Supporting Information for

Hydroxylated and Methoxylated Polybrominated Diphenyl Ethers in a Canadian Arctic Marine Food Web

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MATERIALS AND METHODS

Compilation of physical chemical properties. Molecular weights (MW), octanol-water partition coefficients (K_{OW}'s) and octanol-air partition coefficients (K_{OA}'s) for PCBs and PBDEs were compiled from references *S1-8*. K_{OW}'s and K_{OA}'s of OH-PBDEs and MeO-PBDEs were calculated using KOWWIN and KOAWIN applications within the U.S. EPA's EPI Suite (Estimations Program Interface), which is available online (http://www.epa.gov/oppt/exposure/pubs/ episuite.htm). Table S4 summarizes the compiled MWs, K_{OW}'s and K_{OA}'s

SUPPORTING TABLES

Table S1. Compilation of previously reported OH- and MeO-PBDEs reported in the literature, including samples from laboratory experiments (rats and fish) and field-surveys (i.e. environmental samples: marine sponges, algae, mussels, freshwater and marine fish, marine mammals, birds and humans), including indication of likely sources (i.e., biogenic formation vs. *in vivo* metabolic formation) and potential PBDE precursors.

Compound	Field and Laboratory Observations	Likely Source(s)	Potential Precursor
	OH- and MeO-Br_2 Analogues		
3′-OH-BDE-7	**Environment:** Abiotic media (*S9*)	PBDE Metabolite	BDE-17
	OH- and MeO-Br_3 Analogues		
6′-OH-BDE-17	**Environment:** red algae/mussels (*S10*), abiotic media (*S9*), human milk Spain (*S11*)	Natural Product/ PBDE Metabolite	BDE-28
4′-OH-BDE-17	**Environment**: marine mammals (this study), human milk Spain (*S11*)	PBDE Metabolite	BDE-28 BDE-47
2′-OH-BDE-28	**Environment**: glaucous gulls (*S12*), human milk Spain (*S11*) Laboratory: Rat (*S13*)	Natural Product/ PBDE Metabolite	BDE-47

3′-OH-BDE-28	**Environment:** Abiotic media (*S9*) **Laboratory**: Rat (*S13*)	PBDE Metabolite	
6′-MeO-BDE-17	**Environment**: Human milk Spain (*S11*)	PBDE Metabolite	BDE-28
2′-MeO-BDE-28	**Environment**: red algae/mussels (*S10*), Glaucous gulls (*S12*)	Natural Product/ PBDE Metabolite	BDE-28
4′-MeO-BDE-17	**Environment**: human milk Spain (*S11*)	PBDE Metabolite	BDE-28
4′-MeO-BDE-30	**Environment**: human milk Spain (*S11*)	PBDE Metabolite	BDE-28

OH- and MeO-Br_4 Analogues

4-OH BDE-42	**Environment**: Freshwater fish (*S14*) polar bears (*S12*) **Laboratory**: Rat (*S15*), fish (*S16*)	PBDE Metabolite (Confirmed)	BDE-47
4′-OH-BDE-49	**Environment***:* Abiotic media (*S9*), freshwater fish (*S14*) glaucous gulls *(S12),*polar bears *(S12)* **Laboratory:** Rat (*S15*), fish (*S16*)	PBDE Metabolite (Confirmed)	BDE-47
6′-OH-BDE-49	**Environmen**t: Abiotic media (*S9*), glaucous gulls (*S12*)	Natural Product	-

