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Brominated flame retardants in the Arctic environment — trends and new candidates ☆

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) containing two to 10 bromines are ubiquitous in the Arctic, in both abiotic and biotic samples. Hexabromocyclododecane (HBCD) is also ubiquitous in the Arctic, with the γ-HBCD isomer predominating in air, the α-HBCD isomer predominating in biota and similar concentrations of α-, β- and γ-HBCD found in marine sediments. Other brominated flame retardants (BFRs) found in some Arctic samples are polybrominated biphenyls (PBBs), tetrabromobisphenol A (TBBPA), 1,2-bis (2,4,6-tribromophenoxy)ethane (BTBPE), hexabromobenzene (HxBBz), pentabromoethylbenzene (PBEB), pentabromotoluene (PBT), and 1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane (TBECH). Temporal trends of tetra- to heptaBDEs and HBCD show increasing concentrations or a tendency to levelling off depending on the matrix (air, sediment, biota) and location, but no uniform picture for the Arctic emerges. BDE-209 concentrations are increasing in air. PBDEs and HBCD spatial trends in seabirds and marine mammals are similar to those seen previously for polychlorinated biphenyls (PCBs), with highest concentrations found in organisms from East Greenland and Svalbard. These trends indicate western Europe and eastern North America as important source regions of these compounds via long range atmospheric transport and ocean currents. Latitudinal trends showed lower concentrations and fluxes of PBDEs at higher latitudes. The tetra-hexaBDEs and α-HBCD biomagnify in Arctic food webs. Results for BDE-209 are more conflicting, showing either only low or no biomagnification potential. PBDE and HBCD concentrations are lower in terrestrial organisms and higher in marine top predators such as some killer whale populations in Alaska and glaucous gulls from the Barents Sea area. Higher concentrations are seen near populated areas indicating local sources. Findings of BTBPE, HxBBz, PBEB, PBT and TBECH in seabirds and/or marine mammals indicate that these compounds reach the Arctic, most probably by long range atmospheric transport and accumulate in higher trophic level organisms and that increasing use as PBDE replacements will lead to increasing concentrations.

1. Introduction

In a previous review of this topic, results from studies up to mid-2005 showed that several brominated flame retardants (BFRs) were reaching the Arctic (de Wit et al., 2006). The review covered the polybrominated diphenyl ethers (PBDEs), including the fully brominated BDE-209 in the DecaBDE product, hexabromocyclododecane (HBCD) and tetrabromobisphenol A (TBBPA). PBDEs with two to seven bromines were found to be ubiquitous in Arctic biota, from zooplankton to polar bears and humans, as well as in abiotic samples such as air, soil and sediments. HBCD, polybrominated biphenyls (PBBs) and TBBPA were also found when they were included in analyses, but there were fewer data available on these BFRs.

Generally, spatial trends of PBDEs and HBCD in top predators such as polar bears were similar to those seen for polychlorinated biphenyls (PCBs) indicating western Europe and eastern North America as source regions. Temporal trends indicated increasing concentrations of tetra-decaBDEs in several Arctic species and the tetra-hexaBDEs were found to biomagnify. The concentrations of the various BFRs were generally lower in the Arctic compared to more southerly regions and were lower than concentrations of legacy chemicals such as PCBs. The results indicated that most of these BFRs undergo long range transport to the Arctic.

The Stockholm Convention on Persistent Organic Pollutants (POPs), which went into force in May, 2004, states that chemicals may qualify as POPs if they are found far from sources and show evidence of long range transport. This has made the Arctic an important indicator region for assessing persistence and bioaccumulation properties of POPs and for identifying potential new POPs. Based on the previous review, several of the BFRs were considered to have characteristics that qualify them as POPs according to the Stockholm Convention.

☆ This paper is a contribution to the AMAP POPs assessment.

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As part of the Arctic Monitoring and Assessment Programme's (AMAP) ongoing work with updates on POPs in the Arctic, we update the previous review of this topic and also consider new candidate BFRs that may have potential for long range transport to the Arctic. The review also includes studies reporting the presence of hydroxy- and methoxy-PBDEs in Arctic wildlife, which may be metabolites and/or naturally-occuring compounds. This review is based on new data from peer-reviewed publications, in a few cases from extended abstracts from conferences and workshops as well as from published government scientific reports. The extended abstracts come mainly from the annual International Symposium on Halogenated Persistent Organic Pollutants (Dioxin 2006, Dioxin 2007, Dioxin 2008, published as Organohalogen Compounds) and the 4th International Workshop on Brominated Flame Retardants (BFR 2007). The scientific reports present data from specific national surveys or national monitoring programs within AMAP, where defined quality assurance and quality control criteria have been followed.

2. Brominated flame retardants (BFRs)

Brominated flame retardants (BFRs) are chemicals used in materials to make them more fire-resistant. Typical uses are in polyurethane foam, plastics used in electric and electronic equipment, printed circuit boards, expanded and extruded plastic (such as Styrofoam), textile back-coating in furniture, various textiles used in public environments (curtains, furniture coverings, carpets), rubber for coating wire, etc. Many countries have legislated high fire safety standards, which has led to an increase in the use of flame retardants.

Most brominated flame retardants, such as PBDEs, HBCD and PBBs, are additives that are mixed directly into the product during manufacture. Because they do not react with the material, they may migrate out of the product and be released into the environment. Some flame retardants, such as TBBPA, are reactive and are chemically bound to the material and thus do not migrate out of the material. However, polymerization may not be complete and the unreacted BFR can then migrate out of the material. Table 1 gives the estimated annual market demand for the major BFRs for the years 2001–2003. Physicochemical properties of PBDEs and HBCD have been studied to some extent (Wania and Dugani, 2003; Hayward et al., 2006) but little such data is available for other BFRs. In general, most BFRs are hydrophobic and accumulate in organic material such as lipids.

The toxicity of PBDEs, PBBs, HBCD and TBBPA has been reviewed (de Wit, 2002; Darnerud, 2003, 2008; Birnbaum and Staskal, 2004). For some of the PBDEs, neurotoxic effects (Eriksson et al., 2001), effects on the thyroid hormone system (Fowles et al., 1994; Hallgren and Darnerud, 2002) and effects on sex hormones and reproduction (reviewed in Darnerud, 2008) have been shown in rodents. There are indications that HBCD exposure can affect the liver and thyroid hormone system (van der Ven et al., 2006), and might cause neurobehavioral alterations (Eriksson et al., 2006). TBBPA has also been shown to have effects on thyroid hormones, neurological function and reproduction (Lilienthal et al., 2008; van der Ven et al., 2008). A more detailed discussion of the toxicology of BFRs can be found in Letcher et al. (2010-this issue).

Table 1
Estimated annual worldwide market demand of BFRs in 2001 by region, and total estimated demand in 2002 and 2003 (metric tons) (BSEF, 2006).

	PentaBDE	OctaBDE	DecaBDE	TBBPA	HBCD
Americas [a]	7100	1500	24,500	18,000	2800
Europe	150	610	7600	11,600	9500
Asia	150	1500	23,000	89,400	3900
Total (2001)	7500	3790	56,100	119,700	16,700
Total (2002)	–	–	65,700	150,600	21,400
Total (2003)	–	–	56,400	145,100	22,000

[a] Americas includes North and South America, but North America, particularly the USA, is the major user.

2.1. PBDEs

There are three technical PBDE products that have been or are still in use as additive flame retardants, known as PentaBDE, OctaBDE and DecaBDE. PentaBDE contains primarily tetra- (BDE-47) (Fig. 1), penta- (BDE-99, -100) and hexaBDE (BDE-153, -154) congeners, OctaBDE contains primarily a heptaBDE (BDE-183) plus hexa- (BDE-153, -154) and octaBDEs and DecaBDE consists primarily of the fully brominated BDE-209 (La Guardia et al., 2006). PentaBDE has primarily been used in polyurethane foam (mattresses, furniture, pillows) and in adhesives. OctaBDE is used in hard plastics (acrylonitrile butadiene styrene-ABS) such as computer casings and computer monitors. Penta- and OctaBDE were banned in the European Union (EU) in 2004 as well as in Norway. Production of Penta- and OctaBDE was voluntarily discontinued in the USA at the end of 2005. PentaBDE and OctaBDE are also on the proposed list of new chemicals to be included in the Stockholm Convention and the POPs Protocol to the UN ECE Convention on Long-Range Transboundary Air Pollution.

DecaBDE is used in plastics such as high impact polystyrene (HIPS) in electrical and electronic equipment, such as the back covers of TVs, but also in rubber coating for wiring, as well as in textile back-coating in furniture. It is also widely used in the transportation, construction and building sectors. There are currently no restrictions on the production or use of DecaBDE in most countries. However, according to the EU Directive on the restriction of use of certain hazardous substances in electrical and electronic equipment (RoHS Directive), manufacturers must substitute PBBs and PBDEs in new equipment. DecaBDE was exempted from the directive, but this exemption was challenged in the EU courts and recently overturned (April 1, 2008). The use of DecaBDE in the EU has thus been banned since 1 July 2008 according to this ruling (European Court of Justice, 2008).

2.1.1. Hydroxy- and methoxy-PBDEs (OH-PBDE, MeO-PBDE)

Hydroxylated (OH-PBDE) and methoxylated PBDEs (MeO-PBDE) have been reported in blood from wildlife (Hakk and Letcher 2003). Laboratory studies support the formation of some OH-PBDEs from parent PBDEs via metabolism, whereas there are indications that others may be produced by algae and sponges as natural products. Thus, the presence of OH-PBDEs may be due to uptake and metabolism of PBDEs and/or the accumulation of naturally-occurring compounds (Hakk and Letcher, 2003; Malmberg et al., 2005; Malmvärn et al., 2005; Marsh et al., 2006). The MeO-PBDEs, such as 6-MeO-BDE-47 and 2'-MeO-BDE-68, have been shown to be of natural origin (Teuten et al., 2005).

To date, there is no evidence that metabolic formation of MeO-PBDEs occurs in animals exposed to environmentally relevant parent BDEs, and they are likely to be of natural origin. The structure of 6-MeO-BDE-47 is shown in Fig. 1.

2.2. HBCD

Technical HBCD consists primarily of γ-HBCD (more than 70%), but also contains α- and β-HBCD and some impurities (Law et al., 2005). HBCD is used as an additive flame retardant in expanded (EPS) and extruded polystyrene (XPS) foams, which are used as insulation in buildings and in roads to prevent frost-heaving. HBCD is also used in textile back-coating in furniture. There are no restrictions on the production or use of HBCD. A risk assessment of HBCD is currently being completed within the EU. HBCD has been proposed for inclusion in the Stockholm Convention but is only at the proposal stage and a risk profile will not be prepared until 2010. The structure of HBCD is shown in Fig. 1.

2.3. TBBPA

TBBPA is used primarily as a reactive flame retardant in printed circuit boards, and is thus found in electrical and electronic equipment such as TVs, computers, printers, fax machines, cell phones, videos,

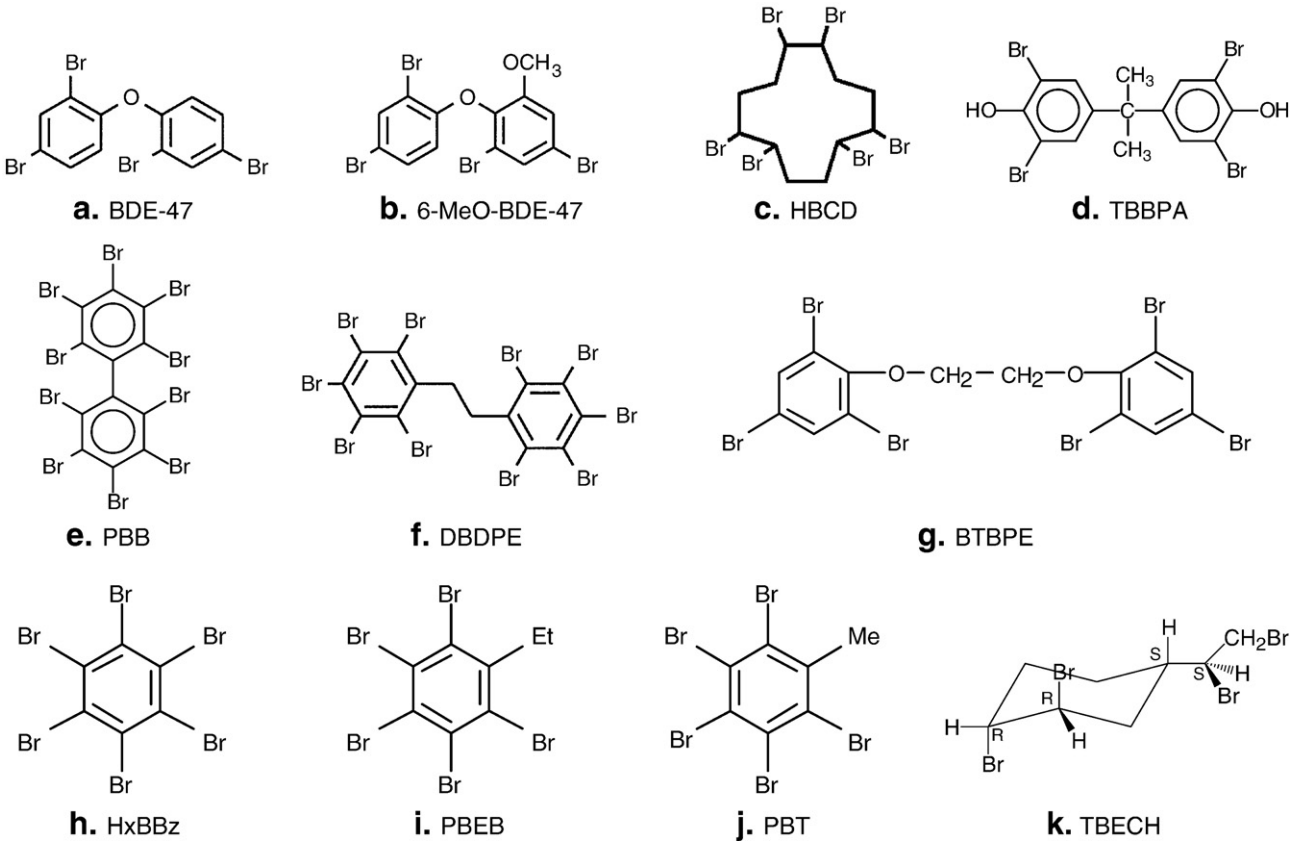

Fig. 1. The structure of a) 2,2′,4,4′-tetrabrominated diphenyl ether (BDE-47); b) a methoxylated PBDE (6-MeO-BDE-47); c) hexabromocyclododecane (HBCD); d) tetrabromobisphenol A (TBBPA); e) polybrominated biphenyls (PBB); f) decabromodiphenyl ethane (DBDPE); g) 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE); h) hexabromobenzene (HxBBz); i) pentabromoethylbenzene (PBEB); j) pentabromotoluene (PBT); and k) β-1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane (tetrabromoethylcyclohexane or TBECH).

washing machines etc. However, TBBPA is also increasingly being used as an additive flame retardant in ABS plastics, as a replacement for banned BFRs. There are no restrictions on the production or use of TBBPA. A risk assessment has been performed within the EU and was recently closed in June 2008. The assessment concluded that, generally, no health or environmental risks were identified when TBBPA is used reactively (European Union, 2008). However, there is a need for further information and testing in aquatic and terrestrial ecosystems to determine if TBBPA degrades to bisphenol A in sediments or forms dimethylated TBBPA. The dimethylated derivative of TBBPA (Me-TBBPA) may be formed by anaerobic bacteria in sediments (Allard et al., 1987). The risk assessment also concluded there is a need for specific measures to limit risks when TBBPA is used as an additive flame retardant. The structure of TBBPA is shown in Fig. 1.

2.4. Polybrominated biphenyls (PBB)

Several polybrominated biphenyl (PBBs) technical products consisting of primarily hexa-, octa- or decabromobiphenyls have been used as additive flame retardants. These were first used in the 1970s, but due to the accidental mixing of a HexaBB formulation into cattle feed and the subsequent widespread contamination of farm products in Michigan, US, in 1973, the hexaBB product was quickly banned. The octa- and decaBB formulations were in continued production until 1979. In Europe, production of decaBB in France ceased in 2000 (Alaee et al., 2003). The general structure of PBBs is shown in Fig. 1.

2.5. Decabromodiphenyl ethane (DBDPE)

Decabromodiphenyl ethane (DBDPE) is marketed as Saytex 8010, by Albemarle Corporation. It has been in use as an additive flame retardant since the early 1990s and has only recently been found in environmental samples (Kierkegaard et al., 2004). It is used in high impact polystyrene (HIPS) and in wire and cable coverings and is considered to be a suitable alternative to DecaBDE (Albemarle Corporation, 2008). The structure of DBDPE is shown in Fig. 1.

2.6. 1,2-Bis(2,4,6-tribromophenoxy)ethane (BTBPE)

BTBPE is marketed as FF-680 (Great Lakes Chemical Corporation) and is used as an additive replacement for OctaBDE (Hoh et al., 2005). It is marketed for use in HIPS, thermoplastics and thermoset resins. Worldwide production/usage was estimated to be 16 710 tons in 2001 (Verreault et al., 2007a). The structure of BTBPE is shown in Fig. 1.

2.7. Hexabromobenzene (HxBBz)

Japan has used HxBBz as an additive flame retardant to paper, plastic and electronic goods and it is still used in low volumes (350 tons in 2001) (Watanabe and Sakai, 2003). The structure of HxBBz is shown in Fig. 1.

2.8. Pentabromoethylbenzene (PBEB)

PBEB was a low production volume chemical (10–1000 tons) in 2002 according to the Oslo-Paris Commission (OSPAR) but is not currently produced or used by any OSPAR signatory states (Verreault et al., 2007a). PBEB is an additive flame retardant suitable for use in thermoset resins in circuit boards, textiles, adhesives, wire and cable coating, polyurethane and thermoplastic resins (US EPA, 1985). The structure of PBEB is shown in Fig. 1.

2.9. Pentabromotoluene (PBT)

PBT is a component of the Flammex 5-BT commercial formulation (Verreault et al., 2007a). PBT is used as an additive flame retardant in textiles, polyester resins and paint emulsions. Production volume information is not publicly available, but it is used and distributed in the US under the trade name FR-105. The structure of PBT is shown in Fig. 1.

2.10. 1,2-Dibromo-4-(1,2-dibromoethyl)cyclohexane (TBECH)

1,2-Dibromo-4-(1,2-dibromoethyl)cyclohexane, also known as tetrabromoethylcyclohexane or TBECH, is used primarily as an additive flame retardant in extruded polystyrene (XPS) but is also used in electrical cable coatings, high impact plastics, fabric adhesives and some types of construction material (Tomy et al., 2008a). TBECH is produced by Albemarle Corporation under the market name Saytex BCL-462. The technical grade TBECH consists of near equimolar amounts of two (of a possible four) diastereoisomers: rac-(1R,2R)-1,2-dibromo-(4 S)-4-((1 S)-1,2-dibromoethyl)cyclohexane (α-TBECH) and rac-(1R,2R)-1,2-dibromo-(4 S)-4-((1R)-1,2-dibromoethyl)cyclohexane (β-TBECH). The structure of β-TBECH is shown in Fig. 1.

3. Emission sources

An Arctic Council Action Plan (ACAP) project has recently inventoried BFR production and use in the eight Arctic countries (ACAP/AMAP, 2007). Seven countries (United States, Canada, Russia, Sweden, Finland, Denmark, Norway) provided information for the inventory.

3.1. Production of BFRs

According to the ACAP project, the US is the only Arctic country that is currently producing BFRs, with production of approximately 40,000 metric tons in 2001. Production is in states south of the Arctic. The US Toxic Release Inventory (TRI) reports releases to air for 2003 of 31 metric tons of DecaBDE and 33 metric tons of TBBPA from BFR production facilities. Russia has previously produced BFRs, approximately 200 metric tons from 2000–2004, primarily HBCD. BFR production has ceased since 2004 (ACAP/AMAP, 2007). Globally, BFRs are also produced in Israel, Jordan, China, Japan, Austria, Belgium, France, Germany, the Netherlands and the United Kingdom.

3.2. Production of BFR-containing products

All seven Arctic countries that took part in the inventory import BFR chemicals for use in the manufacturing of BFR-containing products, for example, in electronic and electrical equipment, textiles, furniture, and building and construction materials. Some of these products remain in the country, others are exported to other countries as finished products. In turn, Arctic countries import goods and products that contain BFRs from other countries. However, information on import and export of BFR-containing products as well as the BFR content in manufactured products is generally lacking. Due to confidentiality reasons, the US could not provide import/export data. Canada calculated that up to 6000 metric tons of DecaBDE and 173 metric tons of TBBPA annually could be imported in products from the US. Canada estimates that it exports 700 metric tons/year of BFRs in BFR-containing products to the US (ACAP/AMAP, 2007).

The production and use of Penta- and OctaBDE are banned in the EU and in Norway, and this includes the import and export of products containing these BFRs. Sweden reported limited use of DecaBDE and HBCD and thus limited import and export but TBBPA is used in the production of electronic and electrical equipment. This means that Sweden exports TBBPA-containing goods. Due to confidentiality reasons, there is no information available on import/export of BFR-containing products from Finland. For Russia, DecaBDE was the main BFR in imported products, up to 1100 metric tons, but small amounts of PentaBDE (20 metric tons) were imported in furniture and circuit boards. Russia also imported products containing OctaBDE, TBBPA and HBCD. For Norway, import was dominated by products containing TBBPA and export was dominated by products containing TBBPA and HBCD.

Using the data provided by the different Arctic countries and other available information, a rough budget of BFR consumption/accumulation was calculated for each country and these are presented in Table 2 (ACAP/AMAP, 2007). Although there are large uncertainties in many of the estimates given in Table 2, some cautious observations can be made. The Scandinavian countries and Russia have lower net usage of BFRs than Canada and the US. The US has the highest net usage of BFRs. If these are converted to a per capita basis, however, the consumption of BFRs in Canada (210 tons/year/million inhabitants) is similar to the US (330 t/y/million inhabitants) (ACAP/AMAP, 2007). Russia has the lowest per capita consumption of around 1 t/y/million inhabitants and the Scandinavian countries lie somewhere in between Russia and North America.

3.3. Releases from flame-retarded products

After manufacture, flame-retarded products release BFRs during their entire lifetime, particularly additive BFRs, which are not chemically bound to the material they are incorporated in. This means releases will occur during use, and as waste after disposal. Thus, although several BFRs have been banned or phased out, such as Penta- and OctaBDE, the long life-times of the products they are incorporated in will lead to their continued release from products and then from disposed waste for several decades to come.

Table 2
Budget (balance of imports and exports) of BFRs "accumulated" within the Arctic countries based on reported available information (for the year 2001, and the period 2001–2004 in the case of Russia).

Country	Production of BFR-containing products (t BFR/y)	Import of BFRs in products (t BFR/y)	Export of BFRs in products (t BFR/y)	Net-usage[a] (t BFR/y) production + import − export
Canada	1190	6180	700	6670
Denmark	90[b]	160[b]	65[b]	190+[b]
Finland	400[cd]	Not available	Not available	Not available
Norway	100	270	90	280
Russia	Not available	320[e]	170[e]	150+[e]
Sweden	250[b]	Not available	Not available	Not available
United States	43000	70000	9000	100000

(Note: the values represented in this table are typically the result of calculations involving gross estimation, and are therefore subject to considerable uncertainties. In cases where ranges were reported, mid-range values have been employed to calculate the balance). Data from ACAP/AMAP (2007).

[a] Net-usage can be considered to represent the amount of BFRs "accumulated" in the county that will ultimately enter the waste stream.
[b] Mainly TBBPA.
[c] Excluding TBBPA.
[d] Approximately 90% of reported HBCD use in Finland (400 t/y) is re-exported in products.
[e] Based on average for reported five-year period.

Some BFRs such as PBDEs and HBCD, have also been found in relatively high concentrations in sewage sludge. A recent study from two research stations in Antarctica is illustrative of the possibility of local release of BFRs from untreated sewage in Arctic communities. Hale et al. (2008) analyzed wastewater sludge and dust from living quarters from the U.S.-run McMurdo Research Base as well as from the New Zealand-run Scott Research Base. These bases are only 3 km apart. They also sampled sediments and biota in a gradient from the McMurdo sewage outfall. The results showed high PBDE concentrations in both dust and sludge samples from both bases, but with the US samples having higher concentrations than those from the New Zealand base, reflecting the PBDE use patterns in the respective countries. PBDE concentrations in fish and invertebrates near the McMurdo outfall were as high as seen in urbanized areas of North America and decreased with distance. These results indicate that similar releases of untreated wastewater sludge from Arctic communities could be a significant local emission source of BFRs to the aquatic environment.

4. Levels of BFRs in the Arctic

Concentrations of brominated flame retardants in various media are summarized in Table S1 (Supplementary data: PBDEs, HBCD, PBBs). A map with an overview of sampling sites discussed is given in Fig. 2.

4.1. Long-range transport in air

In the previous review (de Wit et al., 2006), results from Arctic air samples indicated that the di-heptaBDEs and HBCD were capable of undergoing long-range atmospheric transport. However, there were few data for TBBPA and none for BDE-209 in air, and these could not be evaluated for their long-range transport potential in air.

Wania and Dugani (2003) estimated the long range transport (LRT) potential of PBDEs using four different multimedia and fate models (TaPL3, ELPOS, Chemrange, Globo-POP) and compared these to PCBs. All four models gave similar results and indicated that the lower brominated BDEs (tetra-pentaBDEs) had comparable LRT potential as PCB congeners (hexaCBs) previously known to have significant LRT potential. For example, characteristic travel distances (CTDs) for BDE-47 were 1113 and 2483 km, and for BDE-209, 480 and 735 km. However, when compared to PCB congeners with similar physico-chemical characteristics (e.g. similar molar mass), the lower brominated BDEs had smaller CTDs/Arctic contamination potentials than the PCBs. Wania and Dugani (2003) hypothesize that this is due to the higher reactivity of the BDEs leading to more efficient atmospheric degradation via hydroxyl radical reactions. The LRT of higher brominated BDEs such as BDE-183 and -209 was postulated to be limited due to their low volatility, which leads to these compounds being primarily bound to particulates. This in turn leads to efficient

Fig. 2. Map of the Arctic with an overview of sampling sites discussed in this review.

removal from the atmosphere via deposition. The models indicated that the tri-pentaBDEs would have the highest LRT potential, comparable to PCBs. The higher brominated BDEs would not be subject to significant LRT and the process would be controlled by the transport of particulates (Wania and Dugani, 2003).

The long range transport potential and persistence of BDE-47 and -99 were modeled by the European Modelling and Evaulation Programme (EMEP) as supporting information in the review work of possible new substances to be added to the Convention on Long Range Transboundary Pollution POPs protocol (Vulykh et al., 2006). The model was a multicompartment hemispheric transport model used by the Meteorological Synthesizing Centre - East (MSC-E) in Moscow, Russia. The model estimates transport distances (TD) for contaminants, which are the average distances from the source at which the annual mean atmospheric concentration of a contaminant drops 1000 times compared to the concentration near the source. The calculations were based on transport from a conventional point source located in Europe (France) over one year of emissions. TDs for BDE-47 and -99 were 2300 km and 2800 km, respectively and their estimated atmospheric half-lives 7 and 11 days, respectively. The model predicts spread of both BDE-47 and -99 from the point source and into the Arctic, with BDE-99 having a somewhat higher long-range transport potential. The main removal processes for both congeners determined by the model were wet and dry deposition.

