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Levels and trends of brominated flame retardants in the Arctic

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Abstract

Polybrominated diphenyl ethers (PBDEs) containing two to seven bromines are ubiquitous in Arctic biotic and abiotic samples (from zooplankton to polar bears (*Ursus maritimus*) and humans; air, soil, sediments). The fully brominated decabromodiphenyl ether (BDE-209), hexabromocyclododecane (HBCD), tetrabromobisphenol A (TBBPA) and polybrominated biphenyls (PBBs) are also present in biotic and abiotic samples. Spatial trends of PBDEs and HBCD in top predators are similar to those seen for polychlorinated biphenyls (PCBs) and indicate western Europe and eastern North America as source regions. Concentrations of tetra- to heptaBDEs have increased significantly in North American and Greenlandic Arctic biota and in Greenland freshwater sediments paralleling trends seen further south. For BDE-209, increasing concentrations in Greenlandic peregrine falcons (*Falco peregrinus*) and in dated lake sediment cores in the Canadian Arctic have been seen during the 1990s. BDE-47, -99, -100 and -153 are observed to biomagnify in Arctic food webs. $\sum$PBDE concentrations in Arctic samples are lower than in similar sample types from more southerly regions and are one or more orders of magnitude lower than $\sum$PCB concentrations except for some levels for air. Air and harbor sediment results for PBDEs indicate that there are local sources near highly populated areas within the Arctic. Findings of PBBs on moss and TBBPA on an air filter, and that both are found in biota at high trophic levels indicates that these compounds may also reach the Arctic by long-range atmospheric transport. Based on the evidence of their presence in the Arctic and indications that most if not all are undergoing long-range transport, these brominated flame retardants (BFRs) have characteristics that qualify them as POPs according to the Stockholm Convention.

Keywords: Brominated flame retardants; Polybrominated diphenyl ethers; Decabrominated diphenyl ether; Hexabromocyclododecane; Tetrabromobisphenol A; Polybrominated biphenyls; Arctic

1. Introduction

The Stockholm Convention on Persistent Organic Pollutants (POPs) entered into force on May 17, 2004 (http://www.pops.int/) and the UN ECE POPs Protocol to the Convention on Long-Range Transboundary Air Pollution on October 23, 2003 (http://www.unece.org/env/lrtap/pops_h1.htm). The Stockholm Convention states that chemicals with the characteristics of POPs are those found in locations "distant from sources" and those for which "monitoring data showing that long-range environmental transport of the chemical ... may have occurred". Thus, the Arctic is an important indicator region for assessment of persistence and bioaccumulation of POPs. The Arctic environment is well suited as a region in which to evaluate POPs. Some regions of the Arctic, particularly the Barents Sea area north of Norway and western Russia are relatively close to source regions of POPs. Other parts of the Arctic, however, are very remote, with few activities that constitute local sources of POPs. Cold conditions favor persistence of POPs relative to temperate or tropical environments. The presence of fourth trophic level carnivores (e.g. polar bears (*Ursus maritimus*)), and storage of lipid as an energy source, make Arctic food webs vulnerable to bioaccumulative chemicals. Indigenous people in the Arctic utilizing a traditional diet, which is high in nutritionally beneficial fat, results in their elevated exposure to some POPs (Hansen et al., 1998).

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Recent reviews have demonstrated that polybrominated diphenyl ethers (PBDEs) are widespread in the non-Arctic and that levels are increasing (de Wit, 2002; Hites, 2004). The physico-chemical properties of PBDEs have also recently been reviewed (Wania and Dugani, 2003) and indicate similarities to other POPs that are regulated under the Stockholm and UN ECE POPs Conventions. Using several global fate and transport models, Wania and Dugani (2003) concluded that tetra- and pentabrominated BDEs have comparable Arctic accumulation potential as the hexa- and heptachlorinated biphenyls. This suggests that PBDEs and other brominated flame retardants (BFRs) with similar physical properties will undergo long-range atmospheric transport, deposition and revolatilization (a process known as "the grasshopper effect" (Wania and Mackay, 1993; Gouin et al., 2004)) from urban use areas in the northern hemisphere to the remote arctic areas.

BFRs are not currently included in the regional UN ECE Convention on Long-Range Transboundary Air Pollution or the global UNEP Convention on POPs (Stockholm Convention). PBDEs, in particular the PentaBDE technical product, have been proposed as potential new candidates for inclusion (Peltola and Ylä-Mononen, 2001; Tanabe, 2004). Concern about the risks of BFRs, especially PBDEs, led the European Union (EU) to conduct risk assessments of the three major PBDE technical products, Penta-, Octa- and DecaBDE. Based on the risk assessments, the EU has decided to ban the Penta- and OctaBDE products as of August 2004 (Cox and Ethymiou, 2003). Similar bans of PentaBDE and OctaBDE will become effective in several states of the United States, beginning January 1, 2006, earliest. After ceasing the production of PentaBDE and OctaBDE in Europe, Great Lakes Chemicals also ceased production of these chemicals in North America at the end of 2004. Canada is currently carrying out risk assessments of the three PBDE technical products. The EU risk assessment of DecaBDE (European Commission Joint Research Centre, 2002, 2004) has recently been provisionally finalized and no risk reduction measures are considered necessary. However, concerns about DecaBDE remain and needs for filling research gaps have been suggested. One US state has enacted legislation on DecaBDE that may result in risk management measures beginning January 1, 2008 if a safer, nationally available alternative is identified. The EU risk assessment of HBCD is ongoing. Use and production of the polybrominated biphenyl (PBB) technical product containing HexaBB was discontinued in North America and Europe in the 1970s, and production of the technical decabrominated biphenyl (DecaBB, BB-209) was discontinued in Europe after 2000.

The first indication that BFRs were reaching the Arctic was presented in 1987, when the lower brominated PBDEs and PBBs were detected in muscle tissue from Svalbard Brünnich's guillemots (thick-billed murres) (*Uria lomvia*) (130 ng/g lipid weight – lw) and ringed seal blubber (*Phoca hispida*) (40 ng/g lw) collected in 1981 (Jansson et al., 1987). Whitefish (*Coregonus* spp.) collected from Lake Storvindeln in 1986, a pristine mountain lake in the Swedish mountains near Ammarnäs, had $\sum$PBDE levels of 26 ng/g lw in muscle tissue (Sellström et al., 1993). A high-volume air sample collected in 1990–1991 from Ammarnäs was found to contain BDE-47, -99, -100 ($\sum$PBDE-8.3 pg/m^3) and HBCD (6.1 pg/m^3), indicating long-range transport (de Wit, 2002). Despite these early findings, only recently have the spatial and temporal trends of BFRs been studied in detail in the Arctic.

The purpose of this paper is to review the new data on BFRs in the Arctic and assess whether this information supports the view that PBDEs including DecaBDE (BDE-209) and other BFRs of similar molecular weight are POPs and potential global pollutants. This review is based on a recent assessment of POPs in the Arctic (de Wit et al., 2004) combined with newer data not available for that assessment. These newer data are the best that are currently available and come from recent peer-reviewed publications, as well as from extended abstracts from conferences/workshops and from several published government scientific reports. The extended abstracts from The 24th International Symposium on Organohalogenated Organic Environmental Pollutants and POPs (DIOXIN '04) and The Second (BFR 2001) and Third International Workshops on Brominated Flame Retardants (BFR 2004) are available via the internet (http://www.bfr2004.com). The scientific reports provide data from specific national surveys or from national monitoring programs within the Arctic Monitoring and Assessment Programme (AMAP) where defined quality assurance and quality control criteria have been followed. Many of the extended abstracts from the BFR 2004 workshop are the basis of the articles in this current volume, thus results cited as extended abstracts here may be found as publications in this volume. Unpublished data are cited with the permission of the scientists involved and have been checked for correctness and quality. Unless otherwise noted, $\sum$PBDE denotes the sum of lower brominated congeners. If DecaBDE (BDE-209) was quantified, this is indicated separately. A summary of environmental concentrations in the Arctic is given in Table 1. The question is often raised as to how BFR concentrations, especially the PBDEs, compare to those of better known contaminants such as organochlorines. In order to put the $\sum$PBDE concentrations into perspective, they are therefore compared to $\sum$PCB concentrations in the same samples if this information was available.

1.1. Long-range transport and the atmospheric environment

BFRs were analyzed in archived air samples collected over four-week periods in 1994–1995 from Alert and Tagish (Canadian Arctic) and Dunai (Russian Arctic) (Alaee et al., 2003). PBDEs were measured in the same archived extracts as the polychlorinated naphthalenes (PCNs) and coplanar PCBs (Helm et al., 2004). Mean $\sum$PBDE concentrations (Di-HpBDEs) in air samples were 14 pg/m^3 at

Table 1
Concentrations of PBDEs, HBCD and TBBPA in samples from the Arctic

Sample type	Region	Tissue	Year	Sex	% lipid	*n*	Statistic	BDE-28	BDE-47	BDE-49	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209	∑PBDE	HBCD	TBBPA	BB-153	∑PBB	Reference
Air – high volume																						
	Ammarnäs, Sweden		1990–1991			2	Mean		6.3		1.6	0.4					8.3	6.1				1
	Pallas, Finland		2000–2001			2	Mean											2.5				2
	Bjørnøya, Norway		1999–2001				Range										3–10					3
	Dunai, Russia		1994–1995			11	Mean		3.7		9.3	2.8	1.8				17.6		70			4
	Alert, Canada		1994–1995			14	Mean		64		128	21	17	10			240					4
	Tagish, Canada		1994–1995			14	Mean		112		232	35	29	16			424					4
	Alert, Canada		2000–2001			2	Mean										4					5
	Eureka, Canada		2000–2001			2	Mean										37					5
	Devon, Canada		2000–2001			2	Mean										0.3					5
	Kangiqtugaapik, Canada		2000–2001			2	Mean										0.3					5
	Cape Dorset, Canada		2000–2001			2	Mean										68					5
	Kuujjuak, Canada		2000–2001			2	Mean										30					5
	Iceland		2002			1		0.7	<6.3		<9.8	<1.9	<0.6	<0.7			11					6
	Northern Norway		2002			1		<0.4	<6.3		<9.8	<1.9	0.8	<0.7			11					6
	Svalbard		2002			1		<0.4	6.5		14.5	2.3	1.3	0.87			26					6
	Valvik, Norway (67.38° N)		2000–2002			2	Mean	0.03	0.3	0.1	0.7	0.1	0.1	0.1			1.3					7
	Skoganvarre, Norway (69.83° N)		2000–2002			2	Mean	nd	0.3	0.04	0.5	0.1	0.2	0.1			1.2					7
Air – passive samples																						
	Iceland		2002			1		0.1	<0.95		<1.47	<0.29	<0.09	<0.1			1.7					6
	Northern Norway		2002			1		<0.06	<0.95		<1.47	<0.29	0.12	<0.1			1.6					6
	Svalbard		2002			1		<0.06	0.97		2.17	0.34	0.19	0.13			3.9					6
	Valvik, Norway		2000–2002			2	Mean	<0.15	0.74	0.13	1.7	0.28	<0.54	0.19			3.3					7
	Skoganvarre, Norway		2000–2002			2	Mean	nd	0.84	0.09	1.3	0.2	0.39	0.21			3					7
Soil																						
	Kanchalan, Chukotka, Russia		2000–2001			1			0.1		0.13						0.23					8
	Lavrentiya, Chukotka, Russia		2000–2001			1			0.07		0.09						0.16					8
	Norway, Woodlands		1998			24	Median	0.029	0.25	0.023	0.36	0.58	0.051	0.046	0.025		0.97					9
Vegetation																						
Moss	Skoganvarre, Norway		2002			1		0.016	0.22		0.046	0.021	<8	<8	<12	0.025	0.328	nd	0.019		0.023	10
	Valvik, Norway		2002			1		<0.004	0.015		0.012	<4	<4	0.001	0.001	0.12	0.149		0.14			10
	Several sites, N. Russia		2000–2001				Range		0.007–0.012		0.006–0.011						0.013–0.023					11
Lichen	Several sites, N. Russia		2000–2001				Range		0.021–0.029		0.005–0.01						0.026–0.04					11
Berries	Several sites, N. Russia		2000–2001				Range		0.003–0.022		0.001–0.01						0.004–0.03					11
Terrestrial biota																						
Frogs	Kiruna, Sweden	Liver	1998–2000	m,f	2.8	28	Mean		0.93		nd						0.93					12
	Ammarnäs, Sweden	Liver	1998–2000	m,f	1.5	34	Mean		2.3		5.6						7.9					12

(continued on next page)

Table 1 (*continued*)

Sample type	Region	Tissue	Year	Sex	% lipid	*n*	Statistic	BDE-28	BDE-47	BDE-49	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209	∑PBDE	HBCD	TBBPA	BB-153	∑PBB	Reference
Grouse	Norway	Liver	1990–1993		3.8		Mean	0.12	0.27		0.27	nd	0.073	0.044	nd	0.5	1.3					13
Lynx	Norway	Liver	1993–1994		5.4	7	Mean	0.036	1		0.45	0.023	7.8	0.13	1.1	1	11.5					13
Moose	Norway	Liver	1995		5.0		Mean	0.024	0.31		0.36	0.047	0.105	0.027	0.043	0.8	1.7					13
Reindeer	Ottsjö, Sweden	Suet	1986		56.0	31	Mean		0.17		0.26	0.04					0.47					14
Peregrine falcon	N. Sweden	Egg	1991–1999	–	5.57	18	Mean		360		860	540	1900	400	270	110*	4330	220*		114		15
	N. Norway	Egg	1991–1997		4.8	6	Mean										4700				900	16
	N. Norway	Egg	1993–2000		5	9	Median	nd	140		500	250	1200	230	nd	det	3100		det	140		17
	S. Greenland	Egg	2000–2003		5.2	9	Mean	<2.5	235	5.8	760	390	1920	430	220	48	4000	5.7	380**	640		18
Golden eagle	N. Norway	Egg	1991–1997		5 est	9	Mean										140				180	16
	N. Norway	Egg	1992–2002		4	15	Median	nd	13		18	nd	35	5	nd	det	75		det	20		17
Merlin	N. Norway	Egg	1991–1997		5 est	3	Mean										720				150	16
	N. Norway	Egg	1995–2000		7	9	Median	nd	56		140	31	160	46	nd	det	415			10		17
Gyrfalcon	N. Norway	Egg	1991–1997		5 est	2	Mean										360				60	16
Freshwater sediment																						
	Greenland, 7 lakes		2000			7	Mean		0.021													19
	AX-AJ Lake, Canada		1998–2001													0.075						20
	Romulus Lake, Canada		1998–2001													<0,1						20
	Char Lake, Canada		1998–2001													0.042						20
	Several sites, N. Russia		2000–2001				Range		0.003–0.024		0.001–0.003						0.004–0.027					11
Freshwater fish																						
Whitefish	Lake Storvindeln, Sweden	Muscle	1986		0.66				14		5.6	3.8					23					14
Burbot	Fort Good Hope, NWT, Canada	Liver	1988	m,f	30.2	10	Mean	–	0.75		0.28	0.12	0.1	0.067	–	–	1.3					21
	Fort Good Hope, NWT, Canada	Liver	1999	m,f	35.0	4	Mean	–	1.7		1.1	0.59	0.46	0.45	–	–	4.3					21
	Fort Good Hope, NWT, Canada	Liver	2000	m,f	33.3	11	Mean	–	1.9		0.96	0.54	0.41	0.24	–	–	4.1					21
	Lake Grensefoss, Norway	Muscle	1999/2000	–	11.6	1	–	–	84	–	91	–	–	–	–	–	175					22
Trout	Lake Takvatn, Norway	Muscle	1999/2000	–	1.8	1	–	–	5.50	–	3.1	–	–	–	–	–	8.6					22
	Lake Fjellfrøsvatnet 99, Norway	Muscle	1999/2000	–	1.1	1	–	–	6.8	–	6.3	–	–	–	–	–	13					22
	Lake Grunnvatnet, Norway	Muscle	1999/2000	–	1.3	1	–	–	4.9	–	2.9	–	–	–	–	–	7.90					22
	Lake Store Raudvannet, Norway	Muscle	1999/2000	–	2.5	1	–	–	8.3	–	6.2	–	–	–	–	–	15					22
Arctic char	Lake Ellasjøen, Bjørnøya, Norway	Muscle	1999/2000	–	1.3	1	–	–	640	–	620	–	–	–	–	–	1200					22
	Lake Ellasjøen, Bjørnøya, Norway	Muscle	1998–2003			6											420					23
	Lake Oyangen, Bjørnøya, Norway	Muscle	1998–2003			4											50					23
	Lake Fergusson, S. Greenland	Muscle	2000–2001	m,f	1.9	4	Mean		7.4		4.5	2.1	1	1.4			41					24
	Lake Fergusson, S. Greenland	Liver	2000–2001	m,f	4.29	4	Mean	0.16	11		3.3	3.3	1.6	2.1			23					24
Marine sediment																						
	Tromsø, Norway		2003			2	Mean	<0.01	0.055	<0.01	0.08	0.01	0.005	0.005	<0.01	0.43	1.1	<0.3	1.24			25
	Barrow Strait, Canada		1998			1	–	0.004	0.04	0.002	0.023	0.006	0.01	0.002			0.11					26
	Penny Strait, Canada		1998			1	–	0.002	0.046	0.002	0.027	0.009	0.009	0.002			0.12					26
	Nanisivik, Canada		1998			1	–		0.11								0.30					26

