Marine Pollution Bulletin 78 (2014) 258–266

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Baseline

Organohalogen contaminants and total mercury in forage fish preyed upon by thick-billed murres in northern Hudson Bay

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ARTICLE INFO

Key Words:
Hudson Bay
Fish
Thick-billed murre
Halogenated compounds
Mercury
Biomagnification factors

ABSTRACT

Twelve marine fish species collected from a thick-billed murre (*Uria lomvia*) breeding colony in northern Hudson Bay in the Canadian Arctic during 2007–2009 were analyzed for legacy organochlorines (e.g. PCBs, DDT), polybrominated diphenyl ethers (PBDEs), perfluorinated carboxylates (PFCAs) and sulfonates (PFSAs), and total mercury (Hg). No one species of prey fish had the highest levels across all contaminant groups analyzed. For the two pelagic fish species sampled, concentrations of the major organochlorine groups (e.g. Σ_{21}PCB, ΣDDT, ΣCHL, ΣCBz), ΣPBDE, ΣPFCA and Hg were consistently higher in Arctic cod (*Boreogadus saida*) than in capelin (*Mallotus villosus*). Biomagnification factors from whole fish to thick-billed murre liver across all species were generally higher for Σ_{21}PCB and ΣDDT. ΣPBDE did not biomagnify.

Bioaccumulation and biomagnification of environmental contaminants have been reported for a number of arctic marine food webs, including the Canadian Arctic (e.g. Atwell et al., 1998; Campbell et al., 2005; Fisk et al., 2001a; Hargrave et al., 1992; McKinney et al., 2012; Muir et al., 1988; Powley et al., 2008; Tomy et al., 2004, 2009). To date, only a few studies have looked at contaminants in the marine food webs of Hudson Bay. Mercury was recently examined in a Hudson Bay marine food web with a focus on zooplankton (Foster et al., 2012), and Kelly et al. (2008, 2009) examined concentrations of PCBs, organochlorine pesticides (e.g. *p,p′*-DDE, mirex, dieldrin, *trans*-nonachlor, hexachlorobenzene, β-HCH), brominated flame retardants (BFRs) such as the polybrominated diphenyl ethers (PBDEs), as well as per- and poly-fluoroalkyl substances (PFASs) in an eastern Hudson Bay food web. However, to our knowledge, no study has examined contaminants in prey of seabirds breeding in Hudson Bay.

The thick-billed murre (*Uria lomvia*) is a circumpolar seabird which breeds only in the Arctic and sub-Arctic (Gaston and Hipfner, 2000), with large populations in the eastern Canadian Arctic including Hudson Bay (Gaston and Hipfner, 2000). Thick-billed murres feed on small fish and large zooplankton (Gaston and Hipfner, 2000; Provencher et al., 2012), foraging at depths down to 150 m (Elliott et al., 2009b; Falk et al., 2000). Arctic cod (*Boreogadus saida*) was, until recently, the main prey of thick-billed murres breeding in the Canadian Arctic (Davidson et al., 2008; Gaston and Bradstreet, 1993) and, until the mid-1990s, was the most common prey item found in the diet of nestling murres throughout the Canadian Arctic (Gaston and Jones, 1998). However, dietary studies have shown that there has been a shift from Arctic cod and benthic fish species to capelin (*Mallotus villosus*) and sand lance (*Ammodytes* spp.) in the diet of thick-billed murres at Coats and Digges Islands in northern Hudson Bay between 1980 and 2002 (Gaston et al., 2003, 2012).

Food delivered by adult thick-billed murres to nestlings is carried externally in the bill so that it arrives at the colony essentially whole and readily identifiable (Elliott and Gaston, 2008; Gaston et al., 2003). For a variety of reasons, food brought back to the colony is sometimes left uneaten on the breeding ledges (Elliott and Gaston, 2008). These abandoned prey items present an ideal opportunity to evaluate the contaminants in a variety of small forage fish inhabiting the waters of northern Hudson Bay.

The objectives of this study were to (i) report on concentrations of several persistent halogenated compounds such as PCBs, legacy organochlorine pesticides (e.g. DDT, chlorobenzenes, chlordanes), BFRs including the PBDEs, major bioaccumulative PFASs [perfluorinated carboxylates (PFCAs) and perfluorinated sulfonates (PFSAs) including perfluorooctane sulfonic acid (PFOS)] as well as their

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http://dx.doi.org/10.1016/j.marpolbul.2013.11.003

precursors [fluorotelomer alcohols (FTOHs), fluorotelomer unsaturated acids (FTUCAs), perfluorosulfonamides (FOSAs)] and total mercury (Hg) in fish delivered to nestlings of thick-billed murres breeding at Coats Island in northern Hudson Bay, and (ii) compare biomagnification factors (BMFs) from fish to thick-billed murre among contaminant groups.

Representative samples of small fish were collected opportunistically from the breeding ledges of thick-billed murres at Coats Island (62°98′N, 82°00′W) in northern Hudson Bay, Nunavut, Canada, during 2007–2009 (Fig. 1). Species sampled included Arctic cod, capelin, sand lance, Atlantic poacher (*Leptagonus decagonus*), Arctic shanny (*Stichaeus punctatus*), daubed shanny (*Leptoclinus maculatus*), banded gunnel (*Pholis fasciata*), fish doctor (*Gymnelis viridis*), fourline snake blenny (*Eumesogrammus praecisus*), Arctic staghorn sculpin (*Gymnocanthus tricuspis*), other sculpin (*Triglops* spp.) and snailfish (*Liparis* sp.). Arctic cod and capelin are categorized as being pelagic, sand lance as bentho–pelagic, and the rest as benthic species (Froese and Pauly, 2013). Fresh fish collected from the ledges were identified and measured as described by Elliott and Gaston (2008). Samples were washed, individually wrapped in foil, placed in plastic bags and frozen at -20 °C in the field before being shipped to the National Wildlife Research Centre (NWRC), Ottawa, Ontario, where they were stored at −40 °C prior to chemical analysis.