Breivik et al. (2006) examined the long range transport behavior of several BDEs including BDE-209 using results from dated sediments cores along a latitudinal transect in North America to measure empirical half distances (EHDs) (Breivik et al., 2006). They saw an exponential decline in BDE-209 flux with latitude based on the seven lakes studied and this translated into an EHD of 566 ± 101 km. BDE-47 flux also declined with latitude (although the trend was not statistically significant), which translated into an EHD (± SE) of 1168 ±942 km. EHDs for PCB congeners 138/163, 153, and 180 (range 846–947 km) based on the same lakes were significantly greater ($P = 0.10$) than those for BDE-209 based on paired *t*-tests of the slopes. These EHDs are similar to the predicted CTDs made by Wania and Dugani (2003). Breivik et al. (2006) then compared these EHD results to those determined for BDE-209 and other PBDEs using a multi-media transport and fate model (CoZMo-POP) to predict characteristic travel distances (CTDs) in order to understand the processes that control LRT of BDE-209. Calculations using the CoZMo-POP model overestimated the observed atmospheric mobility of BDE-209 relative to that of the PCBs by 1.5 to 8.5 times depending on the CB congener (Wania et al., 2006). This may be caused by processes of potential significance in limiting LRT that are so far not included in the CoZMo-POP model due to lack of quantitative understanding, notably direct or indirect photolytic degradation of BDE-209 sorbed to atmospheric particles, particle dry deposition and snow scavenging of particles, which could be more efficient than rain scavenging (Breivik et al., 2006). There also may be problems with how the models are parametrized. Based on analysis of the parameters in the CoZMo-POP model, Breivik et al. (2006) for example, found that episodes of strong winds and no precipitation will lead to higher than expected LRT of BDE-209, as BDE-209 removal from the atmosphere is driven primarily by wet deposition. Thus, existing LRT models, which include constant rainfall, fail to predict the long range transport potential of contaminants that are particle-bound in the atmosphere and have sufficiently long atmospheric half-lives, such as BDE-209.

Raff and Hites (2007) performed laboratory experiments to study the atmospheric degradation of PBDEs and found that hydroxyl radical degradation is only important for mono-diBDEs. Photolysis becomes the predominant atmospheric degradation pathway for tri-hexaBDEs when they are in the gas phase. BDE-209 is not subject to photolysis in the atmosphere as it is almost entirely particulate bound and is therefore shielded from sunlight, which in turn lengthens the lifetime of BDE-209 in the air. Using these results, Raff and Hites (2007) modeled the behavior of BDE-47, -99 and -209 and found that photolysis is the most important removal pathway for lower brominated BDEs in the gas phase such as BDE-47 and -99 in air, whereas deposition, particularly wet deposition is the most important removal pathway for BDE-209 in air. This explains the observed depletion of lower brominated BDEs in e.g., Great Lakes sediments compared to BDE-209. Thus, rain events and sunlight are important for predicting the atmospheric behavior of PBDEs. Since the amount and strength of sunlight declines and may be totally absent in some parts of the Arctic during winter, the lower brominated BDEs will be able to undergo LRT to a greater extent at that time.

4.2. Atmospheric environment

4.2.1. Air

The previous review presented data from passive and active (high volume) air sampling at several Arctic sites in Canada, Iceland, Norway (including Svalbard), Sweden and Russia (de Wit et al., 2006). The predominant BFRs studied were the PBDEs and data indicated di-heptaBDEs at concentrations ranging from 0.3–68 pg/m^3. HBCD was also found in air samples from northern Finland and TBBPA was found on only one filter of several air samples from Canada. BDE-209 was not included in any analyses so no data were available. PBDE concentrations were comparable to those found at background sites around the Great Lakes, with the exception of some very high concentrations (up to 424 pg/m^3) found near Arctic sites where local waste incineration was a suspected source.

Shen et al. (2006) reported PBDEs (BDE-47, -99, -100, -153, -154) in XAD-based passive samplers (PAS) deployed for 1 year at 7 locations in the Canadian Arctic in 2000–2001 as well as at numerous other sites in North and Central America. BDE-47 and -99 were the predominant congeners and were found in similar proportions as found in commercial PentaBDE products such as Bromkal 70-5DE and DE-71 (La Guardia et al., 2006; Sjödin et al., 1998). Preliminary data were reported in the previous review, but total PBDE (ΣPBDE) concentrations were now confirmed to range from 0.3 to 68 pg/m^3 in the Arctic samples based on an estimated sampling rate of 0.52 m^3/d. A ΣPBDE concentration of 4 pg/m^3 was found at Alert, the long term atmospheric monitoring site on northern Ellesmere Island. No latitudinal trends were seen when the results for the Arctic sites were compared to those from samples taken further south.

Passive air sampling using polyurethane foam disks (PUFs) as sorbent, was utilized by Pozo et al. (2006) in the global air passive sampling (GAPS) campaign. The samplers were deployed for four months (December 2004–March 2005) and the campaign included three Arctic sites at Alert (Canada), Barrow (Alaska) and Ny Ålesund, Svalbard (Norway). Only BDE-47, -99 and -100 were detected in the samples. Based on a sampling rate of 3.9 m^3/d, ΣPBDE concentrations were 2 pg/m^3 at Alert, 5 pg/m^3 at Barrow, AK and 5.3 pg/m^3 at Ny Ålesund (Svalbard). As in the Shen et al. (2006) study, BDE-47 and -99 were the predominant congeners and were present in similar concentrations in the air samples.

The first evidence of BDE-209 in Arctic air samples comes from the study of Wang et al. (2005). High-volume samplers were used to collect particulates on filters on a research cruise traveling from the Bohai Sea off of China through the Bering Strait and into the Canada Basin of the Arctic Ocean from July to September 2003. A total of 49 samples were collected of which 30 were taken above 66.7°N (Arctic samples). Samples were analyzed for tri-decaBDEs and the predominant congeners found were BDE-47, -99 and -209. The mean ΣPBDE (sum of BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -183, -209) concentration for the Arctic samples was 17.3 pg/m^3, with mean BDE-47, -99 and -209 concentrations of 7.21, 4.95 and 3.01 pg/m^3, respectively. The mean ΣPBDE concentrations for the Far East Asia samples was higher, 58.3 pg/m^3, but for the northern Pacific, the ΣPBDE concentration was lower, 12.8 pg/m^3. Latitudinal fractionation, i.e., decreasing ΣPBDE concentrations with higher latitude, was seen

for the samples going from Far East Asia (37° N) up to the northern Pacific (66° N), but for the Arctic samples, no significant relationship was found between concentrations and latitudes. Compared to the samples from Far East Asia and the northern Pacific, the Arctic samples had a lower proportion of BDE-209 and the authors speculate that this may be due to a combination of photodegradation during long range transport and a higher deposition rate. Considering the results of Raff and Hites (2007), the higher deposition rate is the more likely explanation. They also speculate that the relatively high PBDE concentrations in the Arctic samples may be due to sampling near the North American continent, which may serve as a source of PBDEs to air in this area of the Arctic.

Weekly air sampling with high volume samplers using filters and PUFs has been carried out at Alert, Nunavut, in the Canadian High Arctic since 1992. Su et al. (2007) analyzed 104 air samples taken over the period 2002–2004 for di-decaBDEs (BDE-17, -28/33, -47, -49, -66, -85, -99, -100, -138, -153, -154, -183, -190 and -209). Concentrations (gas and particle phase combined) of 14 BDEs (excluding BDE-209) ranged from 0.40–47 pg/m^3 (mean 7.7 pg/m^3) and 0.09–9.8 pg/m^3 (mean of 1.6 pg/m^3) for BDE-209. The Σ_{14}PBDE concentrations were thus in the range of concentrations seen in Alert using passive samplers (Pozo et al., 2006; Shen et al., 2006). However, BDE-209 was not detected using passive samplers, probably because these devices sample mainly gas phase chemicals. The Σ_{14}PBDE concentrations as well as BDE-209 concentrations were also in the same range as in the air samples taken over the Arctic Ocean using high volume samples but measuring concentrations on filters only (Wang et al., 2005).

BDE-47, -99 and -209 were the predominant congeners in the Alert samples, with BDE-47 and -99 being present in similar concentrations, followed by BDE-28/33, -100, -153, and -154 (Su et al., 2007). These BDE congeners together accounted for 90% of the Σ_{15}PBDE concentrations. Concentrations of the lower brominated congeners (di-hexaBDE) were highest in the summer and significant positive correlations were seen between natural logarithm of concentrations and ambient temperatures. This temperature dependence indicates that volatilization emissions of PBDEs are occurring from source regions during the warmer months. But in wintertime, the concentrations showed no seasonality and varied episodically and there were no correlations with temperature. BDE-209 showed no seasonal trends at all and was also episodic. These episodically elevated concentrations in wintertime are thought to be linked to Arctic haze, which is caused by inputs of aerosols and airborne contaminants from mid-latitude sources to the Arctic. In the cold temperatures of winter, PBDEs are predominantly found on particles (Harner and Shoeib, 2002) and would thus be subject to long range transport with Arctic haze aerosols. These results together are interpreted to mean that the source of BDEs in air at Alert is the usage of PentaBDE and DecaBDE technical products at lower latitudes and the subsequent long range transport via volatilization emissions and on particulates.

High volume air samples collected in 2006 and 2007 from the Zeppelin Station, near Ny Ålesund on Svalbard were analyzed for PBBs, PBDEs and HBCD (isomer-specific) (Manø et al., 2008a,b). Mean BB-153 concentration for 2006 was 0.04 pg/m^3. The mean ΣPBDE (BDE-28, 47, 49/71, 66, 99, 100, 153, 154, 183) concentrations were 8.2 (2006) and 1.2 (2007) pg/m^3. BDE-47 was the predominant congener, followed by BDE-99. BDE-196, 206 and 209 were not detected. For HBCD, mean concentrations were 7.1 (2006) and 6.5 pg/m^3 (2007), and γ-HBCD was the predominant stereoisomer, followed by α-HBCD, with very little β-HBCD.

High volume air samples were collected in 2005 from Nuuk, on southwest Greenland (Bossi et al., 2008). Samples were collected over 14 days, twice per month and were analyzed for 11 PBDEs (BDE-17, -28, -47, -49, -66, -85, -99, -100, -153, -154, -183). BDE-209 was not included due to blank problems. The mean (±standard deviation) ΣPBDE concentration for 2005 was 1.14 ± 0.81 pg/m^3. These concentrations are somewhat lower than those seen for high volume air samples from Arctic Canada (Alert) and Svalbard (Zeppelin), but are similar to results found for passive samplers at these sites. The predominant congeners were BDE-47 and -99, followed by BDE-100, -153 and -28. Seasonal variation was seen with the highest concentrations occurring in the summer. Concentrations of BDE-47 and -99 were positively correlated with temperature, indicating that re-emission from previously contaminated surfaces is important for transport of these compounds to West Greenland.

Excluding BDE209, which has not been previously determined in Arctic air, the results for PBDEs were in good agreement with previous measurements by passive air sampling in northern Norway and Iceland (Jaward et al., 2004).

When considered together, these studies indicate that photolytic degradation is a major elimination pathway for the lower brominated BDEs in the gas phase (Raff and Hites, 2007) but is not important for BDE-209 as it is almost entirely particle-bound. Deposition is a more important removal process from the atmosphere for BDE-209, particularly wet deposition. However, during strong winds and periods of no precipitation, wet deposition of BDE-209 will be minimal and thus it and other particle-bound PBDEs will be able to undergo long range transport to a higher extent than previously thought (Breivik et al., 2006). Even the lower brominated PBDEs are primarily bound to particles at the low temperatures typical of the Arctic (Harner and Shoeib, 2002) meaning that photolytic degradation may play less of a role as a removal process. This will especially be the case during the winter, when there is no sunlight, and the Arctic haze season of stable atmosphere allows for the input of aerosols to the Arctic from mid-latitudes. Thus, PBDEs are expected to have longer lifetimes in the Arctic atmosphere in the winter than in the summer (Su et al., 2007). The results from actual air measurements support the results of Breivik et al. (2006), particularly the episodic winter influx of PBDEs, including BDE-209, shown by Su et al. (2007) indicating that periods of Arctic haze, when atmospheric conditions are stable with no precipitation, lead to long range transport to the Arctic from more southerly source regions.

4.3. Terrestrial environment

4.3.1. Soil

Only tetra-heptaBDEs were previously reported in a few Arctic soils and these were found in low concentrations (ΣPBDEs of 0.16–1 ng/g dry weight (dw)). Latitudinal fractionation of the lower brominated PBDEs was also seen.

A recent report presents some PBDE data for soil samples collected in the summer of 2006 at landfill sites in Iqaluit (Nunavut) and Yellowknife (Northwest Territories), Canada (Danon-Schaffer et al., 2007). Replicate soil samples ($n = 3$–5) representing 0–20 cm depth were collected from four locations around the Iqaluit landfill, one upgradient (slightly higher elevation) and three downgradient (progressively lower elevations) from the landfill. In Yellowknife, soil samples ($n = 1$–4) were collected from 0–15 cm depth from four locations, one background site and three downgradient sites, but also from several sites within the landfill where various types of waste are collected. All samples were analyzed for a suite of mono-decaBDEs, but the major BDE congeners found were BDE-47, -85, -99, -153, -154 and -209 and these made up 70–97% (mean of 88%) of the ΣPBDE concentrations. BDE-209 was the predominant congener in all soil samples. The ΣPBDE concentrations (all detected congeners) for soil samples outside both of the landfills were low, ranging from 0.19 to 2.7 ng/g dw, which are similar to previous studies of background soils in the Arctic (de Wit et al., 2006). Interestingly though, the soil concentrations were elevated in the samples taken within the Yellowknife landfill near some types of waste. For example, a soil

sample taken near discarded refrigerators had a ΣPBDE concentration of 52 ng/g dw, of which 47 ng/g dw was BDE-209, indicating that discarded consumer products that are flame-retarded are leaching PBDEs into the surrounding environment.

4.3.2. Vegetation

In the previous review, low concentrations of PBDEs, including BDE-209, as well as TBBPA and PBBs were found on vegetation far from emission sources, indicating deposition after long-range transport (de Wit et al., 2006).

Mariussen et al. (2008) determined BDEs (BDE-28, -47, -49, -66, -85, -99, -100, -119, -153, -154, -183, -196, -206 and -209) in moss (*Hylocomium splendens*) from twelve sites in Norway, three of them above 67°N, collected in summer 2004 using GC-HR-EIMS. ΣPBDE concentrations (BDE-28, -47, -99, -100, -153, -154, -183) ranged from 0.03 to 0.109 ng/g dw, with additional concentrations of BDE-209 ranging from 0.052 to 0.64 ng/g dw (contributing ca. 80% of the sum of 8 congeners). The low presence of BDE-196 and BDE-206 suggests that these compounds are not present as a result of significant debromination of BDE-209. This study shows that BDEs, including BDE-209, can spread into the terrestrial environment and foodchains involving herbivores. The considerable presence of BDE-209 is probably a result of particle transport and deposition from the atmosphere. A significant decrease in the content of the lower brominated BDEs was seen with increasing latitude, and the concentrations of BDE-209 also decreased from south to north.

4.3.3. Terrestrial animals

Concentrations of ΣPBDE in terrestrial animals at lower trophic levels were found to be low in the previous review, whereas they were much higher in terrestrial birds of prey, particularly in peregrine falcons (de Wit et al., 2006). The PBDE congener patterns differed as well, with herbivores having a pattern more similar to the PentaBDE product (BDE-47, -99), and terrestrial birds of prey having a predominance of BDE-153, unlike piscivorous birds of prey where BDE-47 predominates. BDE-209 was found both in herbivores and birds of prey and HBCD, TBBPA and some PBBs were found in predatory birds. Several BDEs, including BDE-209, showed increasing temporal trends in Greenland peregrine falcons.

Mariussen et al. (2008) reported PBDE data for liver samples from moose (*Alces alces*) and grouse (*Lagopus lagopus*) collected in 2000 from Arctic parts of northern Norway, north of 67°N (provinces of Nordland, Troms). BDE-47 and -99 were the major BDE congeners detected in moose liver samples and were found at comparable median levels of 0.24–0.26 and 0.26–0.34 ng/g lw, respectively. BDE-100 and -209 could be detected in some of the samples with maximum levels of 0.08 and 3.2 ng/g lw, respectively. Median ΣPBDE concentrations (excluding BDE-209) in moose were 0.76 and 0.88 ng/g lw for Troms and Nordland Provinces, respectively. Median ΣPBDE concentrations in the grouse liver samples were 0.51 and 0.25 ng/g lw for Troms and Nordland, respectively, and primarily BDE-47 was detected.

No additional new data for BFRs in terrestrial organisms could be found for the observed timeframe for this review (2006–mid 2008). However, data on sledge dogs (*Canis familiaris*) and Arctic fox (*Alopex lagopus*) have been published recently, and the respective results are described under the marine mammal section as both sledge dogs and Arctic foxes feed in the marine food chain.

In summary, little is known about the BFR contamination of the terrestrial ecosystem in the Arctic. Recent studies show low but measurable concentrations of PBDEs in background soils from Arctic Canada, mosses, moose and grouse from northern Norway. BDE-209 is the predominant BDE congener in soil and moss samples, and, when present, in moose samples. The presence of these BFRs is added evidence of their ability to reach the Arctic via long-range atmospheric transport and deposition processes.

4.4. Freshwater environment

4.4.1. River water

Data on mono-octaBDEs in river water from the Ob and Yenisey Rivers in the Russian Arctic were published by Carroll et al. (2008). Both rivers together contribute 37% of the riverine freshwater inputs to the Arctic Basin and thus represent an important pathway for the land-Arctic ocean exchange of contaminants. Sampling was carried out at a number of sampling sites upstream in the Yenisey (2003) and Ob (2005) Rivers, into their estuaries and further out into the Kara Sea. Contaminant analyses were performed on sample extracts taken from filtered large volume water samples (50–100 L). PBDEs, like other hydrophobic organic contaminants, tend to associate highly to particulate matter in water, with only a small fraction actually dissolved in the water (freely dissolved fraction). Filtering eliminates larger particles and suspended solids and these water samples are thus more representative of the dissolved fraction of PBDEs, which is also the fraction that is bioavailable to water-breathing organisms.

PBDEs were detected at all sampling locations. Among the 43 measured PBDE congeners, 20 were above detection limits but at concentrations less than 1 pg/L; only levels of BDE-37, -47, and -99 exceeded 1 pg/L. BDE-47 and -99 were predominant congeners in all water samples and were found in proportions similar to those seen in technical PentaBDE products, indicating these as the major sources of PBDEs to these rivers. Values of ΣPBDE ranged from 1.8 to 10.8 pg/L. Mean ΣPBDE in the Gulf of Ob (6.7 pg/L) was significantly higher than in the Yenisey Bay (3.2 pg/L), but not statistically different from the Kara Sea (4.3 pg/L). The estimated fluxes of ΣPBDE flowing to the Kara Sea were 1.92 kg/yr for the Yenisey River discharges and 1.84 kg/yr for the Ob River. The estimated contaminant fluxes for PCBs and HCHs from these rivers are similar to those reported for major Canadian rivers, confirming expectations that the Ob and Yenisey are also major point sources for the Arctic basin. Di-triBDEs not found in the commercial mixtures were detected in all three regions in differing proportions relative to ΣPBDE, indicating possible compositional changes via debromination of PBDEs during atmospheric transport.

4.4.2. Freshwater sediments

In the previous review on BFRs in the Arctic, increasing concentrations with time were reported in freshwater sediment cores from the Canadian Arctic and from Greenland (de Wit et al., 2006). Fluxes of BDE-47 were slightly higher in the Canadian lakes, ranging from <0.5 to 8 ng/m^2/year, in contrast to 0.05–1.3 ng/m^2/year in Greenland, and differed with regard to their maximum, which occurred in the late 1980s in Canada and late 1990s in Greenland. BDE-209 was detected in Char Lake (Canada) approximately ten years after the first occurrence of BDE-47 and increased up to the most recent layer (1995–1998), at a maximum flux of approximately 8 ng/m^2/year. PBDE concentrations in the top layer were low, 0.04–0.07 ng/g dw (tetra-decaBDEs) in Canada and 0.007–0.05 ng/g dw (BDE-47) on Greenland. Additional sediment data were available from lakes and rivers of the Russian Arctic, covering concentrations between 0.004–0.027 ng/g dw for the sum of BDE-47 and BDE-99, i.e., 90–600 times lower than PCB concentrations in the same samples.

PBDEs (BDE-28, -47, -99, -100, -138, -153, -154, -183) and HBCD (on a diastereoisomer-specific basis) were analyzed in sediment cores collected in 2001 from Lake Ellasjøen from the Norwegian island Bjørnøya (Bear Island) in the central Barents Sea (74°30′N, 10°00′E) (Evenset et al., 2007). The lake had previously been shown to be a "hot spot" for contamination as it receives large amounts of guano from seabird colonies, possibly a transport medium for contaminants from the marine to the limnic environment. In addition, the precipitation rate is high, leading to high deposition of air-borne contaminants. The ΣPBDE concentration for the congeners detected (BDE-28, -47, -99, -100, -153) peaked in surface sediments, dated to be deposited between 1987 and 2001, at 0.73 ng/g dw or 12.2 ng/g organic carbon

(OC). This concentration is higher than seen in freshwater sediments from the Arctic previously and is in the lower end of what has been reported from the industrialized areas of the Great Lakes (Song et al., 2004, 2005), possibly confirming the "contamination hot spot" character of Lake Ellasjøen. This PBDE concentration was approximately 65 times lower than the ΣPCB concentration in the same core layer. However, this ratio had diminished over time, due to different temporal trends in the concentration development (see Section 5 Temporal trends).The PCB composition indicated large proportions of highly chlorinated congeners and thus resembled the composition typically found in biota. The authors concluded that input from seabird guano, possibly in combination with relative geographic proximity to emission sources, could explain this pattern. The significance of seabird guano might also be valid for PBDEs, and is supported by the predominance of BDE-47 in the sediment samples.

HBCD was detected only in the depth interval from 1–2 cm (median age 1980) (Evenset et al., 2007). Only the α- and γ-isomers were detected, at 0.43 and 3.9 ng/g dw, respectively (7.3 and 66 ng/g OC), which is an order of magnitude higher than ΣPBDE in the same depth slice. The authors found it surprising that no HBCD was found in the top layer of the sediment, given the ongoing use of HBCD. Consistent with other studies on HBCD isomers in sediments (Birnbaum and Staskal, 2004), γ-HBCD accounted for about 90% of ΣHBCD, while β-HBCD was not detected.

In addition to Lake Ellasjøen, PBDEs were analyzed in surface sediments of 41 other lakes in northern Norway (Nordland, Troms and Finnmark Provinces) and on Svalbard, as part of the Norwegian 2004–2006 National Lake Survey (Christensen et al., 2008). ΣPBDE included BDE-17, -28, -47, -49, -66, -71, -85, -99, -100, -138, -153, -154, -183, -190, -206, -207, -208 and -209. The mean and median ΣPBDE concentrations for all 42 lakes were 0.63 ng/g dw and 0.11 ng/g dw (range: 0.025 to 9.6 ng/g dw). The highest ΣPBDE concentrations were above those found in industrialized areas, e.g. in sediments from the Great Lakes (Song et al., 2004, 2005), and were found in two lakes on Svalbard (Arresjøen — 2.4 ng/g dw; Åsøvatn — 0.96 ng/g dw), Lake Ellasjøen on Bjørnøya (0.57 ng/g dw) and several lakes in the province of Finnmark (up to 9.6 ng/g dw) with a mean of 0.81 ng/g dw for Svalbard and 0.88 ng/g dw for Finnmark sediments (Christensen et al., 2008). These means are about 10 times higher than the means of 0.084 ng/g dw and 0.096 ng/g dw found in Nordland and Troms lake sediments, respectively. It is noteworthy that the lowest ΣPBDE concentration of the survey also was found on Svalbard (0.025 ng/g dw, Lake Kongressvatn), another example of substantially different concentration levels within a relatively small area. The authors explained that the high concentrations in lakes from Svalbard might be due to seabird activities, i.e., the deposition of guano into the lakes, similar to the case in Lake Ellasjøen on Bjørnøya. No explanation could be given for the high levels in lakes from Finnmark. In some of the samples with high concentrations, ΣPBDE exceeded the concentration of ΣPCB. BDE-99, -47 and -153 were the main congeners found in the sediments, whereas BDE-209 was not detected in any of the samples.

PBDE fluxes were determined in sediment cores in Alaska from Lake McLeod (63°38′N) and Lake Wonder (63°48′N) (Denali National Park and Preserve), Lake Matcharak (67°75′N) (Gates of the Arctic National Park and Preserve) and Lake Burial (68°43′N) (Noatak National Preserve) (Landers et al., 2008). Denali National Park is located approximately 300 km south of Gates of the Arctic National Park and Noatak National Preserve. Cores were sampled in 2004 and sliced into 1.0 or 0.5 cm sections for age determination and chemical analysis. ΣPBDE included the congeners BDE-47, -99, -100, -153 and -154. Concentrations were multiplied by the mass sedimentation rate and normalized to focusing factors, to arrive at the focus-corrected flux (ng/m^2/year). In the surficial sediment, PBDEs were detected in less than half of the samples. Fluxes of approximately 12.5 ng/m^2/year for Denali National Park and 6.5 ng/m^2/year for the other two parks were given, calculated using half the detection limits. For the sake of comparisons, fluxes of approximately 1000 ng/m^2/year were found for surficial sediments from the Rocky Mountain National Park (Colorado) and the Sequoia and Kings Canyon National Parks (California).

The new data have added information on PBDE levels in lake sediments from Alaska, Svalbard and northern Norway, including Lake Ellasjøen on the island Bjørnøya. PBDE concentrations in Lake Ellasjøen seem to be influenced by the input of seabird guano. For Bjørnøya, concentrations are high in an Arctic context, and the PBDE pattern reflects that of biota rather than what has previously been reported for sediments. Similar influences were assumed for some lakes from Svalbard with high PBDE concentrations, compared to the Arctic in general and other lakes in the immediate vicinity. High concentrations, exceeding levels in industrialized regions, were also found for lake sediments in the Norwegian province of Finnmark. While PBDE concentrations peaked in the surface sediment layer in Lake Ellasjøen, HBCD – mainly consisting of γ-HBCD – was found to have the highest concentrations around 1980. This is the first report of HBCD in Arctic freshwater sediments. Sediment fluxes in lakes from Alaska contained several values below detection limits and were generally classified as low, compared with lakes from lower latitudes. However, fluxes in the lakes from the more southerly Alaskan site (Denali) were twice as high as those from the two lakes in more northerly sites (Gates of the Arctic and Noatak). Previous sediment fluxes determined for lakes in Arctic Canada are similar to the fluxes for the northern Alaskan lakes, whereas sediment fluxes on Greenland are 5–10 times lower. The higher fluxes in Denali compared to northern Alaska and Canada may be due to its closer proximity to source areas in North America.

4.4.3. Invertebrates and fish

The contaminant hot-spot character of Lake Ellasjøen on Bjørnøya in the Barents Sea is also reflected in the PBDE data available from freshwater biota. The previous review reported concentrations in land-locked Arctic char (*Salvelinus alpinus*) to be an order of magnitude higher in Lake Ellasjøen compared with the 5 km distant Lake Øyangen on the same island or lakes in Greenland (de Wit et al., 2006). Similarly, burbot (*Lota lota*) from Lake Grensefoss in northern Norway exhibited unexpectedly high concentrations, possibly influenced by the 100 km distant, highly industrialized town of Nikel in Russia. Generally speaking, ΣPBDE concentrations were similar across the Arctic within the same fish species, i.e., geographical trends seem to be less pronounced than in the marine environment. The concentrations were lower than generally observed for marine fish, and the PBDE pattern more similar to the PentaBDE technical product.

Evenset et al. (2005) analyzed zooplankton, chironomid larvae (*Chironomidae* sp.), tadpole shrimps (*Lepidurus arcticus*) and small and large individuals of landlocked Arctic char collected from Lake Ellasjøen and Lake Øyangen for BDE-33, -37, -47, -66, -71, -75, -99, -119 and -153, and in addition, a selection of the samples were analyzed for BDE-12, -13, -15, -30, -32, -35, -77, -85 and -100. As expected, the concentrations in biota differed considerably between the two lakes, with all but two samples of Arctic char below the limit of detection in Lake Øyangen. In these two samples, one small and one large individual, ΣPBDE concentrations (BDE-33, -47, -71, -99, -100, -119, -153) were 79 and 71 ng/g lw (1.6 and 1.3 ng/g ww), respectively. The mean ΣPBDE concentrations in Lake Ellasjøen were 52 ng/g lw (0.78 ng/g ww) in zooplankton, 44 ng/g lw (1.62 ng/g ww) in chironomid larvae, 480 ng/g lw (15.1 ng/g ww) in small Arctic char and 410 ng/g lw (17.5 ng/g ww) in large Arctic char. The PBDE concentrations in Arctic char from Lake Ellasjøen were comparable to results from Lake Vättern in Sweden (Sellström et al., 1993) i.e., from industrialized areas. However, compared with DDTs and PCBs, analyzed in the same samples, PBDE concentrations were 10–100 times lower. PBBs were also analyzed in large char from both lakes

(BB-15, -49, -52, -101 and -153). BB-153 was the only detectable congener, with means of 22 ng/g lw (0.90 ng/g ww) in Arctic char from Lake Ellasjøen and below detection in Lake Øyangen. Thus, the BB-153 concentration was approximately 20 times lower than ΣPBDE in the same samples.

