Supporting Information

Temporal trends of hexabromocyclododecane, polybrominated diphenyl ethers and polychlorinated biphenyls in ringed seals from East Greenland

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Quality assurance and quality control

The laboratory is accredited according to ISO17025 for the analysis of PBDEs and OCs in biota, i.e. these analyses followed accredited procedures which include regular participation in the QUASIMEME proficiency testing scheme. Control charts indicated too low values for BDE-99 and BDE-100 in one of the 2006 batches. These values are included in the further data analysis, but have to be considered as indicative. The tri- to heptaBDEs had LOQs between 0.13 and 0.66 ng/g lw, the latter primarily applicable to BDE-183. For PCBs, the LOQs varied between 0.32 and 2.6 ng/g lw depending on individual response and instrumental performance.

As far as possible, the same QA/QC elements were applied to HBCD analyses as well. Each batch contained minimum one duplicate analysis, one procedural blank and four samples of the laboratory reference material (fish oil). As the native content of β- and γ-HBCD is very low in these reference samples, two of them were spiked with 4.7 ng of each of the three diastereoisomers. The results did not exceed warning and action limits of the control charts and none of the diastereoisomers were detected in the procedural blanks. The LOQ was set to the lowest calibration standard, corresponding to approximately 2.5 ng/g lw.

Table S1: Number and mean age (range) of ringed seals included in the analysis of HBCD, PBDEs and PCBs.

Year	N	Mean age (Range)
HBCD		
1986	5	3.4 (1-6)
1994	5	4.2 (3-5)
1999	5	2.2 (1-4)
2000	5	2.6 (1-5)
2001	5	1.6 (0-3)
2002	5	3.2 (1-4)
2003	5	3.0 (2-4)
2004	5	1.8 (0-4)
2006	5	2.0 (1-3)
2008	5	6.0 (4-10)
PBDE and PCB		
Age<5 years old (1986-2008)		
2006	14	1.3 (0-4)
2008	19*	2.6 (1-4)
Age ≥ 5 years old (1994-2008)		
2006	16	12.9 (6-24)
2008	11	7.2 (5-10)

* one individual without age determination, but classified as juvenile.

Table S2: Arithmetic mean concentrations (ng/g lipid weight, lw) of HBCD in archived and new blubber samples of ringed seals from East Greenland. Numbers in brackets are median concentrations. Numbers in italics are ranges. LOQ was approximately 2.5 ng/g lw for all samples and diastereoisomers. Concentrations below LOQ are treated as zero in calculations of mean and median.

Year	α-HBCD	β-HBCD	γ-HBCD
1986	2.27 (2.02) *< LOQ – 4.15*	< LOQ	< LOQ
1994	3.03 (3.86) *1.99 – 4.42*	< LOQ	< LOQ
1999	4.51 (4.04) *3.17 – 7.98*	< LOQ	< LOQ
2000	8.11 (5.96) *3.03 – 17.9*	< LOQ	< LOQ
2001	7.19 (6.56) *4.31 – 11.2*	< LOQ *< LOQ – 1.32*	< LOQ *< LOQ – 4.07*
2002	6.36 (5.45) *4.55 – 10.7*	< LOQ	< LOQ
2003	6.19 (6.61) *4.07 – 7.69*	< LOQ	< LOQ
2004*	6.34 (6.16) *4.02 – 9.01*	< LOQ	< LOQ
2006	7.18 (7.08) *5.60 – 9.92*	< LOQ	< LOQ *< LOQ – 1.87*
2008	9.56 (8.71) *8.49 – 12.4*	< LOQ	< LOQ

* N=4 because of too low recovery in one of the samples.

Table S3: Arithmetic mean concentrations (ng/g lipid weight, lw) of PBDE congeners in blubber samples of ringed seals from East Greenland. Numbers in brackets are median concentrations. Numbers in italics are ranges. LOQ was between 0.13 and 0.66 ng/g lw for the lower brominated congeners and between 0.5 and 1 ng/g lw for BDE-209. Concentrations below LOQ are treated as zero in calculations of mean, median and ΣPBDE.