2′-OH-BDE-68	**Environment**: Marine sponge (*S17*,*S18*,*S19*,*S20*), red algae/mussels (*S10*), Abiotic media (*S9*), freshwater fish (*S14*), marine fish (*S21*), glaucous gulls (*S12*)	Natural Product	None
3-OH-BDE-47	**Environment:** Abiotic media (*S9*), freshwater fish (*S14*), glaucous gulls (*S12*) **Laboratory:** Rat (*S15*), fish (*S16*)	PBDE Metabolite (Confirmed)	BDE-47
5-OH-BDE-47	**Environment:** Abiotic media (*S9*), freshwater fish (*S14*)	PBDE Metabolite	BDE-47
6-OH-BDE-47	**Environment:** Marine sponge (*S18*,*S19*,*S20*), red algae/mussels (*S10*), abiotic media (*S9*), freshwater fish (*S14*), marine fish (*S21*), glaucous gulls (*S12*), human milk Spain (*S11*), human plasma Sweden (*S22*) **Laboratory:** Rat (*S15*), fish (*S16*)	PBDE Metabolite (Confirmed)	BDE-47
2′-OH-BDE75	**Environment:** human milk Spain (*S11*)	PBDE Metabolite	BDE-100
2′-OH-BDE74	**Environment**: human milk Spain (*S11*)	PBDE Metabolite	BDE-99
6′-OH-BDE-66	**Environment:** human milk Spain (*S11*) **Laboratory:** Rat (*S15*), fish (*S16*)	PBDE Metabolite (Confirmed)	BDE-47
6′-MeO-BDE-49	**Environment**: Glaucous gulls (*S12*)	Natural Product	-

2′-MeO-BDE-68	**Environment:** Marine sponge (*S23*,*S24*), red algae/mussels (*S10*), freshwater fish (*S14*); marine fish (*S21*), glaucous gulls (*S12*), cetaceans (*S25*,*S26*), seals (*S27*) polar bears (*S12*), human milk (*S28*)	Natural Product	None
3-MeO -BDE-47	**Environment:** glaucous gulls (*S12*), polar bears (*S12*)	PBDE Metabolite	BDE-47
6-MeO-BDE-47	**Environment:** Marine sponge (*S29*), red algae/mussels (*S10*), freshwater fish (*S14*), marine fish (*S21*); glaucous gulls (*S12*), cetaceans (*S25*,*S26*), seals (*S27*), polar bears (*S12*)	Natural Product/ PBDE Metabolite	BDE-47
2′-MeO-BDE-75	**Environment**: human milk Spain (*S11*)	PBDE Metabolite	BDE-47
2′-MeO-BDE-74	**Environment**: human milk Spain (*S11*)	PBDE Metabolite	BDE-47
6′-MeO-BDE-66	**Environment**: human milk Spain (*S11*)	PBDE Metabolite	BDE-47
	OH- and MeO-Br_5 Analogues		
6-OH-BDE-90	**Environment:** Red algae/mussels (*S10*), abiotic media (*S9*), freshwater fish (*S14*), marine fish (*S21*)	Natural Product	None
6-OH-BDE-99	**Environment:** Marine sponge (*S30*), red algae/mussels (*S10*), abiotic media (*S9*), freshwater fish (*S14*), marine fish (*S21*)	Natural Product	BDE-99

2-OH-BDE-123	**Environment:** Marine sponge (*S31*), red algae/mussels (*S10*), abiotic media (*S9*), freshwater fish (*S14*)	Natural Product	None
6-OH-BDE-85	**Environment:** Abiotic media (*S9*), freshwater fish (*S14*)	Natural Product	BDE-99
6 MeO-BDE-85	**Environment:** red algae/mussels (*S10*)	Natural Product	None
6-MeO-BDE-90	**Environment:** glaucous gulls (*S12*), polar bears (*S12*)	Natural Product	BDE-99
6-MeO-BDE-99	**Environment**: glaucous gulls (*S12*), polar bears (*S12*)	Natural Product/ PBDE Metabolite	BDE-99
	OH- and MeO-Br_6 Analogues		
6-OH-BDE-137	**Environment**: Marine sponge (*S17*), red algae/mussels (*S10*), abiotic media (*S9*)	Natural Product	None
6-MeO-BDE-137	**Environment**: algae/mussels (*S10*)	Natural Product	None

Table S2. List of OH- and MeO-PBDEs, relative retention times (RRTs, relative to BDE-47) on standard 30 m DB-5 column (relative to BDE-47) for GC-HRMS analysis.