	Kola Bay, Russia		1998						0.05		0.06	0.02	0.03				0.14				27
	Guba Zapadnaya Litsa, Kola, Russia		1998						0.05		0.06	0.02	0.01				0.16				27
	Polyarnyy, Kola, Russia		1998						19		176	33	13				241				27
Marine invertebrates																					
Zooplankton	Svalbard																1				28
Blue mussels	Svolvaer, Lofoten, Norway	Muscle	2002	–	0.935	20	Mean	<1	7.4		4.6	2.1	<1	<1	<1	33	47.1	12 α	na		25
	Varangerfjorden, Norway	Muscle	2002	–	1.68	50		<0.6	1.2		<0.6	<0.6	<0.6	<0.6	<0.6	3.6	4.8	3.6 α	na		25
	Usuk, S. Greenland	Muscle	2000	–	2	20	Mean		5		1	<1	<2.5				5.5				29
Marine fish																					
Atlantic cod	Lofoten, Norway	Liver	–	–	67.7	1	–	<0.01	22.0	–	1.1	–	0.89	–	–	–	24				30
	Svolvaer, Lofoten, Norway	Liver	2002		73.2	21	Mean	1.3	8.1		0.34	1	0.04	0.44	0.03	0.99	12	6.6 α	0.5		25
	W. coast, Norway	Liver	–	–	75.2	1	–	<0.04	26	–	1.10	–	1.30	–	–	–	28.4				30
	Varangerfjorden, Norway	Liver	1998, 2002		56.8	21	Mean	2.5	11.7		0.25	1.1	<0.04	0.44	<0.02	0.98	17	7.7 α	2.5		25
	SW Greenland	Liver	2000	–	57.6	3	Mean	–	3.1	–	0.56	–	–	–	–	–	8.9				31
	SW Greenland	Muscle	2000	–	0.7	2	Mean	–	30	–	480	–	–	–	–	–	510				31
Polar cod	Svalbard						Range										10–15	5–25			28
	Svalbard		2001			3	Geom mean		2.1		0.1	0.31		0.16			3.60				32
	Bjørnøya, Norway	Liver	2003	–	15–20	4–6	Mean	nd	9.9			1.9		nd			12	14.7			33
Arctic cod/uvak	Usuk, S. Greenland	Liver	2000	m,f	39.4	10	Mean		20		1.5	3.9	<0.63				25				29
Greenland halibut	SW Greenland	Liver	2000	–	36.3	3	Mean		2.9	–	0.5	–		–	–	–	7.5				31
Tusk	Norwegian Sea	Liver	–	–	60.4	1	–	<0.05	60	–	2.5	–	2.6	–	–	–	65				30
Spotted wolffish	Usuk, S. Greenland	Liver	2000	m	15.5	1	–		7.9		<0.84	<0.84	<1.6				7.9				29
Starry ray	Usuk, S. Greenland	Liver	2000	m	42	1	–		2.6		0.19	0.52	<0.55				3.3				29
Shorthorn sculpin	Qaqortoq, S. Greenland	Liver	2000	f	18.9	5	Mean		41		<0.69	2.1	<1.3				43				29
	Igaliko, S. Greenland	Liver	2000	m,f	16.9	10	Mean		17		0.33	0.77	<1.4				18				29
	Usuk, S. Greenland	Liver	2000	m,f	18.3	9	Mean		11		<0.66	0.6	<1.4				12				29
	Ittoqqortoormiit, E. Greenland	Liver	2001	f	17.5	20	Mean										9.6				34
Arctic char	Ittoqqortoormiit, E. Greenland	Muscle	2001	f	2.2	13	Mean										26				34
	Ittoqqortoormiit, E. Greenland	Muscle	2001	m	3.2	7	Mean										46				34
Seabirds																					
Glaucous gull	Bjørnøya, Norway	Liver	1999	m,f	5.9	15	Median		39		15						54				35
	Bjørnøya, Norway	Plasma	2002–2004	m,f	1.4	89	Mean	7.1	610		180	84	230	120	23	410	1400	45		15	36
	Bjørnøya, Norway	Eggs	2002–2004		9.61	4 pools	Mean/range									23–53	549	142		2.8	36
	Bjørnøya, Norway	Muscle	1998–2003			3	Mean		620		250	100	420				1400				37
	Svalbard	Liver			6 est	20	Range									nd-170					36
Great black-backed and herring gulls	Alta, Norway	Eggs	2001	–	8.3	20		–	460		34	–	–	–	–	–	490				38
	Kongsfjord (Finmark), Norway	Eggs	2001	–	8.4	20		–	490		44	–	–	–	–	–	530				38
	Sommarøy, Norway	Eggs	2001	–	7.7	20		–	670		22	–	–	–	–	–	690				38
	Vardø, Norway	Eggs	2001	–	8.3	20		–	600		54	–	–	–	–	–	650				38
Black-legged kittiwake	Bjørnøya, Norway	Muscle	1998–2003			3	Mean		100		20	10	30				150				37
	Prince Leopold Island, Canada	Eggs	1993		4.3	1 pool	Mean										70				39
Northern fulmar	Prince Leopold Island, Canada	Eggs	1993		4.4	1 pool	Mean										15				39
Little auk	Bjørnøya, Norway	Muscle	1998–2003			3	Mean		200		80	30	50				360				37

(continued on next page)

Table 1 (*continued*)

Sample type	Region	Tissue	Year	Sex	% lipid	*n*	Statistic	BDE-28	BDE-47	BDE-49	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209	∑PBDE	HBCD	TBBPA	BB-153	∑PBB	Reference
Thick-billed murre	SW Greenland	Liver	1999	–	5.1	4	Mean	–	7.1		5.1	–	–	–	–	–	34					31
	Prince Leopold Island, Canada	Eggs	1993		3.3	1 pool	Mean										14					39
Black guillemot	SW Greenland	Liver	1999	–	4.9	5	Mean	–	23		4.5	–	–	–	–	–	61					31
	Qeqertarsuaq, W. Greenland	Liver	2000	m,f	6.2	10	Mean										25					40
	Qeqertarsuaq, W. Greenland	Eggs	2000	–	10.3	7	Mean										25					40
	Ittoqqortoormiit, E. Greenland	Liver	2000	m,f	7.2	8	Mean										73					40
	Ittoqqortoormiit, E. Greenland	Eggs	2001	–	11	10	Mean										83					40
Marine mammals																						
Ringed seal	Svalbard	Blubber	1987		88	7	Mean		47		1.7	2.3					51					14
	Svalbard	Blubber					Range										50–100	15–35				28
	Svalbard	Blubber	1999			9	Geom mean		16.8		0.66	0.39		nd			18.3					32
	Holman, W. Canada	Blubber	1996	m	96 est	8	Mean	0.16	2.8	0.07	0.18	0.22	0.05	0.03	–	–	3.6					41
	Holman, W. Canada	Blubber	2000	m	91	7	Mean	0.14	4.1	0.14	0.21	0.28	0.08	0.04	–	–	5.1					41
	West Greenland	Muscle	2000	–	10.5	2	Mean	–	44	–	3.5	–	–	–	–	–	60					31
	West Greenland	Blubber	2000	–	88.1	9	Mean	–	2.7	–	0.45	–	–	–	–	–	4.1					31
	NE Greenland	Blubber	2000	–	106	8	Mean	–	40.6	–	4.6	–	–	–	–	–	58.0					31
	Ittoqqortoormiit, E. Greenland	Blubber	2001	ad	95.2	5	Mean										38					34
	Ittoqqortoormiit, E. Greenland	Blubber	2001	sub-ad	93.8	14	Mean										35					34
	Pangnirtung, Baffin Isl., E. Canada	Blubber	1993	f	94	3	Mean	–	0.35	–	0.12	–	–	–	–	–	0.53					42
	Pangnirtung, Baffin Isl., E. Canada	Blubber	1993	m	95	2	Mean	–	0.8	–	0.095	–	–	–	–	–	1					42
	Pangnirtung, Baffin Isl., E. Canada	Blubber	1999–2002	f	89.3	12	Mean	0.11	1.8		0.29	0.10	0.039	0.078	0.001		2.8					43
	Sachs Harbour	Blubber	2001	f	90.3	10	Mean	0.16	3.3		0.73	0.19	0.012	0.058	0.02		4.7					43
	Grise Fiord	Blubber	2003	f	81.3	7	Mean	0.11	2.0		0.22	0.11	0.012	0.034	nd		3.1					43
	Resolute	Blubber	2000	f	88.3	5	Mean	0.04	0.7		0.17	0.04	0.009	0.023	nd		1.3					43
	Inkujuaq	Blubber	2002	f	75.8	8	Mean	0.73	18.3		2.1	2.7	1.3	1.3	0.03		30					43
	Hudson Strait (Nunavik)	Blubber	2002	f	85.1	12	Mean	0.30	7.8		2.0	1.0	0.67	0.47	0.06		13					43
	Onega Bay, White Sea, Russia	Blubber	2001	m,f	91.2	24	Mean	0.12	6.6		0.56	0.42	0.18	0.11	<0.01		8					44
	Vaygach Island, Barents Sea, Russia	Blubber	2002	m,f	89.8	8	Mean	0.43	10.7		0.22	0.54	0.10	0.08	<0.01		12.1					44
	Dikson Island, Kara Sea, Russia	Blubber	2002	m,f	90.6	6	Mean	0.37	11.3		0.20	0.63	0.11	0.10	0.01		12.7					44
Minke whale	Barents Sea	Muscle	–	–	0.47	1	–	<2.1	26	–	6.4	–	<11	–	–	–	32					45
	Barents Sea	Muscle		–	1.03	1	–	<0.97	34	–	9.7	–	<3	–	–	–	44					45
	Norwegian Sea	Muscle	–	–	0.72	1	–	<1.4	290	–	130	–	9.7	–	–	–	430					45
	Norwegian Sea	Muscle		–	3.62	1	–	<0.28	300	–	96	–	<1.4	–	–	–	400					45
Beluga	Cumberland Sound, E. Canada	Blubber	1997	m	90	17	Mean		10.5		0.8	1.6	–	0.9			15.6					46
	Kangiqsujuaq, E. Canada	Blubber	2001	m,f	83	9	Mean	0.63	18.8		2.3	2.5	3.4	0.40	0.07		30.5	18				43
	Nastapoka, E. Canada	Blubber	2001	m,f	85	10	Mean	0.45	17.4		2.8	2.9	3.7	0.56	0.08		30.9	9.8				43
	Hendrickson Island, W. Canada	Blubber	2001	m	93	11	Mean	–	9.4	1.4	1.8	1.6	0.39	0.68	–	–	17					46
	Svalbard	Blubber	1998	m,f	4–90	10	Mean (ww)	–	77.8	–	5.15	–	nd	–	–	–	93					47
	Svalbard	Blubber	1998	m		2	Mean		70		3.0	7.3		nd			161					32
	Svalbard	Blubber	1998	f		2	Mean		17		nd	nd		nd			29					32

