

Supplemental Material

Hydroxylated Metabolites of Polybrominated Diphenyl Ethers (PBDEs) in Human Blood Samples from the United States

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Supplemental Material, Table 1

Full names of the PBDEs and hydroxylated PBDEs mentioned in this paper:

BDE-28	2,4,4'-tribromodiphenyl ether
BDE-47	2,2',4,4'-tetrabromodiphenyl ether
BDE-71	2,3',4',6-tetrabromodiphenyl ether
BDE-77	3,3',4,4'-tetrabromodiphenyl ether
BDE-99	2,2',4,4',5-pentabromodiphenyl ether
BDE-100	2,2',4,4',6-pentabromodiphenyl ether
BDE-153	2,2',4,4',5,5'-hexabromodiphenyl ether
BDE-154	2,2',4,4',5,6'-hexabromodiphenyl ether
BDE-166	2,3,4,4',5,6-hexabromodiphenyl ether
4'-HO-BDE-17	4'-HO-2,2',4-tribromodiphenyl ether
2'-HO-BDE-28	2'-HO-2,4,4'-tribromodiphenyl ether
4-HO-BDE-42	4-HO-2,2',3,4'-tetrabromodiphenyl ether
3-HO-BDE-47	3-HO-2,2',4,4'-tetrabromodiphenyl ether
5-HO-BDE-47	5-HO-2,2',4,4'-tetrabromodiphenyl ether
6-HO-BDE-47	6-HO-2,2',4,4'-tetrabromodiphenyl ether
4'-HO-BDE-49	4'-HO-2,2',4,5'-tetrabromodiphenyl ether
2'-HO-BDE-66	2'-HO-2,3',4,4'-tetrabromodiphenyl ether
2'-HO-BDE-68	2'-HO-2,3',4,5'-tetrabromodiphenyl ether
4-HO-BDE-90	4-HO-2,2',3,4',5-pentabromodiphenyl ether
5'-HO-BDE-99	5'-HO-2,2',4,4',5-pentabromodiphenyl ether
6'-HO-BDE-99	6'-HO-2,2',4,4',5-pentabromodiphenyl ether
5'-HO-BDE-100	5'-HO-2,2',4,4',6-pentabromodiphenyl ether
4'-HO-BDE-101	4'-HO-2,2',4,5,5'-pentabromodiphenyl ether
4'-HO-BDE-103	4'-HO-2,2',4,5',6-pentabromodiphenyl ether

Statistical analysis of the measured concentrations: The following three tables give the measured PBDE and HO-PBDE concentrations in (a) picograms per gram (pg/g) of plasma (“wet weight”) separated by the neonatal and maternal samples, (b) pg/g lipid pg/g separated by the neonatal and maternal samples, and (c) pg/g lipid in which the neonatal and maternal samples have been combined. In all cases, the column averages, standard errors, and medians are given. The % of total rows are the average concentration relative to the total of the PBDEs or phenolic compound concentrations. In the first two cases, the mean concentrations (C_1) for the neonates have been compared to the mean concentrations (C_2) in the mothers using the two-tail Student’s t-test:

$$t = \frac{Abs(C_1 - C_2)}{\sqrt{S_1^2 + S_2^2}}$$

where S_1 and S_2 are the standard errors of the neonate and maternal concentrations, respectively.

The first two tables show that virtually all of the t -values correspond to probabilities well below 0.05, indicating that the concentrations in the neonatal and maternal samples cannot be distinguished from one another. Thus, the data have been combined in the third table. Summary results from these tables are presented in Table 1 of the paper.

Possible dietary exposure to HO-PBDEs: *Ortho*-hydroxylated PBDEs occur naturally; for example, 6-HO-BDE47 is known to be produced by algae, sponges, and fish (Valters et al. 2005). In fact, this compound was by far the most abundant OH-PBDE in the plasma of 13 pelagic- and benthic-feeding fish in the Detroit River, and an anthropogenic source (e.g., wastewater effluent) was suspected. We wondered if the HO-PBDEs we observed in our human subjects could have been coming from this source rather than from metabolism. We suggest that this dietary source is negligible for the following reasons:

First, there is no evidence to that HO-PBDEs are present in the lipids of the fish people eat. Since blood accounts for only a small portion of a fish’s body weight and concentration ratios of total HO-PBDEs to total PBDEs ranged from 0.05-2% in plasma from fish from the Detroit River (Valters et al. 2005), we conclude that, compared to metabolism of PBDEs, this source of HO-PBDEs is negligible.