Fish were analyzed for various persistent organohalogen pollutants and total Hg either individually or as composite samples (pools) comprised of 2–8 fish (see Table S1). Pooled samples were created by taking equal aliquots from each fish. In some cases, only sagittal sections were available after the other half of the fish was used for other analyses but it was assumed that the sagittal sections were representative of the whole fish.

Homogenized samples were analyzed for organochlorines (OCs) including chlorobenzenes (ΣCBz = 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene), hexachlorocyclohexanes (α-, β- and γ-hexachlorocyclohexane), chlordane-related compounds (ΣCHL = oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor and heptachlor epoxide), DDT and its metabolites (ΣDDT = *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT), octachlorostyrene (OCS), mirex, photomirex, dieldrin and PCB congeners (ΣPCB). Of the 74 PCB congeners analyzed, only 21 congeners were detected: 31/28, 44, 52, 66, 70/76, 74, 95, 99, 101/90, 110, 118, 138, 149, 151, 153, 179, 180 and

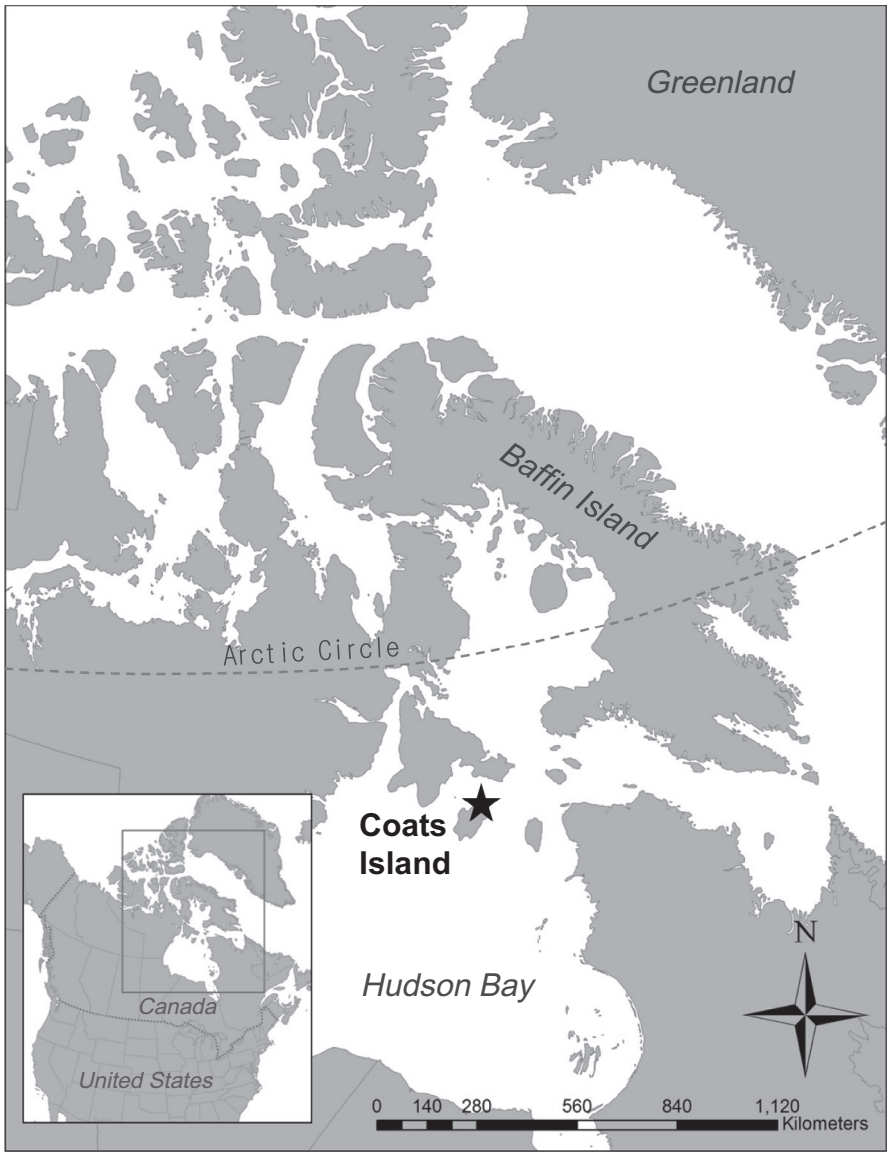

Fig. 1. Location of study site in northern Hudson Bay.

187 as identified according to IUPAC numbers (Ballschmiter et al., 1992).

Samples were analyzed for OCs by gas chromatography using a mass selective detector (GC/MSD) and lipids were determined by gravimetric methods. Chemical extraction and cleanup of PCBs and OC pesticides followed the procedures of Lazar et al. (1992). Chemical analysis was performed using a capillary gas chromatograph (Agilent 6890 N) coupled with a mass selective, single quadrupole detector (Agilent 5973 N) operated in positive electron impact (EI) mode. PCBs and OC pesticides were determined using an internal standard method. For each batch of samples, one duplicate extraction, one duplicate injection, two/three method blanks, one/two in-house reference materials (Reference Egg Pools DCCOQA-2011-01, or DCCOQA-2011-06 and DCCOQA-2011-07) and one certified reference material (Lake Michigan Fish Tissue NIST1947) were run for quality control. All reported residue levels were corrected for internal standard recoveries. The nominal detection limit was 0.1 ng g^{-1} wet weight.