Harbor porpoise	Iceland	Blubber	1997–2001	m,f		10	Range										25–95		48
Long-finned pilot whales	Hvannasund, Faroe Islands, Denmark	Blubber	1994	f	82	9 pools	–	–	500	–	200	–	39.0	–	–	–	840		47
	Vestmanna, Faroe Islands, Denmark	Blubber	1996	m,f	66–79	4 pools	Mean	–	970	–	420	–	65	–	–	–	2200		49
	Torshavn, Faroe Islands, Denmark	Blubber	1997	m,f	52–85	12	Mean	–	420	–	90	–	8	–	–	–	940		47,49
	Leynar, Faroe Islands, Denmark	Blubber	1997	m,f	83–86	4 pools	Mean	–	270	–	55	–	4.6	–	–	–	610		47,49
	Sandavagur, Faroe Islands, Denmark	Blubber	1997	m,f	64–80	4 pools	Mean	–	500	–	89	–	4.3	–	–	–	1100		47,49
	Hvalvik, Faroe Islands, Denmark	Blubber	1998	m,f	66–83	4 pools	Mean	–	250	–	47	–	6.1	–	–	–	740		47,49
	Faroe Islands, Denmark	Blubber	2000	f,?	2–83	6	Mean	–	180	–	31	–	4.9	–	–	–	360		47
Polar bears	Amundsen Gulf	Adipose		f	95	10	Mean	<0.1	11	–	2.5	0.68	1.5	0.05	0.05	–	16	–	50
	Foxe Basin/ Gulf of Boothia	Adipose		f	89	8	Mean	<0.1	7.6	–	2.3	0.41	0.68	0.06	0.12	–	7	–	50
	Lancaster Sound	Adipose		f	89	6	Mean	<0.1	8.9	–	2.3	0.50	0.69	0.06	0.06	–	13	–	50
	NE Baffin	Adipose		f	96	9	Mean	<0.1	13	–	2.4	0.65	1.7	0.20	0.19	–	19	–	50
	SE Baffin	Adipose		f	85	17	Mean	<0.1	14	–	2.3	0.67	3.6	0.06	0.12	–	22	–	50
	Western Hudson Bay	Adipose		f	56	15	Mean	<0.1	9.9	–	1.9	0.49	1.2	1.08	0.09	–	14	–	50
	Svalbard	Adipose		f	91	15	Mean	<0.1	41	–	2.3	0.87	3.2	<0.01	<0.01	<1	50	44	50
	Bering/Chukchi Sea	Adipose		f	60	8	Mean	<0.1	4.9	–	0.78	0.20	0.11	0.03	<0.01	–	6.7	0.4	50
	Eastern Greenland	Adipose		f	90	44	Mean	0.013	51	<0.01	3.9	1.5	12	<0.1	<0.01	<1	70	45	50
	Svalbard	Adipose	1998	m,f	5–67	20	Range	–		–	nd	–	nd	–	–	–	14–144		47
	Svalbard	Adipose	2001–2003													1			51
	Svalbard						Range										15–30	5–15	28
	Svalbard	Adipose		m		10	Geom mean		27.4		nd			nd			27.4		32
	Svalbard	Adipose		f		10	Geom mean		45.6		nd			nd			45.6		32
Humans																			
	Nunavik (N. Quebec), Canada	Milk	1996–2000	f		7	Median		3.4								6.2		52
	Faroe Islands, Denmark	Milk	1998–1999	f		1 pool	Mean		1.7		1.0	1	3.6				7.2		53
	Krasnoshchelie, Kola, Russia	Plasma	2000–2001	m,f	0.40	3	Geom mean										0.93		11
	Lovozero, Kola, Russia	Plasma	2000–2001	m,f	0.40	4	Geom mean										0.44		11
	Nelmin-Nos, Pechora Basin, Russia	Plasma	2000–2001	m,f	0.30	6	Geom mean										0.41		11
	Khatanga, Taymir, Russia	Plasma	2000–2001	m,f	0.30	10	Geom mean										0.12		11
	Kanchalan, Chukotka, Russia	Plasma	2000–2001	m,f	0.30	9	Geom mean										0.31		11
	Uelen, Chukotka, Russia	Plasma	2000–2001	m,f	0.30	6	Geom mean										0.23		11
	Chukotsky district, Chukchi, Russia	Milk	2000–2001	f	2.96	10	Mean	0.015	0.22		0.016	0.017	0.022	nd	nd		0.31		11
	Anadyrsky district, Chukchi, Russia	Milk	2000–2001	f	2.52	7	Mean	0.007	0.083		0.019	0.009	nd	nd	nd		0.11		11

Values for high volume air samples are given in pg/m^3, passive samples in ng/g sample, soil and sediment samples in ng/g dw, vegetation in ng/g ww and biota samples in ng/g lw unless otherwise noted.

Nd – not detected, est – estimated, det – detected.

References: 1. de Wit (2002); 2. Remberger et al. (2004); 3. Kallenborn et al. (submitted for publication); 4. Alaee et al. (2003); 5. Shen et al. (in press); 6. Jaward et al. (2004a, sum calculated using half the detection limits); 7. Jaward et al. (2004b); 8. RAIPON/AMAP/GEF Project (2001); 9. Hassanin et al. (2004); 10. Schlabach et al. (2002); 11. AMAP (2004); 12. ter Schure et al. (2002); 13. Mariussen et al. (2004); 14. Sellström et al. (1993); 15. Lindberg et al. (2004, *8 eggs analyzed); 16. Herzke et al. (2001); 17. Herzke et al. (2005); 18. Vorkamp et al. (2005, **dimethyl TBBPA); 19. Malmquist et al. (2003); 20. Muir et al. (2003); 21. Stern et al. (2001); 22. Schlabach et al. (2001); 23. Herzke et al. (2004); 24. Vives et al. (2004); 25. Fjeld et al. (2004); 26. Stern and Evans (2003); 27. Chernyak et al. (2003); 28. Jenssen et al. (2004); 29. Christensen et al. (2002); 30. Herzke (2002a); 31. Johansen et al. (2004); 32. Wolkers et al. (2004); 33. Bytingsvik et al. (2004); 34. Vorkamp et al. (2004a); 35. Herzke et al. (2003); 36. Verreault et al. (2004); 37. Herzke et al. (2004); 38. Pusch et al. (2005); 39. Braune et al. (2003); 40. Vorkamp et al. (2004b); 41. Ikonomou et al. (2002); 42. Muir et al. (1999); 43. Muir et al. (2004); 44. Savinova et al. (2004); 45. Herzke (2002b); 46. Law et al. (2003); 47. van Bavel et al. (2001); 48. Thron et al. (2004); 49. Lindström et al. (1999); 50. Muir et al. (2006); 51. Skaare (2004); 52. Pereg et al. (2003); 53. Fängström et al. (2004).

Dunai (range not detected–62 pg/m^3), 240 pg/m^3 at Alert (range 10–868 pg/m^3) and 424 pg/m^3 at Tagish (range 27–2127 pg/m^3). The mean concentrations at Alert and Tagish were higher than air concentrations reported by Strandberg et al. (2001) for the city of Chicago (mean of 52 pg/m^3 for 1997–1999) and much higher than concentrations at air monitoring sites in the Great Lakes (means of 5–23 pg/m^3 at three sites). The spatial trends for Arctic PBDE concentrations from high volume air sample and passive air sample measurements discussed in this section are shown in Fig. 1.

Mono-, di- and triBDEs were found in air samples from Alert, which may indicate photodegradation (debromination) of PBDEs during long-range atmospheric transport (Alaee et al., 2003). The high $\sum$PBDE concentrations at Alert and Tagish, two sites that have been used for studies of PCBs, polyaromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) in air (Fellin et al., 1996; Halsall et al., 1997, 1998) illustrates a potential problem with measurements of PBDEs at background sites, i.e. remote sites far from major emission sources. Open burning is common in Arctic and sub-Arctic communities in Canada, because municipal waste is not recycled, incinerated under controlled conditions or cannot always be suitably landfilled. This may cause local emissions of some chemicals found in consumer products. Indeed, Halsall et al. (1998) noted higher combustion-related PAHs (phenanthrene, retene) during the summer months at Tagish and Alert, which may be related to local burning. Farrar et al. (2004) have recently demonstrated that PBDEs were elevated in air during "bonfire" events in the UK and that the combustion profile is enriched in BDE-99, -153 and -154, compared to the PentaBDE technical product, which has similar concentrations of BDE-47 and -99 and lower concentrations of BDE-100, -153 and -154 (Sjödin et al., 1998).

A significant fraction of the PBDEs (10–25% for BDE-47; 10–90% for BDE-99) in the Arctic samples was present in the particulate phase during the winter months. Maximum concentrations were seen in samples taken in the summer months. BDE-47 predominated in air samples in Sweden (de Wit, 2002) as well as in all Great Lakes samples (Strandberg et al., 2001). The major congeners observed at

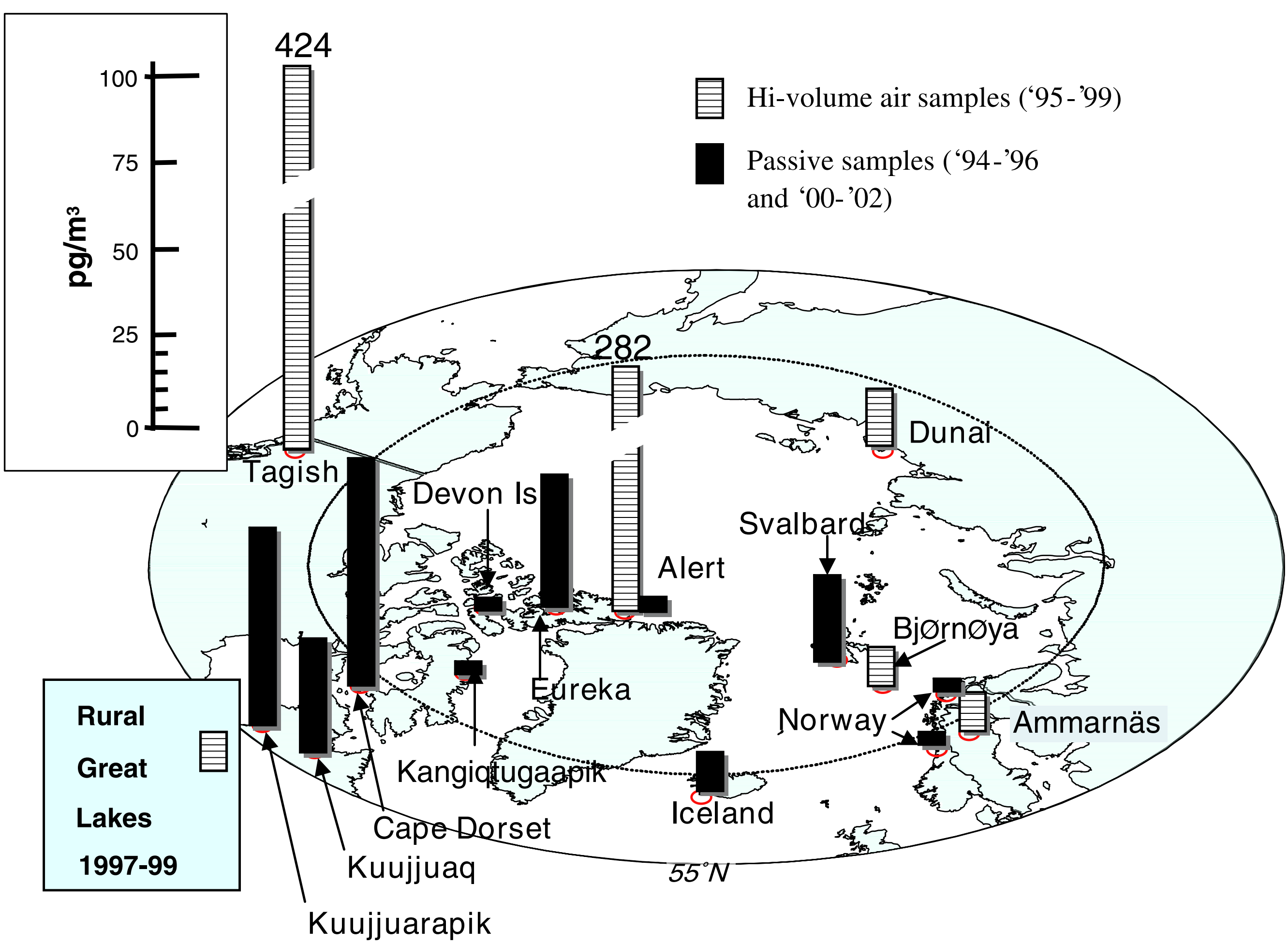

Fig. 1. $\sum$PBDE concentrations in Arctic air samples. Results for passive samplers have been converted to pg/m^3. Results for Arctic samples are from de Wit (2002), Alaee et al. (2003), Jaward et al. (2004a,b), Kallenborn et al. (submitted for publication) and Shen et al. (in press); results for the Great Lakes are from Strandberg et al. (2001).

the Canadian and Russian Arctic sites were BDE-47, -99, -100, -153 and -154 with BDE-99 having the highest concentrations.

Mean ∑PCB concentrations in air from Dunai (34 pg/m^3) are somewhat higher than for ∑PBDE. But at Alert and Tagish, ∑PBDE concentrations in air are higher than for ∑PCBs (22 and 32 pg/m^3; Stern et al., 1997) at the same sites. The PBDE results, if typical, imply significant air contamination by these compounds. Preliminary analyses of some samples from Alert, Dunai and Tagish show HBCD and BDE-209 concentrations below detection limits but tetrabromobisphenol A (TBBPA) was detected on the filter of one Dunai sample at 70 pg/m^3 (Alaee et al., 2003). New PBDE results for Alert were obtained in a recent passive air sampling campaign to investigate levels and spatial trends across North America and to better understand the levels and potential sources at Alert (Shen et al., in press). Passive air samplers were deployed for one year (2000/2001) and then retrieved for analysis. The ∑PBDE (BDE-47, -99, -100, -153, -154) concentration for Alert was 4 pg/m^3, which is probably more representative of average air concentrations, supporting the hypothesis that the high concentrations seen previously may have been due to local emissions sources (burning trash). The BDE congener profile in the new sample was similar to that seen in the PentaBDE technical product (similar concentrations of BDE-47 and -99), unlike the previous samples. ∑PBDE concentrations in passive samples from five other sites in northern Canada (Eureka, Devon, Kangiqtugaapik, Cape Dorset, Kuujjuak) ranged from 0.3 to 68 pg/m^3 (Shen et al., in press).

The air concentrations of ∑PBDEs observed during a short air monitoring campaign on Bjørnøya (situated between northern Norway and Svalbard) ranged from 3 to 10 pg/m^3 for the period of mid-December 1999 to mid-April 2000 (Kallenborn et al., submitted for publication). The same suite of PBDE congeners as at Alert were detected, with BDE-47 predominating. Di- and triBDEs (BDE-13, -15 and -33) also were prominent. These ∑PBDE concentrations were somewhat lower than the ∑PCB levels (mean 34 pg/m^3, range 16–51 pg/m^3). While no summertime concentrations are available from Bjørnøya, the results suggest that PBDE levels are much lower than at Alert or Tagish but similar to levels in rural areas of the Great Lakes (Fig. 1). Thus, the first reported results for Alert and Tagish are probably not typical of concentrations of PBDEs in Arctic air, although they could be typical of low temperature burning which is common in Arctic communities in Canada and Alaska.

Air samples were taken concurrently from June 15 to July 30, 2002, in 22 European countries including Iceland, Norway (remote site) and on Svalbard using passive air samplers (Jaward et al., 2004a). According to the authors, the six-week deployment was equivalent to sampling 130–170 m^3 of air. The ∑PBDE levels (BDE-28, -47, -75, -99, -100, -153, -154) in the samples from these sites were lower (1.7, 1.6 and 3.9 ng/sample, or 11, 11 and 26 pg/m^3, respectively) than at urban sites (up to 42 ng/sample or 280 pg/m^3) but were comparable to many rural or remote European sites. In the sample from Svalbard, BDE-99 dominated the congener pattern, followed by BDE-47, -100, -153 and -154. In the other samples, most congeners were below the detection limits. ∑PCB concentrations (sum of 17 congeners) for the three samples were considerably higher than ∑PBDEs, at 10.5, 3.9 and 8.6 ng/sample (70, 26 and 57 pg/m^3), for Iceland, Svalbard and remote Norway, respectively.

Air was sampled using passive air samplers deployed for two years (2000–2002) on a latitudinal transect of UK and Norway (Jaward et al., 2004b). The ∑PBDE concentrations (BDE-28, -47, -49, -99, -100, -153, -154) for air samples from two sites in northern Norway (latitude 67°38′ N and 69°83′ N) were 3.3 and 3.0 ng/sample (1.3 and 1.2 pg/m^3). Latitudinal fractionation with decreasing concentrations going northwards was seen for BDE-47, -49, -99 and -100 whereas BDE-153 and 154 showed no trend. ∑PCB concentrations were 2–5 times higher depending on the number of congeners included (7 or 31).