OH-PBDEs	**RRT_{BDE-47}**	**MeO-PBDEs**	**RRT_{BDE-47}**
6'-OH-BDE-17	0.983	6'-MeO-BDE-17	0.948
4'-OH-BDE-30	0.984	4'-MeO- BDE-30	0.950
2'-OH-BDE-28	0.996	**2'-MeO- BDE-28**	**0.966**
3'-OH-BDE-28	1.010	3'-MeO- BDE-28	0.986
4'-OH-BDE-17	**1.010**	**4'-MeO- BDE-17**	**0.986**
6'-OH-BDE-49	1.039	6'-MeO- BDE-49	1.024
2'-OH-BDE-68	1.050	2'-MeO- BDE-68	1.038
2'-OH-BDE-75	**1.057**	**2'-OMe-BDE-75**	**1.041**
6-OH-BDE-47	**1.061**	**6-MeO -BDE-47**	**1.054**
4'-OH-BDE-69	1.064	4'-MeO- BDE-69	1.058
3-OH-BDE-47	1.074	**2'-OMe-BDE 74**	**1.063**
2'-OH-BDE-66	1.075	3-MeO-BDE-47	1.072
5'-OH-BDE-47	1.078	2'-MeO-BDE-66	1.073
4'-OH-BDE-49	1.080	5'-MeO-BDE-47	1.077
2'-OH-BDE 74	**1.081**	**6'-MeO-BDE-66**	**1.079**
6'-OH-BDE 66	**1.089**	4'-MeO-BDE-49	1.079
4'-OH-BDE-121	1.109	4'-MeO-BDE-121	1.118
4-OH-BDE-42	1.111	4-MeO-BDE-42	1.121
6-OH-BDE-90	1.122	6-MeO-BDE-90	1.136
6-OH-BDE-99	1.125	6-MeO-BDE-99	1.140
4-OH-BDE-90	1.150	4-MeO-BDE-90	1.173
2-OH-BDE-123	1.156	2-MeO-BDE-123	1.181
6-OH-BDE-85	1.167	6-MeO-BDE-85	1.197

Table S3. Frequency of detection (% of samples above MDL) in Arctic marine sediments and biota.

	Sediments	Macro-algae	Bivalves	Cod	Sculpin	Salmon	Eider Ducks	White winged Scoter	Ringed Seal Blubber
5-OH-BDE-47	0	0	0	0	0	0	0	0	0
6′-OH-BDE-49	0	0	0	0	0	0	0	0	0
2′-OH-BDE-68	0	0	0	0	0	0	0	0	0
2′-OH-BDE-75	0	0	0	0	0	0	0	0	0
6-OH-BDE-90	0	0	0	0	0	0	0	0	0
6-MeO-BDE-17	0	0	100	0	0	0	0	0	0
2′-MeO-BDE-28	0	0	70	0	0	100	0	0	100
4-MeO-BDE-42	0	0	0	0	0	0	0	0	80
5-MeO-BDE-47	0	0	0	0	0	0	0	0	70
6-MeO-BDE-47	0	0	100	100	100	100	100	100	100
6′-MeO-BDE-49	0	0	100	0	0	75	0	80	90
6′-MeO-BDE-66	0	0	70	80	0	100	0	0	40
2′-MeO-BDE-68	0	0	100	100	100	100	80	80	100
6-MeO-BDE-90	0	0	0	0	0	0	90	0	0
6-MeO-BDE-99	0	0	0	0	0	0	0	0	0

Table S3 continued.

	Male Beluga Blood	Male Beluga Liver	Male Beluga Blubber	Female Beluga Blood	Female Beluga Milk	Female Beluga Blubber	Beluga Calf Blubber
5-OH-BDE-47	0	0	70	0	0	60	0
6′-OH-BDE-49	0	0	80	0	70	100	100
2′-OH-BDE-68	0	0	70	0	100	100	100
2′-OH-BDE-75	0	0	100	0	80	20	30
6-OH-BDE-90	0	0	80	0	0	60	30
6-MeO-BDE-17	70	100	80	70	0	0	0
2′-MeO-BDE-28	70	100	100	70	100	100	100
4-MeO-BDE-42	0	80	100	0	100	100	100
5-MeO-BDE-47	0	80	80	0	80	80	60
6-MeO-BDE-47	100	100	100	100	100	100	100
6′-MeO-BDE-49	0	100	100	0	100	100	100
6′-MeO-BDE-66	50	80	100	0	80	80	60
2′-MeO-BDE-68	100	100	100	100	100	100	100
6-MeO-BDE-90	0	100	100	0	100	100	100
6-MeO-BDE-99	0	80	100	0	80	100	100

Table S4. Compilation of molecular weight ($g \cdot mol^{-1}$), logK_{OW}'s and log K_{OA}'s for Br_2-Br_{10} PBDEs, Br_2-Br_9 OH-PBDEs and Br_2-Br_9 MeO-PBDEs.