Homogenized samples were also analyzed for polybrominated diphenyl ethers (PBDEs), polybrominated biphenyls (PBBs) and total (α) hexabromocyclododecane (HBCDD). Sample extraction and clean-up were the same as for the OCs except that tissue homogenates were spiked with internal standards (BDE-30, BDE-156, labeled $^{13}C_{12}$-BDE-209). Chemical analysis for 14 PBDE congeners (BDE-17, -28, -47, -49, -66, -85, -99, -100, -138, -153, -154 (co-elution with BB-153), -183, -190 and -209), BB-101 and total-(α)-HBCDD was performed using an Agilent 6890 gas chromatograph (GC) equipped with a 5973 quadrupole mass spectrometer (MS) detector run in electron capture negative ionization (ECNI) mode. For method details, see Chen et al. (2012). For each batch of samples, one duplicate extraction, one duplicate injection, two/three method blanks, and the same in-house and certified reference materials identified for the OC analysis were run for quality control. Quantification of the brominated compounds was performed using an internal standard method (see Chen et al., 2012). The method limit of quantification (MLOQ) was 0.1 ng g^{-1} ww for all brominated compounds except for HBCDD (1 ng g^{-1} ww) and BDE-209 for which the MLOQ was 5 ng g^{-1} ww for the 2007 samples and 1 ng g^{-1} ww for the 2009 samples. All reported residue levels were corrected for internal standard recoveries.

BDE-154 generally occurs at lower or similar concentrations to BDE-153 in biota (e.g., Elliott et al., 2005; Hites, 2004; Norstrom et al., 2002). Only one of our samples had a quantifiable BDE-153 concentration and BDE-154/BB-153 was either not detected or only at trace, unquantifiable levels. Therefore, BDE-154/BB153 was not included in the calculation of ΣPBDE concentrations. ΣPBDE was standardized to the sum of BDE-17, -28, -49, -47, -66, -100, -99, -85, -153, -138, -183, -190 and -209.

The extraction, cleanup and analysis of the per- and polyfluoroalkyl substances (PFASs) have been described in Chu and Letcher (2008) and Gebbink et al. (2009). Analysis of PFCAs (C_6–C_{15} chain lengths: PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFTrA, PFTeA and PFPA, respectively), PFSAs [C_4 (PFBS), C_6 (PFHxS), C_8 (PFOS), C_{10} (PFDS)] and FTUCAs (6:2, 8:2 and 10:2) was done using negative electrospray ionization (ESI^-), and the FTOHs (6:2, 8:2 and 10:2) and FOSAs [perfluorooctanesulfonamide (PFOSA), methylated perfluorooctanesulfonamide (N-Me-FOSA)] were analyzed by negative atmospheric pressure photoionization ($APPI^-$). Quantification was performed using an internal standard approach. See Table S2 for a complete listing of all of the above PFASs as well as the ^{13}C- or ^{18}O-enriched internal standards used. Since an isotope dilution quantification approach was used, concentrations were inherently recovery-corrected. Recoveries of the FTOH and FOSA internal standards averaged 65% and, for the PFCAs, PFSAs and FTUCAs, recoveries averaged 50%. For every block of 10 samples, a blank sample and a NWRC in-house reference material (spiked pork liver) was analyzed. See Table S2 for limits of detection (LODs) and method quantification limits (MQLs) for the measured PFASs.

Total mercury (Hg) was analyzed by direct combustion of the homogenized sample in an oxygen-rich atmosphere using an Advanced Mercury Analyzer (AMA-254) equipped with an ASS-254 autosampler for solid samples (see EPA Method 7473; Salvato and Pirola, 1996). Analytical accuracy was determined using three standard reference materials [DOLT-3 and TORT-2 (Canadian National Research Council); Oyster Tissue 1566b (National Institute of Standards and Technology)] plus 11 random samples analyzed in replicate. Recovery of reference materials was within the confidence interval of the certified values and the nominal detection limit for total Hg was 0.006 μg g^{-1} dry weight sample.

Stable-nitrogen isotope assays were performed by continuous-flow stable isotope ratio mass spectrometry according to method described in Chambellant et al. (2013) for the 2007 samples, and in Gebbink et al. (2011) for the 2008 and 2009 samples. Data were normalized using international standards for calibration, and quality control was maintained through sample duplicates. Measurements are reported in standard δ notation in parts per thousand (‰) relative to the AIR international standard. Analytical precision, based on repeat measures of internal standards, was ±0.2‰ for the 2007 samples and ±0.3‰ for the 2008 and 2009 samples.

Non-detect values were set to zero for calculation of the sums of major halogenated groups (e.g. Σ_{21}PCB, ΣDDT, ΣPBDE, ΣPFCA). Unquantifiable trace values for PCB and PBDE congeners were set to one-half the detection limit for calculation of Σ_{21}PCB and ΣPBDE. For the PFCAs, values below the method quantification limit (MQL) were set to one-half the MQL for calculation of ΣPFCA. Total OC (ΣOC) concentrations were calculated as the sum of ΣCBz + ΣHCH + ΣCHL + ΣDDT + dieldrin + Σ_{21}PCB. ΣPFCA was the sum of the C_6–C_{15} chain lengths. For pooled samples, mean values for species were weighted to account for the number of fish in each sample. The tabulated data are presented as arithmetic means, weighted as appropriate, in concentration units of ng g^{-1} lipid weight (lw) for the OCs and PBDEs, ng g^{-1} wet weight (ww) for the PFASs, and μg g^{-1} dry weight (dw) for total Hg. The Spearman Rank Correlation (Statistica for Windows Version 7.0, StatSoft Inc., Tulsa, OK) with a significance level of $p < 0.05$ was used to examine relationships between trophic position ($\delta^{15}N$) and various contaminant groups using the weighted species means. Biomagnification factors (BMFs) were calculated for the major contaminant groups by dividing the mean liver concentration data for female thick-billed murres sampled from Coats Island by weighted mean concentration data for whole fish by species.