Air and deposition samples were collected on two occasions (September/October 2000 and January 2001) from an air monitoring station at Pallas, northern Finland, and analyzed for total HBCD using GC–ECD (Remberger et al., 2004). Air concentrations were 3 pg/m^3 in the autumn sample and 2 pg/m^3 in the winter sample. These concentrations are similar to those seen previously in Sweden (de Wit, 2002). Deposition was 13 ng/m^2 day in the autumn sample (21 mm precipitation) and 5.1 ng/m^2 day in the winter sample (0.4 mm precipitation).

The results from BFR studies in Arctic air support the conclusions of Wania and Dugani (2003) that a number of BFRs are capable of undergoing long-range air transport. These include the di-heptaBDEs, HBCD and possibly TBBPA. No data are available for BDE-209. ∑PBDE concentrations in Arctic air are generally comparable to those found at monitoring sites in the Great Lakes and in many cases, the congener profile is similar to the PentaBDE technical product. An exception to this is the case where high ∑PBDE concentrations are found at sites where local incineration is suspected to be a source and these samples are often enriched in BDE-99.

1.2. Terrestrial environment

1.2.1. Soil

Samples of surface soil were collected from four regions of the Russian Arctic: Chukotka (Kanchalan and Lavrentiya); Kola Peninsula; Pechora Basin; and, Taymir Peninsula (Dudinka), in 2000–2001 (RAIPON/AMAP/GEF Project, 2001; AMAP, 2004). Samples were collected in the same locations as plants and terrestrial mammals, and covered a wide geographical area. Each sample analyzed was a pool of the samples collected. Low concentrations (0.16–0.23 ng/g dry weight – dw) of the sum of BDE-47, -99 and -100 (detection limits approximately

0.5 ng/g in soil) were observed in three soil samples from the two sites in Chukotka. ∑PCB concentrations were 20–80 times higher.

In a study of remote and rural soils collected on a latitudinal transect of UK and Norway, the median ∑PBDE concentration (sum of 20 Te-HpBDEs) for Norwegian woodland soils was 0.97 ng/g dw (1.5 ng/g organic matter) (Hassanin et al., 2004). BDE-47, -99, -100, -153 and -154 were the predominant congeners and the ∑PBDE concentration for these was 0.71 ng/g dw (1.1 ng/g organic matter). BDE-99 was present in the highest concentrations, followed by BDE-47, -100, -153 and -154. The pattern was similar to that of the PentaBDE technical product. BDE-183 was present at low concentrations (median 0.025 ng/g dw). ∑PBDE concentrations declined going north and latitudinal fractionation was seen with higher relative contribution of BDE-47 to ∑PBDE going northwards, while the relative proportions of BDE-99, -153 and -154 decreased. Median ∑PCB concentration in the same soil samples was 4.6 ng/g dw, five times higher than the ∑PBDE levels (Meijer et al., 2002).

Based on the little soil data that is available, ∑PBDE concentrations are quite low, and are probably reflective of the lower air concentrations that are also seen in these areas. The BDE congener profile is also reflective of that seen in air samples from these regions, i.e. similar to the PentaBDE technical product. Results showing latitudinal fractionation of lower brominated PBDEs are indicative of their long-range transport in air.

1.2.2. Vegetation

Moss samples (*Hylocomium splendens*) from several sites in Norway were collected in July, 2002, to determine deposition of several BFRs (Schlabach et al., 2002). Two sites were in the Arctic, at Valvik (67°38′ N) and Skoganvarre (69°83′ N). The ∑PBDE (BDE-28, -47, -99, -100, -153, -154, -183) concentration at Valvik was 0.029 ng/g wet weight (ww), BDE-209 concentration was 0.12 ng/g ww and TBBPA concentration was 0.14 ng/g ww. For Skoganvarre, the ∑PBDE (BDE-28, -47, -99, -100, -153, -154, -183) concentration was 0.31 ng/g ww, BDE-209 concentration was 0.025 ng/g ww, TBBPA concentration was 0.019 ng/g ww and the three HBCD isomers were below the detection limit. PBBs (BB-15, -49) were only detected at Skoganvarre, the more northerly site with ∑PBB concentration of 0.023 ng/g ww. The BDE congener profile was similar to the PentaBDE technical product in the moss samples from Valvik whereas, at Skoganvarre, the more northerly site, the profile was dominated by BDE-47. At Valvik, BDE-209 and TBBPA had the highest concentrations whereas at Skoganvarre, BDE-47 concentrations were highest.

Samples of mosses, lichens and berries were collected from four regions of the Russian Arctic: Chukotka (Kanchalan and Lavrentiya); Kola Peninsula; Pechora Basin; and, Taymir Peninsula (Dudinka and Khatanga), in 2000–2001 (AMAP, 2004). Each sample analyzed was a pool of the samples collected from each area. Low concentrations (0.004–0.04 ng/g dw) of the sum of BDE-47 and -99 were observed in all three sample types. ∑PCB concentrations were 90–600 times higher.

Contaminant concentrations on vegetation are indicative of deposition from air, and when sites are far from emission sources, provide evidence of long-range transport. The various BFR concentrations are quite low, but indicate that not only the lower brominated BDEs undergo long-range transport but also BDE-209, TBBPA and PBBs.

1.2.3. Terrestrial animals

Common frogs (*Rana temporaria*) are semiterrestial amphibians that feed on snails, slugs, worms and insects, i.e. in the terrestrial food web (Beebee and Griffiths, 2000). Frogs were collected in Sweden from 1998 to 2000 along a 1500 km long latitudinal gradient and livers analyzed for BDE-47 and -99 (ter Schure et al., 2002). BDE-47 showed a latitudinal gradient with decreasing concentrations going northward. Mean BDE-47 concentrations were 2.3 and 0.93 ng/g lw (0.034 and 0.026 ng/g ww) in frogs from Ammarnäs (subarctic) and Kiruna (arctic), respectively. BDE-99 concentration was 5.6 ng/g lw (0.084 ng/g ww) in the Ammarnäs sample and was not detected in the Kiruna frogs. These concentrations were similar to those of individual PCB congeners (CB-52, -183, -201) but lower than for CB-153, which was 10 times higher.

∑PBDE (BDE-47, -99, -100) concentration in reindeer (*Rangifer tarandus*) suet from Ottsjö, Sweden (collected in 1986) was 0.47 ng/g lw (Sellström et al., 1993). ∑PBDE concentrations (BDE-28, -47, -99, -100, -153, -154, -183) in liver samples from grouse (*Lagopus* sp.), moose (*Alces alces*) and lynx (*Lynx lynx*) from Norway were 0.64, 0.87 and 9.8 ng/g lw (0.025, 0.04 and 0.54 ng/g ww), respectively (Mariussen et al., 2004). Samples were collected between 1990 and 1995. BDE-153 was the predominant congener in lynx (79% of ∑PBDE) (Fig. 2). Reindeer and moose have

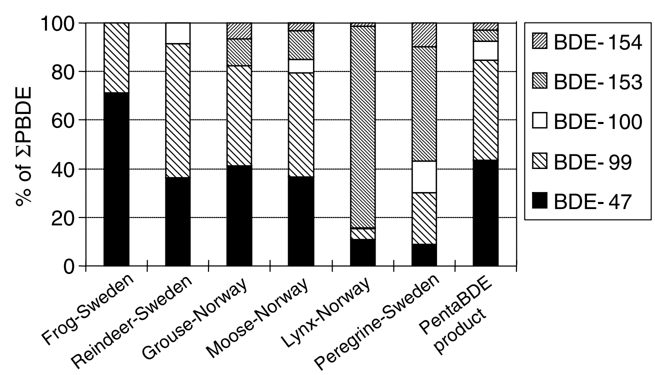

Fig. 2. PBDE congener profile in arctic and subarctic terrestrial animals compared to the technical PentaBDE product. Results for frogs are from ter Schure et al. (2002), for reindeer from Sellström et al. (1993), for grouse, moose and lynx from Mariussen et al. (2004), for peregrine falcons from Lindberg et al. (2004) and for PentaBDE from Sjödin et al. (1998).

congener profiles of the lower brominated BDEs that are similar to the PentaBDE product. No BDE-100 or -183 was found in grouse and very little BDE-100 was found in lynx, which may indicate that these are metabolized. BDE-209 was also found at concentrations of 0.5, 1.0 and 0.8 ng/g lw for grouse, moose and lynx, respectively. Lynx samples from 2002 had ∑PBDE concentrations of 50 ng/g lw and for BDE-209, 4 ng/g lw, but variation was high so no firm conclusions about temporal trends could be made.

Samples of liver from Arctic hares (*Lepus timidus*) and kidney from reindeer were collected from four regions of the Russian Arctic: Chukotka (Kanchalan and Lavrentiya); Kola Peninsula; Pechora Basin; and, Taymir Peninsula (Dudinka and Khatanga), in 2000–2001 (AMAP, 2004). Each sample analyzed was a pool of the samples collected from each area. Low concentrations (0.13–0.27 ng/g ww, no information on lipid content available) of BDE-47 and -99 were observed in both sample types. ∑PCB concentrations were 3–5 times higher.

High BFR concentrations have been found in eggs of predatory birds feeding on terrestrial mammals and birds, particularly peregrine falcons (*Falco peregrinus*) in northern Sweden collected between 1991 and 1999 (Lindberg et al., 2004) and Norway (collected 1993–2000) (Herzke et al., 2001) and more recently (2000–2003) at several sites in South Greenland (Vorkamp et al., 2005). The mean ∑PBDE (BDE-47, -99, -100, -153 and -154) in the Swedish, Norwegian and Greenland populations were 4500, 4700 and 4000 ng/g lw (224, 260 and 230 ng/g ww), respectively, with levels ranging from 680 to 39 000 ng/g lw (43–1580 ng/g ww) in the Swedish population and 810 to 13 000 ng/g lw (43–890 ng/g ww) in the Greenland population. The Swedish and Greenlandic peregrine falcons also had measurable levels of BDE-183 and Swedish, Norwegian and Greenlandic falcons all contained BDE-209 (DeBDE) (Lindberg et al., 2004; Herzke et al., 2005; Vorkamp et al., 2005). The predominant BDE congener in all peregrine falcons was BDE-153 (Fig. 2).

The mean ∑PBB level in Norwegian peregrine falcons was 900 ng/g lw (49 ng/g ww) and the BB-153 level was 114 ng/g lw (5.7 ng/g ww) in Swedish peregrines. HBCD concentrations analyzed by GC–NCIMS ranged from 34 to 2400 ng/g lw (2.2–28 ng/g ww) in the Swedish peregrine falcons but were much lower in the Greenlandic peregrine falcons, <0.1–27 ng/g lw. TBBPA was also detected in Norwegian peregrine falcon eggs (Herzke et al., 2005), but in Greenlandic peregrines only dimethylated TBBPA was found, at concentrations ranging from 0.1 to 900 ng/g lw (mean of 380 ng/g lw). Mean ∑PCB levels in the Swedish peregrine falcons were 52 times higher than the PBDE levels, in the Norwegian falcons, mean ∑PCB levels were 40 times higher (Herzke et al., 2002) and in the Greenlandic falcons mean ∑PCB levels were 20 times higher.

Temporal trends were also determined for South Greenland peregrine falcon eggs collected between 1981 and 2003 (Vorkamp et al., 2005). BDE-47, -49, -85, -99, -100, -153, -183 and -209 were all found to have statistically significantly increasing trends. For example, BDE-99 concentrations are increasing at the rate of approximately 10% per year. Other congeners are increasing at rates between 5% and 10% per year.

Norwegian golden eagles (*Aquila chrysaetos*), gyrfalcons (*Falco rusticolus*) and merlins (*Falco columbarius*) feed on terrestrial birds and mammals. The mean ∑PBDE levels in eggs collected in the period 1991–2002 were approximately 140, 360 and 720 ng/g lw (7, 18 and 36 ng/g ww) for golden eagle, gyrfalcon and merlin, respectively (Herzke et al., 2001). Median levels were somewhat lower and BDE-209 was also present in these species (Herzke et al., 2005). The congener pattern was dominated by BDE-153, just as for the peregrine falcons. The mean ∑PBB levels were 180, 60 and 150 ng/g lw (9, 3 and 7.5 ng/g ww) in golden eagle, gyrfalcon and merlin, respectively (Herzke et al., 2001). TBBPA was also detected in golden eagle eggs (Herzke et al., 2005). Mean ∑PCB levels were 200 times higher than ∑PBDE levels in golden eagle, 111 times higher in gyrfalcon and 44 times higher in merlin (Herzke et al., 2002).

The congener pattern in all these bird species is quite different from that seen in fish and fish-eating birds and mammals, and is dominated by BDE-153 and -99. The burden of PBDEs in peregrine falcons may be linked to their migratory habits as the northern population in Sweden overwinters along the coast and estuaries of central and southern Europe (Fransson and Pettersson, 2001). In addition, many of the birds they prey on in the Arctic are also migratory and may have PBDE burdens from their overwintering sites further south.

Terrestrial animals at lower trophic levels have relatively low ∑PBDE concentrations whereas birds of prey have much higher levels, with highest levels in the top predator, peregrine falcon. The congener pattern in herbivores is more similar to the PentaBDE technical product, whereas in predators such as lynx and birds of prey, the pattern is shifted to a predominance of BDE-153. BDE-209 was found in both terrestrial mammals (moose, lynx), ground-dwelling grouse and in birds of prey, indicating its presence in the terrestrial food web. HBCD, TBBPA including the metabolite dimethylated TBBPA, and some PBBs have also been found in some birds of prey. The increasing levels of PBDEs including BDE-209 seen in Greenlandic peregrine falcons from 1981 to 2003 are probably reflective of increased use of these BFRs.

1.3. Freshwater environment

1.3.1. Sediments

Dated sediment cores were collected from Canadian lakes on a north-south transect from southern Ontario to Ellesmere Island (Muir et al., 2003). The lakes were uninhabited or had a history of very limited human disturbance. Slices of the cores representing different depths were analyzed for PBDEs (Te-DeBDE). BDE-209 was detected in recent slices of most of the cores and concentrations in the three Arctic lakes were 0.075 (AX-AJ Lake), <0.1 (Romulus Lake) and 0.042 (Char Lake) ng/g dw. These concentrations were much lower than at the more southerly sites. The Arctic lakes have much lower fluxes (ng/m^2 year) than more southerly lakes with somewhat later appearance of BDE-209. This is consistent with BDE-209 having lower mobility due to it most likely being sorbed to and transported on atmospheric aerosols (Wania and Dugani, 2003). The fluxes in the sediment slices of the cores indicate increasing inputs of BDE-209 with time (Fig. 3). BDE-47 flux in the sediment core from Char Lake indicates increasing inputs up to the late 1980s, with a lower flux in the slice from 1995. Wania and Dugani (2003) estimated a characteristic travel distance (CTD) of BDE-209 of 480 km. This compares to a "half distance" based on BDE-209 in sediment cores of about 580 km (Muir et al., 2003). By comparison Muir et al. (1996) reported a half distance of 750 km for PCBs over a mid-continental latitudinal gradient.