	MW	**Log K_{OW}**	**Log K_{OA}**
PBDEs			
Br_2-PBDEs	328	5.8	9.0
Br_3-PBDEs	407	6.7	10.2
Br_4-PBDEs	486	6.8	10.7
Br_5-PBDEs	565	7.7	12.0
Br_6-PBDEs	644	8.6	13.3
Br_7-PBDEs	722	9.4	14.6
Br_8-PBDEs	801	10.3	15.8
Br_9-PBDEs	880	11.2	17.1
Br_{10}-PBDEs	959	12.1	18.4
OH-PBDEs			
Br_2-OH-PBDEs	344	4.5	11.6
Br_3-OH-PBDEs	423	5.4	12.9
Br_4-OH-PBDEs	502	6.3	14.2
Br_5-OH-PBDEs	581	7.2	15.5
Br_6-OH-PBDEs	660	8.1	16.8
Br_7-OH-PBDEs	739	9.0	18.0
Br_8-OH-PBDEs	817	9.9	19.4
Br_9-OH-PBDEs	896	10.7	20.6
MeO-PBDEs			
Br_2-MeO-PBDEs	342	5.5	8.6
Br_3- MeO-PBDEs	421	6.4	9.9
Br_4- MeO-PBDEs	500	7.3	11.2
Br_5- MeO-PBDEs	579	8.2	12.5
Br_6- MeO-PBDEs	658	9.1	13.8
Br_7- MeO-PBDEs	737	10.0	15.1
Br_8- MeO-PBDEs	816	10.9	16.4
Br_9- MeO-PBDEs	894	11.8	17.6

Table S5. Regression results and trophic magnification factors (TMFs) of PBDEs and PCBs in the E. Hudson Bay marine food web.

	Log K_{OW} 25°C [a]	$\log[C_B] = mx + b$	R^2	*P* value	TMF	95 % CI
PBDEs						
BDE-28	6.9	-0.02·(TL) + 0.44	0.0003	0.88	0.96	0.57-1.6
BDE-47	7.3	0.20·(TL) + 0.17	0.095	0.0006	1.6	1.2-2.0
BDE-99	7.6	-0.12·(TL) + 0.90	0.02	0.07	0.76	0.57-1.0
BDE-100	7.4	-0.02·(TL) + 0.37	0.0004	0.80	0.96	0.72-1.3
BDE-153	7.9	-0.55·(TL) + 0.13	0.003	0.59	0.88	0.54-1.4
BDE-154	7.8	-0.08·(TL) + 0.37	0.005	0.46	0.81	0.47-1.4
MeO-PBDEs						
2′-MeO-BDE-28	6.4	–0.12·(TL) – 0.14	0.02	0.38	0.75	0.4-1.4
6-MeO-BDE-47	7.3	0.42·(TL) – 0.11	0.1	0.06	2.6	0.9-7.2
2′-MeO-BDE-68	7.3	0.35·(TL) – 0.43	0.2	0.09	2.3	0.89-5.9
6′-MeO-BDE-49	7.3	– 0.11·(TL) – 0.35	0.01	0.62	0.8	0.3-2.1
6′-MeO-BDE-66	7.3	– 0.3·(TL) – 0.39	0.15	0.03	0.98	0.72-1.3
PCBs						
PCB-28	5.0	0.45·(TL) – 1.4	0.54	6.1×10^{-18}	2.9	2.4-3.4
PCB-52	5.9	0.92·(TL) – 2.4	0.57	1.6×10^{-17}	8.3	6.1-11
PCB-101	6.4	0.99·(TL) – 2.3	0.58	2.9×10^{-12}	9.8	6.8-14
PCB-138	6.8	1.0·(TL) – 2.1	0.70	3.0×10^{-17}	10	7.6-13
PCB-153	6.9	1.0·(TL) – 2.2	0.76	6.0×10^{-18}	11	8.6-14
PCB-180	7.5	1.0·(TL) – 2.7	0.60	4.1×10^{-17}	10	7.2-14
PCB-195	7.8	0.65·(TL) – 2.7	0.25	2.6×10^{-4}	4.3	2.0-9.2
PCB-206	8.1	0.71·(TL) – 2.8	0.26	4.3×10^{-5}	5.1	2.4-11
PCB-209	8.4	0.59·(TL) – 2.6	0.20	2.7×10^{-5}	4.0	2.1-7.4

SUPPORTING FIGURES

Figure S1. Map of the Eastern Hudson Bay region of the Canadian Arctic where field sampling was conducted.