Slightly lower ΣPBDE concentrations in Arctic char were found in analyses in the Norwegian National Lake Survey 2004–2006 which, besides Lakes Ellasjøen and Øyangen on Bjørnøya, covered two lakes on Svalbard (Richardvatn and Åsøvatn) (Christensen et al., 2008). Of the 18 BDE congeners analyzed in this survey (including BDE-209), only ten were detected in fish samples (BDE-28, -47, -49, -66, -71, -99, -100, -153, 154 and -183). ΣPBDE concentrations in pooled fish samples were highest for Lake Ellasjøen (113 ng/g lw, 1.92 ng/g ww), which is only about 30% of the concentrations found by Evenset et al. (2005). Likewise, fish from Lake Øyangen had much lower ΣPBDE levels (8.1 ng/g lw, 0.28 ng/g ww) than previously reported (Evenset et al., 2005). Lipid-normalized ΣPBDE concentrations were 24 and 39 ng/g lw (0.42 and 1.2 ng/g ww) in fish from Richardvatn and Åsøvatn, respectively. In addition to the pooled samples, 5 individuals were analyzed from the Lakes Richardvatn and Åsøvatn, respectively. Although the trend with higher levels in Lake Åsøvatn was confirmed, the absolute concentrations were higher in the individual samples, with ΣPBDE mean concentrations and standard deviations of 58 ± 46.6 ng/g lw for Arctic char from Lake Richardvatn and 86 ± 96.9 ng/g lw for fish from Lake Åsøvatn. The individual fish were larger than the individuals of the pooled samples, which, according to the authors, can explain the concentration difference from the pooled samples.

Levels similar to those established for Lake Ellasjøen by the Norwegian National Lake Survey 2004–2006 have been reported in landlocked char from Resolute, Char and Amituk Lakes on Cornwallis Island, Nunavut, Canada. For Resolute Lake, ΣPBDE concentrations were approximately 80 ng/g lw in 2007, of which about 50% was BDE-47. Concentrations of about 110 ng/g lw were found for samples from Char Lake in 2007. Results from Lake Amituk in 2007 were approximately 30 ng/g lw (D. Muir, Environment Canada, personal communication).

In addition to Arctic char, the Norwegian National Lake Survey 2004–2006 included analyses of lake trout (*Salvelinus namaycush*) in three lakes from northern Norway, in the provinces of Finnmark (Lake Langvatn), Nordland (Lake Valnesvatn) and Troms (Lake Holmvatn) (Christensen et al., 2008). Perch (*Perca fluviatilis*) from Lake Gavdnajavri (Finnmark, Norway) were analyzed as well. ΣPBDE levels (BDE-28, -47, -49, -66, -71, -99, -100, -153, 154 and -183) in the pooled samples ($n = 8–12$) were comparable for both species and all locations, with 19, 16, and 11 ng/g lw (0.29, 0.57 and 0.26 ng/g ww) for trout from Lakes Langvatn, Valnesvatn and Holmvatn, respectively, and 14 ng/g lw (0.03 ng/g ww) for perch from Lake Gavdnajavri. However, the lipid content in perch was only 0.2%, meaning that the wet weight concentrations in perch were an order of magnitude lower than those in lake trout. In almost all fish samples from this survey, BDE-47 was the main BDE congener. Only trout from Lake Holmvatn had a higher content of BDE-99 compared with BDE-47. BDE-183 was detected occasionally, while BDE-209, although included in all analyses, was consistently below the detection limit (Christensen et al., 2008).

PBDEs and, to some extent, HBCD have also been analyzed in lake trout from three lakes in the Yukon, Canada – Lake Laberge, Lake Kusawa and Lake Quiet – in order to assess temporal and spatial trends (Stern et al., 2007). Trout from Lake Laberge had the highest mean ΣPBDE (BDE-47, -49, -99, -100, -153 and -154) concentrations of 8800 ng/g lw (88 ng/g ww) in 2006. Intermediate levels of 4700 ng/g lw in 2003 (7.5 ng/g ww) and 190 ng/g lw (3.4 ng/g ww) in 2006 were found in trout from Lake Kusawa. Mean ΣPBDE levels in trout from Lake Quiet were 240 ng/g lw in 2003 (0.29 ng/g ww) and were thus 1–2 orders of magnitude lower than means for Lake Laberge trout. The higher PBDE concentrations in the Lake Laberge trout agrees with previous studies showing elevated concentrations of organochlorine compounds in lake trout from this lake, which has been attributed to higher bioaccumulation due to a longer food web (Kidd et al. 1995). BDE-47 accounted for 14–40% of ΣPBDE. Interestingly, BDE-99 was the predominant congener in the three lakes, with very few exceptions.

HBCD was also analyzed in the lake trout from Lake Laberge and Lake Kusawa caught in 2006 (Stern et al., 2007). However, only 1 individual was available from Lake Laberge. The levels of α- and γ-HBCD in this particular fish were 5300 and 1600 ng/g lw, respectively (54 and 16 ng/g ww), and thus almost three orders of magnitude higher than those of the Lake Kusawa fish (mean of α- and γ-HBCD concentrations: 2.5 and 3.4 ng/g lw, 0.045 and 0.061 ng/g ww, respectively). The α-HBCD/γ-HBCD ratio is <1 in the trout from Lake Kusawa. The authors explained the large differences observed between trout from the different lakes as being primarily linked to biological factors, such as age, weight and lipid content of the fish. They have also observed changes of lipid content and body masses of trout over time within each of the lakes, which might be related to fluctuating conditions, e.g. plankton productivity, climate variations and degree of fishing (Stern et al., 2007).

Lake trout were collected in 2004 from three lakes in Alaska: Lake Burial in the Noatak National Preserve, Lake Matcharak in the Gates of the Arctic National Park and Preserve and Lake Wonder in Denali National Park and Preserve (Ackerman et al., 2008; Landers et al., 2008). Furthermore, four individual burbot and two individual round whitefish (*Prosopium cylindraceum*), respectively, from Lake McLeod in Denali National Park were analyzed. A total of 34 BDE-congeners were determined, but only BDE-47, -99, -100, -153 and -154 were consistently above the detection limits. For some samples, quality assurance did not allow quantification of all congeners. If BDE-47 was not among the quantifiable congeners, we excluded the sample from subsequent data analysis. Median ΣPBDE values of 11, 5.9 and 24 ng/g lw were found for lake trout from Lake Matcharak, Lake Burial and Lake Wonder, respectively. The corresponding mean values of 15, 22 and 21 ng/g lw were not statistically significantly different from each other. Similar to the Yukon results, BDE-47 and BDE-99 had comparable concentrations in most fish. ΣPBDE concentrations in the two burbot samples varied by a factor of 2: 8.3 and 15 ng/g lw. The range of ΣPBDE concentrations in the four whitefish samples from Denali National Park covered an order of magnitude, with median and mean values of 32 and 88 ng/g lw, respectively. The authors commented that PBDE concentrations in fish varied less than most other organic contaminants, e.g., current or historical use pesticides and PCBs, both within and between lakes. Concentrations were lowest in Alaska, compared with more southerly National Parks in the Western USA. On average, ΣPBDE concentrations were about three times higher in Alaskan fish than in fish from high-elevation lakes in Europe (Ackerman et al., 2008; Landers et al., 2008; Vives et al., 2004). This finding was different from most organochlorines (HCB, DDTs, HCHs), which had 2–9 times lower concentrations in fish from the Western USA compared with European mountain fish (Ackerman et al., 2008).

Studies on burbot from the Mackenzie River, Fort Good Hope, Northwest Territories, Canada, have continued since the first review (de Wit et al., 2006) and new data have become available for PBDEs and HBCD (Stern and Tomy, 2007). Mean ΣPBDE (BDE-47, -49, -99, -100, -153 and -154) concentrations in burbot collected in 2006 were 24 ng/g lw (5.2 ng/g ww). BDE-47 was the most predominant congener in burbot liver, contributing approximately 35–50% to the ΣPBDE (BDE-47, -99, -100, -153, -154), while BDE-99 accounted for about 25%. PCBs analyzed in the same individuals had concentrations about an order of magnitude above those of PBDEs. Very similar HBCD concentrations of 1.32 ng/g lw (0.25 ng/g ww) and 1.30 ng/g lw (0.39 ng/g ww) were found for 2005 and 2006, respectively. The α-HBCD/γ-HBCD ratio was 24 in 2005 and 19 in 2006.

The new Arctic char data confirm previous results of Lake Ellasjøen being a "contaminant hot spot", with PBDE levels clearly above those of nearby Lake Øyangen, but possibly comparable with concentrations from the Canadian Arctic. PBDE concentrations in lake trout seemed to be highest in lakes from the Yukon, and much lower in lakes from northern Norway and Alaska. While the Yukon study in particular demonstrated large intra- and inter-lake variations, possibly due to biological factors, only small differences were found between the Norwegian lakes. Levels in burbot are similar in the Mackenzie River and in Alaskan lakes. The Canadian and Alaskan data seem to confirm the previous finding of BDE patterns similar to the technical PentaBDE mixture (i.e. almost equal percentages of BDE-47 and BDE-99), but BDE-47 was the predominant congener in the Norwegian data.

HBCD data continue to be sparse. For lake trout from the Yukon, HBCD concentrations seem to follow the same concentration trends as PBDEs, with higher concentration in fish from Lake Laberge. In direct comparisons, ΣHBCD ($\alpha+\gamma$) concentrations are lower than ΣPBDE, in the same species at the same site. The α-HBCD/γ-HBCD ratio is not constant, but was greater than 1 in most cases.

4.5. Marine environment

4.5.1. Marine sediments

In the previous review on BFRs in the Arctic, lower brominated PBDE concentrations were reported in sediment samples from the Canadian, Norwegian and Russian Arctic (de Wit et al., 2006). BDE-209 was only analyzed in one sediment sample from Tromsø (0.43 ng/g dw). Except for one Russian site where local contamination was suspected (Polyarnyy, Kola — 241 ng/g dw) the ΣPBDE concentrations were generally low, covering a range from 0.04–0.25 ng/g dw, but were higher than concentrations found in freshwater sediments. Where PCB data were available, these generally exceeded the PBDE concentrations, however, the ratio differed considerably. BDE-47, -99 and, when present, -209 were the most predominant congeners in all cases.

In the meantime, data have become available on PBDEs, BB-153, TBBPA and HBCD in sediment samples from the Barents Sea area, including coastal sediments from northern Norway (Bakke et al., 2008). The samples originated from two projects which focused on the Russian Barents Sea and Pechora Sea and on the Tromsøflaket region, outside the coast of northern Norway near Tromsø, respectively. Samples were collected from the Barents Sea and Pechora Sea in June 2006, while samples from Tromsøflaket originated from cruises in May/June 2006 and spring and autumn of 2007. ΣPBDE concentrations (BDE-28, -47, -99, -100, -153, -154 and -183) ranged between 0.0055 and 0.0067 ng/g dw in the Tromsøflaket sediments, whereas the Barents Sea and Pechora Sea samples had higher concentrations (0.0015–0.298 ng/g dw). On the other hand, BDE-209 was not detected in the Barents and Pechora Sea samples, but was present in concentrations ranging from 0.023 to 0.7 ng/g dw in the Tromsøflaket sediments (Bakke et al., 2008). Thus the sediments clearly fall into two categories with regard to BDE composition. The samples from the Tromsøflaket contained primarily BDE-209, which had concentrations 1–2 orders of magnitude higher than other BDE congeners and the second highest concentrations in these sediments were found for BDE-206, which is an unusual finding as this nonaBDE is only found in trace amounts in the technical DecaBDE product. These two congeners, however, were below detection limits in the samples from the Barents and Pechora Seas, which contained the highest concentrations of BDE-47 or BDE-100 and in some cases, BDE-99. The authors discussed several reasons for this marked difference in BDE composition. As the analyses were performed at two laboratories, differences in analytical methods cannot be ruled out. Alternatively, different sources can be of importance of which, however, no information was available. In general, these ΣPBDE levels are similar to those reported previously for marine sediments from the Canadian, Norwegian and Russian Arctic from background sites and lower than seen previously at the more contaminated site of Polyarnyy, Kola, Russia (de Wit et al., 2006).

TBBPA was below the detection limits of approximately 0.0006 ng/g dw in all sediment samples, while all three isomers of HBCD were detected in all but two samples (both from the Barents/Pechora Sea) (Bakke et al., 2008). One of these, however, had artificially high detection limits, due to matrix interferences. Again, the two sample groups seem to differ, here in terms of their variance: the Tromsøflaket samples had similar ΣHBCD concentrations of 0.026–0.069 ng/g dw, while the samples from the Barents and Pechora Sea ranged between below the detection limits to 0.191 ng/g dw. About 80% of this high concentration was γ-HBCD, in a sample collected south of Novaja Zemlja. No predominance of any of the isomers was found in the other samples. BB-153 was only included in the analysis of the Barents and Pechora Sea samples and concentrations were <0.00023 ng/g dw in all samples.

4.5.2. Invertebrates

At the time of the previous review, only one study from Svalbard had reported levels of PBDEs in zooplankton species, all of which were very low, approximately 1 ng/g lw. New results indicate that some zooplankton species may be exposed to higher concentrations of BFRs, probably as a result of a higher trophic position in the food web.

Calenoid copepods (mainly *Calanus glacialis*) and krill (*Thysanoessa inermis*) collected around Svalbard in 2003 had detectable concentrations of BDE-47 and BDE-99 with ΣPBDE concentrations of 0.16 and 0.26 ng/g lw (0.02 and 0.03 ng/g ww), respectively (Sørmo et al., 2006).

Zooplankton (mixed species, predominantly copepods), shrimp (*Pandalus borealis* and *Hymenodora glacialis*) and clam (*Mya truncate* and *Serripes groenlandica*) were included in a bioaccumulation study on PBDEs and HBCD in the eastern Canadian Arctic (Tomy et al., 2008b). Zooplankton and clam samples were collected from Frobisher Bay in May 2002, while shrimp samples were collected from Davis Strait in October of 2000 and 2001. ΣPBDE includes BDE-47, -85, -99, -100, -153, -154 and -209. Diastereomers of HBCD were analyzed, but β-HBCD was below the detection limits in all samples. Zooplankton had the highest PBDE concentrations of all species in this study, including marine mammals, with a geometric mean (including BDE-209) of 73 ng/g lw. The authors discussed this result in the light of other findings and concluded that there are broad geographic differences in PBDE contamination at the base of Arctic marine food webs. Shrimp and clam had similar geometric mean ΣPBDE concentrations of 16 and 21 ng/g lw, respectively. Interestingly, ΣHBCD did not show the same tendencies as ΣPBDE, with similar geometric mean concentrations of 1.1, 1.9 and 1.4 ng/g lw for zooplankton, shrimp and clams, respectively.

The PBDE pattern also differed considerably between shrimps and clams on the one hand and zooplankton on the other hand (Tomy et al., 2008b). While BDE-47 accounted for approximately 20% of ΣPBDE in shrimps and clams, it was <5% of ΣPBDE in zooplankton. Shrimps and clams also contained considerably more BDE-99 than zooplankton, which in turn had higher percentages of BDE-153. All three groups had in common that BDE-209 contributed less than 25% to the overall ΣPBDE concentrations.

With regard to HBCD, only α- and γ-HBCD were detected, but their pattern differed between species. Geometric mean concentrations of α- and γ-HBCD in zooplankton were 0.6 and 0.5 ng/g lw, respectively, in shrimp, 1.4 and 0.5 ng/g lw, respectively, and in clams, 0.2 and 1.2 ng/g lw. Thus, clams and zooplankton had a similar isomer pattern of approximately 75% γ-HBCD and 25% α-HBCD, while the composition was 25% γ-HBCD and 75% α-HBCD in shrimps. This difference was explained by the physical-chemical properties of the individual isomers. Given the low water solubility of γ-HCBD, the authors discussed diffusion from the water phase into zooplankton, which

may be considered floating-globules of lipid. The predominance of the γ-isomer in sediments could lead to a specific exposure to this isomer by benthic feeders such as clams. The authors noted that it remained unclear why the HBCD diastereoisomer profile was dominated by the α-isomer in shrimp (Tomy et al., 2008b).

Thirteen pooled samples of blue mussels (*Mytilus edulis*) collected in 1994 and 2006 from 9 coastal stations in northern Norway were analyzed for PBDEs (Bakke et al., 2008). ΣPBDE (BDE-28, -47, -99, -100, -153, -154 and -183) concentrations were generally very low, and only a few congeners were above detection limits. Mean ΣPBDE concentration was 1.19 ng/g ww in mussel samples from a harbour (Skrouva Harbour) collected in 1994. Three other samples had mean ΣPBDE of 0.13–0.14 ng/g ww (no information was given on lipid content), among these mussel samples collected near the Skrouva Harbour location in 2006. The remaining samples had ΣPBDE concentrations of <0.10 ng/g ww. For two stations, data were available for mussel samples collected in 1994 and 2006. There is a tendency towards higher concentrations, i.e. more congeners above detection limits, in the samples from 2006.

Bakke et al. (2008) also reported results of PBDEs and HBCD in 5 pooled samples of shrimp (*P. borealis*) collected in the Barents Sea in 2007. Prior to analysis, the shrimp had been peeled and boiled, and only the edible parts were used for the analysis. ΣPBDE (BDE-28, -47, -99, -100, -153, -154 and -183) concentrations ranged from 0.1–0.85 ng/g lw (0.002–0.017 ng/g ww). According to the authors, these levels were about 10 times lower than those found in the North Sea and Skagerrak, probably due to the proximity to PBDE sources of the latter. BDE-47 was the only congener above the limit of detection in all five samples. BDE-153 was detected twice, and BDE-66, BDE-99, BDE-119 were detected once. HBCD was below the limit of detection (about 0.2 ng/g ww) in all samples.

Svendsen et al. (2007) reported concentrations and patterns of PBDEs, HBCD and organochlorines in the benthic amphipod species *Eurythenes gryllus* collected northeast of Svalbard in 2002. Of the 15 PBDE congeners analyzed in this study, 12 congeners (BDE-17, -28/33, -47, -66, -71, -85, -99, -100, -138, -153, -154 and -209) and HBCD were detected. ΣBFR (PBDE + HBCD) concentrations ranged between 115 and 493 ng/g lw, with mean and median values of 303 ng/g lw and 271 ng/g lw, respectively. Unfortunately, individual concentrations were not reported, but on average, BDE-47 accounted for 60% of ΣBFR (approximately 180 ng/g lw), HBCD accounted for 15% of ΣBFR, (approximately 45 ng/g lw) and BDE-99 and -100 each accounted for 8% of ΣBFR (approximately 24 ng/g lw). The concentration of BDE-209 was not specified, but seemed to be below 2% of the summed concentration. These concentrations were comparable to those in higher trophic level animals, such as ringed seals (*Phoca hispida*) from East Greenland and glaucous gulls (*Larus hyperboreus*) from the same area (Verreault et al., 2005; Vorkamp et al., 2004a), i.e. not typical of benthic organisms feeding at low trophic levels, such as blue mussels. Additional data on organochlorines and stable isotopes in other amphipod species indicated that *E. gryllus* scavenges on pelagic carrion, which indeed makes their trophic position more comparable to that of glaucous gulls than to that of blue mussels. The ΣBFR concentrations were about 10 times lower than PCB concentrations in the same samples. ΣBFR, ΣPCB and ΣDDT were significantly positively correlated to each other, which probably reflects the same source of contamination; however, there was no correlation between any of the compound groups and amphipod size.

Relatively high BFR concentrations were also found in the ice-associated omnivorous amphipod *Gammarus wilkitzkii* collected north of Svalbard in 2003, with a ΣPBDE concentration of 15 ng/g lw (Sørmo et al., 2006). About half of this concentration originated from BDE-209, while BDE-47 and BDE-99 contributed with 16% and 27%, respectively and BDE-154 with <2%. The authors noted, however, that the homogenized sample might have included small particles from the digestive system or the body surface, i.e. not reflected truly absorbed BDE-209. Besides algae and zooplankton, *G. wilkitzkii* feeds on detritus, which might expose this species to higher BFR concentrations than the other invertebrates included in this study. The amphipod, *Themisto libellula,* also collected around Svalbard in 2003, only had detectable concentrations of BDE-47, BDE-99 and BDE-100 with a ΣPBDE of 0.53 ng/g lw. The congener pattern could not directly be compared between the samples: BDE-28, BDE-153 and HBCD were analyzed in *G. wilkitzkii*, but could not be detected, while BDE-153, BDE-209 and HBCD were not analyzed in *T. libellula*.

In another study, samples of *G. wilkitzkii* from the Barents Sea east of Svalbard were collected in 2004 (Haukås et al., 2007). PBDEs were analyzed (BDE-28, -47, -71, -77, -99, -100, -138, -153, -154, -183) but concentrations were below the detection limits.

For marine invertebrates, ΣPBDE concentrations are generally low. However, species that are scavengers, such as *E. gryllus*, feed at a higher trophic level and have much higher concentrations. ΣPBDE concentrations in zooplankton, shrimp, blue mussels and the amphipod *T. libellula* from northern Norway/Svalbard are similar and low, ranging from 0.1–0.5 ng/g lw, which is similar to the concentration range found in zooplankton and krill previously (de Wit et al., 2006). Higher ΣPBDE concentrations were seen in zooplankton, shrimp and clams from Canada, ranging from 16–73 ng/g lw. BDE-209 was only included in one study but made up 50% of the total PBDE concentration in *G. wilkitzkii* from Svalbard. HBCD was found in zooplankton, shrimp, clams and *E. gryllus*.

4.5.3. Marine fish

In the previous review, data on marine fish were presented from Greenland and northern Norway, covering several cod species (Atlantic cod, *Gadus morhua*; polar cod, *Boreogadus saida*; anduvak, *Gadus ogac*; Arctic cod; *Cadus callarias*), tusk (*Brosme brosme*), Greenland halibut (*Reinhardtius hippoglossoides*), shorthorn sculpin (*Myoxycephalus scorpius*) and starry ray (*Raja radiata*). The main BDE congener was always BDE-47, accounting for 90–95% of the ΣPBDE concentration, which ranged from a few ng/g ww to approximately 50 ng/g ww (3–480 ng/g lw). Results from southwest Greenland suggested an influence of human settlements on PBDE concentrations in local fish. BDE-209, α-HBCD, TBBPA and methoxylated TeBDEs were detected in cod liver from northern Norway and Svalbard.

Anadromous (sea-run) char were collected from five communities in the Canadian Arctic in 2005 and four communities in 2006. ΣPBDEs ranged from 2.70–6.48 ng/g lw (0.0.16–0.61 ng/g ww) in 2005 and 6.98–23.77 ng/g lw (0.27–2.13 ng/g ww) in 2006 (M. Evans, Environment Canada, personal communication).

As part of a temporal and spatial trend study, Evans and Muir (2007) reported PBDE concentrations in fillets of anadromous char collected at three locations in the Canadian Arctic: Pond Inlet (Nunavut), Kangiqsualujjuaq (Quebec) and Cambridge Bay (Northwest Territories). About 20 individuals were collected at Pond Inlet and Cambridge Bay in August 2006, and at Kangiqsualujjuaq in June and December 2006. No information was given on the BDE congeners included in the analyses. At Pond Inlet, mean ΣPBDE concentration was 24 ng/g lw (2.13 ng/g ww) in 2006, which was statistically different from the concentration found in 2005 (3.4 ng/g lw; 0.32 g/g ww). For most of the organochlorine compounds analyzed in the same samples, a concentration decrease was noted from 2005 to 2006. Fish from 2006 were older and larger than those from 2005 and had a lower lipid and water content, but none of these differences were statistically significant.

At Kangiqsualujjuaq large differences were found in mean ΣPBDE concentrations for the two sampling times, 41 ng/g lw (2.23 ng/g ww) and 3.6 ng/g lw (0.30 ng/g ww) for fish collected in June and December, 2006, respectively, thus reflecting concentrations in fish immediately upon entering the sea and after their summer feeding period and return inland (Evans and Muir, 2007). The fish caught in December 2006 were heavier and had a significantly lower water, but

higher lipid content than those from June 2006. The concentrations observed at Kangiqsualujjuaq were in the same range as those at Pond Inlet. The lowest mean ΣPBDE concentrations were found in fish from Cambridge Bay, with 3.5 ng/g lw (0.14 ng/g ww) and 6.25 ng/g lw (0.8 ng/g ww) in 2004 and 2006, respectively. The authors assumed that the samples from this location originated from different stocks in the two years of sampling. The concentrations differed significantly between the two sampling times for all three locations, but with no conclusive upward or downward trend.

Wolkers et al. (2006a) analyzed BDE-47 and -99 in Greenland halibut and polar cod collected in 1998–99 from Svalbard. These are important prey species of beluga (*Delphinapterus leuca*) and narwhal (*Monodon monoceros*). BDE-47 concentrations in muscle tissue (fillets) were found to be significantly higher in Greenland halibut than in polar cod (mean concentrations of 0.16 ng/g ww and 0.066 ng/g ww, respectively), while BDE-99 was more similar in the two fish species (0.0089 ng/g ww in Greenland halibut and 0.0119 ng/g ww in polar cod). The number of fish analyzed in this study was not given. The concentration difference was attributed to different exposure situations as Greenland halibut are benthic and polar cod are pelagic fish. The same tendency of higher concentrations in halibut than in cod was found for PCBs, DDTs, toxaphene and chlordane-related pesticides analyzed in the same samples.

Polar cod (whole fish) from Svalbard were also analyzed for PBDEs (BDE-28, -47, -99, -100, -154, -209) and HBCD (Sørmo et al., 2006). The mean ΣPBDE concentration in 7 individuals was 1.99 ± 1.19 ng/g lw. On a wet weight basis, the concentrations were somewhat higher than those seen by Wolkers et al. (2006a): 0.097 ng/g ww for BDE-47 and 0.02 ng/g ww for BDE-99. BDE-209 and ΣHBCD were also analyzed in polar cod and found to be 0.20 ± 0.13 ng/g lw and 1.89 ± 0.56 ng/g lw, respectively. Thus, the ΣHBCD concentration was very similar to the ΣPBDE concentration. As HBCD was analyzed by GC-MS, no information is available on the isomer composition.

Polar cod were collected from the Barents Sea east of Svalbard in 2004 and liver samples analyzed for BDE-28, -47, -71, -77, -99, -100, -153, -154 and -183 (Haukås et al., 2007). Only BDE-28 and -47 were detected in all samples at mean concentrations of approximately 1.0 and 2.5 ng/g lw, respectively.

Polar cod and redfish (*Sebastes mentella*) collected from Davis Strait in October of 2000 and 2001 were analyzed as part of a bioaccumulation study in the Eastern Canadian Arctic (Tomy et al., 2008b). Geometric mean ΣPBDE (BDE-47, -85, -99, -100, -153, -154 and -209) concentrations were eight times higher in polar cod than in redfish (23 and 3 ng/g lw, respectively). Similarly high concentrations in polar cod were found by Morris et al. (2007) (23 ng/g lw), while Sørmo et al. (2006) only detected 2.0 ng/g lw in polar cod from a similar geographic region, when BDE-209 is included. The authors therefore concluded that broad geographical differences in PBDE contamination occurred at the base of the Arctic marine food web. In both fish species, BDE-209 was the predominant BDE congener, accounting for 75% and 60% of ΣPBDE in polar cod and redfish, respectively (Tomy et al., 2008b). The authors discussed possible exposure of these fish to zooplankton and limited metabolic capacities in metabolizing or eliminating BDE-209 as possible explanations for the predominance of BDE-209. Geometric mean ΣHBCD concentrations determined using LC-MS-MS were slightly higher in redfish (2.0 ng/g lw) than in polar cod (0.42 ng/g lw), and the diasteromeric composition differed with about 90% of ΣHBCD in redfish being α-HBCD, while the percentage of α-HBCD in polar cod was about 70% .