Second, let’s do a simple mass analysis. Valters et al. (2005) reported that in the plasma of fish from the Detroit River, the concentration ratios of total PBDEs to total HO-PBDEs ranged from 50 to 2000. Given that the blood usually accounts for 1.5~3% of the body weight, that lipid accounts for 1~10% of the body weight of fishes, and that PBDEs are far more lipid soluble than HO-PBDEs, we conclude that in fish, the total mass of PBDEs are several orders of magnitude higher than that of HO-PBDEs. In other words, on the assumption that the uptake efficiencies

of PBDEs and HO-PBDEs from food are the same, a fish diet will contribute several orders of magnitude more PBDEs than HO-PBDEs. In this study, however, HO-PBDEs and PBDEs were roughly at the same concentration levels, indicating that HO-PBDEs from diet can be neglected.

Third, before fish are eaten, most of the blood is removed, and it is this blood that contains the relatively high levels of HO-PBDEs.

Fourth, dietary exposure to PBDEs is generally considered less important than inhalation/digestion of indoor dust.

For all these reasons, in this paper, we neglected potential dietary exposure to HO-PBDEs, and concluded that these compounds were produced from the metabolic degradation of PBDEs in people.

Supplemental Material, Table 2
Concentrations on wet weight basis (pg/g plasma)