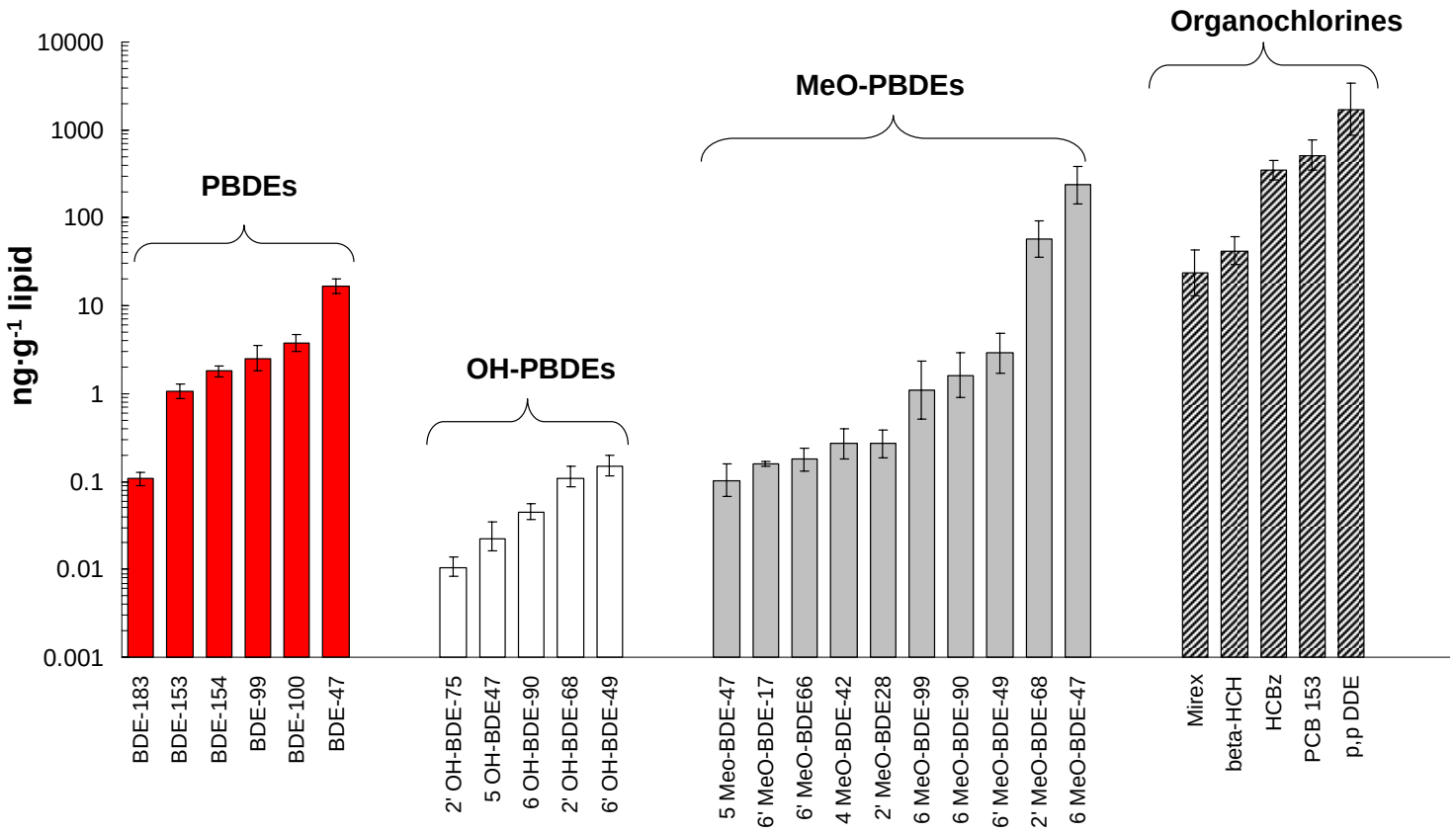

Figure S2. Concentrations (ng·g^{-1} lipid) of PBDEs, OH-PBDEs, MeO-PBDEs and several organchlorines (Mirex, beta-HCH, HCBz, PCB-153 and p,p DDE) in E. Hudson Bay beluga whale blubber. Data are geometric means and error bars represent the range of 1 standard deviation.