PBDEs were also analyzed in Atlantic cod from the northern Norwegian coast and capelin (*Mallotus villosus*) from the open Barents Sea (Bakke et al., 2008). Twenty-five individuals of capelin were sampled in February 2007 and pooled into five samples as whole fish, representing different locations. For analysis of Atlantic cod liver, 25 individual fish were collected from four stations in 2005/2006: Bjørnerøya (68.2°N, 14.8°E), Leisundet (70.3°N, 21.4°E), Revsbotn (70.7°N, 24.5°E) and Varangerfjorden (69.9°N, 29.7°E). In capelin, ΣPBDE (BDE-28, -47, -99, -100, -153, -154 and -183) concentrations ranged from 0.047–0.18 ng/g ww with a mean of 0.13 ng/g ww (no information on lipid reported). The lowest concentrations were found in fish from the two stations in the open sea, while fish from coastal stations had higher concentrations. Besides their low trophic position in the food chain, age can be an important factor explaining the low concentrations in capelin, as individuals in the Barents Sea do not usually exceed 4–5 years of age (Bakke et al., 2008). The authors point out that direct comparisons with other species or other analyses are difficult as whole fish were analyzed. ΣPBDE levels in Atlantic cod liver were higher than those in capelin and ranged from 3.4 ng/g ww at Leisundet to 9 ng/g ww at Bjørnerøya. BDE-47 was the main congener in all samples and accounted for 83–89% and 61–79% of ΣPBDE in capelin and Atlantic cod, respectively. BDE-28 -66, -99 and -154 were the only other congeners above the limits of detection in capelin. One sample of capelin differed in its composition from the other samples, as it did not contain BDE-28 or BDE-154, but BDE-66 was present in detectable concentrations.

A study from Alaska included sablefish (*Anoplopoma fimbria*), Pacific halibut (*Hippoglossus stenolepsis*), Chinook salmon (*Oncorhynchus tshawytscha*) and pink salmon (*Oncorhynchus gorbuscha*) in 16, 3, 2 and 2 individuals, respectively (G. Ylitalo, National Oceanic and Atmospheric Administration, personal communication). The samples were collected in southeast Alaska in 2003 (Chinook and pink salmon) and 2003/2004 (sablefish and Pacific halibut). ΣPBDE concentrations were below detection limits in all *Oncorhynchus* individuals, and ranged from below detection limits to 13 ng/g lw (2.6 ng/g ww) and 22 ng/g lw (0.6 ng/g ww) in sablefish and Pacific halibut, respectively. The mean concentrations and standard deviations in sablefish and Pacific halibut were 6.2 ± 1.1 ng/g lw (1.2 ± 0.23 ng/g ww) and 7.3 ± 9.0 ng/g lw (0.20 ± 0.24 ng/g ww), respectively.

Shaw et al. (2008) analyzed farmed and wild salmon from different regions, among these 3 individuals of wild Chinook salmon from the Aleutian Islands, Alaska. Of the nine BDE congeners included in this study (BDE-28, -47, -66, -85, -99, -100, -138, -153 and -154), only BDE-47, -99 and -100 were above the detection limits in the wild salmon samples. BDE-47 accounted for 38–58% of ΣPBDE in all samples of salmon. Congeners below the limit of detection were treated as half the limit of detection. Mean ΣPBDE concentration in the wild salmon was 9.3 ng/g lw (0.71 ng/g ww) and thus not statistically different from the mean concentration of all samples of farmed salmon (0.92 ng/g ww). The authors noted that Chinook salmon fed at a higher trophic level than Atlantic salmon, which could explain the lack of difference from farmed salmon. The concentration in the wild salmon was slightly higher than previously reported for Alaskan Chinook salmon (0.5–0.6 ng/g ww; Easton et al., 2002; Hites et al., 2004). The removal of skin led to slightly lower PBDE concentrations in the wild salmon, which, however, was not consistent for the entire set of samples. The lipid content was reduced considerably by removing the skin, and lower concentrations of PCBs had previously been shown in skin-off samples (Shaw et al., 2006). This result indicated different mechanisms and binding affinities for PBDEs and PCBs (Shaw et al., 2008). In general, the authors did not find any clear correlation between lipids and PBDEs or between PCBs and PBDEs. The PCB/PBDE ratios in the wild and farmed salmon were 8 and 18, respectively, possibly reflecting the differences in exposure history, migratory patterns and habitats.

GC-MS and LC-MS-MS were applied in a screening study of HBCD, TBBPA and dimethyl-TBBPA in biota from the marine environment of Greenland and the Faroe Islands (Frederiksen et al., 2007). GC-MS analysis leads to conversion of HBCD isomers to one isomer yielding results for total HBCD whereas LC-MS-MS separates the different isomers and provides isomer-specific HBCD results. Ten liver samples of shorthorn sculpin collected in central East and West Greenland in 2002, respectively, were pooled into two samples of 5 individuals for

each location. The results are considered preliminary as ^{13}C-labelled HBCD standards were not available at the time of the study and are presented as minimum values, as no recovery correction was performed. For the same extracts, higher ΣHBCD concentrations were found for the GC-MS than for the LC-MS-MS determination: For the two pools from East Greenland, GC-MS analysis gave 3.79 ng/g lw and 4.92 ng/g lw, versus 1.82 ng/g lw and 1.78 ng/g lw obtained by LC-MS-MS. Neither β- nor γ-HBCD were detected, i.e., α-HBCD accounted for 100% of ΣHBCD. Samples from West Greenland had lower HBCD concentrations and these were often below the detection limit except for one pooled sample from West Greenland analyzed by GC-MS (1.21 ng/g lw). Thus, there seem to be indications of a similar east>west gradient for HBCD to that consistently found for organochlorine contaminants and PBDEs (e.g. Riget et al., 2004; Vorkamp et al., 2004a). The difference in the measured concentrations between the two methods was attributed to uncertainty in the instrumental analysis, i.e. low responses and broad peaks in the GC-MS analysis and the risk of ion suppression in the LC-MS-MS method. TBBPA and dimethyl-TBBPA were not detected.

These new results increase the spatial coverage of BFRs in marine fish to Alaska and Canada as well as increase information for Greenland and Svalbard. More species have also been included. PBDE concentrations ranged from 2–41 ng/g lw in the different species. Polar cod from Canada had 10 times higher concentrations than from Svalbard. PBDE concentrations were higher in Greenland halibut than in polar cod from the same location in Svalbard, probably due to their exposure as benthic and pelagic feeders, respectively. PBDE concentrations in wild salmon from Alaska were not statistically significant from the mean PBDE concentration of a group of farmed salmon from different regions. BDE-209 was found in several species when included in the analysis, and in the case of redfish and polar cod from Canada, it was the predominant BDE congener. More data on HBCD in marine fish have become available. HBCD concentrations are similar in polar cod from Canada and Svalbard, redfish from Canada and shorthorn sculpins from Greenland (range 0.42–1.8 ng/g lw). HBCD concentrations were similar to ΣPBDE concentrations for redfish from Canada and polar cod from Svalbard, but were much lower for polar cod from Canada. Where isomer-specific HBCD analysis was performed, α-HBCD was the only detectable isomer. HBCD concentrations were higher in shorthorn sculpin from East Greenland than from West Greenland, possibly indicating a similar east>west gradient as found for organochlorines.

4.5.4. Seabirds

Arctic marine birds have been in focus in several previous studies investigating BFRs in the Arctic (Braune et al., 2005; Braune et al., 2007). Since contaminant burdens in the egg reflect residues assimilated by the female, seabird eggs have often been used to monitor trends and patterns of contaminants in the Arctic marine environment. In the previous review, BDE-47 was found to be the predominant BDE congener in seabirds in the Arctic (de Wit et al., 2006). ΣPBDE concentrations generally ranged from 20–100 ng/g lw, but glaucous gulls from Bjørnøya had the highest ΣPBDE concentrations, up to 1400 ng/g lw. Higher ΣPBDE concentrations were found in black guillemots (*Cepphus grylle*) from East Greenland compared to West Greenland, and higher concentrations were also seen around Svalbard and northern Norway compared to Arctic Canada. BDE-209, HBCD and PBBs were also found in glaucous gulls.

Yolk sacs of newly hatched chicks of Brünnich's guillemot (also known as thick-billed murres) (*Uria lomvia*) and common eider (*Somateria mollissima*) from Kongsfjorden, Svalbard (79°N) collected in 2002 were analyzed for PBDEs (BDE-28, -47, -99, -100, -153, -154) and HBCD (Murvoll et al., 2007). ΣPBDE concentrations were approximately 40 times higher in Brünnich's guillemot (mean of 90 ng/g lw, 25 ng/g ww) compared to common eider (means of 2.1 ng/g lw, 0.4 ng/g ww). A different PBDE congener pattern was observed, with higher proportions of BDE-47 in Brünnich's guillemot and higher proportions of BDE-153 in the common eider. The mean HBCD concentration in Brünnich's guillemot was 35.4 ng/g lw (10 ng/g ww), whereas in common eider, it was detected in only one sample at 6.23 ng/g lw (1.3 ng/g ww). These differences may be due to differences in diet as guillemots are piscivores, whereas the eiders are benthic feeders.

In a recent report, levels of PBDEs and HBCD in eggs of Brünnich's guillemot collected at Bjørnøya and Kongsfjorden, Svalbard in 2007 are presented (Bakke et al., 2008). BDE-47 was the most abundant congener in all samples. ΣPBDE (BDE-28, -47, -100, -99, -138, -153, -154, -183, -206, -207, -208, -209) concentration was 16.7 ng/g lw for the samples collected in the Kongsfjorden and 17.2 ng/g lw for the samples collected at Bjørnøya. BDE-209 could be detected in the samples from Kongsfjorden and Bjørnøya at low ng/g lw levels. HBCD was analyzed in the same sample set as well. HBCD concentrations were 12 and 29 ng/g lw at Kongsfjorden in 2002 and 2007, respectively, and on Bjørnøya, 22 and 17 ng/g lw in the years 2003 and 2007, respectively. No spatial trends between Bjørnøya and Kongsfjorden could be detected.

Eggs ($n=15$) of northern fulmars (*Fulmarus glacialis*) and thick-billed murres (Brünnich's guillemot) were collected in 2006 on the basis of one egg per nest from Prince Leopold Island (74°02′N, 90°05′W) in Lancaster Sound, Canada (Braune, 2007). The samples were analyzed in pools of 3 eggs each (15 eggs per collection = 5 pools of 3 eggs each for each species). Mean ΣPBDE concentrations (BDE-17,-28, -49, -47, -66, -100, -99, -85, -153, -138, -183) were approximately 12 ng/g lw in northern fulmars and 25 ng/g lw in thick-billed murres. HBCD concentrations were 28 ng/g lw in northern fulmars and 7.4 ng/g lw in thick-billed murres. The ratio of total HBCD to ΣPBDE was considerably higher in the northern fulmars than in the thick-billed murres, which may be related to differences in overwintering habits. Northern fulmars may overwinter in the northeast Atlantic, which is influenced by European emissions, whereas thick-billed murres overwinter along southwestern Greenland, which may be more influenced by North American emissions. BDE-209 was analyzed for but not detected in any of the samples. BDE-47 was the predominant BDE congener in both species.

Fängstrom et al. (2005) determined PBDEs (BDE-47, -77, -99, -100, -138, -153, -154 and -209) in eggs, fat and muscle tissue of northern fulmars collected in 2000–2001 from the Faroe Islands. BB-153 was also determined. The ΣPBDE concentration range (including BDE-209 when detected) was 11–42 ng/g lw (geometric mean — 21 ng/g lw) in fulmar eggs, 5.8–110 ng/g lw (geometric mean — 16 ng/g lw) in adult fulmar muscle, 4.7–68 ng/g lw (geometric mean — 11 ng/g lw) in juvenile fulmar muscle and 11–78 ng/g lw (geometric mean — 19 ng/g lw) in the subcutaneous fat of juvenile fulmars. BB-153 concentration ranges were 3.7–51, 6.2–44, 0.26–2.6 and 0.34–3.3 ng/g lw, respectively, in the same samples. The geometric means of the ΣPBDE concentrations in fulmar eggs were similar in both 2000 and 2001, 22 and 21 ng/g lw, respectively. BDE-209 was either undetectable or present at very low concentrations in all samples. These results indicate only limited PBDE contamination in Faroe Island seabirds.

PBDEs (BDE-28, -47, -99, -100, -153, -154/BB-153, -183 and -209) in fulmar eggs collected at Viðareiði on the Faroe Islands in 2003 were also studied by Karlsson et al. (2006). The levels of BDEs determined were similar to those reported earlier by Fängstrom et al. (2005), with ΣPBDE concentrations ranging from 16 to 43 ng/g lw and with a mean of approximately 25 ng/g lw. BDE-153 and -154 were the predominant congeners. BTBPE was detected in 8 of 9 samples at a maximum concentration of 0.17 ng/g lw, and BDE-209 was detected in only 1 of 9 samples at 7.2 ng/g lw (Karlsson et al., 2006).

Knudsen et al. (2007) collected blood and liver samples from northern fulmars breeding on Bjørnøya in the Norwegian Arctic for analysis of PBDEs (BDE-28, -47, -66, -49 + 71, -77, -85, -99, -100, -119, -138, -153, -154, -183, -196, -200, -209) as well as a suite of other

halogenated contaminants. Liver samples were also analyzed for HBCD, PBBs (BB-15, -153) and tribromoanisole. Only one liver sample contained quantifiable amounts of PBDE and the only congener detected was BDE-209, exhibiting very high levels of 5255 ng/g lw (Knudsen et al., 2007). This concentration was similar to that for ΣPCB, and the authors suggest that this high BDE-209 concentration may be due to analytical errors. Liver samples also had mean HBCD, PBB and tribromoanisole concentrations of 14.8, 6.6 and 0.6 ng/g lw, respectively. Only BDE-47 and -99 were analyzed in plasma samples and the concentrations ranged from not detected to 39.5 ng/g lw.

Concentrations of polybrominated compounds (PBDEs, HBCD, OH-PBDEs and MeO-PBDEs) were analyzed in common guillemot (*Uria aalge*) eggs collected from sampling locations in the Faroe Islands (Sandøy in 2003), Iceland (Vestmannaeyjar in 2002), Norway (Bjørnøya, Hjelmsøya, Sklinna in 2005) and the Baltic Proper (St. Karlsö in 2003) (Jörundsdóttir et al., 2008). The samples from Iceland and the Faroe Islands had somewhat higher mean ΣPBDE (BDE-47, -99, -153) concentrations (59 and 37 ng/g lw, respectively), compared to Norwegian samples (11–15 ng/g lw), but these were much lower than concentrations in samples from the Baltic Proper (155 ng/g lw). BDE-209 was detected in some eggs from Iceland (3.2 ng/g lw) and the Faroe Islands (2.4 ng/g lw) but was otherwise below the detection limit. The predominant BDE congener in all eggs was BDE-47. Mean HBCD concentrations in eggs were fairly similar for all sites (22–42 ng/g lw) except in the Baltic Proper, where concentrations were much higher (340 ng/g lw). Both 2′-OH-BDE-68 and 6-OH-BDE-47 were detected in eggs from all sites, with concentrations in the eggs from Iceland, the Faroe Islands and Norwegian sites ranging from 0.21–0.39 ng/g lw for 2′-OH-BDE-68 and 2–10 ng/g lw for 6-OH-BDE-47. Interestingly, the 6-OH-BDE-47 concentrations are as high as those for BDE-47, a very strong indication that this compound is not a BDE-47 metabolite. The samples from the Baltic Sea have higher 6-OH-BDE-47 concentrations than the samples from the North Atlantic, and the difference between the two areas is much greater than for the parent compounds. 2′-MeO-BDE-68, 6-MeO-BDE-47 and 6-MeO-BDE-90 were only detected in eggs from Baltic Sea guillemots, which may be related to massive algal blooms that occur each year in the Baltic Sea due to eutrophication.

Several seabird species were collected from the Seven Islands Archipelago off the coast of the Kola Peninsula of the Russian Arctic (68°N, 37°E) and liver samples were analyzed for PBDEs (Savinova et al., 2007). Mean ΣPBDE concentrations (BDE-17, -28, -47, -66, -71, -99, -100, -138, -153, -154, -183, -190) were 20 ng/g lw in common guillemot, 66 ng/g lw in Brünnich's guillemot, 170 ng/g lw in black-legged kittiwake (*Rissa tridactyla*), 190 ng/g lw in Atlantic puffin (*Fratercula arctica*) and 550 ng/g lw in razorbill (*Alca torda*). The predominant BDE congener found in all samples was BDE-47. PBDE concentrations found in the common guillemot liver samples were comparable to those seen in eggs of the same species from northern Norway and Bjørnøya (Bakke et al., 2008). However, ΣPBDE concentrations in the Brünnich's guillemot liver samples from the Kola Peninsula were somewhat higher than seen in eggs from the same species from Bjørnøya.

Liver from black guillemots collected from the Barents Sea east of Svalbard in 2004 were analyzed for PBDEs (BDE-28, -47, -71, -77, -99, -100, -138, -153, -154, -183) (Haukås et al., 2007). The mean ΣPBDE concentration (BDE-28, -47, -99, -100, -154) was 51 ng/g lw and BDE-47 was the predominant congener. These concentrations are similar to those found in black guillemot liver samples collected in 2000 from East Greenland (73 ng/g lw), but are higher than those seen for black guillemot from West Greenland (25 ng/g lw) (Vorkamp et al., 2004b).

A screening study with the purpose of determining the presence of HBCD, TBBPA, and Me-TBBPA in various types of marine biota was performed by Frederiksen et al. (2007). The samples analyzed in this study included liver and subcutaneous fat from northern fulmars from the Faroe Islands collected in 1998/1999 as well as liver and egg samples of Greenland black guillemots collected in 2000. The sampling process included pooling of individual samples (typically pools of five subsamples based on equal amounts). α-HBCD was the only HBCD isomer detected in quantifiable amounts in black guillemot liver from East Greenland (8.4 and 11.20 ng/g lw) and in black guillemot eggs from East Greenland (1.9 and 4.10 ng/g lw). No HBCD could be found in the samples from West Greenland. However, in subcutaneous fat of female and male northern fulmars collected on the Faroe Islands, concentrations of 19.20 and 41.9 ng/g lw, respectively, of α-HBCD, as well as 6.6 and 2.4 ng/g lw, respectively, of γ-HBCD were reported. When analysing the livers of the same individuals, only α-HBCD was found (10.3 and 23.1 ng/g lw, respectively for females and males). No TBBPA or Me-TBBPA was detected in any of the samples.

Eggs from common guillemots (murres), thick-billed murres, glaucous-winged (*Larus glaucescens*) and glaucous gulls from numerous sites around Alaska, collected in 2002–2005, were analyzed for 33 PBDEs within the Seabird Tissue Archival and Monitoring Project (STAMP) (Roseneau et al., 2007). Currently, the STAMP study area consists of 33 sampling sites in the Bering and Chukchi seas, Gulf of Alaska, and northeastern Pacific Ocean. Common guillemot eggs were collected from St. Lazaria, Middleton, East Amatuli, Duck and Gull Islands (Gulf of Alaska) in 2003 and 2004 as well as from five sites in the Bering Sea (Bogoslof Island, Cape Pierce, Sledge Island, Bluff, Cape Denbigh) in 2005. An east–west spatial trend was observed within the Gulf of Alaska with highest mean ΣPBDE values (BDE-47, -99, -100, -153, -154) found in common guillemot samples from St. Lazaria Island (220 and 110 ng/g lw; 23 and 12 ng/g ww for 2003 and 2004, respectively) compared to Middleton Island (21 and 72 ng/g lw; 2.1 and 7.4 ng/g ww for 2003 and 2004, respectively) and East Amatuli Island (19 ng/g lw; 1.9 ng/g ww for 2003), which is the farthest west. ΣPBDE concentrations in common guillemot eggs from Gull and Duck Islands collected in 2004 had intermediate ΣPBDE concentrations of 35 and 69 ng/g lw (3.6 and 6.9 ng/g ww), respectively. Common guillemot eggs from the Bering Sea sites had similar ΣPBDE concentrations, ranging from 2.8 to 23 ng/g lw (0.31 to 2.3 ng/g ww). These concentrations are similar to those seen at Amatuli Island, also indicating an east–west geographic trend for PBDEs in guillemots, with lowest concentrations in the Bering Sea and highest concentrations on the southwest Alaskan coast. The birds on the southwest Alaskan coast are probably more affected by their proximity to populated areas and emission sources.

Thick-billed murre eggs were collected from St. Lazaria Island in the Gulf of Alaska and three sites in the Bering Sea: St. George Island, St. Lawrence Island and Cape Lisburne (Roseneau et al., 2007). As for the common guillemots, ΣPBDE concentrations were higher in the St. Lazaria Island thick-billed murre eggs (170 ng/g lw, 18 ng/g ww) than in the Bering Sea thick-billed murres (range of 7.7–11 ng/g lw, 1.0–1.2 ng/g ww), confirming higher PBDE concentrations in the Gulf of Alaska. BDE-47 was the predominant congener in both common guillemot and thick-billed murre eggs.

Eggs of glaucous gulls were collected from the Chuckchi Sea (Noatak River Delta) and the Bering Sea (Hooper Bay, Penny River Delta) and from glaucous-winged gulls from the Bering Sea (Ualik Lake, Shaiak Island) and the Gulf of Alaska (Viesokoi Rock, Middleton Island) in 2005 (Roseneau et al., 2007). Mean ΣPBDE concentrations were 150 ng/g lw (12 ng/g ww) and 55 ng/g lw (4.3 ng/g ww) in glaucous gull eggs from the Chukchi Sea and Bering Sea sites, respectively. Glaucous-winged gull eggs had mean ΣPBDE concentrations of 13 ng/g lw (0.95 ng/g ww) in the Bering Sea sites but highest ΣPBDE concentrations were found in the gull eggs from the Gulf of Alaska sites (1200 ng/g lw, 91 ng/g ww). BDE-28, -47 and -66 were abundant in the eggs sampled in the Bering Sea, and BDE-99 and -100 were also detected in the eggs from the Chuckchi Sea. In the samples from the Gulf of Alaska, BDE-28, -47, -66, -99, -100, -138, -153, -154, -183 were found with BDE-28, -47, -66, -99, and -100 detected in all

eggs analyzed ($n=6$). BDE-47 was the predominant congener in all gull eggs.

Liver samples from glaucous gulls collected in 2004 from the Barents Sea east of Svalbard were analyzed for PBDEs (BDE-28, -47, -71, -77, -99, -100, -138, -153, -154, -183) (Haukås et al., 2007). The mean ΣPBDE (BDE-28, -47, -71, -99, -100, -153, -154) concentration was 950 ng/g lw. BDE-47 was the predominant congener.

Verreault et al. (2007b) reported PBDEs, PBBs, HBCD, and PBDE metabolites and/or naturally occurring compounds with similar structures (MeO- and OH-PBDEs) in liver, blood and whole body homogenate samples of adult glaucous gulls collected in 2002 from Svalbard. Mean ΣPBDE (BDE-28, -47, -66, -85, -99, -100, -138, -153, -154/BB-153, -183, -190, -209) concentrations were 9400 ng/g lw (51.5 ng/g ww) in blood and 12,500 ng/g lw (522 ng/g ww) in liver. ΣPBDE concentrations in whole body homogenates were similar or lower, 9500 ng/g lw (178 ng/g ww) with feathers and 4400 ng/g lw (202 ng/g ww) without feathers. HBCD and PBB-101 were detected in all samples analyzed as well. HBCD concentrations were 600 ng/g lw (3.3 ng/g ww) in blood, 1800 ng/g lw (76 ng/g ww) in liver. HBCD concentrations in whole body homogenates concentrations were higher, 6300 ng/g lw (117 ng/g ww) with feathers and 2000 ng/g lw (91 ng/g ww) without feathers. BB-101 concentrations were lower, 51, 49, 35 and 32 ng/g lw (0.28, 2.0, 0.65 and 1.5 ng/g ww) for blood, liver, whole body with and without feathers, respectively. MeO-PBDE and OH-PBDE were detected as well. Mean concentrations of MeO-PBDEs were 2.8 and 32 ng/g ww in blood and liver, respectively. Mean OH-PBDE concentrations were 3.5 and 3.6 ng/g ww in blood and liver, respectively.

In an additional study, Verreault et al. (2007a) investigated HxBBz, BTBPE, PBEB, PBT, and total-(α)-HBCD, in egg yolk and plasma of male and female glaucous gulls collected in 2006 from Bjørnøya in the Norwegian Arctic. Also examined were BDE-209 and 38 tri-nonaBDE congeners and BB-101. HxBBz, BTBPE, PBEB, and PBT had high detection frequencies and highly variable concentrations in male and female plasma and egg yolk samples, and their concentrations ranged from non-detectable (<0.02–0.27 ng/g ww) to 2.64 ng/g ww. The detection frequencies and concentrations of non-BDE BFRs were generally highest in plasma of males relative to females. ΣPBDE concentration ranges were 2.49–54.5 ng/g ww in plasma (males and females) and 81.2–321 ng/g ww in egg yolk. Mean ΣPBDE concentrations were 2400 ng/g lw (males) and 1000 ng/g lw (females) (18.8 and 8.55 ng/g ww, respectively) in plasma and 530 ng/g lw (163 ng/g ww) in egg yolk. BDE-209 was not detected, whereas six octaBDEs (i.e., BDE-196, -197, -201, -202, -203, and -205), as well as three nonaBDEs (i.e., BDE-206, -207, and -208, and potential BDE-209 debromination products) were found sporadically in plasma and egg yolk. HBCD concentrations were highest among the non-PBDE BFRs with mean concentrations of 220 (males) and 250 (females) ng/g lw (1.73 and 2.07 ng/g ww, respectively) in plasma and 64 ng/g lw (19.8 ng/g ww) in egg yolk. Next highest concentrations were found for HxBBz with mean concentrations in egg yolk of 3.5 ng/g lw (1.1 ng/g ww), which was within a range comparable to the minor PBDEs monitored (e.g., BDE-28, -116 and -155). The results from this study suggest that in addition to PBDEs, several current use, non-BDE BFRs undergo long-range atmospheric transport, bioaccumulate at low levels in glaucous gulls from the Norwegian Arctic and are maternally transferred to eggs.

Savinov et al. (2005) determined BDEs (17 congeners, not fully defined but including BDE-209) in livers of glaucous gulls collected in 2001 from West Spitsbergen, Svalbard. ΣPBDE concentrations in female and male gulls ranged from 3.5–100 ng/g ww (mean — 570 ng/g lw) and 6.1–27 ng/g ww (mean 430 ng/g lw), respectively. The highest concentrations were found for BDE-47, followed by BDE-100, -99, -209, -153 and -154. Although the average concentration of BDE-209 was not high (0.95 ng/g ww, 26 ng/g lw)) the levels in two individuals reached 7.5 and 87 ng/g ww. A similar study was conducted on Bjørnøya in 1995. The median concentration of BDE-47 was 3.6 times higher than in a similar study on Bjørnøya in 1995, but levels of BDE-99 were similar. In this study, 15 MeO-BDE and 15 OH-BDE congeners were also determined in the plasma samples, but less than half were detected in glaucous gulls. The dominant compound was consistently 3-MeO-BDE-47, followed by 4′-MeO-BDE-49 and 6-MeO-BDE-47 (due to a lack of standard materials, it should be noted that 3-MeO-BDE-47 may actually be 2′-MeO-BDE-66, or a combination of 3-MeO-BDE-47 and 2′-MeO-BDE-66). 3-OH-BDE47, 4′-OH-BDE-49 and 6-OH-BDE-47 were also detected in plasma of glaucous gulls.

Murvoll et al. (2006) investigated the occurrence of PBDEs (BDE-28, -47, -99, -100, -153, -154) and HBCD as well as PCBs and OCPs in yolk sacs of black-legged kittiwake hatchlings from Kongsfjorden at Svalbard (79°N) and from Runde, an island on the coast of Norway (62°N) collected in 2002. No differences between the two populations of kittiwake hatchlings were found regarding levels of ΣPBDEs (528 and 461 ng/g lw, for Runde and Kongsfjorden, respectively) indicating long-range transport as the main source of exposure. BDE-47 was the predominant congener at both sites. HBCD levels were significantly lower in hatchlings from Kongsfjorden (118 ng/g lw) compared to those in hatchlings from Runde (260 ng/g lw). Higher levels of PCBs and OCPs were found in hatchlings from Kongsfjorden than in hatchlings from Runde. Svalbard kittiwake eggs have higher PBDE concentrations compared to kittiwake liver samples from the Kola Peninsula.