For all fish samples analyzed, mean values of $\delta^{15}N$ ranged from 12.9‰ to 17.8‰. Banded gunnel ($n = 3$), Atlantic poacher ($n = 2$), fourline snake blenny ($n = 6$) and Arctic cod ($n = 18$) had the highest mean $\delta^{15}N$ values (17.8‰, 16.1‰, 15.7‰, 15.1‰, respectively), and sand lance ($n = 16$), capelin ($n = 21$) and Arctic staghorn sculpin ($n = 2$), the lowest (12.9‰, 13.4‰, 13.6‰, respectively). Mean $\delta^{15}N$ values for the other fish species ranged from 14.2‰ to 15.0‰.

Arctic cod, Atlantic poacher and fourline snake blenny had the highest mean ΣOC concentrations, and Arctic staghorn sculpin, Arctic shanny, capelin and sand lance, the lowest (Table 1). The ΣOC profile was dominated by ΣCHL (26%), ΣCBz (25%) and α-HCH (21%) in most fish species except for snailfish and banded gunnel which had less α-HCH (7.3% and 7.3%, respectively) and more dieldrin (34% and 19%, respectively), Arctic shanny which had less ΣCHL (4.5%) and more dieldrin (30%), and Arctic staghorn sculpin which had no detectable α-HCH but greater amounts of ΣDDT (27%) and ΣPCB (25%). The contribution of ΣDDT and Σ_{21}PCB to ΣOC averaged 12% and 13%, respectively, over all fish species. CB-153, -138 and -99 comprised over 70% of the Σ_{21}PCB

Table 1
Concentrations (ng g^{-1} lw) of organochlorines and polybrominated diphenyl ethers (PBDEs) as well as $\delta^{15}N$ (‰) in forage fish sampled during 2007–2009 from Coats Island in northern Hudson Bay. Mean values are weighted to account for pooled samples comprised of 2–8 fish. The total number of fish analyzed (*n*) includes fish analyzed both individually and as pooled samples (see Table S1). Minimum and maximum values are given in brackets below the mean, where appropriate, and may include both individual fish and pooled samples.

	Arctic cod	Capelin	Sand lance	Atlantic poacher	Arctic shanny	Daubed shanny	Banded gunnel	Fish doctor	Fourline snake blenny	Arctic staghorn sculpin	Sculpin (*Triglops* spp.)	Snailfish
n	14	13	7	2	1	2	1	6	3	1	7	1
Length (mm)[a]	95–153	87–134	125–150	142, 151	130	121, 220	199	123–195	113–132	117	103–138	124
$\delta^{15}N$	15.4 (15.0, 16.0)	13.7 (12.7, 14.0)	13.3 (12.1, 13.7)	16.1	14.6	14.2	17.7	14.9 (13.7, 15.1)	15.8 (15.7, 15.9)	12.9	15.0 (14.9, 15.1)	14.5
% lipid	2.0 (1.4, 2.8)	6.9 (4.4, 7.6)	4.6 (3.8, 4.8)	4.0	1.7	2.2	3.2	2.4 (1.0, 2.7)	1.4 (1.0, 2.3)	1.0	4.7 (3.0, 6.0)	2.4
Organochorines												
HCB	187 (148, 244)	47.8 (31.1, 52.1)	43.3 (26.8, 48.5)	80.6	27.2	40.3	38.8	46.3 (38.4, 47.8)	62.6 (16.6, 85.6)	26.9	61.3 (55.3, 69.3)	31.9
ΣCBz	201 (158, 272)	52.4 (33.1, 56.4)	43.6 (26.8, 48.5)	86.9	30.1	40.3	41.6	46.3 (38.4, 47.8)	62.4 (16.6, 85.6)	26.9	65.9 (59.9, 73.9)	34.3
α-HCH	72.2 (64.3, 79.5)	46.4 (23.1, 48.9)	57.2 (28.8, 68.0)	71.6	30.7	91.9	18.8	61.7 (16.6, 70.7	88.2 (50.7, 107)	nd	73.6 (72.9, 74.2)	15.6
HE	nd	7.3 (nd, 8.1)	6.8 (nd, 8.2)	nd	nd	nd	nd	nd	nd	nd	nd	nd
Oxychlordane	nd	nd	nd	28.4	nd	nd	nd	nd	nd	nd	nd	nd
cis-Chlordane	53.2 (43.3, 66.2)	14.4 (13.8, 19.1)	9.7 (4.2, 11.0)	49.6	nd	16.3	12.1	16.4 (15.9, 16.5)	21.5 (4.7, 29.9)	nd	27.9 (22.7, 34.9)	14.9
trans-Chlordane	4.5 (nd, 8.1)	1.0 (nd, 1.5)	nd	1.5	nd	nd	3.0	nd	nd	nd	2.8 (2.6, 3.0)	nd
cis-Nonachlor	30.4 (20.6, 40.1)	5.3 (5.0, 5.9)	7.7 (6.0, 8.2)	23.7	nd	10.6	10.3	11.2 (10.8, 13.2)	23.4 (6.9, 31.7)	5.6	12.7 (11.4, 14.6)	5.0
trans-Nonachlor	85.1 (59.5, 109)	15.4 (14.3, 23.8)	23.5 (19.5, 24.3)	78.3	6.7	36.9	43.7	40.2 (33.3, 74.6)	68.6 (20.0, 92.9)	24.1	38.3 (31.9, 46.8)	24.6
ΣCHL	173 (123, 223)	43.4 (42.0, 48.8)	47.7 (30.2, 51.6)	181	6.7	63.9	69.0	67.8 (60.7, 104)	114 (31.7, 155)	29.7	81.7 (68.9, 98.9)	44.6
DDE	71.9 (67.1, 86.7)	16.3 (11.9, 18.5)	18.0 (16.2, 20.2)	62.6	21.7	32.5	34.8	23.6 (21.0, 36.8)	97.0 (26.6, 132)	31.1	27.3 (23.0, 33.1)	23.2
ΣDDT	79.4 (69.5, 97.9)	18.9 (14.4, 20.8)	18.0 (16.2, 20.2)	72.0	21.7	32.5	34.8	23.6 (21.0, 36.8)	97.0 (26.6, 132)	31.1	31.9 (26.6, 39.0)	23.2
Dieldrin	nd	2.9 (nd, 37.8)	3.9 (nd, 27.5)	nd	45.0	nd	48.4	nd	13.8 (nd, 41.3)	nd	nd	72.1
Σ_{21}PCB[b]	95.9 (56.9, 127)	19.0 (8.2, 24.4)	23.6 (16.8, 26.1)	88.1	17.3	25.5	44.4	28.0 (25.2, 42.1)	83.1 (17.5, 116)	28.8	32.5 (27.6, 39.0)	22.3
ΣOC	621 (576, 678)	183 (166, 192)	194 (119, 210)	500	152	254	257	227 (225, 237)	458 (184, 595)	117	286 (257, 324)	212
PBDEs												
BDE 47	4.9 (tr, 6.9)	1.0 (nd, 7.7)	2.5 (tr, 7.1)	2.9	9.0	nd	7.2	5.1 (tr, 21.3)	14.2 (8.0, 17.4)	17.7	tr (tr, tr)	6.9
BDE 99	tr (nd, tr)	tr (tr, tr)	tr (tr, tr)	nd	tr	nd	tr	tr (tr, tr)	tr (tr, tr)	tr	nd	tr
BDE 100	tr (tr, tr)	tr (nd, tr)	tr (tr, tr)	nd	tr	nd	tr	tr (tr, tr)	tr (tr, tr)	tr	tr (nd, tr)	nd
BDE 209	nd	3.0 (nd, 38.6)	31.9 (nd, 185)	nd	68.2	nd	60.2	tr (nd, tr)	18.9 (nd, 56.8)	tr	nd	47.0
ΣPBDE[b,c]	10.6 (6.9, 14.0)	5.8 (1.3, 47.4)	36.6 (3.1, 195)	26.3	83.0	nd	70.5	18.6 (5.5, 84.4)	41.2 (27.2, 69.1)	81.3	1.7 (1.7, 1.7)	55.9

