		Chi		Clev		StPt		SBD		EH	
		a_0	a_0 err								
BDE-47	vapor	2.71	0.11	2.43	0.10	0.38	0.10				
	particle	1.82	0.09	1.97	0.08			-0.61	0.13		
	precip	9.53	0.43	5.73	0.28	3.82	0.40	6.18	0.46	4.46	0.29
BDE-99	vapor	1.08	0.11	1.63	0.10						
	particle	1.40	0.09	2.24	0.10						
	precip	9.22	0.30	5.92	0.23	4.65	0.15	6.41	0.23	5.04	0.16
BDE-209	vapor	0.85	0.17	0.58	0.20	-0.94	0.15	-0.63	0.21	-0.87	0.19
	particle	2.20	0.12	3.14	0.16	1.01	0.12			-0.28	0.14
	precip	6.95	0.25	7.17	0.30	5.66	0.24	5.82	0.21	5.58	0.24
Total PBDEs	vapor	3.16	0.11	3.00	0.11	0.91	0.17	0.54	0.16		
	particle	3.03	0.13	3.87	0.17	1.92	0.14	2.00	0.22	0.63	0.13
	precip	10.41	0.35	7.96	0.24	5.31	0.29	7.28	0.35	6.48	0.23
PBEB	vapor	-0.43	0.11			-2.22	0.11	-3.33	0.14		
	particle	-2.64	0.14	-2.30	0.14	-3.41	0.43	-2.74	0.35		
	precip	1.76	0.35	1.37	0.20	1.41	0.41	0.94	0.40	1.88	0.42
HBB	vapor	-0.40	0.16					-1.74	0.20	-2.01	0.19
	particle	-1.59	0.20	-1.33	0.19	-1.59	0.19	-2.02	0.29	-2.41	0.28
	precip	3.39	0.70	2.70	0.35	2.79	0.52	3.83	0.44	3.64	0.45
TBE	vapor	-0.78	0.17	-1.79	0.63	-1.47	0.17	-2.07	0.23	-2.11	0.37
	particle	-0.53	0.20	-0.47	0.15	-1.37	0.15	-0.67	0.21	-2.19	0.22
	precip	4.19	0.26	4.35	0.19	3.20	0.27	3.42	0.23	3.20	0.26
DBDPE	particle			1.13	0.28					-1.51	0.55
	precip	5.54	0.32	5.67	0.33	4.99	0.34	5.06	0.24	5.32	0.24
TBB	particle	1.08	0.38	1.57	0.46	-1.43	0.44				
	precip	30.32	5.19			14.07	2.62	29.14	4.44		
TBPH	particle										
	precip	12.67	4.19	12.16	3.61			28.96	5.72		

Table S3. The regression results for parameters a_1 and $a_2 \pm$ standard errors for $P \leq 5\%$ for either parameter; otherwise, the cell is blank.

		Chi		Clev		StPt		SBD		EH	
		a_1	err								
BDE-47	vapor	-0.43	0.07	-0.81	0.08	-0.23	0.07	-0.61	0.08	-0.15	0.07
	particle									-0.24	0.07
	precip										
BDE-99	vapor	-0.32	0.07	-0.40	0.08						
	particle										
	precip										
BDE-209	vapor										
	particle										
	precip									0.70	0.15
Total PBDEs	vapor	-0.36	0.08	-0.62	0.08	-0.20	0.08	-0.70	0.09	-0.28	0.12
	particle										
	precip									0.92	0.25
PBEB	vapor	-0.24	0.08	-0.54	0.08	-0.24	0.10	-0.64	0.10	-0.28	0.07
	particle					0.21	0.10	0.50	0.10	-0.42	0.08
	precip										
HBB	vapor	-0.31	0.10	-0.25	0.10					-0.28	0.14
	particle	0.25	0.11	0.52	0.11	0.31	0.11	0.69	0.11	-1.15	0.14
	precip										
TBE	vapor										
	particle										
	precip									0.73	0.17
DBDPE	particle									0.35	0.16
	precip									0.41	0.18
										0.52	0.19
TBB	particle										
	precip										
TBPH	particle										
	precip										

Table S4. The regression results for parameter $a_3 \pm$ standard errors (each $\times 10^{-4}$) for P $\leq 5\%$; otherwise, the cell is blank.