Braune et al. (2007) analyzed eggs of ivory gulls (*Pagophila eburnea*) for PBDE, HBCD and PBBs, collected in 2004 from Seymour Island in the Canadian Arctic. The ΣPBDE concentration (BDE-17, -28, -30, -47, -49, -66, -85, -99, -100, -138, -153) was 45 ng/g lw (4.5 ng/g ww). BDE-183 and -209 were below the limits of detection in the samples. BDE-47 was the predominant congener, followed by BDE-154/BB-153 and BDE-99. The mean concentration of total-(α)-HBCD was 2 ng/g lw (0.2 ng/g ww). The BB-101 concentration was 6.6 ng/g lw (0.7 ng/g ww).

In spring 2001 and 2002, 214 gull eggs were collected at 12 different sites in northern Norway, the Faroe Islands and Svalbard. The eggs were collected from herring gulls (*Larus argentatus*), great black-backed gulls (*L. marinus*), lesser black-backed gulls (*L. fuscus*) and glaucous gulls (Pusch, 2004; Pusch et al., 2005). BDE-47 and -99 were the most abundant PBDEs observed. The highest ΣPBDE (BDE-47, -99) levels were found in herring gull eggs (0.14 ng/g ww), and black-backed gull eggs from Kirkjubøreyni (0.15 ng/g ww). Samples from glaucous gulls, collected at Svalbard showed the lowest levels with ΣPBDE of 0.017 and 0.014 ng/g ww for Ny Ålesund and Isfjord, respectively. Lipid weight values were not reported.

New seabird data have been generated for Alaska and parts of the European Arctic. BDE-47 was the predominant congener in most species, except in northern fulmars, where BDE-153 was the most abundant congener. Lowest ΣPBDE concentrations were found in eiders, which feed on invertebrates, higher concentrations were found in alcids and northern fulmars, and highest concentrations were found in several gull species, which are known to feed at higher trophic levels. Spatial trends were evident both in Alaska, on Greenland as well as across the Arctic (see Section 6 Spatial trends). More data are now available on the presence of PBBs, HBCD and BDE-209 in several more seabird species, and for the first time, several new BFRs, BTBPE, PBEB, PBT and HxBBz, have been found in Arctic seabirds.

4.5.5. Marine mammals

4.5.5.1. Seals. In the previous review, tri-hexaPBDE concentrations in ringed seal blubber were summarized from a number of locations including Russia, numerous sites in Canada, several sites on Greenland and from Svalbard. ΣPBDE concentrations were in the range of 1–100 ng/g lw (de Wit et al., 2006). Highest mean ΣPBDE

concentrations were found in seals from northeast Greenland and Svalbard (30–58 ng/g lw). HBCD was found in ringed seals from Svalbard and several sites in Canada, with concentrations in the range of 1.3–35 ng/g lw, with highest concentrations on Svalbard. BDE-209 was only included sporadically and not found above the limits of detection. PBDE concentrations in larga seals (*Phoca largha*) and bearded seals (*Erignatus barbatus*) from the Russian Arctic were similar to those in ringed seals and were in the range of 0.51–1.9 ng/g ww (no information on lipid weight was available).

Blubber samples of male ringed seals were collected in 2002 and 2003 on Holman Island in the western Canadian Arctic (Ikonomou et al., 2005). The age group covering 0–15 years had mean ΣPBDE concentrations of approximately 5 ng/g lw in 2002 and 3.2 ng/g lw in 2003. However, this last point was only based on two samples. Concentrations were higher in older animals: Individuals of 16–35 years of age had mean ΣPBDE concentrations of approximately 6.8 ng/g lw in 2002 and 5.9 ng/g lw in 2003. BDE-47 seems to account for 60–70% of ΣPBDE in the seal samples, with no apparent difference between the age groups.

Blubber samples from female ringed seals from five locations in the Canadian Arctic (Pangnirtung, Resolute Bay, Sachs Harbour, Arviat and Holman Island) collected in 2006 were analyzed for PBDEs, HBCD, BTBPE and DBDPE as part of a time trend study (Muir et al., 2007). PBDEs were detected in most blubber samples at low ng/g concentrations and BTBPE was detected in about 10% of the samples, while HBCD and DBDPE were below the detection limit in all samples. The mean ΣPBDE concentrations of 5–12 individuals were highest at Arviat (21 ng/g lw) and lowest at Resolute Bay (4.2 ng/g lw). The intermediate mean ΣPBDE concentrations were 4.5 ng/g lw at Sachs Harbour, 6.3 ng/g lw at Holman Island and 9.7 ng/g lw at Pangnirtung. ΣPBDE mainly consisted of BDE-47 and BDE-99. BDE-209 was detected at concentrations near the detection limit (0.1–0.2 ng/g lw). The concentrations of BTBPE ranged between <0.01 to 0.29 ng/g lw.

PBDEs (BDE-17, -28, -47, -49, -66, -85, -99, -100, -153, -154 and -183) were analyzed in blubber from ringed seals from Ittoqqortoormiit (Central East Greenland) collected in 2004 and Qeqertarsuaq (Central West Greenland) collected in 2004 and 2006 (Riget et al., 2006; Vorkamp et al., 2008). ΣPBDE was highest in ringed seals from East Greenland with a mean concentration of 32.5 ng/g lw (median of 27.2 ng/g lw). This concentration was approximately 20 times lower than the summed concentrations of 10 PCB congeners in the same samples. BDE-47 and BDE-99 accounted for approximately 75% and 10%, respectively, of ΣPBDE. In contrast to the relatively high proportion of BDE-153 found in polar bears (Muir et al., 2006b), this congener only contributed 1.7% to ΣPBDE in the ringed seal from East Greenland. A general concentration increase with age was found for both PBDEs and PCBs in the ringed seals from East Greenland. For male animals, the PBDE concentrations seemed to increase until the age of 10, but not beyond, while the accumulation in females was less clear, possibly due to contaminant transfer to the offspring.

The samples from Qeqertarsuaq (West Greenland) had considerably lower concentrations, with BDE-47 being the only congener consistently above the limit of detection (Vorkamp et al., 2008). Assigning a value of half the detection limit to congeners below the detection limit, median ΣPBDE in 2004 and 2006 were 3.94 and 5.15 ng/g lw, respectively. The corresponding median concentrations of BDE-47 were 2.38 and 2.84 ng/g lw in 2004 and 2006, i.e. an order of magnitude below the respective concentrations from East Greenland (Riget et al., 2006) but similar to concentrations from the Western Canadian Arctic (de Wit et al., 2006). BDE-209 was not included in ΣPBDE as it was not analyzed in all samples. Most samples had BDE-209 concentrations below the detection limit of 0.91–1.36 ng/g lw, but BDE-209 was detected in 5 of 20 individuals from 2006, in the range of 0.93–3.25 ng/g lw. No statistically significant differences in ΣPBDE concentrations were observed between male and female ringed seals. As the West Greenland study only included juvenile animals, gender-specific effects on concentrations were assumed to be low (Vorkamp et al., 2008).

Ringed seal samples from Central East and West Greenland were also included in a preliminary screening of HBCD, TBBPA and dimethyl-TBBPA, applying GC-MS and LC-MS-MS methods (Frederiksen et al., 2007). Liver and blubber samples of 10 individuals collected in 2002 were pooled into two samples for each matrix and site. TBBPA and dimethyl-TBBPA could not be detected in any of the samples. GC-MS analysis generally yielded higher HBCD concentrations than the LC-MS-MS method, possibly as a result of uncertainty in the instrumental analysis, e.g. low response, broad peaks (GC-MS) and ion suppression (LC-MS-MS). The authors noted that the concentrations should be regarded as minimum values since no recovery correction had been performed. α-HBCD accounted for close to 100% of ΣHBCD in the ringed seal samples. β- and γ-HBCD were present in a few of the samples, but always below the limits of quantification. For the two East Greenland pooled blubber samples, the HBCD concentrations were 4.69 and 6.92 ng/g lw (GC-MS) or 2.50 and 2.40 ng/g lw (LC-MS-MS). Pooled blubber samples from West Greenland were below limits of quantification in the LC-MS-MS method, and quantified as 1.85 and 1.54 ng/g lw in the GC-MS-analysis. Thus, concentrations were higher in the East Greenland samples, which was also seen for shorthorn sculpin liver and black guillemot liver and eggs in the same study. This means that HBCD seems to follow the same geographical trend as previously described for organochlorines and PBDEs (e.g. Riget et al., 2004; Vorkamp et al., 2004a).

Pooled liver samples gave somewhat unclear results for HBCD, with large variations between the two pools and conflicting findings for GC-MS and LC-MS-MS analysis (Frederiksen et al., 2007). In one pooled liver sample from East Greenland seals, HBCD was not detected in the GC-MS analysis, but had ΣHBCD concentration of 16.7 ng/g lw in the LC-MS-MS run and the second pooled sample had HBCD concentration of 13 ng/g lw (GC-MS) but not detected for LC-MS-MS. The two pooled liver samples from West Greenland had HBCD concentrations of 23 and not detected (GC-MS) and below detection in the LC-MS-MS method.

Ringed seals from Svalbard sampled in 2003 were analyzed for PBDEs (BDE-28, -47, -99, -100, -153, -154 and -209) (Sørmo et al., 2006). ΣPBDE concentrations in blubber from 6 individuals ranged from 42–94 ng/g lw, with a mean concentration of 59 ng/g lw. The median concentration (55 ng/g lw) was very similar to the mean. BDE-209 was only detected in one seal, at a concentration of 0.02 ng/g lw. Of the other BDEs, BDE-47 was found at the highest concentrations and accounted for 83% of ΣPBDE, on average. HBCD was analyzed in the same samples, with mean concentrations of 19.6 ng/g lw and a range of 14.6-35 ng/g lw.

In a study from Alaska, four species of seals, relevant for human consumption in Alaskan coastal communities, were analyzed for their blubber content of 38 mono-octaBDEs: ringed seal, bearded seal, spotted seal (larga seal) and ribbon seal (*Phoca fasciata*), all collected in 2003 (Quakenbush, 2007). Only 11 mono-tetraBDEs were detected. The mean concentrations based on 3–6 individuals increased in the order bearded seal (3.4 ng/g ww)<ringed seal (5.9 ng/g ww)< spotted seal (12.4 ng/g ww)<ribbon seal (16.5 ng/g ww). Except for ribbon seals (Little Diomede only), the animals originated from two locations, Little Diomede (65.7°N, 169.2°W) and Hooper Bay (61.5°N, 166°W), however, the data set was too small to allow for statistical tests of differences between locations. Compared with levels in seals from eastern Canadian or the European Arctic (e.g. Ikonomou et al., 2002; Riget et al., 2006), these BDE levels are relatively low, which is consistent with previous results for organochlorines in biota from Alaska (e.g. Muir et al., 1999). It is particularly interesting and a bit odd that the PBDE composition also seems to differ from that seen in other parts of the Arctic: of 38 mono- to heptabrominated congeners included in the analysis, only 11 were detected, with BDE-47 being the

highest brominated one. While BDE-47 generally is the predominant congener in marine mammals from the Arctic, it was only found in two samples of spotted seal. BDE-30 (2,4,6-triBDE) was the only congener detected in all samples. According to the author, spotted seal fed at the highest trophic level of the animals analyzed. Possible reasons for this deviating PBDE pattern that are discussed by the author are the distance of the Bering Sea from any anthropogenic sources of PBDEs, leading to possible degradation of PBDEs prior to entering the food chain or natural sources of brominated compounds occurring locally (Quakenbush, 2007). Because of the very different congener pattern found in these samples, the results need to be confirmed to exclude methodological errors as the source of these PBDEs.

Based on these most recent publications, the highest PBDE concentrations were found in ringed seals from Svalbard and, slightly lower, but in the same order of magnitude, East Greenland. A pronounced East Greenland>West Greenland difference was found, with West Greenland levels being similar to those from Holman Island in the western Canadian Arctic and Alaska. However, the Alaskan results differed considerably in terms of PBDE profiles, favoring the less brominated and most volatile BDEs. This might be related to long distances from the main anthropogenic sources and possible photodegradation during long range transport. On average, the HBCD concentration was approximately one-third that of ΣPBDE concentrations in ringed seals from Svalbard. HBCD and ΣPBDE concentrations were comparable in West Greenland, while ΣPBDE concentrations were clearly higher than HBCD concentrations in East Greenland ringed seals. HBCD was not detected in Canadian ringed seals. The East Greenland > West Greenland gradient also exists for HBCD, but is less pronounced than for PBDEs. Just as for fish, ΣHBCD consisted of close to 100% α-HBCD when stereoisomer analysis was performed. The new results for ringed seals confirm the levels and trends found in the previous review.

4.5.5.2. Walrus. Only limited data are available on the degree of BFR exposure in walrus (*Odobenus rosmarus*). Wolkers et al. (2006b) collected blubber samples from 17 adult, male walruses from eastern Svalbard in 2002–2004 and analyzed these for chlorinated and brominated contaminants. Concentrations of ΣPBDEs (BDE-28, -47, -99, -100, -153, -154) were relatively low and ranged from 2.1–75.9 ng/g lw, with a mean of 15.6 ng/g lw. BDE-47 was the predominant congener found. ΣPBDE contributed only 0.3% to the overall pollutant load. The wide range of concentrations (which was also seen for PCBs and other OCs) was attributed to higher trophic level feeding by some walrus, possibly on ringed seals, instead of on invertebrates.

PBDEs and HBCD were analyzed in the blubber of walrus collected from Cape Dorset in the eastern Canadian Arctic in 1998 by Tomy et al. (2008b). The samples came from both sexes. The mean ΣPBDE concentration (BDE-47, -85, -99, -100, -153, -154) was 0.4 ng/g lw. BDE-209 was analyzed for but not detected. HBCD was analyzed using LC-MS-MS and the geometric mean concentration of α-HBCD was 0.2 ng/g lw and for γ-HBCD, 0.4 ng/g lw.

4.5.5.3. Whales. Previous studies reported tri-hexaBDEs in beluga from Svalbard and several sites in Canada, with concentrations ranging from 15 to 161 ng/g lw. Highest PBDE concentrations were seen in the Svalbard population (29–161 ng/g lw) and lower concentrations at the Canadian sites (16–31 ng/g lw). HBCD was studied only in Canadian beluga and concentrations were in the range of 10–20 ng/g lw. Tetra-hexaBDEs were also found in harbour porpoise (*Phocoena phocoena*) around Iceland (25–95 ng/g lw), minke whales (*Balaenoptera acutorostrata*) from the Barents Sea (32–44 ng/g lw) and Norwegian Sea (400–440 ng/g lw) and long-finned pilot whales (*Globicephala melas*) from the Faroe Islands (61–3200 ng/g lw). For beluga and long-finned pilot whales, juveniles had higher concentrations than adults.

PBDEs and HBCD were analyzed in beluga blubber collected in the eastern Canadian Arctic from Frobisher Bay near Iqaluit in 1996 (Tomy et al., 2008b). The geometric mean ΣPBDE concentration (BDE-47, -85, -99, -100, -153, -154) was 12 ng/g lw and BDE-47 was the predominant congener. BDE-209 was analyzed for but not detected. HBCD analysis using LC-MS-MS gave geometric mean concentrations of 1.2 ng/g lw for α-HBCD and 0.2 ng/g lw for γ-HBCD.

PBDEs, OH-PBDEs and MeO-PBDEs were determined in the liver of beluga whales from western Hudson Bay in the Canadian Arctic collected in 2002–2003 (McKinney et al., 2006). The mean ΣPBDE concentration (BDE-47, -99, -100, -153, -154) was 53 ng/g lw, which is somewhat higher than seen in east Hudson Bay beluga (30 ng/g lw) (Tomy, 2007). BDE-47 was the predominant congener followed by BDE-100 and -99 (McKinney et al., 2006). Of the 15 OH-PBDEs, only two were detectable, but not quantifiable (notably 2′-OH-BDE-68 and 6-OH-BDE-47). Of the 15 MeO-PBDEs, only 2′-MeO-BDE-68 and 6-MeO-BDE-47 were detected ($n=2$) with concentrations of 43 and 100 ng/g lw. The OH-PBDEs and MeO-PBDEs most likely are of natural origin and accumulate in beluga whales.

Tomy et al. (2007) analyzed PBDEs, HBCD and BTBPE in blubber from Canadian Arctic beluga collected from several sites in 2002–2005. Highest concentrations of ΣPBDEs (17 congeners) were found in beluga blubber from Hudson Bay (Sanikiluaq, 35 ng/g lw) collected in 2003 and the southeastern Beaufort Sea (Hendrikson Island), and lowest from eastern Canadian Arctic stocks (Pangnirtung, Baffin Island, 7 ng/g lw) collected in 2002. However, in a later study, the ΣPBDE concentration was higher in Pangnirtung beluga collected in 2005 (30 ng/g lw) and 2006 (22 ng/g lw) (Tomy, 2007). HBCD concentrations were lower than PBDEs at each site, but showed a similar spatial pattern, with highest concentrations in Hudson Bay (3.9 ng/g lw), lower concentrations at Hendrikson Island (1.5 ng/g lw in 2005, 1.2 ng/g lw in 2006) and at Pangnirtung (1.2 ng/g lw in 2005 and 1.0 ng/g lw in 2006). BTBPE was observed in a few beluga samples from Hendrikson Island (0.1–2.5 ng/g lw) and from the central Canadian Arctic site (Arviat) at concentrations similar to those for HBCD.

Tomy et al. (2008a) investigated the occurrence of a new brominated flame retardant, 1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane (TBECH) in beluga blubber from animals collected from the Canadian Arctic (Hendrickson Island, Igloolik, Resolute and Pangnirtung) 2003–2006. PBDEs and HBCD were also analyzed. Technical grade TBECH consists of near equimolar amounts of α-TBECH and β-TBECH. β-TBECH was the only isomer observed in the samples and was detected in 17 of the 29 samples and from all four sampling sites. Observed concentrations of the β-isomer as measured by HRMS ranged from 1.1 to 9.3 ng/g lw with the following means: Hendrikson Island — 2.7 ng/g lw (only one sample), Igloolik — 6.1 ng/g lw, Pangnirtung — 2.8 ng/g lw and Resolute — 5.2 ng/g lw. In the same samples mean ΣPBDE concentrations were 18, 54, 25 and 36 ng/g lw for Hendrikson Island, Igloolik, Pangnirtung and Resolute, respectively (Tomy, 2007; Tomy et al., 2008a). HBCD was detected at much lower concentrations with similar means at all four sites: 1.3, 1.7, 1.2 and 0.73 ng/g lw at Hendrikson Island, Igloolik, Pangnirtung and Resolute, respectively.

BDE-47 and -99 were included in a study of contaminants in blubber of subadult beluga and narwhal (*Monodon monoceros*) from Svalbard collected in 1998–1999 (Wolkers et al., 2006a). Both are long-lived top predators in the Arctic marine food chain, hence they are exposed to high contaminant concentrations. Based on four samples of beluga and three samples of narwhal, mean concentrations of BDE-47 and -99 were 68.6 and 3.1 ng/g lw, respectively, in beluga, and 172.9 and 22.1 ng/g lw, respectively, in narwhal. Thus, ΣPBDE concentrations in narwhal are approximately three times higher than in beluga. The two species are closely related in terms of feeding habits and distribution, however, narwhal tend to feed on Greenland halibut to a greater extent than beluga (Wolkers et al., 2006a). Being

benthic feeders, Greenland halibut have higher contaminant levels than e.g. polar cod, a pelagic fish species (see Section 4.5.3 Marine fish). The BDE pattern was similar in both species, with BDE-47 being the predominant congener. The concentrations in beluga were similar to those from comparable samples from the same area sampled in 1998 (Wolkers et al., 2004) but about four times lower than in adult males from the St. Lawrence Estuary (Lebeuf et al., 2004). BDE-47 and -99 accounted for <1% of the total load of organohalogen compounds analyzed in the same samples.

PBDEs and HBCD were analyzed in the blubber of male narwhal collected from Broughton Island in the eastern Canadian Arctic in 2000 (Tomy et al., 2008b). The geometric mean ΣPBDE concentration (BDE-47, -85, -99, -100, -153, -154) was 18 ng/g lw. BDE-209 was analyzed for but not detected. Compared to data from Svalbard, narwhal from the eastern Canadian Arctic seem to have lower PBDE concentrations, but the data base is very small. Geometric mean HBCD concentrations were 2.9 ng/g lw for α-HBCD and 0.5 ng/g lw for γ-HBCD.

Frederiksen et al. (2007) analyzed HBCD (total and isomer-specific) and TBBPA in pooled blubber and liver samples from minke whales from West Greenland collected in 1999 and in long-finned pilot whales from the Faroe Islands collected in 2001. Only low concentrations of α-HBCD could be detected in the minke whale blubber (3 ng/g lw) and no HBCD was found in the liver samples of the minke whales. In contrast, liver and blubber samples from the long-finned pilot whales, showed high α-HBCD levels in juvenile and male individuals (80 and 91.1 ng/g lw, respectively) compared to females (26.2 ng/g lw). Liver concentrations showed no clear difference between the sexes and varied between 10.6 and 22.5 ng/g lw. The blubber sample from juveniles also contained β-HBCD at 40 ng/g lw. Due to the lack of labelled internal standards for HBCD, possible incomplete recoveries and/or supression in the LC/MS due to matrix effects the concentrations were not corrected. TBBPA could not be detected in any of the samples.

The mean ΣPBDE concentration (BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -183 and -209) in blubber from a pooled sample of West Greenland minke whale collected in 2004 was 63 ng/g lw (Sonne et al., 2006a).

Killer whale (*Orcinus orca*) populations are characterized by different migratory and dietary habits. In the North Pacific both resident (salmonid fish-eating) and transient (marine mammal-eating) killer whale populations can be found. Transient whales from Alaska can be grouped by geographical collection region and include the Central Aleutian Islands, Eastern Aleutian Islands and Gulf of Alaska populations. A third group belongs to the offshore killer whales. Although offshores have been encountered as far south as Los Angeles mostly in the winter months (Black et al., 1997; Ford et al., 2000), only recently have photo-identification re-sightings of offshore individuals been recorded between Alaska, Washington State and California, thus suggesting a single population with the same individuals moving throughout the northeast Pacific including the polar regions (Krahn et al., 2007). PBDEs were analyzed in blubber biopsy samples from North Pacific killer whales ($n=84$) collected in 2003–2004 (Krahn et al., 2007). The offshore killer whales were shown to consume prey species that were distinctly different from those of sympatric resident and transient killer whales. These probably include long-lived fish species, for example tuna and sharks. Mean concentrations of ΣPBDEs (BDE-28, -47, -49, -66, -85, -99, -100, -153, -154, -183) in offshore killer whales were significantly higher (mean of 3300 ng/g lw) than those found for the transients (mean of 790 ng/g lw) and both were significantly higher than ΣPBDEs for the residents (mean of 76 ng/g lw). Offshore killer whale concentrations are similar to the highest concentrations of PBDEs measured in Arctic marine mammals, seen in Faroe Island long-finned pilot whales (61–3200 ng/g lw) (Lindström et al., 1999).

Wolkers et al. (2007) analyzed 10 BDE congeners in blubber tissue of one subadult and eight male adult killer whales sampled in northern Norway in 2002. Norwegian killer whales feed on fish. Geometric mean ΣPBDEs (BDE-28, -47, -99, -100, -153, -154) were approximately 500 ng/g lw and the predominant congener was BDE-47. This concentration is of the same order of magnitude as found in the transient killer whale population from the North Pacific (Krahn et al., 2007).

Besides more data on beluga whale and minke whale, new data on killer whales and narwhal have been produced for PBDEs. In beluga whale blubber, observed levels were twice as high in the Norwegian Arctic (72 ng/g lw) compared to the eastern and western Canadian Arctic (17 and 22 ng/g lw, respectively). Similar ΣPBDE levels were found in blubber of minke whales from West Greenland and resident killer whales from Alaska (63 and 76 ng/g lw respectively). Narwhal blubber collected at Svalbard had higher ΣPBDE concentrations than from the eastern Canadian Arctic. Highest ΣPBDE concentrations were seen in killer whales, particularly offshore killer whales, which migrate between Alaska and California (3300 ng/g lw). Killer whales from the Norwegian coast and Alaskan transients also had elevated levels of 500–790 ng/g lw. BDE-209 was analyzed for but not detected in beluga and narwhal. HBCD was found in beluga, narwhal, minke whale and long-finned pilot whale. HBCD concentrations were similar in eastern and western Canada and somewhat lower than in Hudson Bay and West Greenland beluga. Isomer-specific analyses showed a predominance of the α-isomer although γ-HBCD was also found in beluga and narwhal and β-HBCD in juvenile long-finned pilot whale. BTBPE and TBECH were detected in beluga as well. The PBDE results for beluga and minke whale are similar to those presented in the previous review.

4.5.5.4. Arctic fox, sledge dogs and sea otters. Fuglei et al. (2007) assessed the concentrations of tetra-hexaBDEs as well as PCBs and several pesticides in adipose tissue of Arctic fox from Svalbard, Norway. The Arctic foxes investigated in the study were mainly feeding within the marine ecosystem, determined by stable carbon isotope measurements. A total of 20 adult Arctic foxes (five fat and five lean individuals of each sex) were collected between November 1998 and March 1999. Animals were trapped at two locations (Kapp Wijk and Austfjordnes) in the Svalbard Archipelago (74–81°N; 10–35°E). PBDEs (BDE-47, -99, -100, -153, -154) were found in all individuals, but were quantitatively of minor importance, making up less than 1% of the total pollutants. Geometric mean ΣPBDE concentrations were 26.3 ng/g lw in subcutaneous adipose tissue and 31.6 ng/g lw in abdominal adipose tissue. There were no statistically significant differences due to age or sex, but lean animals had higher concentrations than fat animals. The most prevalent brominated compound was BDE-154 making up about 47% of the ΣPBDE, followed by BDE-47 (about 17% of ΣPBDE), and BDE-153 (about 8% of ΣPBDE). The authors conclude that coelution issues with BB-153 could explain the high findings of BDE-154.

Verreault et al. (2008) chose domestic Greenland sledge dogs as potentially representative, surrogate species for the East Greenland polar bear. Sledge dogs were amenable to a captive and multigenerational feeding study to investigate the fate, bioaccumulation and biological effects of organohalogen compound (OHC) exposure. Starting in 2004, and carried out over a nearly two-year period, juvenile sledge dogs in West Greenland were fed a controlled diet consisting of pork fat (lard for human consumption) or a naturally OHC-containing diet comprised of blubber obtained from one minke whale harvested in the waters off West Greenland (Sonne et al., 2006a, 2007, 2008a,b,c). Female sledge dogs, two months of age, obtained in the community of Aasiaat (Disco Bay, West Greenland) were assigned to two experimental groups: an exposed (EXP) ($n=8$) and a control (CON) group ($n=8$). The groups were composed of paired sisters, one sister in each group, in order to minimize differences in age and genetics among the EXP and CON groups. The sum of BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -183 and -209

was determined and ten times higher concentrations of ΣPBDE were found in the exposed group (adipose tissue) compared to the control group (73 and 7 ng/g lw, respectively). HBCD behaved similarly with concentrations of 8.3 ng/g lw in the exposed group and 1 ng/g lw in the control group.

PBDEs were included in a study on organic pollutants and trace elements in livers of sea otters (*Enhydra lutris*) stranded on the Pacific coast of the US and the Kamchatka Peninsula (Russia) in 1995–1998 (Kannan et al., 2008). Following legal protection and reintroductions into former habitats, the sea otter populations along the US Pacific coast increased between the 1960s and 1990s, but declined again in the late 1990s. The animals of this study originated from the three subspecies Californian (*E. l. nereis*), Alaskan (*E. l. kenyoni*) and Russian sea otter (*E. l. lutris*). The Alaskan sea otters were found stranded in Prince William Sound and at Adak Island (Aleutians). Russian sea otters were from the southern Kamchatka Peninsula. Livers of 2–6 adult individuals were analyzed from each location. PBDE analysis covered tri-heptaBDEs, but individual congeners were not specified. Sea otters from Arctic locations had mean ΣPBDE concentrations of 14.7 ng/g lw (0.5 ng/g ww) at Kamchatka, 275 ng/g lw (11 ng/g ww) at Adak Island, 1027 ng/g lw (38 ng/g ww) at Prince William Sound compared to 909 ng/g lw (30 ng/g ww) from northern Washington. The highest concentrations were found in animals from California, exceeding the other locations by a factor of 2 (2400 ng/g lw).