nd – not detected.
tr – trace amount but not quantifiable; i.e.<0.1 ng g^{-1} ww except BDE-209 (<1.0 ng g^{-1} ww).
[a] Fish length reported as total length or fork length, as appropriate; i.e. fork length was reported for Arctic cod, capelin and sand lance.
[b] If more than one PCB or PBDE congener showed a trace value for a given sample, the sum (i.e. Σ_{21}PCB, ΣPBDE) included the trace value as ½ detection limit.
[c] ΣPBDE = Sum of BDE-17, -28, -49, -47, -66, -100, -99, -85, -153, -138, -183, -190, -209.

profile averaged across all fish species. ΣCBz was comprised primarily of HCB (95%) and ΣDDT was comprised primarily of *p,p′*-DDE (93%). ΣCHL was comprised of mainly *trans*-nonachlor (49%) followed by *cis*-chlordane (28%) and *cis*-nonachlor (15%). Oxychlordane was detected only in Atlantic poacher, and heptachlor epoxide was detected only in capelin and sand lance (Table 1). 1,2,4,5-tetrachlorobenzene, OCS, β-HCH, γ-HCH, *p,p′*-DDT, mirex and photomirex were not detected in any of the fish samples.

Arctic cod (a.k.a. polar cod) is a key prey species for both marine mammals and seabirds in arctic marine food webs (Rand et al., 2013), and numerous studies have analyzed this species for contaminants (e.g. Atwell et al., 1998; Borgå et al., 2001, 2005; Campbell et al., 2005; Dietz et al., 1996; Haukås et al., 2007; Hoekstra et al., 2003a; Jaeger et al., 2009; Kelly et al., 2008, 2009; Muir et al., 1988; Powley et al., 2008; Rigét et al., 2000; Sørmo et al., 2006; Stern and Macdonald, 2005; Tomy et al., 2004, 2009; Wolkers et al., 2004). However, there are far fewer data published on contaminants in other forage fish in arctic marine food webs. Given the value of thick-billed murres as indicators of contaminants in the arctic marine environment, there is a need to broaden our understanding of contaminants in other important forage fish in their diet.

Sculpins have been analyzed in studies evaluating the environments near Distant Early Warning (DEW) Line military installations in the Canadian Arctic and sub-Arctic (Bright et al., 1995; Brown et al., 2009; Kuzyk et al., 2005) but as those fish were sampled in close proximity to known contaminant sources, comparisons with data from other locations were not appropriate. Of the sculpins sampled from northern Hudson Bay, the *Triglops* spp. had higher concentrations of ΣCHL, ΣCBz and α-HCH than the single *Gymnocanthus tricuspis* analyzed for OCs (Table 1). The reasons for this are unclear but may be related to diet as suggested by the difference in δ^{15}N values (Table 1).