Malmquist et al. (2003) analyzed BDE-47 in sediment cores from seven lakes in West Greenland collected in May 2000. The lakes had very small catchment areas and contaminants in sediments were believed to be entirely due to atmospheric input. BDE-47 concentrations ranged from 0.007 to 0.051 ng/g dw in the top layer (0–1 cm) of the cores and generally showed increasing concentrations in recent layers which dated to the period 1990–2000. PCB concentrations (sum of 67 congeners) were 40–135 times higher. Fluxes of BDE-47 ranged from about 0.05 to 1.3 ng/m^2 year which was similar to the range found for BDE-47 in Canadian arctic lake sediments (<0.5–8 ng/m^2 year) (Muir et al., 2003) (Fig. 3).

Samples of lake and river sediments were collected from four regions of the Russian Arctic: Chukotka (Kanchalan); Kola Peninsula; Pechora Basin; and, Taymir Peninsula (Dudinka and Khatanga), in 2000–2001 (AMAP, 2004). Low concentrations (0.004–0.027 ng/g dw) of the sum of BDE-47 and -99 were observed in all samples. $\sum$PCB concentrations were 90–600 times higher.

1.3.2. Fish

In the late 1990s, various fish species were collected from lakes in a north-south transect of Norway and analyzed for BDE-47 and -99 (Schlabach et al., 2001). The sampling included Arctic char (*Salvelinus alpinus*) from the lake Ellasjøen on Bjørnøya. In Lake Grensefoss in very northern Norway, $\sum$PBDE in burbot (*Lota lota*) liver was 175 ng/g lw (20 ng/g ww). For trout (*Salmo trutta*) muscle from four different lakes (Takvatn, Fjellfrøsvatnet 99, Grunnvatnet, Store Raudvannet), the range was 8–14 ng/g lw (0.10–0.36 ng/g ww). For Arctic char muscle from

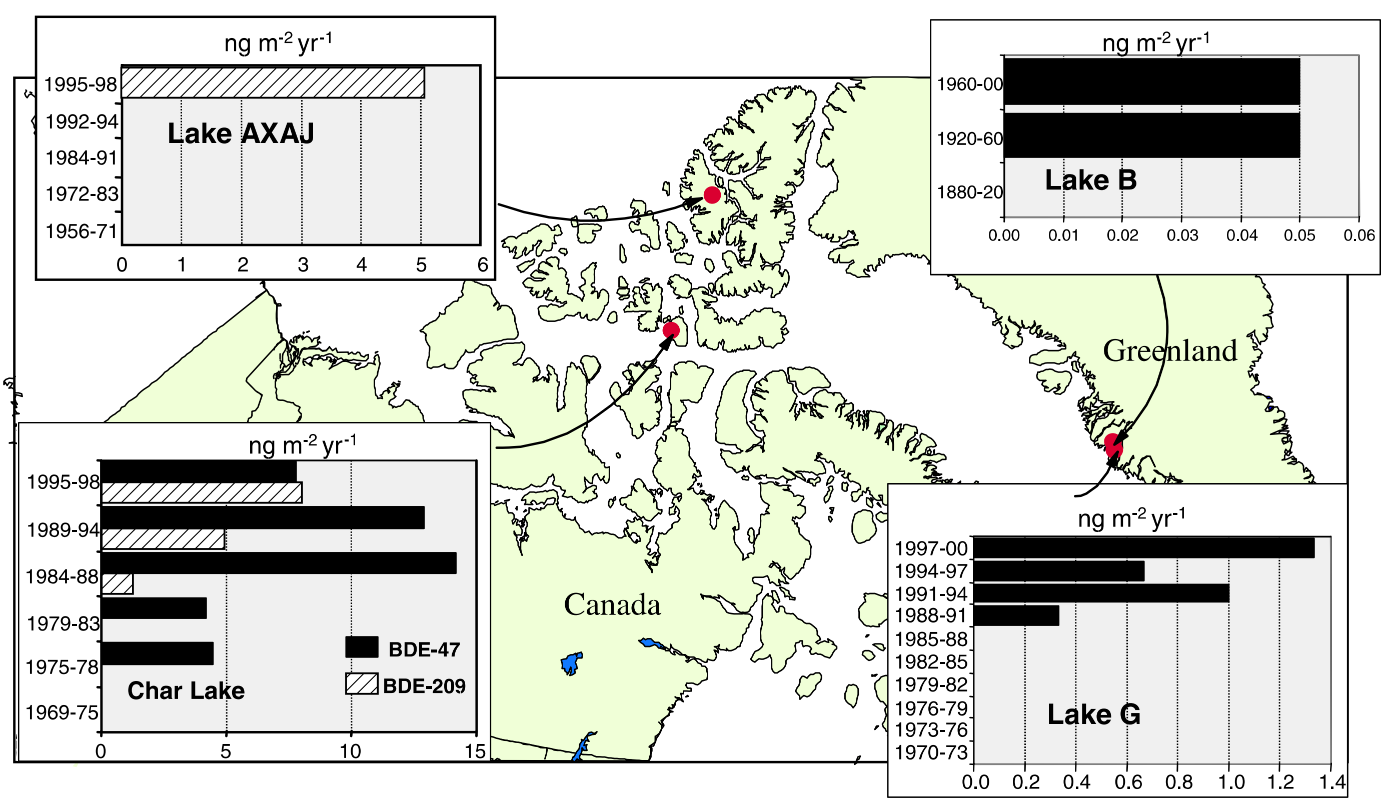

Fig. 3. Profiles and fluxes (ng m-2 yr-1) of BDE-47 and BDE-209 in dated lake sediment cores from lakes in the Canadian Arctic and western Greenland. Results from Malmquist et al. (2003) for BDE-47 in Lakes B and G and from Muir et al. (2003) for BDE-47 and -209 in Char Lake and Lake AXAJ. BDE-47 was undetectable (<0.2 ng/m^2 year) in Lake AXAJ.

Ellasjøen on Bjørnøya, the ∑PBDE concentration was 1250 ng/g lw (16.3 ng/g ww). This lake is impacted by seabird guano from nearby seabird colonies, which has been shown to cause higher organochlorine concentrations in the char (Evenset et al., 2004). A recent study confirms that seabird guano, particularly from kittiwakes and glaucous gulls, is also a major source of BDEs to this lake (Herzke et al., 2004) and that this contributes significantly to the bioaccumulation of BDEs in Arctic char (Gandhi et al., 2004). In this study, Arctic char from Lake Ellasjøen had ∑PBDE concentration (BDE-28, -47, -99, -100, -153) of 420 ng/g lw while char from Lake Øyangen, which is also on Bjørnøya but is not impacted by seabirds, had a much lower concentration of 50 ng/g lw (Herzke et al., 2004).

The congener pattern in all three fish species was evenly distributed between BDE-47 and -99, and resembles the PentaBDE technical product. For burbot and trout, the PBDE levels were 2–10 times lower than ∑PCB levels. For Arctic char at Bjørnøya, ∑PBDE concentrations were 30–40 times lower than ∑PCB concentrations (Schlabach et al., 2001). For burbot and trout, the concentrations found in the northern lakes were much lower than for the same species in more southerly lakes (Schlabach et al., 2001). The explanation for the high ∑PBDE concentration in burbot from Lake Grensefoss is not clear as this lake is in a national park. However, it is only 100 km southwest of the highly industrialized city of Nikel, Russia, which may impact this area.

∑PBDE levels (BDE-28, -47, -99, -100, -153, -154) were determined in fish from 11 high mountain lakes in Europe and one (Lake Fergusson) on Greenland (Vives et al., 2004). Arctic char collected from Lake Fergusson on southern Greenland had ∑PBDE levels of 23 ng/g lw (0.90 ng/g ww) in liver and 41 ng/g lw (0.31 ng/g ww) in muscle. BDE-47 was the predominant congener in muscle (30% of the total) followed by BDE-99, -100, -154 and -153. In liver, BDE-47 was even more predominant (40% of the total) and was followed by BDE-100, -99, -154 and -153. Generally speaking, the concentrations in the different lakes differed little and no statistically significant correlations were seen based on altitude or latitude.

PBDEs (BDE-47, -99, -100, -153, -154) were measured in the livers of burbot collected at Fort Good Hope on the Mackenzie River (Canada) in 1988, 1999 and 2000 as part of a project on temporal trends (Stern et al., 2001). ∑PBDE levels in the burbot liver collected in 1999 and 2000 were 4.3 and 4.1 ng/g lw (1.6 and 1.3 ng/g ww), respectively (Stern et al., 2001). BDE-47 was the predominant congener followed by BDE-99, -100, -153 and -154, but levels are below those observed in the Norwegian fish. ∑PCB levels for 1999 and 2000 were 149 and 151 ng/g lw (62.8 and 54.6 ng/g ww), 35 and 41 times higher than ∑PBDE levels.

Muir and Köck (2003) determined PBDEs in landlocked Arctic char collected from Resolute Lake in the Canadian Arctic annually from 1997 to 2002. BDE-47 and BDE-99 doubled in concentration in Resolute Lake char between 1997 and 2001. This doubling time was similar to observations for Canadian Arctic ringed seals (Ikonomou et al., 2002), thick-billed murres and northern fulmars (*Fulmaris glacialis*) (Braune et al., 2003). Mean concentrations of these PBDEs were significantly higher in 2001 and 2002 than in 1997.

Liver samples from several fish species were collected from three regions of the Russian Arctic: Chukotka (Kanchalan) – pike (*Esox lucius*), inconnu (*Stenodus leucichthys*); Kola Peninsula – whitefish, pike; and Pechora Basin – perch (*Perca fluviatilis*), ide (*Leucisus idus*), in 2000–2001 (AMAP, 2004). Similar but low concentrations (0.22–0.42 ng/g ww, no information on lipid content available) of BDE-47 were observed in all samples and no geographical trend was seen.

With a few exceptions, ∑PBDE concentrations within the same fish species are similar and generally low across the Arctic. Lowest concentrations are seen in burbot and highest in Arctic char. The exceptions are Arctic char from the lake Ellasjøen on Bjørnøya, which have 8–24 times higher concentrations than char from other areas and burbot from Lake Grensefoss, Norway, which have 20–40 times higher concentrations than other sites in Norway and Canada. The higher levels at Ellasjøen are due to inputs of seabird guano, whereas it is unclear what the source is at Lake Grensefoss. The congener profiles in freshwater fish resemble the PentaBDE technical product.

1.4. Marine environment

1.4.1. Marine sediments

∑PBDE concentrations (Di-HxBDEs) in three marine sediments from the High Arctic Canadian archipelago collected in 1998 were 0.107 (Barrow Strait), 0.122 (Penny Strait) and 0.297 ng/g dw (Nanisivik) (Stern and Evans, 2003). BDE-47 was the predominant congener with concentrations of 0.040, 0.046 and 0.11 ng/g dw, respectively. These concentrations were generally much higher than found in freshwater sediments in the Canadian Arctic and western Greenland. ∑PCB concentrations (sum of 102 congeners) were 2–13 times higher than PBDE concentrations.

Two marine sediment samples from Tromsø harbor (northern Norway) collected in 2003 had concentrations of 0.06 and 0.25 ng/g dw (1.8 and 7.4 ng/g total organic carbon – TOC) ∑PBDE (BDE-47, -99, -100, -153, -154), 0.42 and 0.43 ng/g dw (12.4 and 12.6 ng/g TOC) of BDE-209, 1.24 ng/g dw (36.5 ng/g TOC) of TBBPA and no detectable levels of HBCD (Fjeld et al., 2004). BDE-183 was not detected. BDE-209 was the most abundant congener followed by BDE-99, then BDE-47.

Sediments from three sites along Kola Bay, northern Russia, were collected in 1998 and analyzed for Te-HxBDEs (Chernyak et al., 2003). ∑PBDEs for Kola Bay and Guba Zapadnaya Litsa were 0.14–0.16 ng/g dw, but at Polyarnyy, a closed city with a navy base, they were 241 ng/g dw, with BDE-99 dominating the congener profile. PCB concentrations were 1000 times higher in Kola Bay and Guba Zapadnaya Litsa samples, but only 36 times higher in the Polyarnyy sediment sample (Savinova et al., 2000).

Based on the limited data available, ∑PBDE concentrations are quite similar in marine sediments from remote sites in Canada, Norway and Russia, but are generally higher than in freshwater sediments. The exception is at the Russian site of Polyarnyy, where local contamination from the naval base is indicated.

1.4.2. Zooplankton

In a food web study from Svalbard, PBDEs and HBCD were analyzed in three species of pelagic zooplankton – *Calanus glacialis*, *Thysanoessa inermis* and *Parathemisto libellula* (Jenssen et al., 2004). Only BDE-28, -47 and -99 were quantifiable and the ∑PBDE concentrations were in the range of 1 ng/g lw.

1.4.3. Blue mussels

PBDEs (BDE-47, -99, -100, -153) and PCBs (13 congeners) were studied in blue mussels (*Mytilus edulis*) collected in 2000 from western Greenland at Usuk, a background station located about 3–5 km from the village of Igaliko (Christensen et al., 2002). ∑PBDE concentrations were 5.5 ng/g lw (0.11 ng/g ww) and were seven times lower than for PCBs. The congener pattern was dominated by BDE-47 (90% of the total), followed by BDE-99. The other congeners were below detection limits. Blue mussels from two sites at Lofoten (Svolvaer) and one site at Varanger (northern Norway) collected in 2002 had ∑PBDE (BDE-47, -99, -100) levels of 5.9–23 and 1.5 ng/g lw (0.06–0.19 ng/g and 0.02 ng/g ww), respectively (Fjeld et al., 2004). BDE-209 concentrations in the mussels were 1.9–640 ng/g lw (0.02–5.24 ng/g ww) at Lofoten and 3.5 ng/g lw (0.06 ng/g ww) at Varanger. α-HBCD (analyzed using LC–MS) levels at Lofoten were 12 ng/g lw (0.1 ng/g ww) and at Varanger, 3.6 ng/g lw (0.06 ng/g ww). No β- or γ-HBCD was detected in any mussel samples.