Thus, the mean ΣPBDE concentration in the sea otters was significantly correlated with the population density. PCBs followed the same geographical trend, but exceeded PBDE concentrations by factors of 7 (California) to 56 (Kamchatka). Interestingly, the PBDE concentrations in Alaskan sea otters were two orders of magnitude higher than those reported in livers of polar bears from Alaska (Kannan et al., 2005) possibly caused by their proximity to urban sources and their diet based on benthic organisms (Kannan et al., 2008). BDE-47 was the predominant congener in all sea otters, ranging from approximately 40% in sea otters from Prince William Sound to 100% of ΣPBDE at the other Arctic locations. Sea otters from Kamchatka and Adak contained only BDE-47. Sea otters from Prince William Sound contained a high proportion of BDE-99, suggesting current exposure to PBDEs in this region of Alaska.

4.5.5.5. Polar bears. Previous studies reported PBDEs in polar bears (*Ursus maritimus*) from Alaska, Canada, Greenland and Svalbard, ranging from 7–144 ng/g lw. ΣPBDE concentrations were highest in polar bears from East Greenland and Svalbard and the predominant congeners were BDE-47, -99 and -153. BDE-209 was detected at low concentrations (1 ng/g lw) from Svalbard bears. HBCD was also found (range 5–110 ng/g lw) with highest concentrations from East Greenland and Svalbard.

Sørmo et al. (2006) analyzed PBDEs and HBCD in four polar bears from Svalbard collected in 2002–2003. Mean ΣPBDE concentration (BDE-28, -47, -99, -100, -153, -154, -209) in polar bear adipose tissue was 30 ng/g lw (range 21–45 ng/g lw). Very low concentrations of BDE-209 were found (range — 0.03–0.16 ng/g lw). Mean HBCD concentration was 11.5 ng/g lw (range 5.3–16.5 ng/g lw).

Verreault et al. (2005) determined PBDEs in plasma of 15 female Svalbard polar bears collected in April 2002. ΣPBDE concentrations (BDE-28, -47, -66, -85, -99, -100, -138, -153, -154, -183, -209) ranged from 270 to 970 ng/g lw and consisted predominantly of BDE-47 although BDE-209 was detected in 7% of the samples (range <0.06–0.10). Total HBCD could be detected in 14% of the samples at concentrations of <3–85 ng/g lw.

Polar bears from the Ittoqqortoormiit area in central East Greenland were sampled in 1999–2001 for a number of different studies which are summarized below. Adipose tissue from 92 East Greenland polar bears was analyzed for 32 PBDE congeners to determine seasonal patterns (Dietz et al., 2007). The mean ΣPBDE concentration (BDE-28/33, -47, -99, -100, -153, -101/154) was 70 ng/g lw (range 22–192 ng/g lw) with BDE-47, -153, -99 and -100 comprising 99.6% of the total ΣPBDE concentration. A seasonal pattern of median ΣPBDE concentrations with 1.2 to 1.8 times higher concentrations in March to July than the rest of the year was reported. Changes in BDE-47/ΣPBDE (decrease) and BDE-153/ΣPBDE (increase) ratios with age were also observed. No age/sex relationship with PBDE concentrations was found in this study. However, Sonne et al. (2006b) did find some differences with comparable ΣPBDE concentrations (BDE-17, -28, -47, -66, -85, -99, -100, -138, -153, -154, -183, -190 and -209) in subadult male and female as well as adult male bears, (53, 59 and 56 ng/g lw respectively), but lower concentrations of 40 ng/g lw in adult females. Gebbink et al. (2008a,b) reported the tissue-specific composition of sum classes of brominated contaminants and metabolic/degradation byproducts determined in adipose tissue, brain, liver and blood in adult male and female polar bears. Significantly ($p<0.05$) higher wet weight concentrations of ΣPBDEs (BDE-17, -28, -47, -66, -85, -99, -100, -138, -153, -154, -183, -190 and -209) were found in the adipose (92 ng/g lw; 83 ng/g ww) and liver tissues (400 ng/g lw; 40 ng/g ww) relative to whole blood (92 ng/g lw; 1.2 ng/g ww) and brain (14 ng/g lw; 2.9 ng/g ww). Among the BDE-47, -99, -100, -153 and -154 congeners, the trend showed a significant brain localization preference for BDE-47 ($p<0.03$), a significant preference of BDE-99 to accumulate in the liver ($p<0.001$), and a significant accumulation preference of BDE-153 in the subcutaneous adipose tissue ($p<0.001$). Total HBCD was only detected in adipose tissue with a mean of 46 ng/g lw (41 ng/g ww). In contrast, ΣOH-PBDE concentrations were significantly higher ($p<0.05$) in whole blood than in adipose tissue (2.9 ng/g ww compared to 0.9 ng/g ww, respectively). The 6-OH-BDE-47 metabolite was found only in the adipose tissue, while 3-OH-BDE-47 was found mainly in the blood but also in adipose tissue in addition to a total of three MeO-PBDE congeners, which differed among the tissues.

A more recent collection and analysis of polar bear samples from East Greenland confirmed the predominance of BDE-47 and -153 (BDE-47: 21.6 ng/g lw, BDE-153: 11.9 ng/g lw). BDE-209 was not detected (Letcher, 2007). Considerable amounts of BB-153 were detected as well. Total HBCD was a major BFR contaminant, and low levels of HxBBz were detected in all bears.

Muir et al. (2006b) determined PBDEs in adipose tissue of adult and subadult female polar bears sampled between 1999 and 2002 from sub-populations in Arctic Canada, eastern Greenland, and Svalbard, and in males and females collected from 1994 to 2002 in northwestern Alaska. Only 4 congeners (BDE-47, -9, -100, and -153) were consistently identified in all samples. BDE-47 was the major PBDE congener representing from 65% to 82% of the ΣPBDEs. Age was not a significant covariate for individual PBDEs or ΣPBDE. Higher proportions of BDE-99, -100, and -153 were generally found in samples from the Canadian Arctic than from Svalbard or the Bering-Chukchi Sea area of Alaska. Geometric mean ΣPBDE concentrations were highest for female polar bear fat samples collected from Svalbard (50 ng/g lw) and East Greenland (70 ng/g lw). Significantly lower ΣPBDE concentrations were found in fat of bears from Canada and Alaska (means ranging from 7.6 to 22 ng/g lw). For the entire dataset, ΣPBDE concentrations were correlated with ΣPCBs. Higher HBCD concentrations were found in fat of bears from East Greenland (44.5 ng/g lw) and Svalbard (44 ng/g lw) than in those from Alaska (0.40 ng/g lw). The geographical trends for PBDEs and HBCD parallel those for PCBs implying similar source regions for long range transport to the Arctic and bioaccumulation pathways in the Arctic marine food web.

Frederiksen et al. (2007) analyzed HBCD (total and isomer-specific) and TBBPA in two pooled adipose and liver samples from polar bears from East Greenland ($n=5$) collected in 1999–2001 and 2000–2002, respectively. Only α-HBCD could be detected at low concentrations of 5.4 and 13.2 ng/g lw in the two adipose tissue samples and 2.8 and 3.9 ng/g lw in the two liver samples. Due to the

lack of labelled internal standards for HBCD, possible losses or supression in the LC-MS due to matrix effects were not corrected. TBBPA could not be detected in any of the samples.

Kannan et al. (2005) compared the concentrations of BDE-47 in livers of polar bears from the Alaskan Beaufort Sea and Chukchi Sea sub-populations collected in April 2002. Only BDE-47 could be detected in polar bear liver and concentrations ranged from <0.2 to 2.7 ng/g ww and levels decreased with age. The mean BDE-47 concentrations were 1.01 ng/g ww (12 ng/g lw) for the Beaufort Sea and 0.91 ng/g ww (11 ng/g lw) for the Chukchi Sea sub-populations.

In summarizing data on PBDEs in adipose tissue of polar bears from the different Arctic regions, bears from East Greenland and Svalbard seem to be exposed to higher concentrations of PBDEs and HBCD compared to bears from North America. More studies have included and found BDE-209 and HBCD. A much more thorough investigation was recently carried out on East Greenland polar bears compared to other parts of the Arctic, supplying the scientific community with more detailed data on age, sex and seasonal differences in PBDE concentrations, as well as differences in tissue partitioning.

5. Temporal trends

Su et al. (2007) measured di-decaBDEs in 104 weekly air samples (gas and particulate phases combined) taken over the period of 2002–2004 at Alert, Nunavut. Using digital filtration to remove the seasonal cycling and extract only the interannual trends, they found statistically significantly increasing trends for eight BDE congeners. The doubling times ranged between 2 and 6 years (BDE-28/33: 2.4 y; BDE-47: 5.4 y; BDE-99: 5.2 y; BDE-100: 2.9 y; BDE-153: 2.0 y; BDE-154: 2.7 y; BDE-209: 6.2 y). The doubling times for the lower brominated BDEs are similar to those found previously in biota (Ikonomou et al., 2002).

PBDEs determined in a sediment core from Lake Ellasjøen on the island Bjørnøya (see Section 4.4.2 Freshwater sediments) confirmed the increasing trend observed in biota samples from the Arctic (Evenset et al., 2007). The core covers the time period from 1881 to 2001, divided into intervals of 11–15 years. The first detection of PBDEs at approximately 0.15 ng/g dw (sum concentration of BDE-47, -99 and -100) occurred in the layer representing the years 1946–1959. BDE-28 was detected in the following layer for the first time (1959–1973) and BDE-153 was only found in the surface sediment (1987–2001). Since the first detection in the 1940s/1950s, the summed concentration increased to 0.73 ng/g dw in the surface sediment, now also including BDE-28 and BDE-153. This corresponds to a more than five-fold increase in PBDE concentration in the last 50 years.

No time trend could be established for HBCD in the same sediment core from Lake Ellasjøen (Evenset et al., 2007). HBCD was not detected in the surface sediment, but in the previous layer, representing the years 1973–1987. As HBCD is still in use, this result seemed surprising and could not be explained. Analyses of PCBs and DDTs in the same sediment core showed increasing concentrations until the 1960s and decreases in the following layers, i.e., an opposite trend to PBDEs in these last decades. The authors emphasize that Lake Ellasjøen is not a typical Arctic lake, as it receives large amounts of seabird guano, apparently leading to a contaminant transfer from the marine to the limnic environment. Thus, the PBDE input to this lake is probably not solely due to atmospheric deposition, but intensified by inputs of seabird guano. Since the contaminant development coincides with the main usage of the compounds, as was shown for PCBs and DDT, the authors assume that the guano input has been relatively constant over time.

ΣPBDE concentrations increased in landlocked Arctic char from Resolute Lake (Cornwallis Island) in the Canadian Arctic, from approximately 12 ng/g lw in 1997 to approximately 100 ng/g lw in 2006, with a doubling time of 5 years (Muir et al., 2006c; D. Muir, Environment Canada, personal communication). Interestingly, the 2007 samples had a lower PBDE concentration of about 80 ng/g lw. However, not all time series for Arctic char in the Canadian Arctic showed increasing trends. Samples from Char Lake collected biannually between 1999 and 2005 had rather constant concentrations of about 110 ng/g lw. Results from Lake Amituk showed increasing concentrations for the years 2005–2007, but at a lower level than for the years 2001–2003. Thus, the 2001 and 2007 samples had similar concentrations of 30–40 ng/g lw.

In another study from the Canadian Arctic, a temporal trend for PBDEs was assessed for lake trout from three Yukon lakes, Lake Laberge, Lake Kusawa and Lake Quiet (Stern et al., 2007), but consists at present of only 3–6 data points. A steep increase in the last two years was found for Lake Quiet, with 2.5 ng/g lw in 1992, 8.3 ng/g lw in 2001 and 210 ng/g lw in 2003. The results are less clear for the other two lakes. In Lake Laberge (1993–2006), concentrations ranged between 380 (in 1993) and 8040 ng/g lw (in 2006), but an increasing trend could only be found if the 2005 data point was removed, which was close to the 1993 level. For Lake Kusawa (1999–2006), mean ΣPBDE concentrations ranged from 82 ng/g lw in 2001 to 4300 ng/g lw in 2003, with 150 ng/g lw in 2006. The 2006 result for Lake Kusawa is almost 30 times lower than the 2005 concentration. Thus, the concentration decreases from 1999 to 2001, increases in 2003 and decreases again in 2006. Obviously, no time trend can be established.

Also from the Canadian Arctic, a temporal trend study on burbot in the Mackenzie River, Fort Good Hope, Northwest Territories showed increasing ΣPBDE (BDE-47, -99, -100, -153, -154) concentrations, from 1.3 ng/g lw in 1988 to 24 ng/g lw in 2006 (Fig. 3) (Stern and Tomy, 2007). The last data point is 2.4 times higher than the concentration in 2005 (10 ng/g lw), which indicates a very rapid increase in the past few years, different from the findings of Ikonomou et al. (2005) who have observed stagnating or even decreasing concentrations in ringed seals since 2000. The authors also noted that the 1988–2006 increase was 25.8-fold for BDE-154, but only 8.1-fold for BDE-47. In general, BDE-47 was the predominant congener in burbot liver and accounted for 35–57% of ΣPBDE. Interestingly, the percentage of BDE-47 decreased with time, from 57% in 1988 to 35% in 2006. The other BDE congeners increase accordingly, with comparable slopes for BDE-99 and BDE-100 and a relatively large increase of BDE-154.

Concentrations of α- and γ-HBCD were available from burbot collected in 1986, 1999, 2002, 2003, 2005 and 2006 (Stern and Tomy, 2007). During the study period 1986–2006, the lowest ΣHBCD concentration of 0.72 ng/g lw was found in 1999, and the highest concentration of 1.6 ng/g lw occurred in 2003. The α-HBCD/γ-HBCD ratio varied considerably, from approximately 1 (in 2003) to 24 (in

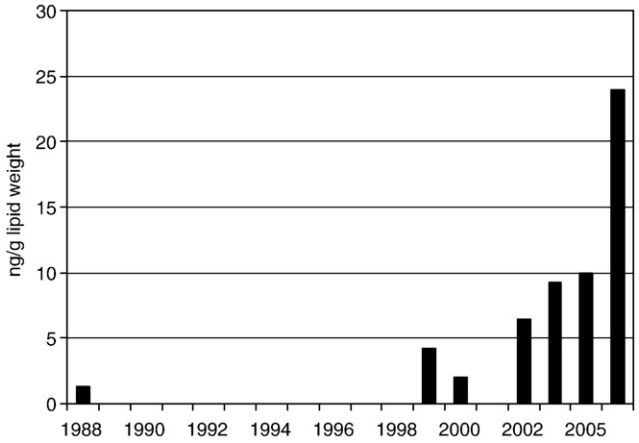

Fig. 3. Temporal trend of ΣPBDE concentrations in burbot liver from Fort Good Hope, Mackenzie River, Yukon, Canada (data from Stern and Tomy, 2007).

2002 and 2005). No temporal trend was reported by the authors for ΣHBCD. Linear regression on natural logarithm-transformed data does indicate a positive slope, which, however, is not significant and has an R^2 of <0.1. Although still not statistically significant at the 5% level, linear regression yields a positive slope and an R^2 of 0.43 if only α-HBCD is considered (Fig. 4). The concentration of γ-HBCD is very variable, thus affecting the ΣHBCD concentrations.

In a study from the Barents Sea, PBDEs were analyzed in liver of 21–25 individuals of Atlantic cod collected from 4 stations in 1994/1995, which were revisited in 2005/2006 (Bakke et al., 2008). The comparisons of data from 1994/1995 and 2005/2006 showed decreases between 16 and 84% in mean ΣPBDE concentrations for all stations. The authors did not state, however, which of the mean values were statistically significant.

Temporal trends of PBDEs and HBCD have been followed in eggs from northern fulmar and thick-billed murres collected from Prince Leopold Island in Lancaster Sound, Canada (Braune, 2006, 2007). Concentrations of ΣPBDE (BDE-17, -28, -49, -47, -66, -100, -99, -85, -153, -138, -183) steadily increased between 1975 and 2003 in both the thick-billed murres and the northern fulmars, but after 2003, ΣPBDE concentrations appear to have started to decrease in both species (Fig. 5). Concentrations of HBCD increased from 13 ng/g lw in 2003 to 28 ng/g lw in 2006 in northern fulmars whereas concentrations ranged from 1.8 to 7.4 ng/g lw in thick-billed murres. The ratio of total HBCD to ΣPBDE was considerably higher in the northern fulmars than in the thick-billed murres and showed a steady increase from 2003 to 2006 in the fulmars. In the thick-billed murres, BDE-47 was the predominant BDE congener in all years followed by BDE-99 and BDE-100 although the presence of other congeners such as BDE-138, BDE-153 and BDE-85 has increased in recent years. BDE-47 was the predominant BDE in northern fulmars in most years, followed by BDE-99 and BDE-153.

Braune et al. (2007), analyzed eggs of ivory gulls for PBDEs, HBCD and PBBs, collected in 1976, 1987 and 2004 from Seymour Island in the Canadian Arctic. Concentrations of the PBDEs increased steadily between 1976 and 2004, driven primarily by increases in BDE-47 (ΣPBDE level of 45 ng/g lw in 2004). Except for a peak in 1987, concentrations of BDE-154/BB-153 and BB-101 did not change greatly between 1976 and 2004. BDE-183 and -209 were below the limit of detection in all analyzed samples. However, the percent contribution of BDE-47, as well as a number of the minor congeners (e.g. BDE-28, -153, -17, -49, -66, -85) to ΣPBDE increased between 1976 and 2004, while proportional contributions of BDE-99, BDE-138 and BDE-100 decreased. Concentrations of total-(α)-HBCD decreased from 4 to 2 ng/g lw in the same period.

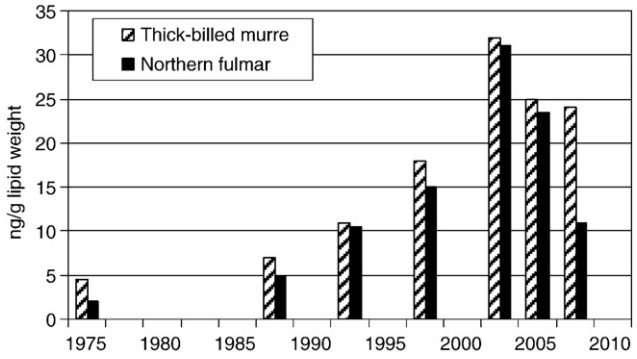

Fig. 5. Temporal trends of ΣPBDEs in thick-billed murre and northern fulmar eggs from the central Canadian archipelago (data from Braune 2006, 2007).

Temporal trends of PBDEs were also studied in eggs of Brünnich's guillemots collected at Bjørnøya and Kongsfjorden, Svalbard in the years 1993, 2002, 2003 and 2007 (Bakke et al., 2008). ΣPBDE (BDE-28, -47, -100, -99, -138, -153, -154, -183, -206, -207, -208, -209) concentrations decreased from 86.7 ng/g lw in 1993 to 47.9 ng/g lw in 2002, then to 16.7 ng/g lw in 2007 for the samples collected in Kongsfjorden. The samples collected at Bjørnøya decreased from 26.9 ng/g lw in 2003 to 17.2 ng/g lw in 2007. BDE-209 could be detected in the 2007 samples from Kongsfjorden and Bjørnøya as well as from 1993 in Kongsfjorden at low ng/g lw levels. Total HBCD levels between 12 and 29 ng/g lw were detected with no time-related trends. Since $\delta^{15}N$ measurements revealed a significant difference in the eggs collected in 2002/2003 compared to 2007, a shift in diet of the mother birds is indicated, potentially concealing changes of concentrations over time and space for both PBDEs and HBCD.

The previous time trend on PBDEs in ringed seals from Holman Island (Northwest Territories, western Canadian Arctic) covering the time period 1981–2000 has been updated with new data points for 2002 and 2003 (Ikonomou et al., 2002, 2005). Mean ΣPBDE concentrations in the samples from 2002 and 2003 (0–15 year age class) were lower than those from 2000 in the same age group (Fig. 6). Analysis of variance, however, showed that the three mean values were not statistically different. It should be noted, however, that the 2003 concentration was only based on two individuals. Similarly, the mean ΣPBDE concentrations in the age class of 16–35 year-old seals were comparable for the years 2002 and 2003 and not statistically different from those from the year 2000. Apparently, the PBDE levels have stabilized since 2000 and might even be decreasing. The authors

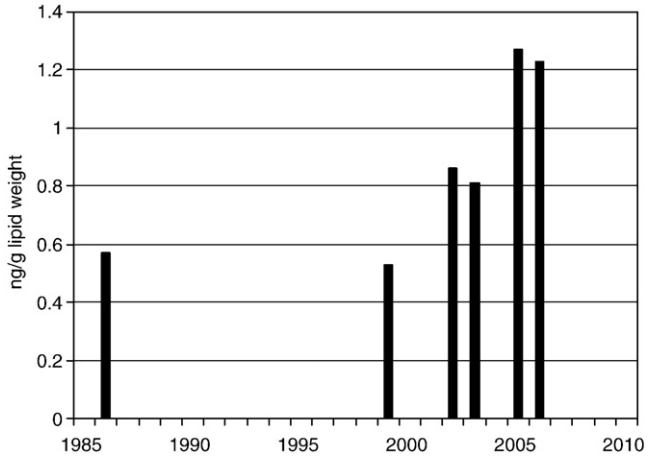

Fig. 4. Temporal trend of α-HBCD in burbot liver from Fort Good Hope, Mackenzie River, Yukon, Canada (data from Stern and Tomy, 2007).

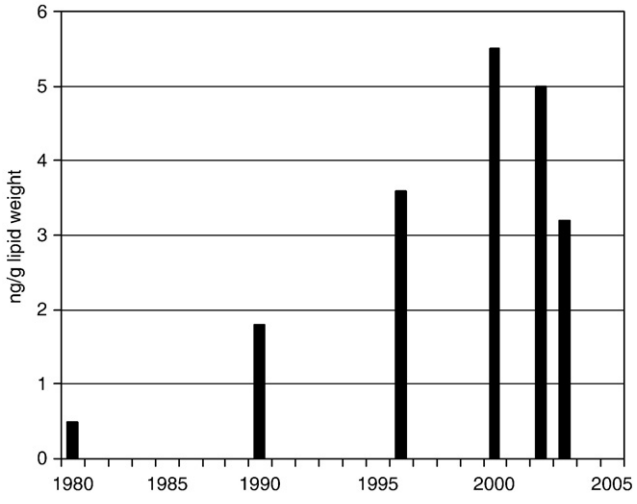

Fig. 6. Temporal trend of ΣPBDE in ringed seals of the 0–15 years-of-age class from Holman Island, western Canadian Arctic. Results from Ikonomou et al. (2005).

point out that another 5–10 years of sampling and PBDE analysis are required to detect statistically significant declines.

In another temporal trend study on ringed seals from the Canadian Arctic, increasing concentrations of ΣPBDEs and HBCD were found in female ringed seal blubber from Arviat (western Hudson Bay) and Resolute (central Canadian archipelago) over the period 1992 to 2005 (Muir et al., 2006a). The study included archived and new samples from the years 1992, 1998, 2003 and 2005 and has since continued with annual sample collections. According to Muir et al. (2007), the 2006 data indicated a stabilization of PBDE levels in ringed seals from these locations.

Temporal trends of PBDEs in ringed seals from Ittoqqortoormiit (Central East Greenland) and Qeqertarsuaq (Central West Greenland) were studied by Riget et al. (2006) and Vorkamp et al. (2008). The statistical time trend analyses followed the procedures suggested by ICES which divide the total variation over time into a linear and a non-linear component (Nicholson et al., 1998). Linear regression analysis is applied to describe the linear component, while a loess smoother is used to describe the non-linear component. Only juvenile animals (≤4 years of age) were included in the study to minimize the influence of covarying factors. For West Greenland, samples were analyzed from the years 1982, 1986, 1994, 2000–2004 and 2006. As most congeners had several values below the limit of detection, the temporal trend was only assessed for BDE-47. A significant annual increase of 5.3% was found (Fig. 7) (Vorkamp et al., 2008). From East Greenland, archived samples were available from 1986, 1994 and 1999–2004. Unlike other temporal trend studies of PBDEs, no significant time trend was found for ΣPBDE in the seals from East Greenland. The three-year running smoother did not result in a significantly better description of the temporal development. The same results were obtained for the individual congeners BDE-28, -47 and -99.

The ΣPBDE concentrations in the East Greenland seals seemed to increase from 1986 until approximately 2002, but the 2003 and 2004 median values were below the 2002 level (Fig. 8). The authors discussed the possibility of the main increase in PBDE concentration occurring prior to 1994, given the relatively high concentrations found in seals from 1986 and 1994. The region might be more strongly affected by developments in Europe than other locations in the Arctic, and the results might reflect the stagnation and decrease in PBDE concentrations that have been found in the European environment in recent years (Johansson et al., 2006; Sellström et al., 2003).

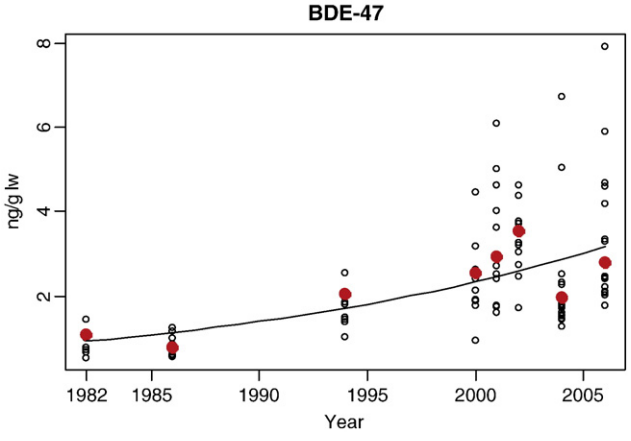

Fig. 7. Temporal trend for BDE-47 in blubber from West Greenland ringed seals. Open circles are individual concentrations and filled circles represent median concentrations. The line represents an exponential curve determined by log-linear regression analysis. Reprinted from Vorkamp et al. (2008). Levels and trends of persistent organic pollutants in ringed seals (*Phoca hispida*) from Central West Greenland, with particular focus on polybrominated diphenyl ethers (PBDEs), Environment International 34, 499–508, with permission from Elsevier Ltd.

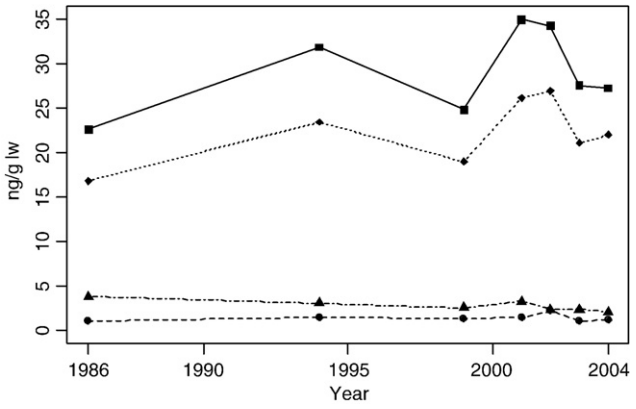

Fig. 8. Temporal trend (medians) for ΣPBDE (squares), BDE-47 (diamonds), BDE-99 (triangle) and BDE-28 (octagons) in blubber of East Greenland ringed seals. Reprinted from Rigét et al. (2006) Temporal trend studies on polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in ringed seals from East Greenland, Journal of Environmental Monitoring 8, 1000–1005, with permission from the Royal Society of Chemistry.