		Chi		Clev		StPt		SBD		EH	
		a_3	a_3 err	a_3	a_3 err	a_3	a_3 err	a_3	a_3 err	a_3	a_3 err
BDE-47	vapor	-3.63	0.73	-2.12	0.69	2.01	0.61	2.72	0.80	2.55	0.71
	particle	-3.60	0.60	-2.06	0.56					11.52	1.99
	precip	-10.72	2.82	5.10	1.92	22.7	2.4				
BDE-99	vapor	2.01	0.74			4.14	0.80	4.37	0.96	4.00	0.98
	particle	-1.88	0.65	-2.09	0.70						
	precip	-18.52	1.98					-5.97	1.57		
BDE-209	vapor			-7.18	1.20						
	particle			-3.60	1.13	-2.24	0.81				
	precip										
Total PBDEs	vapor					2.96	1.15	5.17	1.09	6.31	1.05
	particle			-3.96	1.16	-2.60	0.94	-10.12	1.49	3.17	0.91
	precip	-8.83	2.30			17.32	2.01			7.00	1.59
PBEB	vapor	-1.74	0.79	-4.46	0.92	-3.49	0.75			4.42	1.19
	particle	-3.98	0.97	-2.54	0.97			-11.21	2.18		
	precip										
HBB	vapor	-3.59	1.06	-4.75	1.11	3.79	1.36	-5.05	1.25	-5.01	1.24
	particle							-7.42	2.19	-4.62	1.84
	precip	11.10	4.12	8.46	2.24	18.03	3.14	6.27	2.99		
TBE	vapor					-4.71	1.10				
	particle					-3.80	1.01	-3.73	1.45		
	precip			-4.75	1.31			-3.91	1.57	-5.82	1.73
DBDPE	particle							-11.91	3.44		
	precip									-4.16	1.44
TBB	particle							-22.9	5.7		
	precip	-95.00	23.50	41.96	13.7			-102	20		
TBPH	particle	9.89	2.30	10.37	2.25	5.87	1.82	-12.39	4.21		
	precip							-106	26		

Table S5. Halving (negative) and doubling (positive) times in years with standard errors for all the compounds analyzed in the vapor and particle phases and in precipitation at the five IADN sites. All the numbers reported were calculated from the statistically significant a_3 terms in equation 2 for $P \leq 5\%$; otherwise, the cell is blank.

		Chicago	Cleveland	Sturgeon Point	Sleeping Bear Dunes	Eagle Harbor
BDE-47	Vapor	-5.2 ± 1.1	-8.9 ± 2.9	9.4 ± 2.8	7.0 ± 2.0	7.4 ± 2.1
	Particle	-5.3 ± 0.9	-9.2 ± 2.5			
	Precip	-1.8 ± 0.5	3.7 ± 1.4	0.8 ± 0.1		1.6 ± 0.3
BDE-99	Vapor	9.5 ± 3.5		4.6 ± 0.9	4.3 ± 1.0	4.7 ± 1.2
	Particle	-10.1 ± 3.5	-9.1 ± 3.0			
	Precip	-1.0 ± 0.1			-3.2 ± 0.8	
BDE-209	Vapor		-2.6 ± 0.4			
	Particle		-5.3 ± 1.7	-8.5 ± 3.1		
	Precip					
Total PBDEs	Vapor			6.4 ± 2.5	3.7 ± 0.8	3.0 ± 0.5
	Particle		-4.8 ± 1.4	-7.3 ± 2.6	-1.9 ± 0.3	6.0 ± 1.7
	Precip	-2.1 ± 0.6		1.1 ± 0.1		2.7 ± 0.6
PBEB	Vapor	-10.9 ± 4.9	-4.3 ± 0.9	-5.4 ± 1.2		4.3 ± 1.2
	Particle	-4.8 ± 1.2	-7.5 ± 2.9		-1.7 ± 0.3	
	Precip					
HBB	Vapor	-5.3 ± 1.6	-4.0 ± 0.9	5.0 ± 1.8	-3.8 ± 0.9	-3.8 ± 0.9
	Particle				-2.6 ± 0.8	-4.1 ± 1.6
	Precip	1.7 ± 0.6	2.2 ± 0.6	1.1 ± 0.2	3.0 ± 1.4	
TBE	Vapor			-4.0 ± 0.9		
	Particle			-5.0 ± 1.3	-5.1 ± 2.0	
	Precip		-4.0 ± 1.1		-4.9 ± 1.9	-3.3 ± 1.0
DBDPE	Particle				-1.6 ± 0.5	
	Precip					-4.6 ± 1.6
TBB	Particle				-0.8 ± 0.2	
	Precip	-0.2 ± 0.0	0.5 ± 0.1		-0.2 ± 0.0	
TBPH	Particle	1.9 ± 0.4	1.8 ± 0.4	3.2 ± 1.0	-1.5 ± 0.5	
	Precip				-0.2 ± 0.0	

Table S6. The dates \pm standard errors on which the maximum concentrations were observed. These dates and errors were calculated from a_2 and a_3 using the methods given in Venier and Hites [*Environ. Sci. Technol.* **2010**, *44*, 8050-8055]. If either parameter was not significant, the cell is blank.