Several marine fish species [Arctic cod, capelin and shorthorn sculpin (*Myoxocephalus scorpius*)] have been sampled from marine food webs around Greenland (see Rigét et al., 2004) but fish liver was generally analyzed for contaminants making direct comparisons with our data for whole fish difficult. ΣPCB levels in Arctic cod from Baffin Bay (125 ng g^{-1} lw; Borgå et al., 2005), as well as capelin from Cumberland Sound (323 ng g^{-1} lw; McKinney et al., 2012) and southwest Greenland (340 ng g^{-1} lw; Rigét et al., 2004), were much higher than in comparable species from our study (Table 1). Although Arctic sculpin (*Myoxocephalus scorpioides*) from Cumberland Sound (McKinney et al., 2012) was a different species from the sculpins sampled in our study, it, too, had a much higher ΣPCB level (1482 ng g^{-1} lw) than found in the sculpins sampled in our study (Table 1). However, fish size may be a confounding factor in these comparisons (see Cleeman et al., 2000; Stange and Klungsøyr, 1997); e.g. Arctic sculpin from Cumberland Sound and Arctic cod from Baffin Bay were larger than their counterparts sampled from northern Hudson Bay and the sculpins were a different species.

There may also be regional differences. It has been noted that marine species from East Greenland had consistently higher levels of ΣPCB, ΣDDT and ΣHCH than marine species from West Greenland (Rigét et al., 2004). Additionally, chlorobenzene and chlorinated pesticide concentrations in Greenland biota, including marine fish, tended to be similar to those from Svalbard and Iceland but lower than in the same organisms from the Canadian Arctic (Vorkamp et al., 2004). A comparison of contaminants in marine food webs sampled during 1998–1999 in the European Arctic (central Barents Sea) and the Canadian Arctic (northern Baffin Bay) found similar OC concentrations in Arctic cod from the two regions except for ΣHCH which was higher in the northern Baffin Bay food web, possibly due to its closer proximity to sources in eastern Asia (Borgå et al., 2005).

Only α-HCH was detected in the fish sampled in our study. β-HCH is moderately lipophilic but, due to its low K_{OW}-high K_{OA} properties, respiratory elimination is relatively efficient for water-respiring organisms (Kelly et al., 2009) which may explain why it was not detected in our study. However, Moisey et al. (2001) found both β-HCH and γ-HCH in addition to α-HCH in Arctic cod sampled in 1998 from northern Baffin Bay, as did Hoekstra et al. (2003a) in Arctic cod sampled in 1998–2001 from the Beaufort-Chuckchi Seas, and McKinney et al. (2012) in Arctic sculpin and capelin sampled in 2007–2008 from Cumberland Sound. This is likely due to the different transport pathways from historical source areas in Asia from which the α- and γ-isomers reached the Arctic mainly through atmospheric transport while β-HCH was transported mainly by ocean currents via the Bering Strait (Li and Macdonald, 2005). The difference in measured HCH isomers between Baffin Bay and northern Hudson Bay could be related to the easterly flow of ocean currents through the Canadian Arctic Archipelago versus the small flow actually entering into Hudson Bay (Bidleman et al., 2007).

As found in our study, *trans*-nonachlor was also the major chlordane compound found in Arctic cod sampled during 1992–1993 from Resolute Bay (Wiberg et al., 2000) and in Arctic cod sampled during 1998–2002 from the Beaufort-Chuckchi Seas (Hoekstra et al., 2003b). However, the ΣCHL composition in Arctic sculpin and capelin sampled in 2007–2008 from Cumberland Sound differed from our study in that *trans*-nonachlor, oxychlordane and heptachlor epoxide were the major chlordane-related compounds found in Arctic sculpin, and heptachlor epoxide was the major chlordane-related compound in the capelin (McKinney et al., 2012). Given their limited capacity to metabolize chemical compounds, it has been suggested that exposure rather than biotransformation is more important in determining contaminant profiles in fish and invertebrates (Borgå et al., 2004). However, since oxychlordane is a metabolite of *cis*- and *trans*-chlordane, it would appear that some fish species, including Atlantic poacher (this study, see Table 1), may have some capacity to metabolize chlordane compounds.

Mean species trophic position (δ^{15}N) was significantly and positively correlated with ΣCHL ($n = 12$, $r_s = 0.78$, $p = 0.003$), ΣDDT ($n = 12$, $r_s = 0.73$, $p = 0.007$), Σ_{21}PCB ($n = 12$, $r_s = 0.69$, $p = 0.014$) and ΣOC ($n = 12$, $r_s = 0.80$, $p = 0.002$) in our study. No significant correlations were found for ΣCBz or α-HCH. Kelly et al. (2008, 2009) found a high degree of biomagnification of PCBs and other OC pesticides except β-HCH in a Hudson Bay aquatic piscivorous food web. Hallanger et al. (2011) and Hop et al. (2002) also found that trophic magnification factors were generally lower in a food web consisting of only cold-blooded organisms (i.e. aquatic piscivorous food web) compared with a food web including seabirds. For the OCs, BMFs from fish to thick-billed murre were generally highest for Σ_{21}PCB and ΣDDT and lowest for ΣCHL in our study (Table 2). This pattern for BMFs has been observed in other studies which included Arctic cod and thick-billed murres in the Barents Sea (Borgå et al., 2001) and in northern Baffin Bay in the Canadian Arctic (Fisk et al., 2001a). The lower BMFs for ΣCHL may be attributed to their lower concentrations in the thick-billed murres because of their capability to metabolize these compounds (Fisk et al., 2001b).

PBDEs were detected in all fish species except for daubed shanny (Table 1). The major PBDE congeners found in most species were BDE-47, -99, -100 and -209 although only trace levels of BDE-99 and -100 were detected. BDE-209 predominated in the banded gunnel, Arctic shanny and snailfish. BDE-183 was the major PBDE congener found in Atlantic poacher. BDE-17, -28, -85 and -138 were not detected in any samples. Total-(α)-HBCDD was also not detected (<1 ng g^{-1} ww) in any of the samples and BB-101 was not detected (<0.1 ng g^{-1} ww) in any of the samples except for

Table 2
Biomagnification factors from fish to thick-billed murre (TBMU) at Coats Island based on weighted mean concentrations of the major organochlorine groups (ng g^{-1} lw), ΣPBDE (ng g^{-1} lw), ΣPFCA (ng g^{-1} ww) and total Hg (μg g^{-1} dw).