Temporal trend analyses for BFRs were carried out in beluga from Hendrickson Island on the southeastern Beaufort Sea (Hendrikson Island: 1984, 1993, 1995, 2001, 2005 and 2006) and Pangnirtung, Baffin Island (1982, 1986, 1992, 1995, 2002, 2005 and 2006) (Tomy, 2007). There was no statistically significant difference in concentrations of HBCD or BDEs (sum of 17 congeners) in Hendrikson Island beluga blubber between 1993 to 2001 but there were significant declines in concentrations of both contaminants in animals from 2001 and 2006 (one-way ANOVA: $p<0.05$, in both cases) (Fig. 9). For both compounds, concentrations peaked in 2001, 23.7 ng/g lw for ΣPBDEs and 2.2 ng/g lw for HBCD. In Pangnirtung beluga, blubber HBCD concentrations have remained steady between 1982 and 2006. However, BDE concentrations increased exponentially between 1982 to 2005 ($R^2=0.539$, $p<0.05$) with a peak concentration of 29.7 ± 4.9 ng/g, lw in 2005 and a doubling time of ~11 years (Fig. 10).

A number of temporal trend studies on PBDEs in the Arctic have been conducted, with very different results. Most of the studies, analyzing air, freshwater sediment, freshwater fish, bird eggs, ringed seal and beluga and covering various locations in Canada, Bjørnøya (Norway) and East and West Greenland, report increases of PBDEs in the Arctic. In some of these studies, however, the most recent data points were below the maximum concentrations, but within the uncertainty of the calculated time trend indicating stagnation or possible declines in PBDE concentrations in the last 2–5 years, for example in ringed seals (western and eastern Canada, East Greenland) and thick-billed murre and northern fulmar eggs (Canada). PBDE concentrations in ringed seal from Hudson Bay and West Greenland continue to increase. PBDE concentrations in marine fish and bird eggs from the Barents Sea have decreased since the middle of the 1990s, and a decrease has also been found in beluga from Hendrickson Island (western Canada), while the concentrations in beluga from Pangnirtung (eastern Canada) have increased since 1982. In studies on Arctic char and lake trout from Canada, some locations showed no trend at all, while others clearly showed an increase of PBDEs and burbot from one site in the Yukon also showed increases. Some of the authors of the primary publications discussed these changes in light of the possible effects of restrictions or bans of Penta- and OctaBDE products in the EU and US. It seems likely that these measures are having an effect on the observed changes in concentrations observed in the Arctic. At present, however, the data compiled in this review do not present a uniform picture for the Arctic, neither in terms of species nor locations. As an interesting detail, some authors describe different time trends for the individual congeners, for example a more rapid decrease in BDE-153 in recent years. BDE-209 concentrations are increasing in air and no other time trends for this

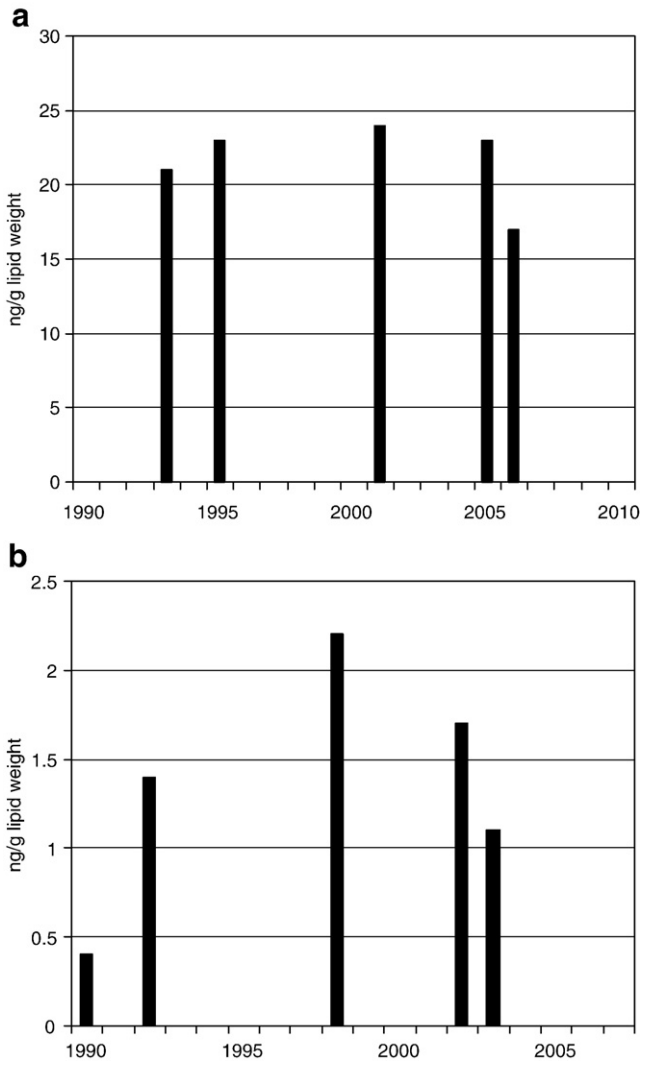

Fig. 9. Temporal trends for ΣPBDE (a) and HBCD (b) in beluga from Hendrickson Island, western Canada. Data from Tomy (2007) and Tomy et al. (2007).

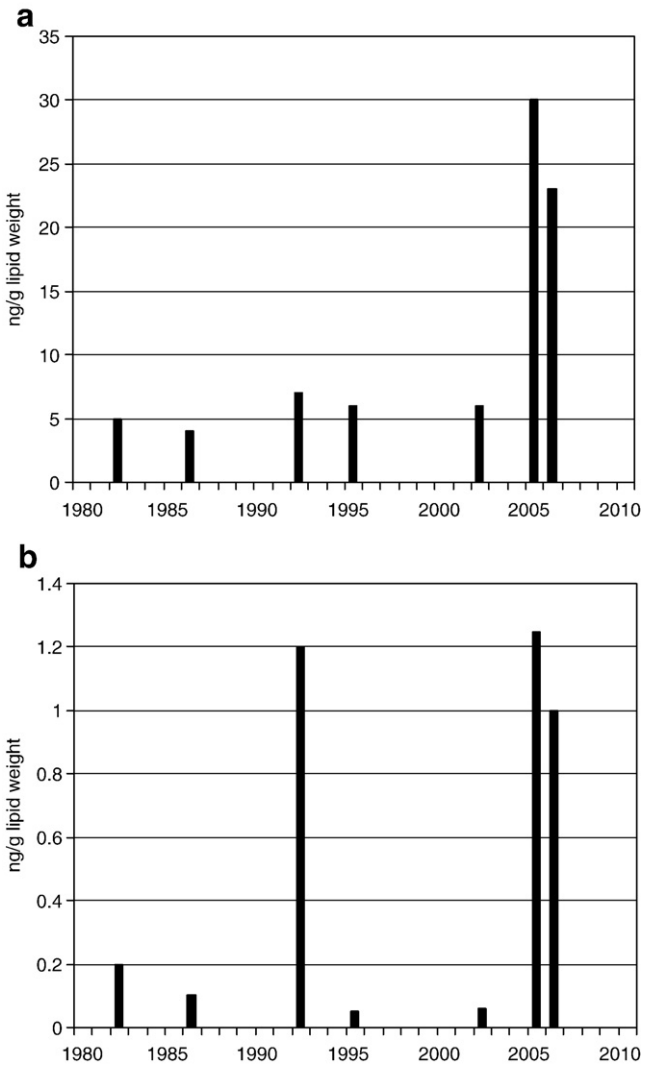

Fig. 10. Temporal trends for ΣPBDE (a) and HBCD (b) in beluga from Pangnirtung, eastern Canada. Data from Tomy (2007) and Tomy et al. (2007).

compound are available. Eight time trend studies have included HBCD, but most of them could not identify any clear trends, as the HBCD concentration was very variable. Increases were found in northern fulmar eggs (Canada) and ringed seal from several sites in Canada, while decreases were reported for ivory gull eggs (Canada) and beluga (Canada). The study on burbot (Canada) indicated that the concentration development might be different for the individual HBCD isomers.

6. Spatial trends

In the previous review, latitudinal trends were seen in PBDE concentrations with indications of declining concentrations and fluxes going northwards. There were also indications of increasing proportions of lower brominated BDEs and decreasing proportions of higher brominated BDEs with increasing latitude. The limited PBDE data for seabirds, beluga, ringed seals and polar bears indicated comparable spatial trends within the Arctic with highest concentrations on East Greenland and Svalbard, similar to spatial trends seen previously for PCBs. However, the amount of data available at that time for spatial analyses was limited and these spatial trends were considered preliminary.

Latitudinal trends in air concentrations of PBDEs, with decreasing concentrations going north were seen from Far East Asia to the northern Pacific, but for the Arctic samples, no significant relationship was seen between concentrations and latitudes (Wang et al., 2005). These new air results fit well with those presented in the previous review (de Wit et al., 2006) and confirm the previous spatial trend. A latitudinal trend with decreasing concentrations going north was also seen for PBDEs in moss samples from northern Norway, including for BDE-209 (Mariussen et al., 2008). Freshwater sediment fluxes of tetra-hexaBDE also decreased with increasing latitude in Alaska with a flux of 12.5 ng/m^2/year for Denali National Park and 6.5 ng/m^2/year for two parks further north (Landers et al., 2008). These can be compared to sediment fluxes from Canadian lakes, which ranged from <0.5 to 8 ng/m^2/year, and Greenland, 0.05–1.3 ng/m^2/year, indicating possibly that the fluxes in Denali lakes are more influenced by nearness to PBDE emission sources.

ΣPBDE concentrations (excluding BDE-209) in marine sediments are higher in the Barents and Pechora Seas and along the northern Norwegian coast than previously seen in Canadian, Norwegian and Russian sediments. Highest ΣPBDE concentrations are seen in the Barents and Pechora Sea sediments. On the other hand, BDE-209 concentrations seem to be higher in the samples from Tromsøflaket, Norway (Bakke et al., 2008).

Clear regional as well as circumpolar spatial trends are seen for seabirds based on concentrations in eggs. Around Alaska, lowest PBDE concentrations are seen in thick-billed murres and common

guillemots from offshore sites or coastal areas of the Bering Sea area and highest PBDE concentrations are seen in the Gulf of Alaska, with a clear west-east gradient in this region (Fig. 11) (Roseneau et al., 2007). A similar spatial trend is seen for Alaskan gulls (Fig. 12). The higher PBDE concentrations in the Gulf of Alaska seabirds are probably due to their proximity to highly populated areas, which are also sources of PBDE emissions. Similar to PBDE concentrations, there are also higher HBCD concentrations in black guillemots on East Greenland than on West Greenland.

When all data for PBDEs except for the Gulf of Alaska are compared on a circumpolar basis, PBDE concentrations in alcids and/or northern fulmars are generally lower in the Bering Sea, intermediate in Canada, W. Greenland, Bjørnøya, northern Norway and the Faroe Islands and highest on Iceland, E. Greenland and Svalbard (Fig. 11). For gulls, although there is less data, the same general spatial trend is seen, with lower levels in Alaska and Canada and highest concentrations at Svalbard and Bjørnøya (Fig. 12). For HBCD in seabird eggs, again, the highest concentrations are seen at Svalbard and around the Barents Sea area and lowest concentrations are seen in Canada and Greenland, with a clear difference between East and West Greenland (Fig. 13).

An even larger difference in PBDE concentrations was found in more recent studies in ringed seals from Ittoqqortoormiit (central East Greenland) and Qeqertarsuaq (central West Greenland). Most BDE congeners were below the limit of detection in the West Greenland samples, i.e., lower than what had previously been detected in animals collected in East Greenland in the same years. Thus, only BDE-47 could be compared reliably and was found to be highest in East Greenland, exceeding the West Greenland concentrations by an order of magnitude (Riget et al., 2006; Vorkamp et al., 2008). More recently higher HBCD concentrations have been found in East Greenland shorthorn sculpin, black guillemot and ringed seals compared to West Greenland (Frederiksen et al., 2007). It is an interesting result that HBCD seems to follow the same occurrence pattern as organochlorines and PBDEs, which may indicate similar transport pathways for all three compound groups.

When compared to PBDE patterns in other seal species sampled at more southerly latititudes in British Columbia and Nova Scotia, the PBDE pattern in Holman Island ringed seals shows a higher proportion of lower brominated BDEs and a lower proportion of higher brominated BDEs (Ikonomou and Addison, 2008). This indicates latitudinal fractionation and is consistent with differential atmospheric transport of the more volatile BDEs to the Arctic.

A circumpolar comparison of PBDE concentrations in ringed seals was possible in the previous review (de Wit et al., 2006). ΣPBDE seemed to decrease in the order East Greenland ≥ Svalbard (30–50 ng/g lw) > Russia (7–11 ng/g lw) > West Greenland (4 ng/g lw) > Canada (1–30 ng/g lw). However, samples originated from different years, and results from Canada showed a large internal variation, with the highest concentrations found for Hudson Bay and the lowest concentrations for the western Canadian Arctic. HBCD did not follow the same trend as PBDEs in the Canadian samples, with highest concentrations (4.7 ng/g lw) in the western Canadian Arctic. New results for PBDEs in ringed seals from Alaska, Canada, Greenland and Svalbard add to and confirm the spatial trends seen previously, with ΣPBDE concentrations in new studies decreasing in the order Svalbard (59 ng/g lw) > East Greenland (33 ng/g lw) > Hudson Bay (21 ng/g lw) > eastern Canada = western Canada = Alaska = West Greenland (3–10 ng/g lw) (Fig. 14). For HBCD, the concentrations decreased in the order Svalbard (20 ng/g lw) > East Greenland (5–7 ng/g lw) > West Greenland (1.5–1.8 ng/g lw). Thus, just as for organochlorines, PBDE and HBCD concentrations are highest in ringed seals from Svalbard and East Greenland. While the PBDE concentrations found in ringed seals from East Greenland and Svalbard were very high in an Arctic context, they were about 10 times lower than concentrations in ringed seals from the Baltic Sea (de Wit, 2002; Riget et al., 2006).

Previous PBDE data for beluga indicated a spatial trend similar to that of ringed seals with highest concentrations around Svalbard (29–161 ng/g lw), followed by Hudson Bay (30 ng/g lw) and with lowest concentrations in eastern and western Arctic Canada (16–17 ng/g lw). The new data confirm this spatial trend with highest PBDE concentrations in Svalbard beluga (69 ng/g lw) followed by Igloolik, Nunavut (54 ng/g lw) and Hudson Bay (30, 35 and 53 ng/g lw) and lowest concentrations from western (Hendriksen Island, 17 and 22 ng/g lw) and eastern Arctic Canada (Baffin Island, 22 and 30 ng/

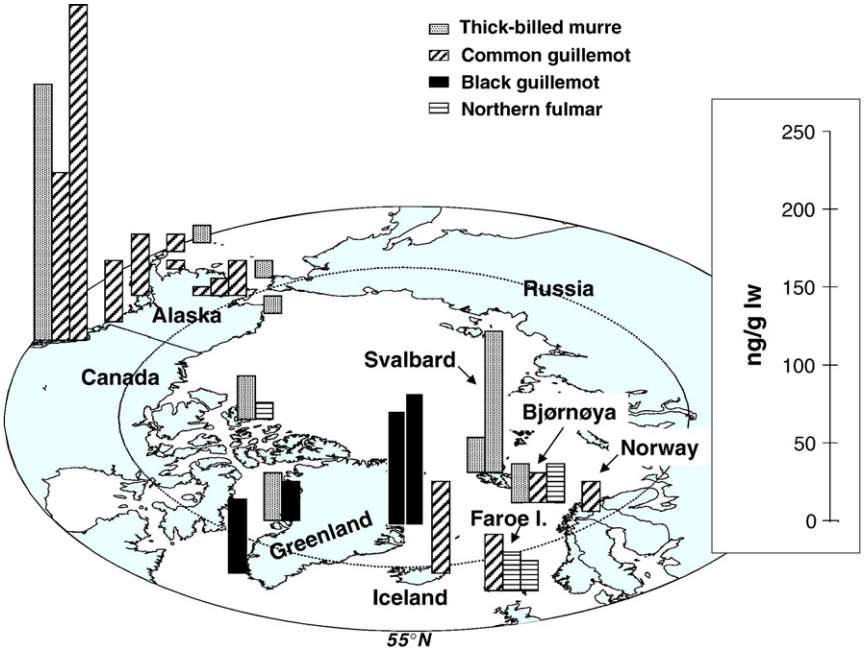

Fig. 11. ΣPBDE concentrations in eggs from common guillemot, thick-billed murre, black guillemot and northern fulmar (plasma for Bjørnøya) (ng/g lw). Results from Johansen et al. (2004), Vorkamp et al. (2004a,b), Fängstrom et al. (2005), Karlsson et al. (2006), Braune (2007), Knudsen et al. (2007), Murvoll et al. (2007), Roseneau et al. (2007), Bakke et al. (2008), Jörundsdóttir et al. (2008).

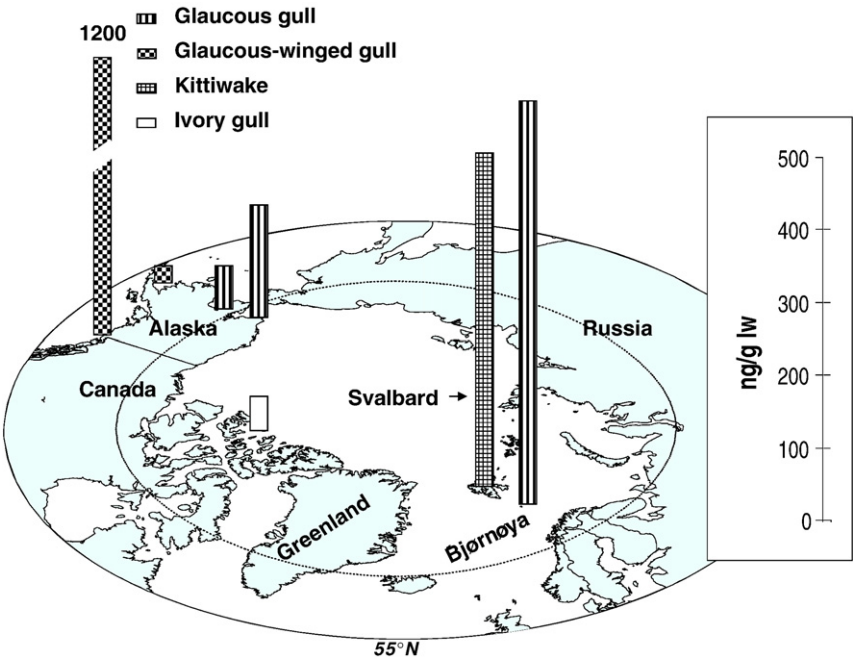

Fig. 12. ΣPBDE concentrations in eggs from glaucous gulls, glaucous-winged gulls, black-legged kittiwakes and ivory gulls (ng/g lw). Results from Murvoll et al. (2006), Braune (2007), Roseneau et al. (2007), Verreault et al. (2007b).

g lw) (Fig. 15). As for ringed seals, PBDE concentrations in Arctic beluga are as much as 100 times lower than in beluga from the St. Lawrence Estuary (Lebeuf et al., 2004; Ikonomou et al., 2005).

New data on PBDE concentrations in polar bears are available for Alaska (Chukchi and Beaufort Seas, 11–12 ng/g lw), East Greenland (22–192 ng/g lw) and Svalbard (30 ng/g lw). These new data fit in well with the spatial trend presented in the previous review (de Wit et al., 2006) and confirm the highest PBDE concentrations in Svalbard and East Greenland polar bears and the lowest concentrations in polar bears from Alaska. This same trend was seen previously for HBCD as well, with highest concentrations in East Greenland and Svalbard polar bears, and lowest concentrations in Alaskan polar bears. Only limited new HBCD data were available for East Greenland (5.4–46 ng/g lw) and Svalbard (<3–85 ng/g lw), but concentrations were comparable to those presented previously (44 ng/g lw at both sites) and do not contradict the spatial trend seen previously.

The new data on spatial trends for PBDEs do not conflict with the data presented in the previous review (de Wit et al., 2006). The same general trends are seen in abiotic media, with lower concentrations and fluxes at higher latitudes and higher proportions of lower brominated congeners at higher latitudes. Circumpolar spatial trends in PBDE concentrations are also found in seabirds, ringed seals, beluga and polar bears, with highest concentrations in populations on East Greenland and Svalbard, lower concentrations in the Russian and Canadian Arctic (except for Hudson Bay) and lowest concentrations in Alaska, if data from the previous review are included. Similar spatial

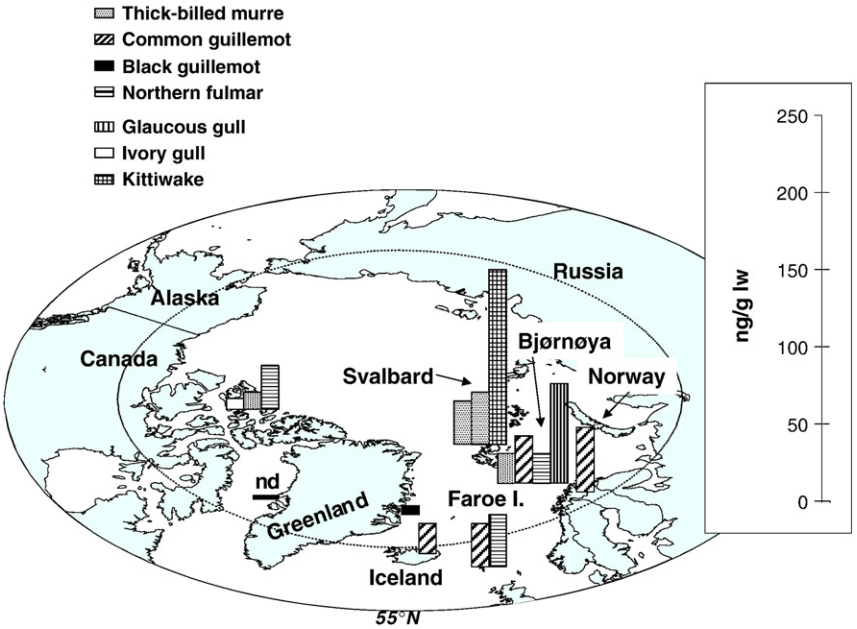

Fig. 13. HBCD concentrations in eggs from thick-billed murre, common guillemot, black guillemot, norther fulmar (liver for Bjørnøya), glaucous gull, ivory gull and black-legged kittiwake. Results from Murvoll et al. (2007), Bakke et al. (2008), Braune (2007), Frederiksen et al. (2007), Verreault et al. (2007b), Knudsen et al. (2007).

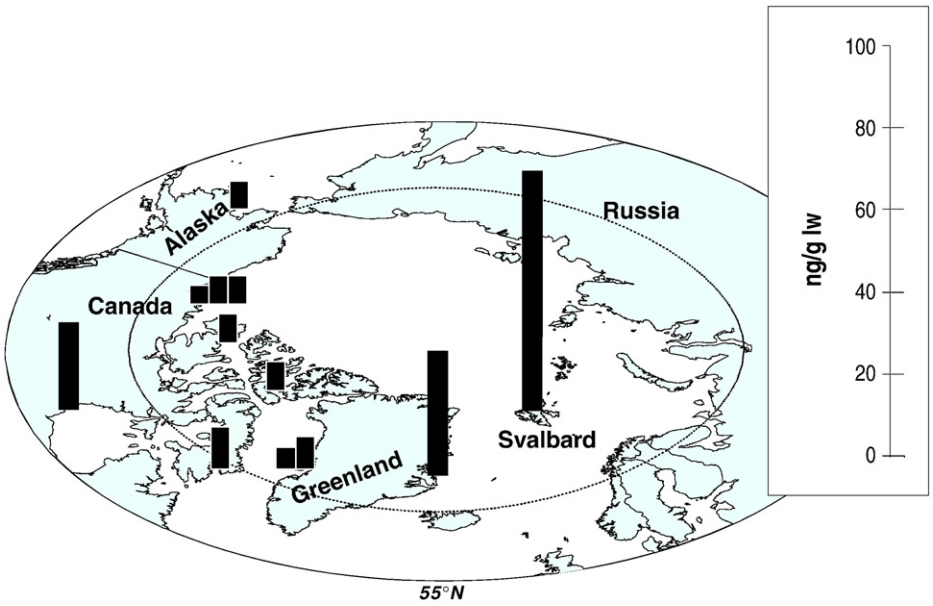

Fig. 14. ΣPBDE concentrations in ringed seal blubber. Results from Ikonomou et al. (2005), Rigét et al. (2006), Sørmo et al. (2006), Muir et al. (2007), Quakenbush (2007) and Vorkamp et al. (2008).

trends are seen for HBCD although the data are more limited. These new data confirm the spatial trends seen in the previous review and indicate that concentrations are higher in the European Arctic and East Greenland compared to the North American Arctic. These spatial trends are also similar to those seen for organochlorine compounds and indicate eastern North America and western Europe as source regions of these compounds to the Arctic via long range atmospheric transport and ocean currents.

7. Bioaccumulation/biomagnification

Several studies have investigated the bioaccumulation and biomagnification properties of PBDEs and in some cases, HBCD, in recent years, primarily in the marine ecosystem. Evenset et al. (2005) studied biomagnification in Lake Ellasjøen and Lake Øyangen, on Bjørnøya, by analyzing zooplankton, chironomid larvae (*Chironomidae sp.*), tadpole shrimps (*Lepidurus arcticus*) and small and large individuals of landlocked Arctic char for PBDEs. To measure the trophic level of the different species, δ^{15}N was determined. The mean ΣPBDE concentrations in Lake Ellasjøen were 52 ng/g lw in zooplankton, 44 ng/g lw in chironomid larvae, 480 ng/g lw in small Arctic char and 410 ng/g lw in large Arctic char. The concentrations were positively correlated with δ^{15}N, also on a lipid–weight basis, indicating accumulation throughout the food web. The number of PBDE congeners detected also increased with trophic level: while only BDE-47, -99 and -100 were detected in zooplankton, Arctic char contained measurable concentrations of BDE-33, -47, -71, -99, -100, -119 and -153. The δ^{15}N values were significantly lower in Lake Øyangen, further confirming different trophic states of the lakes, probably induced by the input of seabird guano to Lake Ellasjøen.

A multichemical food web model was used to estimate biomagnification of a range of contaminants in the Lake Ellasjøen food web, including PBDEs (Gandhi et al., 2006). The data used came from the study by Evenset et al. (2005) and included BDE-47, -99, 100 and -153.

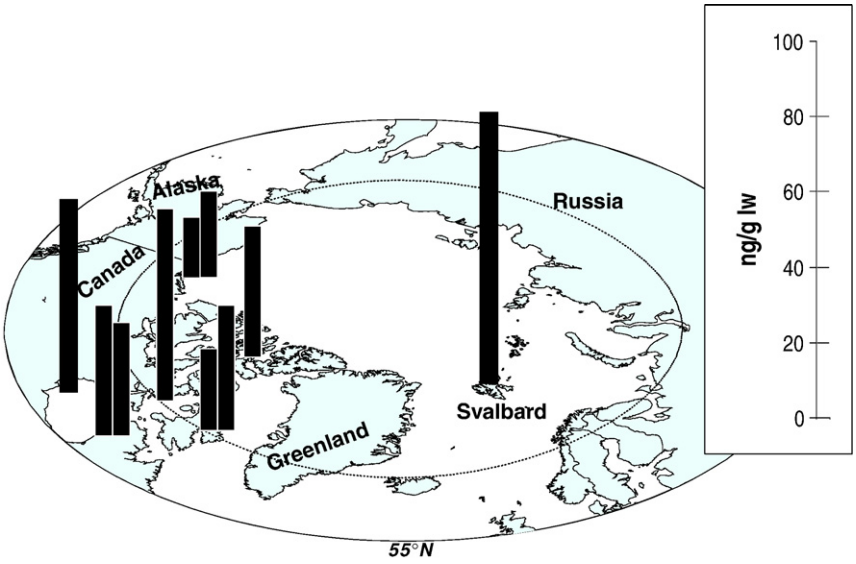

Fig. 15. ΣPBDE concentrations in beluga blubber. Results from Wolkers et al. (2006a), Tomy (2007) and Tomy et al. (2007, 2008b).

The model parameterized PBDE concentrations in various abiotic media (water, sediment), as well as small and large Arctic char and prey items such as insect larvae. Large char were assumed to cannibalize smaller char as well. The model results, with and without biotransformation as a variable, were then compared to measured concentrations found in Arctic char. The model without biotransformation underestimated the measured BDE-47 concentration by 30% but overestimated BDE-99 and -153 concentrations by 75 and 250%, respectively. When biotransformation via metabolic debromination of BDE-99, -100 and -153 to BDE-47 was included in the model, the estimates were closer to the measured values. The authors concluded that biotransformation was a significant factor in explaining PBDE concentrations in the food web i.e. elevated BDE-47 was best explained by metabolic debromination of more highly brominated congeners.