1.4.4. Marine fish

Atlantic cod (*Gadus morhua*) liver from Lofoten (N. Norway) and the west coast of Norway had ∑PBDE levels (BDE-47, -99, -153) of 24 and 28 ng/g lw (16.4 and 21.3 ng/g ww) and tusk (*Brosme brosme*) liver from the Norwegian Sea had 65 ng/g lw (40 ng/g ww) (Herzke, 2002a). Atlantic cod liver from Lofoten (collected in 2002) and Varanger (collected in 1998 and 2002), Norway, had ∑PBDE levels (BDE-28, -47, -49+71, -99, -100, -119, -153, -154, -183) of 11.7–16.3 and 15–25 ng/g lw (8.8–12.2 and 7.8–14.5 ng/g ww), BDE-209 levels of <0.57–1.95 and 0.58–1.60 ng/g lw (<0.43–1.47 and 0.35–0.83 ng/g ww), α-HBCD levels of 6.6 and 7.7 ng/g lw (4.9 and 4.0 ng/g ww) and TBBPA levels of 0.5 and 2.5 ng/g lw (0.35 and 1.31 ng/g ww), respectively (Fjeld et al., 2004). Atlantic cod from southwest Greenland had liver ∑PBDE (BDE-47, -99) levels of 8.9 ng/g lw (5.1 ng/g ww) and muscle ∑PBDE levels of 480 ng/g lw (3.4 ng/g ww) (Johansen et al., 2004). Greenland halibut (*Reinhardtius hippoglossoides*) from southwest Greenland had liver ∑PBDE levels of 7.5 ng/g lw (2.7 ng/g ww) (Johansen et al., 2004). In a food chain study, polar cod (*Boreogadus saida*) were collected in 2001 from eastern Svalbard and analyzed for 22 Te-HxBDE congeners (Wolkers et al., 2004). Only BDE-47, -85, -99, -100, -154 and a methoxylated TeBDE (MeO-TeBDE) were found. The ∑PBDE concentration was 3.6 ng/g lw. The MeO-TeBDE concentration was 0.72 ng/g lw. In a similar food web study on Svalbard, polar cod were found to have ∑PBDE (BDE-28, -47, -99, -100, -153) concentrations of 5–25 ng/g lw and HBCD concentrations of 10–15 ng/g lw (Jenssen et al., 2004). Polar cod from around Bjørnøya collected in 2003 had mean ∑PBDE (BDE-28, -47, -100, -154) concentrations of 12 ng/g lw and HBCD concentrations of 14.7 ng/g lw (Bytingsvik et al., 2004).

The distribution of persistent halogenated organics, including BDE-47, -99, -100 and -153, was studied during 2000 near the western Greenland villages of Qaqortoq (3500 inhabitants), Igaliko (30 inhabitants) and Usuk (background site 3–5 km from Igaliko) (Christensen et al., 2002). Livers were collected from shorthorn sculpin (*Myoxocephalus scorpius*), uvak or Arctic cod (*Gadus ogac*), spotted wolffish (*Anarhichas minor*), and starry ray (*Raja radiata*). A spatial trend in sculpins was apparent with the highest ∑PBDE concentrations observed in samples from Qaqortoq (43 ng/g lw, 8.2 ng/g ww) followed by Igaliko (19 ng/g lw, 3.1 ng/g ww) and Usuk (11 ng/g lw, 2.1 ng/g ww). The concentrations in the other fish species ranged from 3.3 to 34 ng/g lw (1.2–12 ng/g ww). The levels of PBDEs were 15–24 times lower than PCB levels measured in the same individuals, except for shorthorn sculpin collected at Qaqortoq, where the level of ∑PCBs was 40 times higher. The results suggest that villages are sources of PBDEs to the nearby marine environment.

Arctic char and shorthorn sculpin were collected near Ittoqqortoormiit (Scoresby Sound), East Greenland in 2001 and analyzed for 11 BDEs (BDE-17, -28, -47, -49, -66, -85, -99, -100, -153, -154, -183) and several organochlorine compounds (Vorkamp et al., 2004a). Several BDEs were below detection limits. The mean ∑PBDE (BDE-28, -47, -99, -100, -154) concentration in shorthorn sculpin liver was 9.6 ng/g lw (1.7 ng/g ww) and in Arctic char muscle, the mean concentrations were 26 and 46 ng/g lw (0.57 and 1.5 ng/g ww) in females and males, respectively. ∑PCB levels were 19–28 times higher in the sculpin and char samples. The ∑PBDE levels in shorthorn sculpin from East Greenland are similar to those found at the background site of Usuk, western Greenland.

Two methoxylated TeBDEs (MeO-TeBDEs) were found in Arctic cod (*Cadus callarias*) liver from northern Norway collected in 1987, 1988, 1989, 1991, 1992, 1993, 1994, 1995 and 1998 (Sinkkonen and Rantalainen, 2004). Highest concentrations were found in 1992 and 1993 samples. The 1998 samples contained 0.08 and 0.91 ng/g lw of the two

MeO-TeBDEs. The origin of these compounds is unclear at present.

For all marine fish samples, BDE-47 is the predominant congener, usually making up 90–95% of the ∑PBDE concentration. The increasing ∑PBDE concentration with proximity to human communities indicates these as local sources to the marine environment. ∑PBDE levels in sea-run Arctic char from Greenland are similar to those seen in Greenlandic land-locked char, indicating similar inputs to the freshwater and marine ecosystems, probably from long-range transport and deposition. BDE-209, α-HBCD and TBBPA were also present in cod liver from two sites in northern Norway.

1.4.5. Seabirds

PBDEs (BDE-47, -99, -100, -153, -154) were measured in livers and eggs from thick-billed murres, northern fulmars and black-legged kittiwakes (*Rissa tridactyla*) collected in 1993 from Prince Leopold Island in Lancaster Sound, Canada (Braune et al., 2003; Braune and Simon, 2004). ∑PBDE levels in egg and liver samples of kittiwakes were 3 ng/g ww or about 60–70 ng/g lw, and 14 and 15 ng/g lw in the murre and fulmar samples, respectively. BDE-47 was the major PBDE congener in all the samples. PCB concentrations in eggs from the three species from 1993 were 60–300 times higher than for PBDEs (Braune et al., 2001).

PBDEs in thick-billed murres and northern fulmars collected in 1975, 1988, 1993 and 1998 from Prince Leopold Island, Canada, show rapidly increasing trends, from 2–4 ng/g lw in 1975 to 18–20 ng/g lw in 1998 (Braune et al., 2003) (Fig. 4a). The highest level reported is an order of magnitude lower than levels reported for Swedish common guillemots (*Uria aalge*) from the Baltic Sea in 1999 (143 ng/g lw) (Sellström et al., 1993, 2003; Sellström, 1999), but PBDE concentrations in Baltic Sea guillemots have declined since their peak in the mid-1980s. The PBDE levels in Canadian seabirds are higher than levels reported for Canadian marine mammals (Law et al., 2003). Temporal trends of PBDEs in marine mammals indicate increasing levels in the Canadian Arctic during the period of decline in the Baltic Sea, implying that current concentrations in Canadian Arctic seabirds may be approaching those of guillemot in the Baltic Sea.

∑PBDE levels (BDE-28, -47, -99, -100, -153, -154) were determined in the liver of thick-billed murre and black guillemot (*Cepphus grylle*) collected from southwestern Greenland in 1999 (Johansen et al., 2004) and in black guillemot livers and eggs from East and West Greenland in 2000 (Vorkamp et al., 2004b) and East Greenland (eggs) again in 2001 (Vorkamp et al., 2004a). The East Greenland eggs also contained BDE-49 and -66. Concentrations of ∑PBDE in the thick-billed murre (32 ng/g lw; 1.7 ng/g ww) and black guillemot (46 ng/g lw; 3.0 ng/g ww) from southwestern Greenland were similar to levels observed in Canadian Arctic seabird eggs. The ∑PBDE levels in black guillemot liver and eggs from West Greenland were the same, 25 ng/g lw. ∑PBDE levels were 73 ng/g lw in liver and 83 ng/g in eggs from East Greenland black guillemot. PCB levels in liver and eggs from West Greenland were 44 and 26 times higher than PBDEs and for liver and eggs from East Greenland, PCB levels were 30 and 77 times higher.

BFRs were measured in the liver and intestinal contents of 15 glaucous gulls (*Larus hyperboreus*) collected

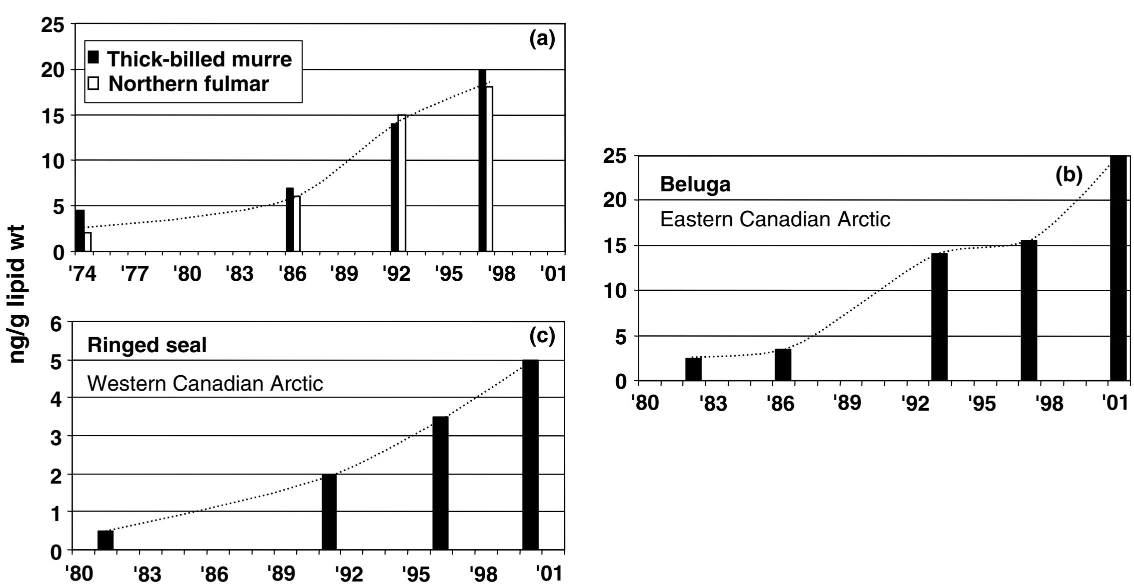

Fig. 4. Temporal trends of ∑PBDEs in (a) thick-billed murre and northern fulmar eggs, (b) beluga whales, and (c) ringed seals in the Canadian Arctic. Results for seabirds from the central Canadian Arctic archipelago from Braune et al. (2003) and for ringed seals from the western Canadian Arctic from Ikonomou et al. (2002). For beluga results from the eastern Canadian Arctic are from Law et al. (2003) for 1982–1998, combined with results from samples collected in 2001 from Muir et al. (2004).

on Bjørnøya in 1999 (Burkow et al., 2001; Herzke et al., 2003). Only two PBDE congeners were detected, BDE-47 and -99, at concentrations between 27 and 450 ng/g lw (2 and 25 ng/g ww). Analysis of the samples using gas chromatography/high resolution mass spectroscopy revealed a number of other PBDE and PBB congeners. ∑PBDE levels in glaucous gulls are up to 1000 times lower than ∑PCB levels. For most glaucous gulls, these ∑PBDE concentrations are similar to what has been measured in seabird eggs from the Canadian Arctic (Braune et al., 2003). However, a few individual glaucous gulls had ∑PBDE levels (480–560 ng/g lw) (Burkow et al., 2001), which were higher than levels seen in Baltic Sea guillemots for the same year (Sellström et al., 2003). Levels of BDE-47 and -99 were up to 100 times lower than levels of CB–153 in the glaucous gulls.

Muscle samples from glaucous gulls from Bjørnøya collected in 1998–2003 have higher ∑PBDE levels (BDE-47, -99, -100, -153) of 1400 ng/glw (Herzke et al., 2004). Although BDE-47 is the dominant congener (620 ng/g lw), high concentrations of BDE-153 (420 ng/g lw) were also found. Verreault et al. (2004) found similar ∑PBDE (BDE-28, -47, -99, -100, -153, -154, -183, -209) levels in plasma from Bjørnøya glaucous gulls collected in 2002 and 2004, 1400 ng/g lw (19 ng/g ww). HBCD concentrations ranged from 6.1 to 120 ng/g lw (mean of 45 ng/g lw; 0.6 ng/g ww) and BB-101 concentration was 14 ng/g lw (0.2 ng/g ww). BDE-209 was found in 30% of the plasma samples, with concentrations ranging from 200 to 1100 ng/g lw, and a mean of 410 ng/g lw (5.7 ng/g ww). BDE-209 was the second congener of importance after BDE-47. BDE-209 concentrations were also determined in four pooled egg samples and ranged from 23 to 53 ng/g lw. HBCD and BB-101 were also detected in the egg samples with a mean of 142 ng/g lw (13.3 ng/g ww) and 2.8 ng/g lw (0.26 ng/g ww), respectively. BDE-209 has also been found in Svalbard glaucous gull liver, ranging from not detected to 10 ng/g ww (T. Savinova, cited in Verreault et al., 2004). Glaucous gulls at Bjørnøya winter in the north Atlantic Ocean and are not considered to be exposed to local sources. Therefore, Verreault et al. (2004) suggest that BDE-209 may have bioaccumulation potential in glaucous gulls.

In a recent study to quantify seabird guano inputs to Lake Ellasjøen on Bjørnøya, muscle and guano samples from kittiwakes, glaucous gulls and little auks (*Alle alle*) were analyzed for BDE-28, -47, -71, -77, -99, -100, -138, -153, -154 and -183 (Herzke et al., 2004). Only BDE-28, -47, -99, -100 and -153 were found and ∑PBDE concentrations in muscle were approximately 1400, 400 and 200 ng/g lw for glaucous gull, little auk and kittiwake, respectively. BDE-47 dominated the muscle and guano congener profiles.

PBDEs have been analyzed in herring gull (*Larus argentatus*) and great black-backed gull (*Larus marinus*) eggs from northern Norway (Pusch et al., 2005). Eggs were collected in 2001 from four sites: Alta, Kongsfjord (Finmark), Sommarøy and Vardø. Twenty eggs from each site were pooled into one sample. Concentrations of PBDEs (BDE-47, -99) were similar between regions, although levels were somewhat lower in Alta. ∑PBDE levels were approximately 500–700 ng/g lw (41–55 ng/g ww) (Pusch et al., 2005). PBDE levels in these seabirds are slightly higher than measured in Bjørnøya glaucous gulls (Burkow et al., 2001; Herzke et al., 2003).

BDE-47 dominates the PBDE congener pattern in most seabirds. Highest ∑PBDE levels are found in glaucous gulls. BDE-209, HBCD and PBBs are also present in glaucous gulls, indicating that these are present in the Arctic marine food web. The high BDE-209 levels found in these gulls indicates that it may have bioaccumulation potential. ∑PBDE levels in black guillemots from East Greenland are higher compared to West and southwest Greenland, and there is a tendency to higher ∑PBDE levels in seabirds in general around Svalbard and northern Norway compared to Canada.

1.4.6. Marine mammals

PBDE data are available for seals and whales from the Canadian and European Arctic. In general, concentrations of PBDEs are orders of magnitude less than legacy OCs such as PCBs and DDTs. However, PBDEs consist of a much smaller number of individual compounds, so the difference between individual PBDE and individual PCB congeners is less. BDE-47 is the most common congener measured, followed by BDE congeners -99 and -153. Other BDE congeners, such as -100 and -49 have been measured in Canadian Artic beluga (*Delphinapterus leucas*) (Law et al., 2003), but others, such as BDE-85 and -138, have been reported as non-detectable in Svalbard beluga and Faroe Island pilot whales (*Globicephala melas*) (van Bavel et al., 2001). These congeners are found at lower concentrations in the technical PBDE products, but may also be less prevalent due to biotransformation.