Predator/prey[a]	ΣCBz	ΣCHL	ΣDDT	Σ_{21}PCB	ΣPBDE	ΣPFCA	Hg
TBMU/Arctic cod	4.8	0.8	15	15	0.8	4.0	19
TBMU/capelin	18	3.2	62	77	1.4	12	36
TBMU/sand lance	22	2.9	65	62	0.2	6.4	22
TBMU/Arctic shanny	32	20	54	84	0.1	29	10
TBMU/daubed shanny	24	2.1	36	57	–	32	15
TBMU/banded gunnel	23	2.0	34	33	0.1	5.8	20
TBMU/fish doctor	21	2.0	50	52	0.4	9.2	12
TBMU/fourline snake blenny	15	1.2	12	18	0.2	42	6.6
TBMU/Arctic staghorn sculpin	36	4.6	38	50	0.1	31	20
TBMU/sculpin (*Triglops* spp.)	15	1.7	37	53	4.8	4.1	27
TBMU/snailfish	28	3.1	51	65	0.1	14	13

[a] Ratio of mean concentration for thick-billed murre liver ($n = 5$) sampled from Coats Island in 2007 (Braune et al., Environment Canada, unpublished data) to weighted species mean for whole fish (from Tables 1, 3 and 4).

non-quantifiable trace amounts in Atlantic poacher and one pool of sculpin (*Triglops* spp.) from 2007. The highest mean concentrations of ΣPBDE were found in the Arctic shanny, Arctic staghorn sculpin and banded gunnel, although the highest ΣPBDE concentration measured (195 ng g^{-1} lw) was in a sand lance collected in 2009. The higher ΣPBDE level found in the Arctic staghorn sculpin compared with the *Triglops* spp. as well as other fish species was primarily due to the inclusion of trace values, particularly BDE-209, in the sum calculation.

The mean ΣPBDE concentration in Arctic cod in our study (10.6 ng g^{-1} lw) was very similar to that found in Arctic cod sampled in eastern Hudson Bay (9.8 ng g^{-1} lw) during 1999–2003 (Kelly et al., 2008) but an order of magnitude lower than the mean level (205 ng g^{-1} lw) in Arctic cod (minus the liver) sampled in the Amundsen Gulf in the western Canadian Arctic in 2004–2005 (Tomy et al., 2009). Concentrations of ΣPBDE in capelin (18 ng g^{-1} lw) and Arctic sculpin (73 ng g^{-1} lw) sampled during 1999–2003 in eastern Hudson Bay (Kelly et al., 2008) were higher than the mean ΣPBDE concentrations measured in capelin and some sculpin (*Triglops* spp. but not *Gymnocanthus tricuspis*) in our study (Table 1). The variability found among species and regions may be due to different exposures to PBDEs by region and diet as well as their relatively rapid rate of depuration through biotransformation in arctic marine organisms (Kelly et al., 2008). As a result of this high capacity for biotransformation, Kelly et al. (2008) found that PBDEs exhibited negligible biomagnification in the Hudson Bay food web, an observation which was reflected in the extremely low BMFs (<1 in most cases) for ΣPBDE from fish to thick-billed murre in our study (Table 2). It may also explain why we found no significant correlation between mean species trophic position (δ^{15}N) and ΣPBDE among the fish species in our study. Elliott et al. (2009a) also found that PBDEs did not increase with trophic position in nestling bald eagles (*Haliaeetus leucocephalus*) fed marine prey from North America's west coast.

BDE-47, the primary constituent of the commercial Penta-BDE mixture which was phased out in 2005 (de Wit et al., 2010), was also the major PBDE congener found in Arctic cod in northern Hudson Bay (this study), in eastern Hudson Bay (Kelly et al., 2008) and in the Amundsen Gulf (Tomy et al., 2009). BDE-209, the main constituent of the commercial Deca-BDE product, is less environmentally mobile and bioavailable due to its physical–chemical properties (Boon et al., 2002; Wania and Dugani, 2003) but is still widely used (de Wit et al., 2010) which may explain its detection in the fish sampled in our study (Table 1).

PFOS was the major PFSA detected in the daubed shanny, Arctic staghorn sculpin and capelin, and PFDS predominated in the banded gunnel (Table 3). Given the higher MQL (3.1 ng g^{-1} ww) for PFOS relative to the other PFSAs in conjunction with the high number of detected but non-quantifiable values for PFOS, it was not possible to determine the major PFSA for the other species. PFOSA was detected at non-quantifiable levels (<0.7 ng g^{-1} ww) in some samples and N-Me-FOSA was not detected (<0.2 ng g^{-1} ww) in any samples. No fluorotelomer alcohols (6:2 FTOH, 8:2 FTOH, 10:2 FTOH) were detected (<0.6 ng g^{-1} ww, <0.6 ng g^{-1} ww and <0.5 ng g^{-1} ww, respectively) in any of the samples, nor were fluorotelomer unsaturated acids (6:2 FTUCA, 8:2 FTUCA, 10:2 FTUCA) detected (<0.01 ng g^{-1} ww) in any of the samples.

Table 3
Concentrations (ng g^{-1} ww) of perfluorinated sulfonates (PFSAs) and perfluorinated carboxylates (PFCAs) as well as δ^{15}N (‰) in forage fish sampled during 2007–2009 from Coats Island in northern Hudson Bay. Mean values are weighted to account for pooled samples comprised of 2–8 fish. The total number of fish analyzed (n) includes fish analyzed both individually and as pooled samples (see Table S1). Minimum and maximum values are given in brackets below the mean, where appropriate, and may include both individual fish and pooled samples.