		Chicago	Cleveland	Sturgeon Point	Sleeping Bear Dunes	Eagle Harbor
BDE-47	Vapor Particle Precip	July 28 \pm 5	July 21 \pm 6	Aug 2 \pm 14		
BDE-99	Vapor Particle Precip	Aug 8 \pm 8				
BDE-209	Vapor Particle Precip				Feb 19 \pm 10	Feb 5 \pm 16
Total PBDEs	Vapor Particle Precip	July 31 \pm 6	July 16 \pm 6	Aug 8 \pm 15		July 31 \pm 15
PBEB	Vapor Particle Precip	July 24 \pm 8	July 21 \pm 9 Jan 22 \pm 11	Aug 3 \pm 8	Feb 28 \pm 13	
HBB	Vapor particle precip	Aug 21 \pm 15 Jan 25 \pm 11	Jan 23 \pm 8	July 14 \pm 7		Feb 3 \pm 11
TBE	vapor particle precip				Mar 5 \pm 12	Feb 7 \pm 16

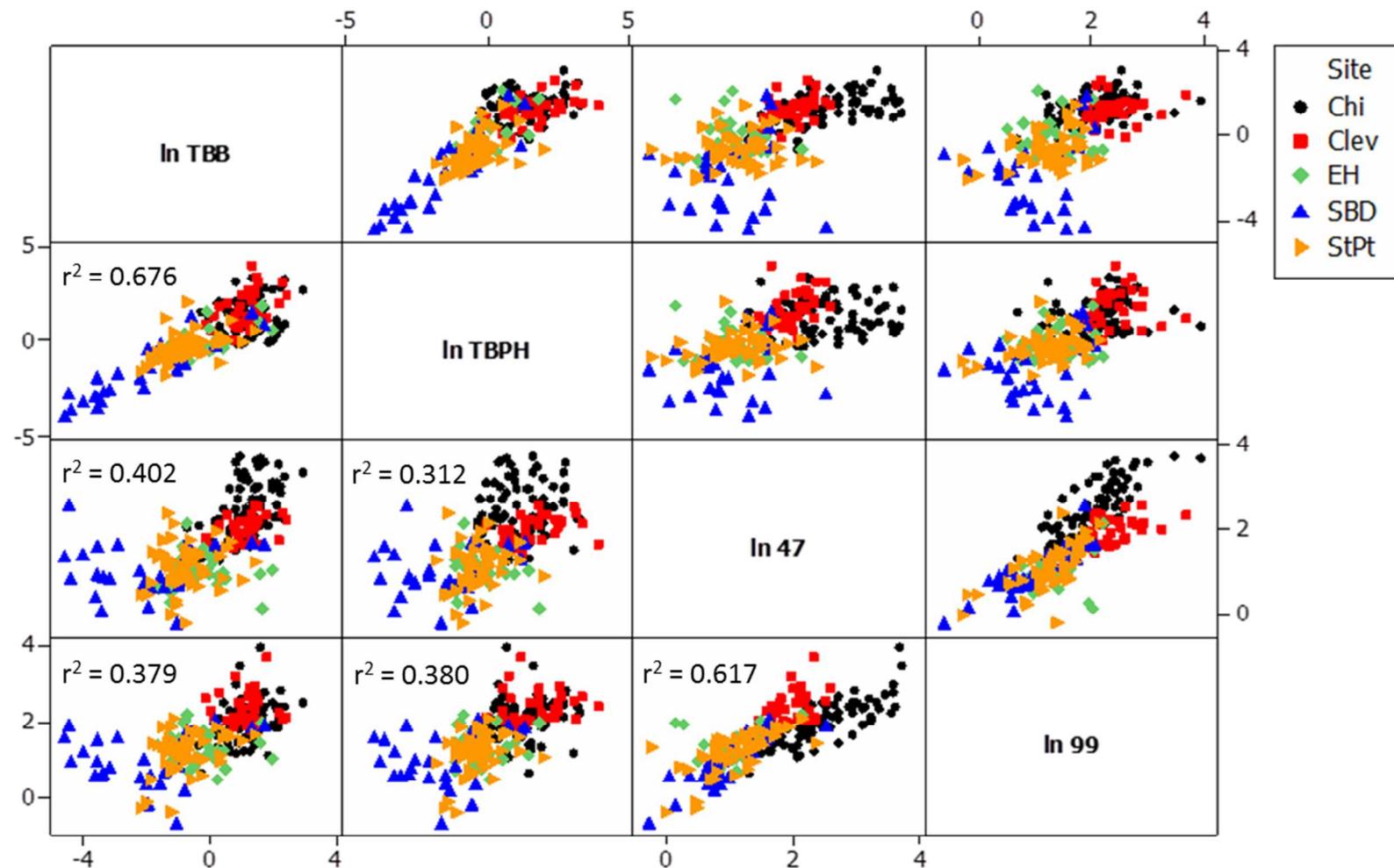


Figure S1. Logarithmically transformed concentrations (in pg/m^3) of TBB, TBPH, BDE-47, and BDE-99 plotted vs. one another (a so-called matrix plot) at the five IADN sites. The color coded site legend is given above right.

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