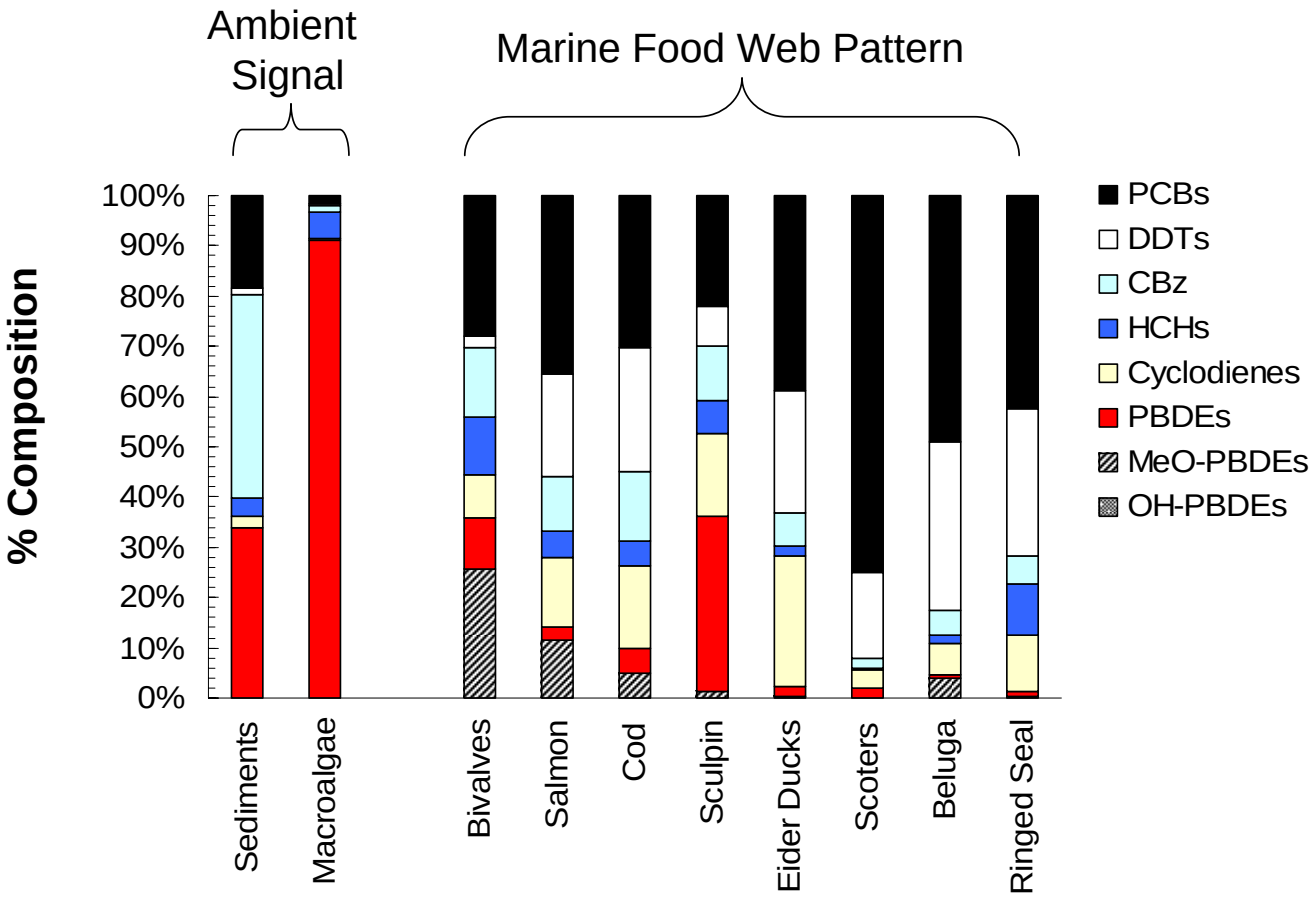

Figure S3. Percentage contribution of ∑PBDEs, ∑OH-PBDEs, ∑MeO-PBDEs and organochlorines to the total organohalogen burdens observed in sampled marine sediments, macroalgae, fish and wildlife from the Canadian Arctic.