Parameter	2006	2008
Age<5 years old (1986-2008)		
BDE-17	< LOQ	< LOQ *(< LOQ – 0.18)*
BDE-28	0.84 (0.79) *0.43 – 1.25*	0.67 (0.68) *0.32 – 1.18*
BDE-47	21.4 (20.9) *10.5 – 33.7*	18.9 (17.7) *9.88 – 27.5*
BDE-49	0.31 (0.33) *< LOQ – 0.52*	0.44 (0.41) *0.17 – 0.77*
BDE-66	0.12 (0.16) *< LOQ – 0.34*	0.16 (0.19) *< LOQ – 0.32*
BDE-85	< LOQ	< LOQ
BDE-99	1.51 (1.59) *0.22 – 2.82*	1.51 (1.43) *0.69 – 2.61*
BDE-100	1.51 (1.54) *0.68 – 2.29*	1.64 (1.70) *0.86 – 2.57*
BDE-153	0.37 (0.41) *< LOQ – 0.74*	0.46 (0.43) *0.26 – 0.76*
BDE-154	0.48 (0.45) *0.17 – 0.86*	0.56 (0.54) *0.32 – 1.06*
BDE-183	< LOQ	< LOQ
BDE-209	0.78 (0.62) *< LOQ – 2.87*	< LOQ
ΣPBDE	27.3 (27.7) *13.6 – 40.0*	24.4 (22.4) *13.2 – 34.8*
Age ≥ 5 years old (1994-2008)		
BDE-17	< LOQ *< LOQ – 0.15*	< LOQ *< LOQ – 0.21*
BDE-28	1.08 (1.08) *0.45 – 2.13*	0.59 (0.55) *0.33 – 0.91*
BDE-47	32.3 (27.8) *5.06 – 64.4*	21.2 (20.4) *13.2 – 35.7*
BDE-49	0.35 (0.32) *< LOQ – 0.69*	0.46 (0.44) *0.22 – 0.66*
BDE-66	0.16 (0.17) *< LOQ – 0.40*	0.15 (0.16) *< LOQ – 0.34*
BDE-85	< LOQ	< LOQ
BDE-99	3.05 (2.61) *0.35 – 7.28*	1.69 (1.41) 0.81 – 3.23
BDE-100	2.30 (1.84) *0.58 – 5.24*	1.74 (1.67) *0.92 – 2.61*
BDE-153	1.24 (1.11) *< LOQ – 2.44*	0.66 (0.68) *0.40 – 1.06*
BDE-154	1.05 (0.79) *0.19 – 3.61*	0.73 (0.69) *0.41 – 1.11*
BDE-183	< LOQ	< LOQ
BDE-209	0.80 (0.78) *< LOQ – 2.08*	< LOQ
ΣPBDE	42.4 (36.4) *8.51 – 82.0*	27.4 (25.3) *16.6 – 45.3*

Table S4: Arithmetic mean concentrations (ng/g lipid weight, lw) of PCB congeners in blubber samples of ringed seals from East Greenland. Numbers in brackets are median concentrations. Numbers in italics are ranges. Concentrations below LOQ are treated as zero in calculations of mean, median and ΣPCB. Σ10-PCB includes the congeners 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180.

Parameter	2006	2008
Age<5 years old (1986-2008)		
CB-28	8.73 (9.06) *6.22 – 10.8*	7.98 (8.20) *3.93 – 11.7*
CB-31	7.39 (7.15) *4.29 – 10.7*	4.38 (4.35) *2.74 – 7.92*
CB-44	2.17 (2.11) *1.27 – 3.92*	2.81 (2.74) *1.49 – 3.95*
CB-49	4.39 (4.06) *2.00 – 8.80*	5.94 (6.06) *3.52 – 9.50*
CB-52	19.66 (17.17) *10.0 - 38.3*	17.1 (16.3) *10.2 – 30.0*
CB-99	69.4 (63.7) *30.1 - 138*	78.3 (79.9) *27.5 – 141*
CB-101	57.9 (59.7) *31.9 – 82.6*	50.7 (48.4) *31.2 – 72.0*
CB-105	36.2 (38.3) *17.6 – 61.0*	20.2 (19.9) *10.7 – 31.9*
CB-110	7.11 (6.52) *3.74 – 13.5*	10.3 (9.53) *6.38 – 15.3*
CB-118	69.6 (75.6) *31.4 - 111*	53.7 (57.1) *24.6 – 90.6*
CB-128	6.24 (5.06) *2.40 – 18.7*	4.93 (4.06) *2.83 – 15.7*
CB-138	121 (114) *53.2 - 205*	86.7 (84.2) *44.5 – 137*
CB-149	16.6 (15.0) *8.43 – 44.7*	11.9 (11.2) *7.48 – 27.7*
CB-151	3.23 (1.93) *1.21 – 15.9*	3.69 (3.06) *1.59 – 10.9*
CB-153	194 (174) *83.4 - 380*	146 (137) *72.7 – 260*
CB-156	7.77 (7.43) *3.47 – 13.9*	5.67 (5.45) *3.25 – 9.40*
CB-170	17.4 (15.8) *7.37 – 30.0*	12.3 (11.6) *5.99 – 26.7*
CB-180	45.0 (42.6) *20.4 – 79.1*	37.0 (36.9) *19.0 – 73.7*
CB-187	25.4 (22.8) *11.8 – 42.6*	18.0 (18.0) *10.7 – 32.4*
CB-188	< LOQ	< LOQ
CB-194	2.72 (2.51) *1.28 – 4.66*	2.62 (2.37) *1.37 – 5.02*
CB-209	< LOQ	< LOQ *< LOQ – 0.74*
Σ10-PCB	567 (576) *281-945*	430 (425) *230 – 675*
ΣPCB	721 (757) *355 - 1154*	581 (579) *307 – 911*