1.5. Seals

Sellström et al. (1993) found BDE-47, -99 and -100 in ringed seals from Svalbard collected in 1981. The ∑PBDE concentration in blubber was 51 ng/g lw (45 ng/g ww).

In more recent studies, highest blubber ∑PBDE concentrations (sum of 42 congeners) are found in ringed seals from northeastern Greenland where Johansen et al. (2004) reported mean concentrations of 58 ng/g lw. Vorkamp et al. (2004a), reported means of 36 and 35 ng/g lw in blubber of adults and subadults, respectively, for ringed seal from Ittoqqortoormiit, East Greenland (BDE-28, -47, -49, -66, -99, -100, -153, -154). On Svalbard, Wolkers et al. (2004) reported means of 18 ng/g lw for the sum of BDE-47, -66, -99, and -100, while Jenssen et al. (2004) found ∑PBDE concentrations (BDE-28, -47, -99, -100, -153, -154) of 15–35 ng/g lw and HBCD concentrations of 50–100 ng/g lw. Lowest ∑PBDE concentrations are found in ringed seals from western Greenland (4 ng/g lw; 3.6 ng/g ww) (Johansen et al., 2004), the eastern (1.3 ng/g lw; Di-HxBDEs) (Muir et al., 2004) and the western Canadian Arctic (5 ng/g lw; 4.6 ng/g ww, BDE-15, -28, -47, -49, -66, -75, -99, -100, -119, -153, -154, -155) (Ikonomou et al., 2002).

Muir et al. (2004) reported PBDEs and HBCD in ringed seals from six locations in the Canadian Arctic. Mean concentrations of ∑PBDEs ranged from 4.2 to 30 ng/g lw with highest concentrations in seals from Hudson Bay and lowest in the western Canadian Arctic. In contrast, mean HBCD concentrations ranged from 1.3 to 4.7 ng/g lw and were highest in seals from the western Canadian Arctic. Intermediate ∑PBDE concentrations (BDE-47, -99, -100, -153, -154) were found in ringed seals from the Russian Arctic (White Sea, Barents Sea, Kara Sea), with levels of 7.6, 10.3 and 11.2 ng/g lw, respectively for the three sites (Savinova et al., 2004). The spatial trend for ∑PBDEs in ringed seals is shown in Fig. 5. BDE-209 was included in the analysis but was not detected above blank levels (0.16–0.24 ng/g ww) in the western Canadian ringed seals (Ikonomou et al., 2002). Samples from Greenland were collected in 2000 and 2001, from Svalbard (Wolkers et al., 2004) in 1999, from the eastern Canadian Arctic in 2000 and in the western Canadian Arctic in 1993. For East Greenland ringed seals, ∑PCB concentrations were 22–36 times higher than ∑PBDEs. ∑PCB concentrations were 80–140 times higher in the Russian ringed seals.

Samples of blubber from ringed, larga (*Phoca largha*) and bearded seals (*Erignatus barbatus*) were collected in the Russian Arctic at Chukotka (Lavrentiya), in 2000–2001 (AMAP, 2004). BDE-47 concentrations were 0.51, 1.9 and 0.24–1.2 ng/g ww (no information on lipid content available) in ringed, bearded and larga seals, respectively. The BDE-47 concentrations in Russian ringed seals are comparable or lower than those seen in ringed seal from the eastern Canadian Arctic, and much lower than those seen in western Arctic Russia. ∑PCB concentrations were 20–500 times higher in the three seal species.

1.6. Whales

Concentrations of ∑PBDE were 29–161 ng/g lw (BDE-47, -66, -99, -100, -154) in Svalbard beluga blubber from

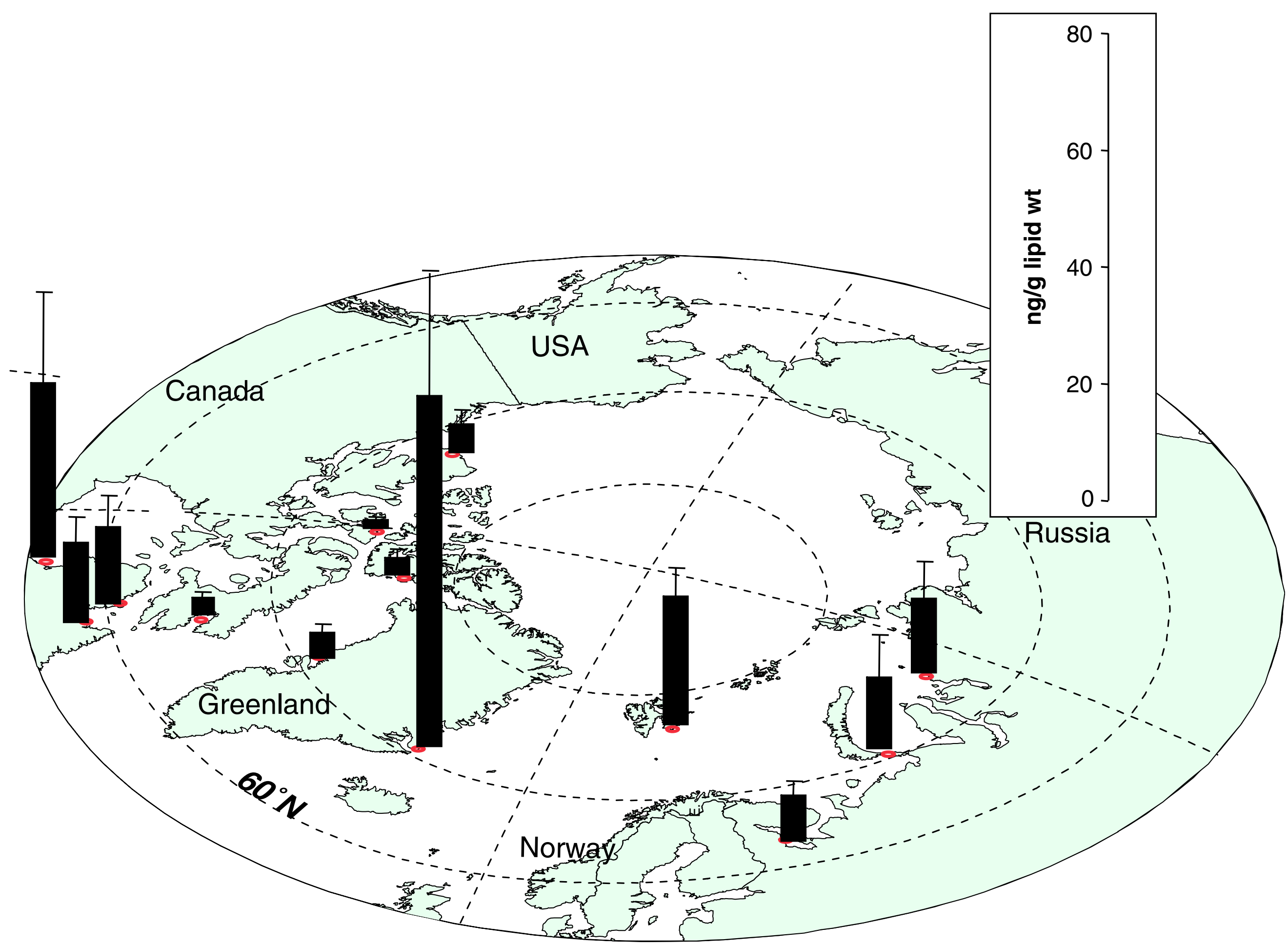

Fig. 5. ∑PBDE concentrations in ringed seal blubber (ng/g lw). Results from Muir et al. (2003, 2004), Johansen et al. (2004), Savinova et al. (2004) and Wolkers et al. (2004).

adults collected in 1998 (Wolkers et al., 2004). These were higher, compared to concentrations of approximately 17 ng/g lw (15.5 ng/g ww; BDE-47, -49, -99, -100, -153, -154) in beluga from the western Canadian Arctic collected in 2001 (Law et al., 2003), 15.6 ng/g lw (14.6 ng/g ww, Tri-HxBDEs, collected in 1997) (Law et al., 2003) in beluga from Cumberland Sound, Baffin Island, and 30–31 ng/g lw in beluga from eastern Hudson Bay and Hudson Strait, respectively, in the eastern Canadian Arctic collected in 2001 (Muir et al., 2004). Svalbard juveniles and calves had even higher ∑PBDE concentrations (174–314 ng/g lw) than adults. The Svalbard belugas also had detectable concentrations of three MeO-TeBDE metabolites, one of which had levels comparable to BDE-47 (Wolkers et al., 2004). HBCD was also detected in eastern Canadian arctic belugas, averaging 9.8 and 18 ng/g lw in Hudson Bay and Hudson Strait animals, respectively (Muir et al., 2004).

Harbor porpoises (*Phocoena phocoena*) collected in 1997–2001 from Icelandic waters had ∑PBDEs (BDE-47, -49, -99, -100, -119, -153, -154) of 25–95 ng/g lw (Thron et al., 2004). Minke whales (*Balaenoptera acutorostrata*) from the Barents Sea had ∑PBDE (BDE-47, -99, -153) levels of 32–44 ng/g lw (0.15–0.45 ng/g ww) in muscle, while minke whales from the Norwegian Sea had higher levels of 400–440 ng/g lw (3.1–15 ng/g ww) in muscle (Herzke, 2002b).

The highest concentrations of PBDEs measured in Arctic marine mammals are those observed in Faroe Island long-finned pilot whales (61–3200 ng/g lw) (Lindström et al., 1999; van Bavel et al., 2001). Concentrations (sum of all Te-, Pe- and HxBDEs, approximately 13 congeners) are an order of magnitude greater than in any other Arctic marine mammal examined to date. The major congeners found are BDE-47, -99 and -153. Due to a general lack of PBDE data in the Arctic, it is difficult to determine if these high levels are due to spatial trends or the behavior of the pilot whales (i.e. trophic level). Additionally, as in beluga whales from Svalbard (van Bavel et al., 2001), surprisingly high levels of three MeO-TeBDEs, almost comparable to BDE-47 levels, were found in the pilot whale. Results for long-finned pilot whales from Torshavn, Faroe Islands also revealed large differences between PBDE concentrations with age and sex (van Bavel et al., 1999). The highest ∑PBDE levels were found in juveniles (402–1246 ng/g lw), with somewhat lower values in adult males (397–669 ng/g lw) and the lowest values in adult females (126–326 ng/g lw) (van Bavel et al., 1999). This indicates transfer of the PBDEs from mother to calf by lactation. Results from a mother and fetus also indicated the presence of a partial transplacental barrier, as the level in the fetus was generally half the concentration of the mother (van Bavel et al., 2001).

Temporal trend studies of ∑PBDEs in Canadian Arctic beluga (1982–1997) (Law et al., 2003; Muir et al., 2004) and ringed seals (1981–2000) (Ikonomou et al., 2002), have shown rapidly increasing concentrations of these compounds (Fig. 4b and c). Law et al. (2003) summarized temporal trends in beluga from the eastern Canadian Arctic (southeastern Baffin Island) and the southern Beaufort Sea (western Canadian Arctic). In the southeast Baffin population, ∑PBDEs increased 6.8-fold in male beluga from 1982 to 1997. More recent analyses of eastern Canadian Arctic belugas collected from Hudson Strait in 2001 (Muir et al., 2004) show higher levels than those reported by Law et al. (2003) suggesting that the increase is continuing (Fig. 4b). Law et al. (2003) reported a 1.3-fold increase in ∑PBDEs in male beluga blubber from the western Canadian Arctic between 1989 and 2001. No significant differences were found between results for 1995 and 2001. For ringed seals from the western Canadian Arctic, a 9-fold increase was found between 1981 and 2000. These trends follow the estimated annual global production of Penta-BDE (Ikonomou et al., 2002).

The trends of ∑PBDEs in seals and beluga in the western Canadian Arctic differ during the 1990s. However, the ringed seals are resident species while the beluga are from the southern Beaufort Sea stock which annually migrates to the Bering/Chukchi Sea and may thus reflect exposure from the Pacific Ocean. Kajiwara et al. (2004) observed a 50% decrease in ∑PBDEs in Pacific fur seals from Japan between 1990 and 1998 coinciding with declining use of PentaPBDE in Japan. Thus, it is possible that western Canadian Arctic beluga are reflecting this decline in emissions to the north Pacific Ocean.

1.7. Polar bears

The polar bear is the apex predator of the Arctic marine ecosystem and has a circumpolar distribution. Levels of recalcitrant organochlorines (OCs) in polar bears are among the highest observed in any Arctic species and are highest in the Kara Sea, Svalbard (Andersen et al., 2001) and East Greenland populations (Norstrom et al., 1998). Mean ∑PBDE concentrations in polar bears from Svalbard were 14–144 ng/g lw (17.5 ng/g ww) in one study (van Bavel et al., 2001) and 13–70 ng/g lw in a more recent study (Wolkers et al., 2004). The congener pattern was dominated by BDE-47 and one MeO-TeBDE (van Bavel et al., 2001). However, a major congener, BDE-153, was not determined in these studies. In a recent food web study from Svalbard, polar bears were found to have ∑PBDE (BDE-28, -47, -99, -100, -153, -154) concentrations of 5–15 ng/g lw and HBCD concentrations of 15–30 ng/g lw (Jenssen et al., 2004).