No.	wet wgt (g)	lipid wgt (g)	BDE 28	BDE 47	BDE 100	BDE 99	BDE 154	BDE 153	Tot PBDE	2,4-DBP	2,4,6-TBP	2,4,5-TBP	6-HO-BDE47	3-HO-BDE47	5-HO-BDE47	4'-HO-BDE49	4-HO-BDE42	6'-HO-BDE99	5'-HO-BDE99	Tot OH-BDE
BABIES																				
HB1-1	9.45	0.027	2.86	58.46	13.98	17.47	4.45	33.04	130.2	10.80	13.98	2.86	2.22	1.27	14.30	ND	0.95	0.95	7.31	54.65
HB1-2	10.34	0.019	1.81	52.86	9.36	19.03	3.32	7.25	93.65	13.59	12.99	5.44	102.1	2.11	81.26	ND	2.42	4.83	39.57	264.3
HB1-4	10.21	0.022	1.53	26.02	6.73	15.92	3.06	12.24	65.50	7.04	11.63	1.53	2.14	0.92	12.55	ND	ND	1.22	7.96	45.01
HB1-5	10.39	0.029	2.10	25.86	4.81	12.03	2.71	3.31	50.82	6.92	9.32	0.90	2.10	0.60	6.01	ND	ND	0.60	2.41	28.89
HB2-1	6.99	0.008	5.52	191.1	75.15	32.27	39.49	72.18	415.7	61.14	11.04	11.89	20.38	3.40	88.31	ND	ND	3.82	24.20	224.2
HB2-2	9.59	0.023	20.44	583.7	58.81	313.2	28.30	23.90	1028	27.36	4.72	4.72	26.10	1.57	20.76	5.35	2.52	3.77	43.08	139.9
HB2-4	6.63	0.016	35.90	1318	161.8	309.5	20.90	62.26	1908	460.3	53.17	234.5	148.6	35.90	601.7	ND	ND	36.35	580.3	2151
HB2-5	8.77	0.025	1.37	21.61	3.09	9.95	ND	ND	36.04	5.49	3.77	2.06	0.34	ND	4.80	1.37	ND	ND	3.77	21.64
HB3-1	10.34	0.023	2.33	32.91	5.83	11.36	ND	25.05	77.49	32.04	16.02	8.16	6.99	1.46	34.37	1.46	ND	2.04	17.19	119.7
HB3-2	11.33	0.028	1.05	17.38	3.42	5.53	1.32	5.79	34.50	8.95	4.48	1.32	2.63	0.53	7.64	ND	ND	1.05	5.27	31.89
HB3-4	13.70	0.026	1.10	21.27	4.82	14.69	2.19	3.51	47.58	10.96	10.31	2.85	0.88	0.22	6.36	ND	ND	0.44	4.82	36.86
HB3-5	12.50	0.027	1.98	45.13	5.21	11.65	0.99	2.73	67.69	17.36	17.85	5.21	1.98	0.74	22.56	ND	ND	1.98	22.81	90.52
HB4-1	13.80	0.033	1.80	49.95	7.65	9.90	4.50	6.53	80.33	17.33	3.60	4.50	4.95	ND	26.10	2.70	ND	1.35	22.73	83.28
HB6-5	12.32	0.028	0.76	13.87	2.52	5.55	0.50	1.77	24.97	7.06	13.62	1.01	2.52	0.76	12.61	1.51	ND	1.77	10.84	51.71
HB7-2	4.20	0.056	2.48	77.85	13.25	43.07	3.31	30.64	170.6	24.85	36.44	9.11	6.63	0.83	47.21	5.80	ND	1.66	32.30	164.8
HB8-2	4.44	0.077	1.81	45.81	8.44	16.28	3.62	6.63	82.58	9.04	10.25	2.41	1.21	0.60	6.63	ND	ND	1.21	3.62	34.98
Average	9.69	0.029	5.30	161.3	24.05	52.96	8.48	19.79	269.6	45.02	14.57	18.65	20.74	3.84	62.07	3.03	1.96	4.20	51.78	221.5
Std err	0.73	0.004	2.36	84.66	10.57	25.33	3.22	5.69	125.8	27.91	3.23	14.41	10.58	2.49	36.54	0.83	0.51	2.32	35.39	129.9
Median	10.28	0.027	1.90	45.47	7.19	15.30	3.32	7.25	78.91	12.28	11.34	3.68	2.58	0.87	17.53	ND	ND	1.66	14.01	68.96
% of total			2.0	59.8	8.9	19.6	3.1	7.3	100.0	20.3	6.6	8.4	9.4	1.6	28.0	1.4	0.9	1.9	23.4	100.0
MOMS																				
HB4-3	7.41	0.041	6.28	161.8	19.80	47.32	6.28	21.25	262.7	14.49	3.38	1.45	2.90	ND	16.42	1.93	ND	1.93	20.76	63.28
HB4-5	6.10	0.053	1.77	70.06	20.02	18.25	11.77	63.58	185.4	5.89	2.94	1.77	0.59	0.59	7.06	1.77	ND	1.77	11.19	33.57
HB7-1	5.51	0.062	5.43	242.1	39.85	116.5	13.28	150.3	567.6	8.45	7.25	2.42	1.81	1.21	6.64	1.21	ND	3.62	23.55	56.16
HB8-1	4.72	0.021	0.75	39.79	11.26	18.77	ND	10.51	81.09	8.26	8.26	1.50	1.50	ND	9.01	1.50	ND	0.75	3.75	34.55
Average	5.94	0.044	3.56	128.4	22.73	50.22	10.45	61.42	274.2	9.27	5.46	1.78	1.70	0.90	9.78	1.60	ND	2.02	14.81	46.89
Std err	0.57	0.009	1.35	45.92	6.06	23.12	2.13	31.78	104.6	1.83	1.34	0.22	0.48	0.31	2.27	0.16		0.60	4.54	7.55
Median	5.81	0.047	3.60	115.9	19.91	33.05	11.77	42.41	224.1	8.36	5.31	1.63	1.66	0.90	8.04	1.63	ND	1.85	15.98	45.36
% of total			1.3	46.8	8.3	18.3	3.8	22.4	100.0	19.8	11.6	3.8	3.6	1.9	20.9	3.4	0.0	4.3	31.6	100.0
t-value			0.64	0.34	0.11	0.08	0.51	1.33	0.03	1.28	2.61	1.17	1.80	1.09	1.43	1.69		0.91	1.04	1.34