Age ≥ 5 years old (1994-2008)		
CB-28	9.15 (8.95) *4.73 – 13.6*	7.27 (7.31) *4.08 – 10.5*
CB-31	10.8 (9.03) *4.44 – 19.2*	4.93 (4.55) *2.49 – 7.94*
CB-44	2.92 (2.89) *1.74 – 4.(0*	3.12 (2.92) *1.88 – 5.80*
CB-49	7.45 (7.1)) *2.85 – 14.8*	6.99 (6.10) *4.47 – 12.7*
CB-52	33.9 (31.5) *11.7 – 73.4*	19.8 (19.0) *11.2 – 33.9*
CB-99	134 (100) *18.7 - 406*	87.0 (86.7) *54.8 - 180*
CB-101	96.8 (82.3) *21.7 - 233*	56.6 (56.6) *28.9 – 95.1*
CB-105	50.4 (37.7) *9.07 - 140*	20.7 (20.3) *13.4 – 30.3*
CB-110	8.69 (8.43) *5.10 - 18.9*	10.5 (9.56) *7.23 – 17.2*
CB-118	93.9 (80.2) *20.2 - 274*	52.6 (49.7) *31.4 – 83.7*
CB-128	13.9 (10.7) *< LOQ - 59.5*	5.64 (4.64) *2.38 – 12.1*
CB-138	248 (175) *26.2 - 824*	104 (91.9) *61.0 – 168.4*
CB-149	25.6 (22.3) *8.57 – 60.7*	13.9 (12.6) *6.01 – 24.4*
CB-151	4.71 (4.34) *1.50 -13.7*	3.98 (3.70) *2.02 – 9.36*
CB-153	375 (256) *40.2 - 1224*	176 (165) *112 - 251*
CB-156	14.2 (11.2) *1.87 – 44.6*	6.57 (6.02) *3.62 – 9.46*
CB-170	40.3 (31.6) *3.48 - 142*	15.5 (15.4) *8.73 – 23.7*
CB-180	114 (94.3) *9.68 - 377*	46.7 (43.8) *26.8 – 70.9*
CB-187	65.4 (53.4) *8.26 - 222*	21.6 (20.9) *10.2 – 39.5*
CB-188	< LOQ	< LOQ
CB-194	9.55 (7.25) *< LOQ – 31.1*	3.67 (3.35) *1.94 – 6.87*
CB-209	1.24 (< LOQ) *< LOQ – 4.68*	< LOQ *< LOQ – 1.26*
Σ10-PCB	1045 (767) *151 - 3098*	495 (480) *318 - 741*
ΣPCB	1358 (1019) *203 - 4066*	667 (627) *425 - 1069*

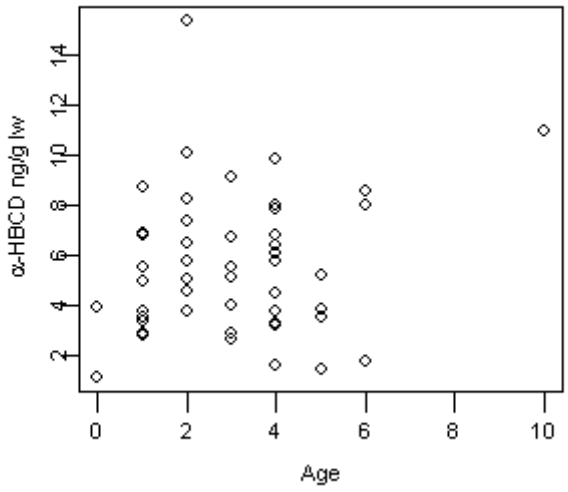

Figure S1: Age-stratified concentrations of α-HBCD (ng/g lw) in individual ringed seals from East Greenland.