In a controlled study, domestic Greenland sledge dogs were used as surrogate species for East Greenland polar bears. Dogs were fed a diet containing either minke whale blubber or pork lard to study accumulation and the possible effects of organohalogenated compounds (Verreault et al., 2008). Biomagnification factors (BMFs) were calculated on a congener-specific basis and compared to polar bear-ringed seal BMFs. For both polar bear and sledge dogs, the BDE congener that showed the highest propensity for biomagnification was BDE-153 with a BMF of 7 for the exposed sledge dogs and 11 for the free ranging polar bears. BDE-99 and -100 showed different BMFs for each species (Fig. 16), indicating that dogs and polar bears have different metabolic capacities.

Arctic fox from Svalbard were analyzed for tetra-hexaBDEs as well as stable carbon and nitrogen isotopes (Fuglei et al., 2007). The stable carbon isotope ratio (δ^{13}C) distinguishes between dietary intake of predominately terrestrial (lower ratio) or marine (higher ratio) origin and the nitrogen isotope ratio (δ^{15}N) provides information on what trophic level the fox are feeding at. Arctic fox on Svalbard feed on a variety of both terrestrial and marine prey depending on time of year and geographic location. The δ^{13}C and δ^{15}N ratios in Arctic fox were found to be significantly correlated to each other, indicating that the fox feeding in the marine food web also feed at a higher trophic level than fox feeding on more terrestrial food. ΣPBDE in abdominal- and subcutaneous tissue were significantly positively correlated to δ^{15}N, indicating that trophic level is directly related to higher PBDE bioaccumulation in arctic foxes on Svalbard.

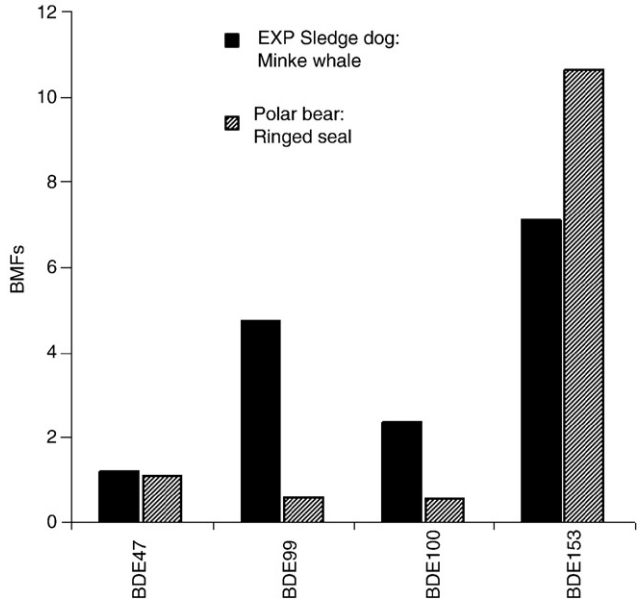

Fig. 16. Biomagnification factors (BMFs) of major PBDE congeners (detected in 60% or more of the samples) based on adipose tissue/blubber concentrations (wet weight basis) from Greenland minke whale blubber to sled dog, and from Greenland ringed seal to Greenland polar bear. Reprinted from Verreault et al. (2008) Comparative fate of organohalogen contaminants in two top carnivores in Greenland: Captive sledge dogs and wild polar bears, Comparative Biochemistry and Physiology, Part C, 147, 306–315, with permission from Elsevier Ltd.

The biomagnification of PBDEs and HBCD was studied in the polar bear food chain at Svalbard, including four invertebrate species (*Calanus glacialis*, *Thysanoessa iermis*, *Themisto libellula* and *Gammarus wilkitzkii*), polar cod, ringed seals and polar bears (Sørmo et al., 2006). The authors chose whole body concentrations (WBCs) over lipid weight concentrations (LWCs) to assess biomagnification, as they suspected bias in estimations of food web transfer if the lipid content varied between organisms. However, the calculation of WBCs of polar bears included some rough estimations of the total lipid content of the bears. Depending on whether WBCs or LWCs were used, the resulting biomagnification factors (BMFs) for ΣPBDE varied by a factor of up to 4, usually with lower LWC-based BMFs. The authors therefore conclude that the use of LWCs may underestimate the actual biomagnification potential of PBDEs. In general, the BMFs indicated biomagnification in the food chain for those BDE congeners that were above the limit of detection. In contrast to BDE-47, BDE-99 did not biomagnify at the lower trophic levels, i.e. from herbivorous copepods to omnivorous amphipods and from pelagic zooplankton to polar cod, which might be related to metabolic debromination of BDE-99 (Sørmo et al., 2006; Stapleton et al., 2004). The largest biomagnification factor was found from zooplankton and polar cod to ringed seal, consistent with the greater energy demand and food intake of mammals. In contrast to the Verreault et al. (2008) study, BDE-47 had the highest BMF. From polar cod to ringed seal, for example, the BMFs of BDE-47 and BDE-99 were 209 and 56.6, respectively, based on WBCs.

Interestingly, most PBDEs were found in lower concentrations in polar bears than in ringed seals, indicating considerable differences in biotransformation of PBDEs in these two species. Higher oxidative biotransformation capacities in polar bears than in seals has previously been shown for PCBs. This may possibly be a species-specific difference or a result of enhanced enzyme induction due to the higher exposure of polar bears (Letcher et al., 1996; Sørmo et al., 2006). However, BDE-153 was an exception, as it was the only congener to biomagnify from ringed seals to polar bears (BMF = 5.2), similar to what was seen in Canadian polar bears (BMF = 71) (Muir et al., 2006b). CB-153 is the most bioaccumulative CB congener, and the similarity in halogen substitution pattern may explain why BDE-153 also biomagnifies to such a large extent (Sørmo et al., 2006).

BDE-209 and HCBD were only analyzed in some of the species in the study by Sørmo et al. (2006). BDE-209 was detected in one sample from ringed seals and all tissue samples from the polar bears, suggesting some uptake and food web transfer of BDE-209 as well. As for the lower brominated BDEs, HBCD biomagnified strongly from polar cod to ringed seal (BMF of 36.4, based on whole body wet weight concentrations), but did not biomagnify from ringed seal to polar bear (BMF of 0.6). As ΣHBCD was reported, the bioaccumulative potential of the individual HBCD isomers could not be assessed.

Haukås et al. (2007) studied biomagnification of PBDEs in a Barents Sea (east of Svalbard) food web, which included *G. wilkitzkii*, polar cod, black guillemot and glaucous gulls. Trophic levels were determined using δ^{15}N ratios. PBDE concentrations were below detection in *G. wilkitzkii*. BDE-28 and -47 biomagnified from polar cod to black guillemot (BMFs of 28 and 45, respectively), from polar cod to glaucous gull (BMFs of 60 and 690, respectively) and from black guillemot to glaucous gull (BMFs of 15 and 110, respectively).

In another study from Svalbard, the transfer of BDE-47 and BDE-99 from fish to beluga and narwhal was studied (Wolkers et al., 2006a). Based on metabolic indices, both congeners accumulated in the two whale species. In general, the metabolic indices were quite similar for beluga and narwhal, but small differences were observed in the contamination pattern of organochlorines, indicating subtle differences

in contaminant metabolism. Regarding the PBDE pattern, no statistically significant difference was found between beluga and narwhal, as BDE-47 accounted for about 90% of ΣPBDE in both species. Organochlorine concentrations were higher in beluga and narwhal than in harp seal although their diets are similar. The authors discuss two opposing effects, which could result in slightly higher concentrations in harp seals than in whales (factor of 1.3). On the one hand, harp seals need more food per body mass than whales, on the other hand, the contaminants are stored in a larger mass-specific blubber depot. As organochlorine concentrations were considerably higher in whales than in harp seals (by more than a factor of 10), the authors conclude that whales have a decreased metabolism relative to seals. Limited PBDE data did not allow for a similar comparison for PBDEs.

Kelly et al. (2007) applied a bioaccumulation model to identify potentially bioaccumulative substances among commercial chemicals. Biomagnification factors for BDE 47, 99 and 209 were included in the calculations for water-respiring and air-breathing organisms assuming no metabolism. In the piscivorous food web, concentrations of non-metabolizing chemicals with log K_{ow} between 5 and 8 biomagnify in top-level predatory fish. BDE-47 and-99 (log K_{ow} of 6.0 and 6.8, respectively) thus had estimated BMFs of 5.1 and 7.5, respectively. BDE-209, with a log K_{ow} of 9.9, is absorbed too slowly and the model predicts that it does not biomagnify in piscivorous food webs. However, in the marine mammalian food web, which includes water-respiring invertebrates and fish as well as air-breathing birds and mammals, poorly metabolized chemicals with a log $K_{ow} \geq 5$ and log $K_{oa} \geq 6$ are predicted to biomagnify, attaining concentrations in top predators (polar bears) up to 10,000 times the concentrations in primary producers. This is predicted to be the case for BDE-47, -99 and -209, with log K_{oa}s of 9.8, 11.2 and 13.1, respectively. In terrestrial food webs, chemicals with a log K_{ow} between 2 and 10 and a log $K_{oa} \geq 6$ are predicted to biomagnify up to 400-fold. In other words, PBDEs are predicted to biomagnify through all parts of the food web, both in marine and terrestrial organisms, leading to exponentially increasing concentrations in top predators. Seabirds, marine mammals and terrestrial carnivores are characterized by the highest BMFs of 19, 49 and 98, respectively, for both BDE-47 and -99. In the model, BDE-209 is biomagnified in marine mammals and terrestrial carnivores with BMFs of 3 and 8.

In another study, Kelly et al. (2008) studied actual biomagnification of PCBs and PBDEs in an Arctic marine food web in Hudson Bay, Canada. The study included analysis of sediment, macroalgae, blue mussels, capelin, polar cod, sculpin, beluga, ringed seal and polar bears. The results from this study showed that, contrary to what was predicted above, only BDE-47 showed any tendency to biomagnify in this food web, with a trophic magnification factor (TMF) of 1.6. No other tri-hexaBDEs had TMFs above 1, whereas all tri-decaCBs did biomagnify (TMFs of 2.9–11). The authors drew the conclusion that the low biomagnification of BDEs is due to their metabolism in the species that were studied in this particular food web.

In a recent paper, Tomy et al. (2008b) reported enantioslective bioaccumulation of HBCD and congener specific accumulation of PBDEs in an eastern Canadian Arctic marine food web. The overall rank order of Σ2HBCD (α- and γ-HBCD) concentrations were narwhal > redfish ≈ clams > shrimp > beluga > zooplankton > walrus > arctic cod. No clear trend in the diastereoisomer profile was observed. However, the α-HBCD diastereoisomer contributes greater than 70% of the total burden in shrimp, redfish, arctic cod, narwhal, and beluga, whereas zooplankton, clams, and walrus contain >60% of the γ-HBCD diastereoisomer. Significant trophic magnification was seen for α-HBCD (TMF of 2.1), primarily for the (−)α-enantiomer (TMF = 2.2), and for BDE 47 (TMF of 2.5) (Fig. 17). Interestingly, BDE 209 and γ-HBCD showed significant negative slopes resulting in TMFs of 0.3 and 0.5, respectively, indicating trophic dilution, i.e. decreases in concentrations with increasing trophic levels for these two BFRs (Fig. 17).

Ikonomou et al. (2005) reported PBDE profiles in various organisms of the Arctic marine food web, in comparison with the Bromkal® PentaBDE formulation. In addition to algae, mussels, eider duck, cod, beluga and seal, sediment and lichens were included. Their PBDE profile was very similar to that of the technical mixture. As had been shown in other food web studies, the percentage of BDE-47 increased with increasing trophic level, eventually accounting for approximately 70% of ΣPBDE in seals. Thus, the PBDE burden of higher trophic level marine animals predominantly consists of lower brominated congeners. The authors attribute the change in PBDE pattern to biomagnification and bioconversion/biodegradation processes, more precisely a debromination along the pathways BDE-153 → BDE-99 → BDE-47 and BDE-183 → BDE-154 → BDE-100. It is interesting that the PBDE profile in eider ducks stands out, with approximately equal shares of BDE-47, -99, -100 , -153 and -154.

Shorthorn sculpin liver, ringed seal blubber and polar bear adipose tissue were collected from the same location in central East Greenland in 2002 (polar bear: 1998–2002) (Frederiksen et al., 2007). While the absolute concentrations of HBCD are uncertain and possibly present minimum concentrations, increasing concentrations were found in the order shorthorn sculpin < ringed seal blubber < polar bear tissue, possibly indicating biomagnification of HBCD. However, analysis of liver samples of ringed seal and polar bear did not confirm this trend for HBCD. As that study had a different objective, more detailed food web studies will be required to assess the biomagnification of HBCD.

Biomagnification studies generally show that the lower brominated BDE congeners and HBCD do biomagnify. BDE-47 and -153 in particular seem to biomagnify highly in comparison with other BDEs. Some differences are seen, however, possibly due to differences in metabolic capacities in some species, such as polar bears. For HBCD, biomagnification is seen mainly for α-HBCD. In conclusion, although there are differences in the magnitude of the biomagnification of single PBDE congeners in general PBDEs, as well as HBCD, have considerable potential to biomagnify in both marine and the terrestrial ecosystems.

8. Conclusions

8.1. PBDEs

As in the previous review, most new data for brominated compounds in the Arctic have been generated for the PBDEs, particularly the components of the PentaBDE technical product, although more data are now available also for the higher brominated BDEs including several octa- and nonaBDEs and BDE-209. The di-heptaBDEs are found in numerous abiotic (air, soil, moss, freshwater and marine sediments) and biotic matrices (zooplankton, invertebrates, fish, terrestrial birds, seabirds, terrestrial and marine mammals) from Alaska to the Barents Sea and the number of species studied has increased. As in the previous review, there are some indications of elevated concentrations near emission sources, such as much higher PBDE concentrations in seabirds from the coast of the Gulf of Alaska, which is more populated, compared to seabirds from the Bering Sea, as well as higher concentrations in soils from inside a landfill compared to outside the landfill. Latitudinal trends for lower brominated PBDEs are seen in moss with declining concentrations of ΣPBDEs with increasing latitude. Higher concentrations of PBDEs are seen in some freshwater lake sediments on Svalbard as well as in Lake Ellasjøen on Bjørnøya, which may be due to inputs of seabird guano. Spatial trends of ΣPBDEs and HBCD in several species indicate that the European Arctic is more contaminated than the North American Arctic, implicating similar atmospheric transport pathways for PBDEs as for organochlorines, i.e. from highly populated eastern North America and western and central Europe.

Lowest ΣPBDE concentrations are generally seen in the terrestrial ecosystem, exemplified by soils, moss, moose and grouse, and highest concentrations are seen in marine seabirds and marine mammals at high trophic levels in the food web. The highest ΣPBDE concentrations are seen in Alaskan offshore killer whales, which are similar to what

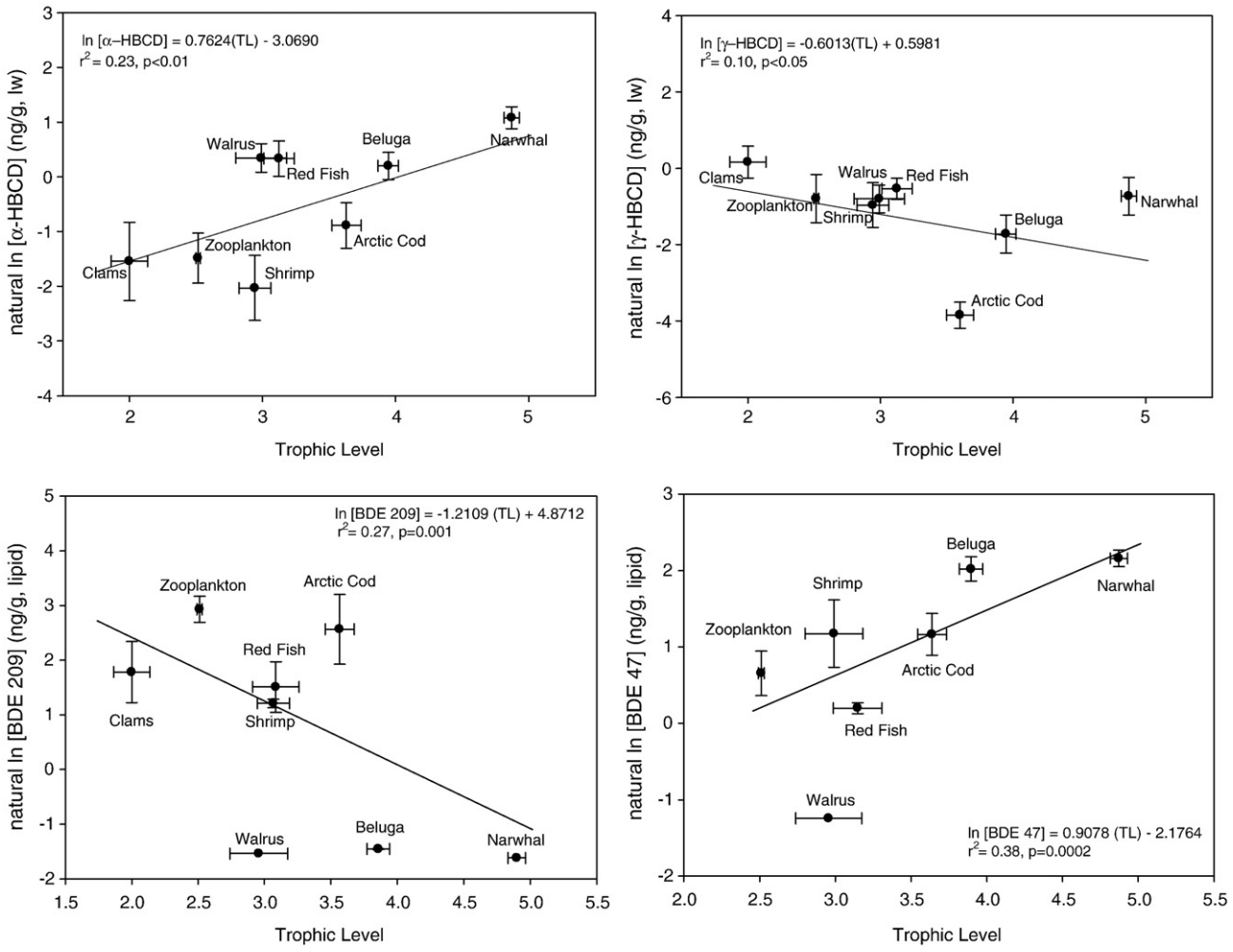

Fig. 17. Mean (±standard error) concentrations of α-HBCD (top left), γ-HBCD (top right), BDE-209 (bottom left panel, ng/g) and BDE-47 (bottom right panel), versus the trophic level relationship (±standard error) for the Eastern Arctic food web. Concentrations were lipid and control corrected. Regression analysis plotted on each panel. Clam purposely omitted from the plot for BDE-47. Reprinted with permission from Tomy et al. (2008b) Enantioselective bioaccumulation of hexabromocyclododecane and congener-specific accumulation of brominated diphenyl ethers in an eastern Canadian Arctic marine food web, Environmental Science & Technology 42, 3634–3639. Copyright (2008) American Chemical Society.

has previously been seen in long-finned pilot whales from the Faroe Islands. Alaskan transient killer whales and northern Norwegian killer whales had somewhat lower concentrations but were still considerably higher than has been seen in other Arctic biota, including polar bears. High PBDE concentrations were also seen in landlocked Arctic char from Lake Ellasjøen on Bjørnøya, which is due to the input of seabird guano from colonies on a nearby cliff. ΣPBDE concentrations in Arctic biota are lower than in biota from more southerly latitudes, and are generally lower than organochlorine compounds. Temporal trends for the lower brominated BDEs indicate continued increases (air, freshwater sediment, land-locked char, lake trout, burbot, ivory gull, West Greenland ringed seal, East Canada beluga) or a tendency to leveling off or possible declines (land-locked char, lake trout, Atlantic cod, thick-billed murres, northern fulmars, East Greenland and western Canada ringed seal, western Canada beluga), depending on the matrix studied and the geographic location. Many of the lower brominated BDEs bioaccumulate and biomagnify, particularly BDE-47 and -153.

BDE-209 had previously been found in a few freshwater sediments and one marine sediment, blue mussels, Atlantic cod, terrestrial birds of prey, glaucous gulls and polar bears, but has now also been found in Arctic air samples, soils, moss, moose, more marine sediments, zooplankton, shrimp, clams, amphipods, redfish, polar cod, Brünnich's guillemot/thick-billed murre, northern fulmars and ringed seals. More data have also been generated for BDE-209 in glaucous gulls and polar bears. Latitudinal trends are seen in moss with declining concentrations of BDE-209 with increasing latitude. The finding of BDE-209 in high volume air samples (filters — particle bound) from Arctic sites and modelling results indicating that periods of stable air conditions and high winds, such as during Arctic haze events, will lead to episodes of long range transport for particulate-bound contaminants, are strong indications that BDE-209 undergoes long-range transport to the Arctic. This is further supported by the findings of BDE-209 as the predominant BDE congener in soils and mosses, probably due to deposition, as well as its presence in biotic samples, including a number of higher trophic level organisms such as glaucous gulls, ringed seals and polar bears. Although it is found in high trophic level biota, BDE-209 appears to undergo trophic dilution instead of accumulation in the marine food web, leading to decreasing concentrations at higher trophic levels. There is an increasing temporal trend in air samples from Canada. Otherwise, there are no new studies presenting spatial or temporal trends for BDE-209 and therefore such studies should be undertaken.

8.2. HBCD

HBCD was previously seen in a few high volume air samples (northern Sweden, Finland), blue mussels, Atlantic (northern

Norway) and polar cod (Svalbard), peregrine falcons (Sweden, Greenland), glaucous gulls (Bjørnøya), ringed seals (Canada, Svalbard), beluga (Canada) and polar bears (Alaska, Canada, Greenland). Considerably more data have been generated for the Arctic, including more isomer-specific data. HBCD has been found in new high volume air samples (Svalbard), freshwater sediments (Bjørnøya), freshwater fish (Canada), marine sediments (Barents and Pechora Seas), zooplankton (Canada), marine invertebrates (Canada, Svalbard), marine fish (Canada, Svalbard, Greenland), seabirds (Canada, Greenland, Svalbard, Bjørnøya, Iceland, Faroe Islands, northern Norway), ringed seals (Greenland, Svalbard), beluga (Canada,), minke whale (Greenland), long-finned pilot whale (Faroe Islands) and polar bears (Svalbard, Greenland). Where isomer-specific analysis was done, γ-HBCD was the predominant isomer in air, α-HBCD predominates in biota, and for marine sediments, similar concentrations of α-, β- and γ-HBCD were found. As for PBDEs, it is now clear that HBCD is ubiquitous in the Arctic, undergoes long range atmospheric transport, bioaccumulates and the α-isomer biomagnifies, whereas the γ-isomer undergoes trophic dilution. Very few temporal trends are available, and indicate no change (eastern and western Canadian beluga, Canadian burbot, Brünnich's guillemot from Svalbard and Bjørnøya) or possibly increasing concentrations (ivory gull, northern fulmars and ringed seals from Canada) of HBCD with time, but more data are needed before any conclusive statements can be made. Spatial trends seem to indicate a similar pattern as for organochlorines and for PBDEs, with higher concentrations in the European Arctic.

8.3. Other BFRs

TBBPA was screened for in marine sediments in one study, and TBBPA and methyl-TBBPA were screened for in shorthorn sculpin, several seabird and marine mammal species in another study but these were below detection limits in all sample types. No other studies included the analysis of TBBPA or Me-TBBPA. TBBPA is a high volume chemical and in the previous review, some studies did show its presence in moss, marine sediments, fish liver and Norwegian peregrine falcon and golden eagle eggs and Me-TBBPA was found in Greenland peregrine falcon eggs. More data are needed to understand possible long-range transport of TBBPA to the Arctic, how prevalent it is in Arctic abiotic and biotic samples, and if there are temporal and spatial trends.

PBBs were only included in a few studies. BB-153 was found in air samples from Svalbard, land-locked Arctic char and northern fulmars from Bjørnøya, northern fulmars from the Faroe Islands and polar bears from Canada and BB-101 was found in glaucous gulls from Svalbard and ivory gulls from Canada. In the GC-MS analysis, BB-153 often co-elutes with BDE-154, which may mask the presence of BB-153, as was shown in studies of ivory gull and Arctic fox. Thus, BB-153 may be more prevalent in the Arctic, but methods to separate it from the BDEs are needed to confirm this.

A range of new BFRs have been screened for and in many cases detected in Arctic biota including BTBPE, DBDPE, HxBBz, PBEB, PBT and TBECH. BTBPE was detected in northern fulmars (Faroe Islands), glaucous gulls (Bjørnøya), beluga (Canada), and ringed seals (Canada). HxBBz, PBEB and PBT were detected in glaucous gulls (Bjørnøya) and HxBBz was detected in East Greenland polar bears. TBECH was detected in beluga (Canada). DBDPE was analyzed for but not detected in ringed seals from Canada. Currently, the presence of several of these BFRs is also being screened for in polar bear samples (Letcher, 2007). The results for these new BFRs indicate that several are reaching the Arctic and can accumulate in higher trophic level organisms. As many of these are being used as replacements for PBDEs that have been banned or discontinued, their presence in the Arctic is a warning sign that these compounds may also undergo long-range atmospheric transport, are potentially bioaccumulative and that increasing use will lead to increasing concentrations in the environment with time. There is thus a need to increase our knowledge of the prevalence of these compounds in Arctic abiotic and biotic samples, spatial and temporal trends and confirmation from air sampling that they are undergoing long-range transport.

8.4. Data gaps

Studies published since mid-2005 have contributed substantial additional information on current levels, trends and biomagnification of BFRs in the Arctic, particularly for marine biota. The presence of BDE-209 at low but readily detectable levels in air, soil and in lake sediments and a variety of organisms including polar bear, illustrates that even very non-volatile chemicals can reach the Arctic and accumulate. Overall, concentrations of BFRs measured to date have been low compared with legacy OCs. However, the number of BFRs determined has been rather limited compared to the number in commerce. There are a number of semi-volatile BFRs including but not limited to tetra-hexaBDE, HBCD, TBECH, (Muir and Howard, 2006) with relatively long predicted atmospheric oxidation half-lives which could be atmospherically transported, and would be good candidates for future surveys of spatial and temporal trends. Recently, Brown and Wania (2008) have used an extensive screening method specifically designed to select chemicals with high likelihood of becoming Arctic contaminants. The method combines models predicting long-range atmospheric transport, accumulation in Arctic marine food webs and bioaccumulation in humans. Besides the PBDEs, they identify a number of brominated flame retardants that would be good candidates for further screening in Arctic samples. The fact that a wide range of BFRs are now being detected in the Arctic illustrates the need for more information on these compounds and others predicted to reach the Arctic.

The use of stable isotopes is a relatively inexpensive method to measure trophic levels in food webs and has been used successfully in a few Arctic bioaccumulation studies. We encourage more studies that include these methods in order to improve the interpretation of data when numerous trophic levels are studied.

Very little data were available from the Russian and Icelandic Arctic for any matrices. Very little data in biota were available from the terrestrial ecosystem in any part of the Arctic.

There is a general lack of recent information on production/use volumes of BFRs, including total volumes and geographical distribution. China is now a known producer of BFRs but production volumes for this country are unknown. Thus, more information on production volumes and the geographical distribution of production and use of BFRs is needed.

In the previous review, the presence of Penta-, Octa-, and DecaBDE, PBBs and HBCD in the Arctic was taken as indications that these compounds should qualify as POPs under the Stockholm Convention (de Wit et al., 2006). The data presented here substantiate the previous review's conclusions, particularly for components of Octa- and DecaBDE as well as HBCD. The data also indicate that other BFRs may need to be considered in the near future.

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Appendix A. Supplementary data

Supplementary data (a table of PBDE, HBCD and PBB concentrations in various Arctic media) associated with this article can be found, in the online version, at doi:10.1016/j.scitotenv.2009.08.037.

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