Supplemental Material, Table 3
Concentration on lipid basis (ng/g lipid)

No.	wet wgt (g)	lipid wgt (g)	BDE 28	BDE 47	BDE 100	BDE 99	BDE 154	BDE 153	Tot PBDE	2,4-DBP	2,4,6-TBP	2,4,5-TBP	6-HO-BDE47	3-HO-BDE47	5-HO-BDE47	4'-HO-BDE49	4-HO-BDE42	6'-HO-BDE99	5'-HO-BDE99	Tot OH-BDE
BABIES																				
HB1-1	9.45	0.027	0.98	20.09	4.80	6.00	1.53	11.35	44.76	3.71	4.80	0.98	0.76	0.44	4.91	ND	0.33	0.33	2.51	18.78
HB1-2	10.34	0.019	0.98	28.72	5.09	10.34	1.81	3.94	50.87	7.38	7.06	2.95	55.46	1.15	44.14	ND	1.31	2.63	21.50	143.6
HB1-4	10.21	0.022	0.71	12.07	3.13	7.39	1.42	5.68	30.40	3.27	5.40	0.71	0.99	0.43	5.82	ND	ND	0.57	3.69	20.88
HB1-5	10.39	0.029	0.75	9.26	1.72	4.31	0.97	1.18	18.19	2.48	3.34	0.32	0.75	0.22	2.15	ND	ND	0.22	0.86	10.33
HB2-1	6.99	0.008	4.68	161.9	63.70	27.35	33.47	61.18	352.3	51.83	9.36	10.08	17.28	2.88	74.86	ND	ND	3.24	20.51	190.0
HB2-2	9.59	0.023	8.60	245.5	24.74	131.8	11.91	10.05	432.6	11.51	1.98	1.98	10.98	0.66	8.73	2.25	1.06	1.59	18.12	58.87
HB2-4	6.63	0.016	15.01	550.9	67.63	129.4	8.74	26.02	797.6	192.4	22.23	98.02	62.12	15.01	251.5	ND	ND	15.20	242.6	899.1
HB2-5	8.77	0.025	0.48	7.63	1.09	3.51	ND	ND	12.74	1.94	1.33	0.73	0.12	ND	1.70	0.48	ND	ND	1.33	7.63
HB3-1	10.34	0.023	1.05	14.83	2.62	5.12	ND	11.29	34.91	14.43	7.22	3.67	3.15	0.66	15.48	0.66	ND	0.92	7.74	53.93
HB3-2	11.33	0.028	0.43	7.10	1.40	2.26	0.54	2.37	14.09	3.66	1.83	0.54	1.08	0.22	3.12	ND	ND	0.43	2.15	13.02
HB3-4	13.70	0.026	0.57	11.12	2.52	7.68	1.15	1.83	24.87	5.73	5.39	1.49	0.46	0.11	3.32	ND	ND	0.23	2.52	19.25
HB3-5	12.50	0.027	0.92	20.97	2.42	5.42	0.46	1.27	31.46	8.07	8.30	2.42	0.92	0.35	10.49	0.69	ND	0.92	10.60	42.76
HB4-1	13.80	0.033	0.74	20.67	3.17	4.10	1.86	2.70	33.24	7.17	1.49	1.86	2.05	ND	10.80	1.12	ND	0.56	9.40	34.45