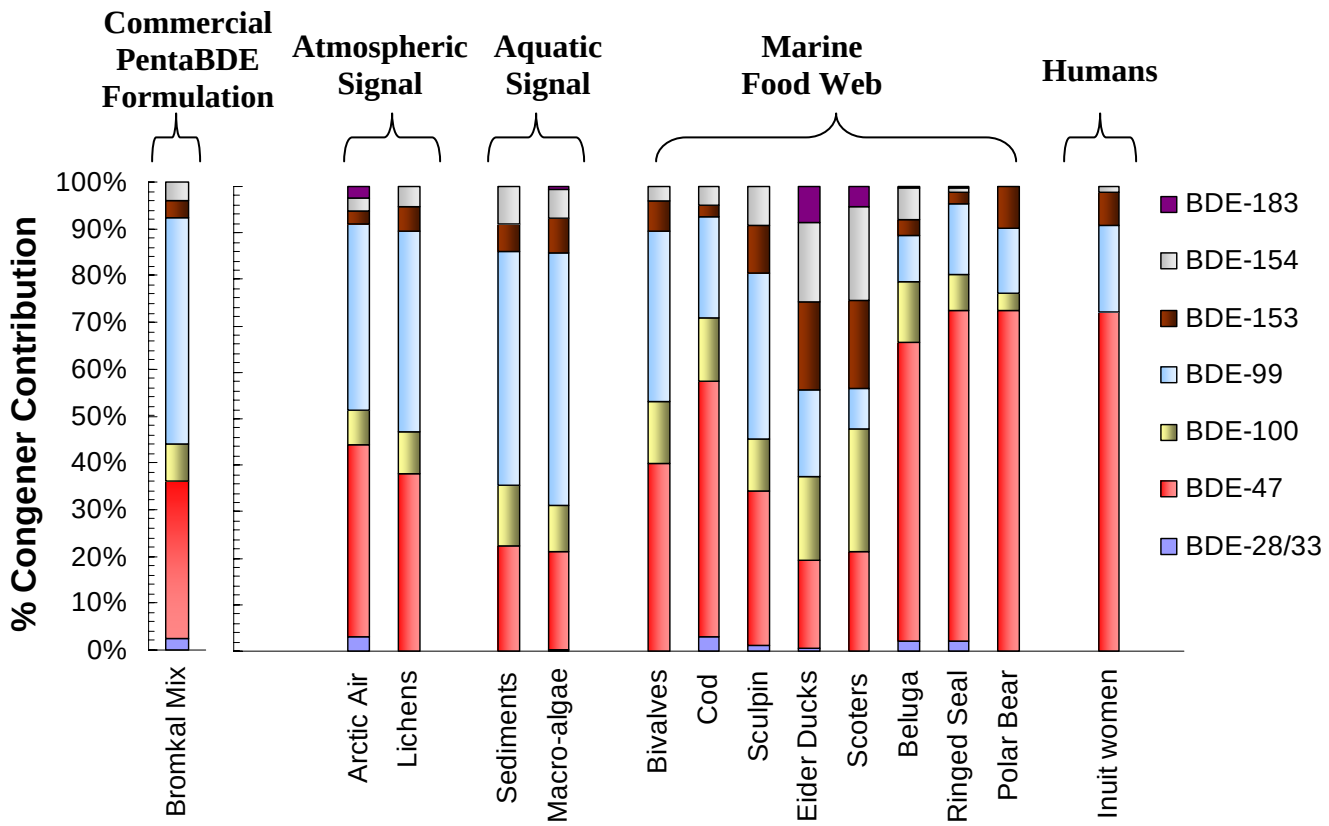

Figure S4. Composition of seven major PBDE congeners (percentage congener contribution) in a commercial PentaBDE technical mixture (Bromkal 70-5DE) compared to the observed congener composition in Arctic air, lichens, sediments, macroalgae, fish, wildlife and humans (breast milk). Previously presented in Kelly et al. (*S32*).