In a spatial study, adipose tissue samples were collected from polar bears by subsistence hunters in the Ittoqqortoormiit/Scoresby Sound area in central East Greenland and in six locations in the Canadian arctic (Amundsen Gulf, Lancaster Sound, Foxe Basin/Boothia Peninsula, Northeast- and Southeast Baffin Island, Western Hudson Bay) (Muir et al., 2006). Fat biopsy samples were collected

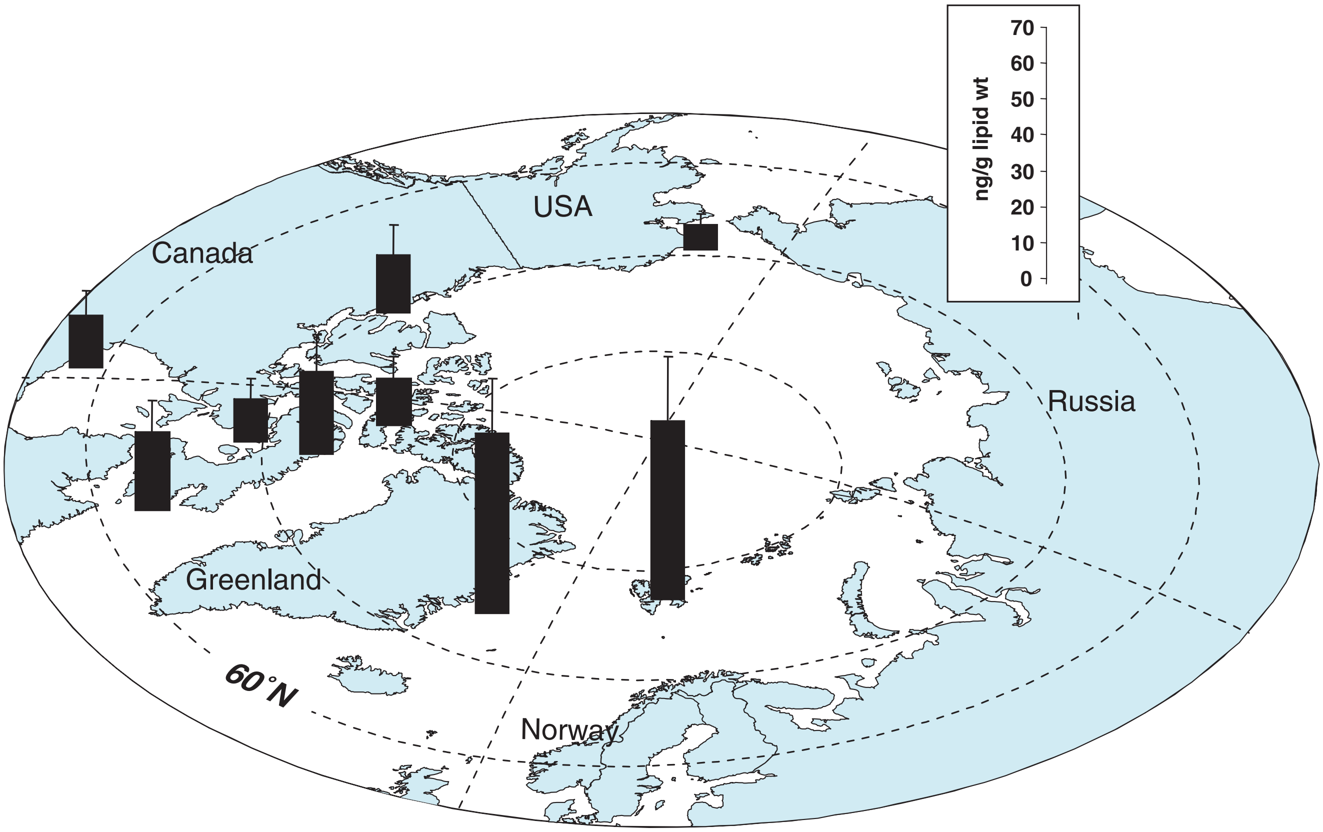

Fig. 6. Geometric mean concentrations of ∑PBDEs in fat of female polar bears (ng/g lw) from nine locations (Muir et al., 2006).

in Svalbard (Norway) as well as in western Beaufort Sea/Chukchi/Bering Sea regions of Alaska. Except for Alaska, samples from female bears were selected to reduce age effects. Both females ($n = 44$) and males ($n = 47$) from East Greenland were analyzed to examine effects of sex and collection season. PBDEs were isolated along with other organochlorines and quantified by GC–ECNI–MS. PBDEs were detected in all 204 fat samples with BDE-47, -99 and -153 predominating. Highest mean concentrations of ∑PBDEs were found in Svalbard, East Greenland and southeast Baffin polar bears (22–70 ng/g lw) (Fig. 6). Lowest ∑PBDE concentrations were found in the samples from northwest Alaska, Foxe Basin and Lancaster Sound (7–13 ng/g lw). Interestingly, BDE-153 concentrations were comparable to those of BDE-47.

HBCD was also detected and was present at similar concentrations to BDE-47. The highest HBCD levels (30–110 ng/g lw) were found in polar bears from East Greenland and Svalbard and lowest levels were found in the Bering/Chukchi population (<0.01–35 ng/g lw) (Gabrielsen et al., 2004; Muir et al., 2006). The geographic trend for PBDE and HBCD concentrations parallels that of PCBs and perfluorinated acids and points to eastern North America and western Europe as source regions. BB-153, was also found in polar bear samples at concentrations comparable to or higher than BDE-47 (Muir et al., 2006). Recently, BDE-209 has also been detected in adipose tissue (approximately 1 ng/g lw) from Svalbard polar bear samples collected in 2001–2003 (SFT, 2004; Skaare, 2004, personal communication).

When comparing PBDE as well as HBCD concentrations with age in Greenland polar bears, no trends were seen (Muir et al., 2006). In contrast, for PCBs, concentrations increase with age in males (Bernhoft et al., 1997). This indicates that the bears are in steady state with respect to BFRs and implies much more rapid elimination compared to PCBs. A similar lack of age trends for PBDEs has been seen in humans (Thomsen et al., 2002).

∑PBDE concentrations in polar bears are more than 100 times lower than ∑PCB levels. This, and the presence of MeO-TeBDEs, which may be metabolites, indicates that polar bears can metabolize PBDEs, and therefore, the concentrations of parent compounds may not give an accurate picture of total exposure.

1.7.1. Humans

There is currently only a limited amount of information on BFRs in humans in the Arctic, however, given the wide use of marine mammals in the traditional diet of the Inuit of eastern Siberia, North America and Greenland, as well as the population on the Faroe Islands, their presence in humans is expected. Median ∑PBDE (Tri-HpBDEs) levels in human milk from Arctic Quebec were 6.2 ng/g lw in 1996–2000, and BDE-47 was the predominant congener (Pereg et al., 2003). This is a 3-fold increase in concentrations since 1989–1991, when concentrations were 2.2 ng/g lw. However, higher ∑PBDE concentrations were found in milk of southern Canadians (22 ng/g lw; Ryan, 2004). Human milk samples from the Faroe Islands collected in 1998–1999 had ∑PBDE (BDE-47, -99, -100, -153) concentrations of 7.2 ng/g lw, with BDE-153 as the predominant congener (Fängström et al., 2004). When compared to samples collected in 1987, 1994–1995 and 1998–1999, PBDE concentrations increase from 1.5 ng/g lw to 7.2 ng/g lw, a similar increase as seen in the Arctic Quebec samples (Fängström et al., 2004).

Plasma samples were collected from adults of indigenous populations in 2000–2001 from four regions of the Russian Arctic: Chukotka (Kanchalan and Uelen); Kola Peninsula (Krasnoshchelie and Lovozero); Pechora Basin (Nelmin-Nos); and, Taymir Peninsula (Khatanga), in 2000–2001 (AMAP, 2004). Geometric mean concentrations of ∑PBDEs (BDE-28, -47, -99, -100, -153, -154, -183) ranged from 0.12 to 0.93 ng/g lw. The highest concentrations were found in the Kola Peninsula samples, and lowest levels in the Taymir Peninsula samples. ∑PCB concentrations were 400–7000 times higher. Samples of breast milk were also collected from two districts in the Chukchi Autonomous Oblast (Chukotsky and Anadyrsky) on the Chukotka Peninsula (AMAP, 2004). Mean ∑PBDE concentrations (BDE-28, -47, -99, -100, -153, -154, -183) were 0.31 at Chukotsky and 0.11 ng/g lw at Anadyrsky, with BDE-47 being the predominant congener.

∑PBDE levels are low in humans, and are lower than seen in more southerly latitudes, unlike the situation for many organochlorines (Hansen et al., 1998).

1.8. Spatial trends

Latitudinal gradients of PBDEs have been seen in air (Jaward et al., 2004b), soil (Hassanin et al., 2004), sediments (BDE-209) (Muir et al., 2003) and frogs (ter Schure et al., 2002). Generally, these show that PBDE concentrations or fluxes decline going northwards. In some cases, increasing proportions of lower brominated BDEs such as BDE-47, and decreasing proportions of higher brominated BDEs, such as BDE-153, are seen going northwards. There are few data for circumpolar spatial trends but there are indications from data in seabirds (PBDEs) (Johansen et al., 2004; Vorkamp et al., 2004a,b), beluga (PBDEs) (Law et al., 2003; Muir et al., 2004; Wolkers et al., 2004), ringed seals (PBDEs) (Fig. 5) and polar bears (PBDEs, HBCD) (Fig. 6) of higher concentrations on E. Greenland and Svalbard. These spatial trends are similar to those that have been found for PCBs. Although it is premature to draw conclusions about spatial trends of PBDEs and HBCD in Arctic marine mammals, there are sufficient data to suggest that concentrations are higher in the European Arctic as compared with the North American Arctic.

Table 2
Comparison of biomagnification factors for marine mammals from the Baltic Sea, North Sea and Arctic on a lipid weight/lipid weight basis

	BDE-47	BDE-99	BDE-100	BDE-153
Baltic grey seal/herring (1)	19	4.3	6.8	NA
Baltic harbor seal/herring (2)	33	33	8.9	110
North Sea harbor porpoise/herring (2)	23	34	26	165
Ringed seal/polar cod (3)	8	6.6	1.3	ND
Beluga (males)/polar cod (3)	33	30	23	ND
Polar bear/ringed seal (3)	1.6	–	–	ND
Polar bear/ringed seal (4)	3.9	5.8	4.7	71

Data from: (1) Sellström et al. (1993); (2) Boon et al. (2002); (3) Wolkers et al. (2004); (4) Muir et al. (2006). NA means not analyzed, ND means not detected.

1.9. Bioaccumulation/biomagnification

Biomagnification of lower brominated PBDEs is seen going from polar cod to ringed seal and polar cod to beluga (Table 2) but little biomagnification was seen in polar bears in the study of Wolkers et al. (2004). The congener-specific BMFs for beluga and ringed seal are comparable to those found for other marine mammals from the Baltic and North Seas (Table 2). In a recent food web study of zooplankton to polar cod to ringed seal to polar bear from Svalbard, both PBDE and HBCD concentrations were found to increase with increasing trophic level, with the notable exception of polar bears, which seem to be able to metabolize some of the compounds (Jenssen et al., 2004). BDE-153 was the most persistent BDE congener in that study. In the study by Muir et al. (2006), the mean BMF for BDE-153 was 10 times higher than that for CB-153. The BMF for BDE-47 is lower than for other marine mammals, the BMFs for BDE-99 and -100 are lower or comparable but the BMF for BDE-153 is similar to that of harbor seal and harbor porpoise (Table 2). A possible explanation for the high BMFs for BDE-153 could be that metabolic debromination of higher brominated BDEs is occurring. For example, metabolic debromination has been shown to occur in fish (Tomy et al., 2004; Stapleton et al., 2004a,b).

2. Conclusions

2.1. PBDEs

The tetra-, penta- and hexaBDEs in the PentaBDE and OctaBDE technical products are ubiquitous in Arctic samples. When analyzed for, the heptaBDE (BDE-183) in the OctaBDE product is often found as well. For air samples, Di-TriBDEs are also present. ∑PBDE concentrations in air samples indicate that trash burning may be a significant local source at some sites. A spatial trend in shorthorn sculpins in western Greenland was related to community population size indicating diffuse sources of PBDEs to the marine environment related to human activities.

Lowest PBDE concentrations are observed in the terrestrial environment, except for predatory birds. Highest PBDE concentrations are generally found in terrestrial predatory birds and in marine seabirds and mammals, all animals at high trophic levels. There are a few exceptions such as the high PBDE concentrations seen in sediments at Polyarnyy, a closed city with a navy base, which are more similar to levels seen at impacted sites in more southerly latitudes and in landlocked Arctic char from Lake Ellasjøen on Bjørnøya due to input from seabird guano from colonies on a nearby cliff.

Temporal trends in both biota and in sediments all show similar increases in $\sum$PBDEs with time.

PBDEs are present in human milk from Nunavik (Arctic Québec), the Faroe Islands and several sites in northern Russia. Temporal trends show rapidly increasing levels with time. However, concentrations of PBDEs in human milk in Nunavik were lower than in human milk samples from residents of southern Canada. This is unlike the trend seen for PCBs, which are higher in Nunavik than southern Canada. This indicates that the traditional diet may not be as significant a pathway for PBDE exposure as it is for PCBs.

$\sum$PBDE concentrations in all sample types from the Arctic are one or more orders of magnitude lower than $\sum$PCB concentrations except for some results for air. PBDE concentrations in Arctic samples are also lower than in similar sample types from more southerly regions. There are large regional variations in concentrations in top predators with highest PBDE and HBCD concentrations in biota from the European arctic (East Greenland to Barents Sea). This has occurred despite the much higher use of Penta-BDE in North America compared to Europe. This implies that some combination of atmospheric and oceanic transport from western Europe and eastern North America must be important pathways for delivering these chemicals to Arctic marine food webs.

MeO-TeBDEs have been found in polar and Arctic cod, beluga, pilot whale and polar bear. The origins of these substances is as yet unclear as they may be metabolites, anthropogenic contaminants or naturally occurring products.

2.2. Other BFRs

While the tetra-, penta- and hexabrominated BDEs are ubiquitous in the Arctic, the presence of BDE-209 in remote Arctic lake sediments, marine sediments, blue mussels, Atlantic cod, moss, several species of birds of prey, grouse, moose, lynx, glaucous gulls and polar bears indicates that even this congener is subject to long-range transport. Data for HBCD showing its presence in air and deposition samples, mussels, Atlantic and polar cod, peregrine falcons, ringed seals and polar bear, also indicates long-range transport. Where isomer-specific analysis has been performed, the α-HBCD isomer dominates. Very few data are presently available for TBBPA. It has been found in air (filter), moss, in marine sediments, in Atlantic cod liver and in Norwegian peregrine falcon and golden eagle eggs. It has also been found as the dimethylated metabolite at relatively high levels in peregrine falcons from Greenland. In principal, TBBPA may also be transported by the same pathways as the Di-DeBDEs, but more data is needed to confirm this. PBBs have been found in moss, seabirds, birds of prey and polar bears, indicating long-range transport.

The lower brominated BDEs have been shown to bioaccumulate and biomagnify, with BDE-153 showing a particularly high biomagnification factor in polar bears. BDE-153 dominates the congener pattern in peregrine falcons, golden eagle, gyrfalcon, merlin, lynx and is also a predominant congener in glaucous gulls, polar bears and human milk from the Faroe Islands. In most other biota types, BDE-47 and/or BDE-99 dominate, whereas in sediments, BDE-209 dominates (when included in the analyses). In the only marine sediment where several BFRs were analyzed (Tromsø, Norway), TBBPA concentrations were highest, followed by BDE-209 and then the $\sum$PBDEs.

There are substantial data gaps for BFRs in the Arctic. More data in all types of matrices are needed for the more poorly studied BFRs: BDE-209, HBCD and TBBPA. No information is available for temporal trends of PBDEs in the European Arctic (except for Greenland) or for temporal trends of other BFRs in any part of the Arctic. Very few data are available for BFRs in the Russian Arctic. More data are needed on biomagnification of individual BDE congeners (including BDE-209) as well as for isomer-specific HBCD, TBBPA and the PBBs. The air and harbor sediment results for PBDEs indicate that there are local sources near highly populated areas within the Arctic and further work is needed to evaluate the influence of such local sources on contaminant levels. The presence of some high values also underscores the need to collect samples using clean techniques that avoid contamination from BFRs in consumer products if results are to be accepted as evidence for presence in remote environments.

2.3. Are BFRs POPs?

Based on the accumulating evidence of their presence in the Arctic and indications of long-range transport for several BFRs, we conclude that Penta-, Octa- and DecaBDE, PBBs and HBCD have characteristics that qualify them as POPs according to the Stockholm Convention. There are indications that TBBPA may also behave as a POP but there are too few data as yet to reach firm conclusions and more research is needed to fill these data gaps.

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