HB6-5	12.32	0.028	0.34	6.18	1.12	2.47	0.22	0.79	11.13	3.15	6.07	0.45	1.12	0.34	5.62	0.67	ND	0.79	4.83	23.04
HB7-2	4.20	0.056	0.19	5.82	0.99	3.22	0.25	2.29	12.76	1.86	2.72	0.68	0.50	0.06	3.53	0.43	ND	0.12	2.42	12.32
HB8-2	4.44	0.077	0.10	2.63	0.48	0.93	0.21	0.38	4.74	0.52	0.59	0.14	0.07	0.03	0.38	ND	ND	0.07	0.21	2.01
Average	9.69	0.029	2.28	70.34	11.66	21.95	4.61	9.49	119.2	19.95	5.57	7.94	9.86	1.61	27.91	0.90	0.90	1.85	21.94	96.87
Std err	0.73	0.004	1.01	36.15	5.46	10.71	2.41	4.08	55.18	11.89	1.30	6.04	4.92	1.05	15.68	0.24	0.30	0.98	14.82	55.02
Median	10.28	0.027	0.75	13.45	2.57	5.27	1.28	2.70	30.93	4.72	5.09	1.24	1.03	0.39	5.72	ND	ND	0.57	4.26	21.96
% of total			1.9	59.0	9.8	18.4	3.9	8.0	100.0	20.6	5.7	8.2	10.2	1.7	28.8	0.9	0.9	1.9	22.6	100.0
MOMS																				
HB4-3	7.41	0.041	1.12	28.90	3.54	8.45	1.12	3.80	46.93	2.59	0.60	0.26	0.52	ND	2.93	0.35	ND	0.35	3.71	11.30
HB4-5	6.10	0.053	0.20	8.02	2.29	2.09	1.35	7.27	21.22	0.67	0.34	0.20	0.07	0.07	0.81	0.20	ND	0.20	1.28	3.84
HB7-1	5.51	0.062	0.49	21.67	3.57	10.43	1.19	13.46	50.80	0.76	0.65	0.22	0.16	0.11	0.59	0.11	ND	0.32	2.11	5.03
HB8-1	4.72	0.021	0.17	8.78	2.48	4.14	ND	2.32	17.90	1.82	1.82	0.33	0.33	ND	1.99	0.33	ND	0.17	0.83	7.62
Average	5.94	0.044	0.49	16.84	2.97	6.28	1.22	6.71	34.21	1.46	0.85	0.25	0.27	0.09	1.58	0.25	ND	0.26	1.98	6.95
Std err	0.57	0.009	0.22	5.10	0.34	1.92	0.07	2.48	8.52	0.46	0.33	0.03	0.10	0.02	0.54	0.06	ND	0.04	0.63	1.65
Median	5.81	0.047	0.34	15.22	3.01	6.30	1.19	5.53	34.07	1.29	0.63	0.24	0.25	0.09	1.40	0.27	ND	0.26	1.69	6.32
% of total			1.4	49.2	8.7	18.4	3.6	19.6	100.0	21.0	12.3	3.6	3.9	1.3	22.7	3.5	0.0	3.7	28.5	100.0
t-value			1.73	1.47	1.59	1.44	1.41	0.58	1.52	1.55	3.52	1.27	1.95	1.45	1.68	0.95		1.59	1.35	1.63