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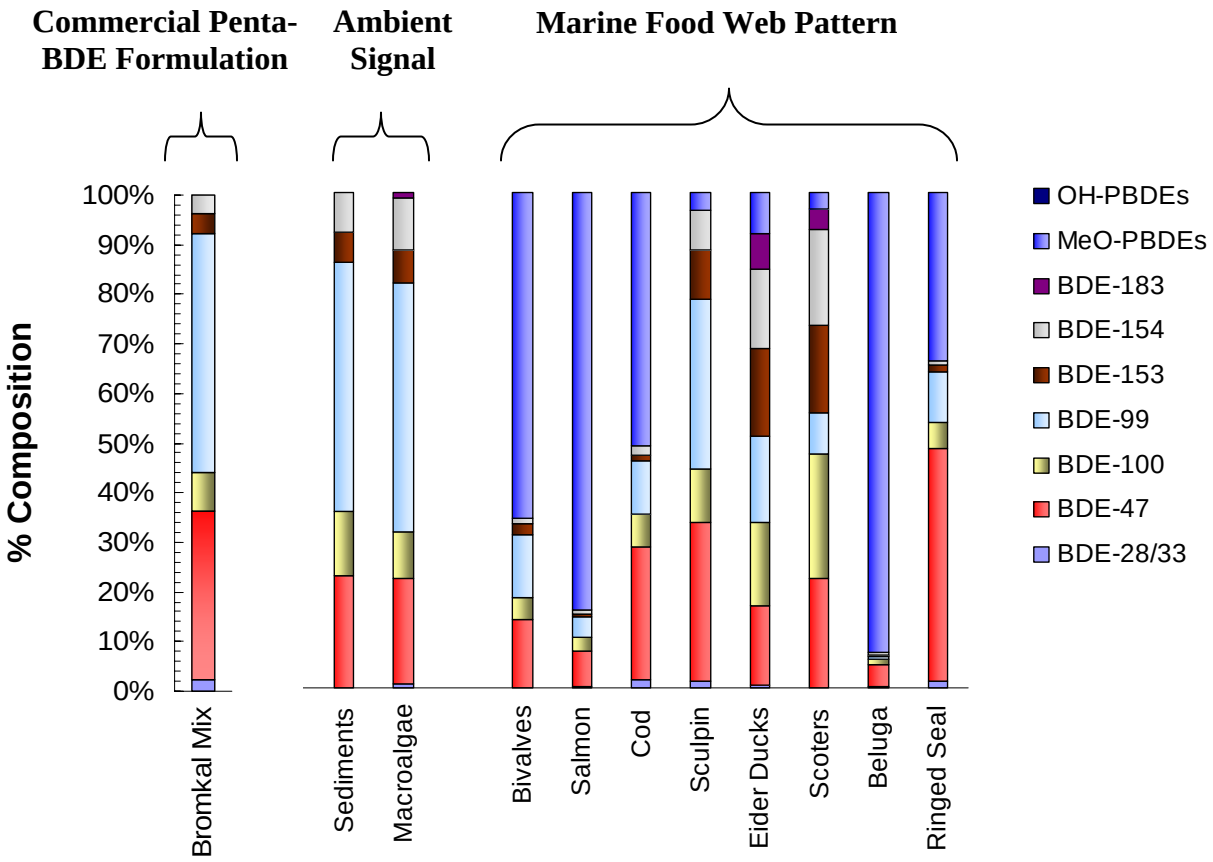

Figure S5. Composition of major Br_3-Br_7 PBDE congeners, ∑OH-PBDEs and ∑MeO-PBDEs (percentage contribution) in a commercial PentaBDE technical mixture (Bromkal 70-5DE) compared to the observed congener composition in sampled air, lichens, sediments, macroalgae, fish, wildlife from the Canadian Arctic. No data are available for OH-PBDEs and MeO-PBDEs in Bromkal 70-5DE or Arctic air.

Supplemental References

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