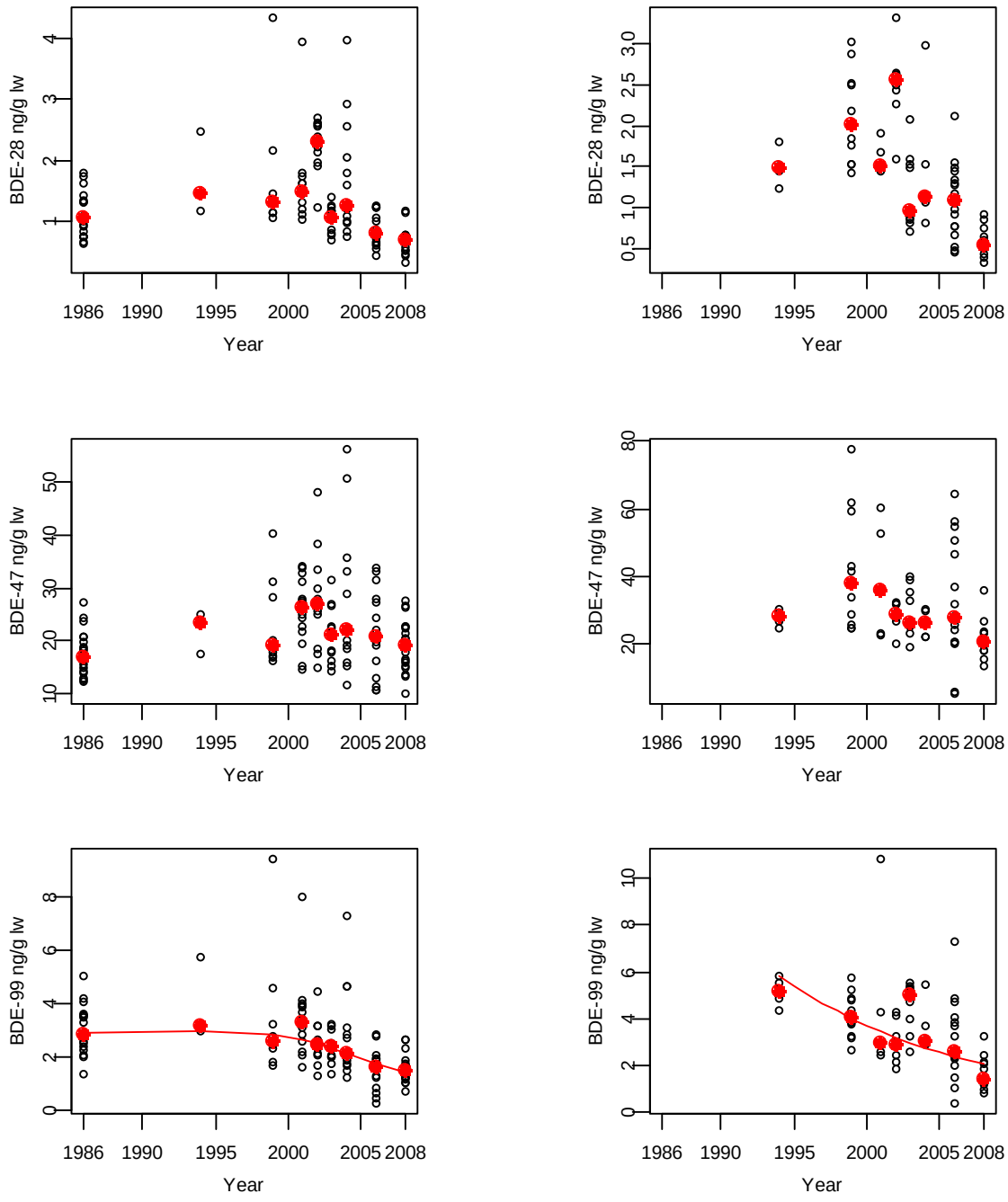

Figure S2: Time trend of BDE-28 (top), BDE-47 (middle) and BDE-99 (bottom) in juvenile seals (< 5 years) to the left and adult ringed seals (≥ 5 years) to the right. The filled dots are median values. The trend lines are shown if significant.