Supplemental Material, Table 4
Combined concentrations on lipid basis (ng/g lipid)

No.	wet wgt (g)	lipid wgt (g)	BDE 28	BDE 47	BDE 100	BDE 99	BDE 154	BDE 153	Tot PBDE	2,4-DBP	2,4,6-TBP	2,4,5-TBP	6-HO-BDE47	3-HO-BDE47	5-HO-BDE47	4'-HO-BDE49	4-HO-BDE42	6'-HO-BDE99	5'-HO-BDE99	Tot OH-BDE
HB1-1	9.45	0.027	0.98	20.09	4.80	6.00	1.53	11.35	44.76	3.71	4.80	0.98	0.76	0.44	4.91	ND	0.33	0.33	2.51	18.79
HB1-2	10.34	0.019	0.98	28.72	5.09	10.34	1.81	3.94	50.87	7.38	7.06	2.95	55.46	1.15	44.14	ND	1.31	2.63	21.50	143.6
HB1-4	10.21	0.022	0.71	12.07	3.13	7.39	1.42	5.68	30.40	3.27	5.40	0.71	0.99	0.43	5.82	ND	ND	0.57	3.69	20.90
HB1-5	10.39	0.029	0.75	9.26	1.72	4.31	0.97	1.18	18.19	2.48	3.34	0.32	0.75	0.22	2.15	ND	ND	0.22	0.86	10.35
HB2-1	6.99	0.008	4.68	161.9	63.70	27.35	33.47	61.18	352.3	51.83	9.36	10.08	17.28	2.88	74.86	ND	ND	3.24	20.51	190.1
HB2-2	9.59	0.023	8.60	245.5	24.74	131.8	11.91	10.05	432.6	11.51	1.98	1.98	10.98	0.66	8.73	2.25	1.06	1.59	18.12	58.87
HB2-4	6.63	0.016	15.01	550.9	67.63	129.4	8.74	26.02	797.6	192.4	22.23	98.02	62.12	15.01	251.5	ND	ND	15.20	242.6	899.1
HB2-5	8.77	0.025	0.48	7.63	1.09	3.51	ND	ND	12.74	1.94	1.33	0.73	0.12	ND	1.70	0.48	ND	ND	1.33	7.66
HB3-1	10.34	0.023	1.05	14.83	2.62	5.12	ND	11.29	34.91	14.43	7.22	3.67	3.15	0.66	15.48	0.66	ND	0.92	7.74	53.94
HB3-2	11.33	0.028	0.43	7.10	1.40	2.26	0.54	2.37	14.09	3.66	1.83	0.54	1.08	0.22	3.12	ND	ND	0.43	2.15	13.04
HB3-4	13.70	0.026	0.57	11.12	2.52	7.68	1.15	1.83	24.87	5.73	5.39	1.49	0.46	0.11	3.32	ND	ND	0.23	2.52	19.27
HB3-5	12.50	0.027	0.92	20.97	2.42	5.42	0.46	1.27	31.46	8.07	8.30	2.42	0.92	0.35	10.49	0.69	ND	0.92	10.60	42.77
HB4-1	13.80	0.033	0.74	20.67	3.17	4.10	1.86	2.70	33.24	7.17	1.49	1.86	2.05	ND	10.80	1.12	ND	0.56	9.40	34.47
HB6-5	12.32	0.028	0.34	6.18	1.12	2.47	0.22	0.79	11.13	3.15	6.07	0.45	1.12	0.34	5.62	0.67	ND	0.79	4.83	23.05
HB7-2	4.20	0.056	0.19	5.82	0.99	3.22	0.25	2.29	12.76	1.86	2.72	0.68	0.50	0.06	3.53	0.43	ND	0.12	2.42	12.33
HB8-2	4.44	0.077	0.10	2.63	0.48	0.93	0.21	0.38	4.74	0.52	0.59	0.14	0.07	0.03	0.38	ND	ND	0.07	0.21	2.03
HB4-3	7.41	0.041	1.12	28.90	3.54	8.45	1.12	3.80	46.93	2.59	0.60	0.26	0.52	ND	2.93	0.35	ND	0.35	3.71	11.32
HB4-5	6.10	0.053	0.20	8.02	2.29	2.09	1.35	7.27	21.22	0.67	0.34	0.20	0.07	0.07	0.81	0.20	ND	0.20	1.28	3.85
HB7-1	5.51	0.062	0.49	21.67	3.57	10.43	1.19	13.46	50.80	0.76	0.65	0.22	0.16	0.11	0.59	0.11	ND	0.32	2.11	5.04
HB8-1	4.72	0.021	0.17	8.78	2.48	4.14	ND	2.32	17.90	1.82	1.82	0.33	0.33	ND	1.99	0.33	ND	0.17	0.83	7.64
Average	8.94	0.032	1.93	59.64	9.93	18.82	3.79	8.90	102.2	16.25	4.63	6.40	7.94	1.26	22.64	0.66	0.90	1.52	17.95	78.90
Std err	0.68	0.004	0.82	29.16	4.41	8.64	1.89	3.24	44.57	9.60	1.12	4.85	4.01	0.82	12.69	0.18	0.30	0.78	11.92	44.50
Median	9.52	0.027	0.73	13.45	2.57	5.27	1.17	3.80	30.93	3.46	3.03	0.72	0.84	0.28	4.22	ND	ND	0.43	3.11	19.03
% of total			1.9	58.4	9.7	18.4	3.7	8.7	100.0	20.6	5.9	8.1	10.1	1.6	28.7	0.8	1.1	1.9	22.7	100.0

Reference

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