Fish species	n	Length[a] (mm)	δ^{15}N	PFBS	PFHxS	PFOS	PFDS	ΣPFCA[b]
Arctic cod	14	95–153	15.4 (15.0, 16.0)	nd	nd	<3.1	0.56 (0.3, 0.9)	1.45 (1.0, 2.1)
Capelin	14	91–134	13.7 (13.2, 14.0)	nd	nd	3.37 (<3.1, 23)	0.11 (nd, 1.2)	0.46 (0.20, 1.0)
Sand lance	6	131–156	13.0	nd	nd	<3.1	0.20	0.90
Benthic fish								
Arctic shanny	3	124–127	14.1	nd	nd	<3.1	0.70	0.20
Daubed shanny	1	169	14.2	nd	0.16	6.24	0.37	0.18
Banded gunnel	1	166	17.9	nd	0.22	nd	0.60	0.99
Fish doctor	6	123–195	15.0 (14.2, 15.1)	nd	nd	nd	nd	0.63 (0.27, 0.70)
Fourline snake blenny	2	117, 132	15.8 (15.7, 15.8)	nd	0.10 (nd, 0.21)	nd	nd	0.14 (0.13, 0.15)
Arctic staghorn sculpin	1	117	12.9	nd	nd	4.27	1.45	0.19
Sculpin (*Triglops* spp.)	7	103–138	15.0 (14.9, 15.1)	nd	nd	<3.1	nd	1.40 (1.0, 1.7)
Snailfish	1	124	14.5	nd	nd	nd	nd	0.42

[a] Fish length reported as total length or fork length, as appropriate; i.e. fork length was reported for Arctic cod, capelin and sand lance.
[b] Trace values (<MQL) were included as ½ MQL for calculation of ΣPFCA.

Kelly et al. (2009) reported PFOS and PFOSA concentrations in Arctic cod muscle sampled during 1999–2003 in eastern Hudson Bay whereas PFOS and PFOSA were not quantifiable in whole Arctic cod in our study. However, given the difference in MQLs between the two studies (3.1 ng g^{-1} versus 0.1 ng g^{-1} ww), it is not possible to say that PFOS levels in Arctic cod were actually different between the two studies. PFOS and PFOSA concentrations have been reported for Arctic cod, capelin and a number of sculpin species from several other arctic food web studies (Bossi et al., 2005; Haukås et al., 2007; Kelly et al., 2009; Martin et al., 2004; Powley et al., 2008; Tomy et al., 2004, 2009). The distinct presence of PFOSA in fish has been noted in other studies, as well, suggesting limited biotransformation capacities for this compound in certain fish (Houde et al., 2006).

PFCAs were detected in all samples in our study. The highest mean ΣPFCA concentrations were found in Arctic cod and sculpin (*Triglops* spp.) (Table 3). PFUnA (C_{11}) was the dominant PFCA in all species followed by PFTrA (C_{13}) in Arctic cod, capelin, sand lance and sculpin (*Triglops* spp.) (Table S3). PFNA (C_9) was found in Arctic cod, fish doctor and sculpin (*Triglops* spp.) samples, and PFHxA (C_6) was found only in the banded gunnel (Table S3). PFOA (C_8) was detected but not quantifiable in samples of capelin, fish doctor, Arctic shanny and sculpin (*Triglops* spp.) (Table S3). However, PFOA has been reported for Arctic cod and capelin from other arctic food web studies (Haukås et al., 2007; Kelly et al., 2009; Tomy et al., 2004, 2009). The number of PFCAs reported varies among studies making comparisons of PFCA profiles difficult. However, examination of PFCA profiles for Arctic cod, capelin and sculpin reported in several studies (Haukås et al., 2007; Kelly et al., 2009; Martin et al., 2004; Powley et al., 2008; Tomy et al., 2009) did not appear to yield any consistent pattern.

We found no significant correlation between mean species trophic position ($\delta^{15}N$) and ΣPFCA in our study. Kelly et al. (2009) also found that biomagnification of PFOS and most PFCAs was negligible in the aquatic piscivorous food web, a finding that they attributed to the relatively high aqueous solubility and hydrophobic nature of those PFAS compounds leading to their efficient respiratory elimination in water-respiring organisms. This may explain the lack of any consistent pattern among studies and may also explain the lack of a significant correlation between trophic position ($\delta^{15}N$) and ΣPFCA among the fish species in our study. However, Kelly et al. (2009) also found that PFOS and most PFCAs were highly bioaccumulative and increased with increasing trophic level in a Hudson Bay marine mammal food web, a result consistent with the BMFs found for ΣPFCA in our study (Table 2) which suggest biomagnification from fish to thick-billed murre. Other studies have also shown biomagnification of PFOS and some PFCA compounds from Arctic cod to upper trophic level, air-breathing species such as marine mammals and seabirds (Haukås et al., 2007; Tomy et al., 2004).

Total Hg concentrations were detected in all samples analyzed. The highest mean Hg concentrations were found for fourline snake blenny and the lowest for capelin and sculpin (*Triglops* spp.) (Table 4). There has not been much information published on Hg levels in forage fish other than Arctic cod from the Canadian Arctic. Total Hg in a single whole Arctic cod sampled from the Northwater Polynya in Baffin Bay in 1998 was 0.2 μg g^{-1} dw (assuming 80% water content for fish; Campbell et al., 2005) compared with 0.11 μg g^{-1} dw for Arctic cod in our study (Table 4). Stern and Macdonald (2005) reported mean total Hg concentrations of 0.097 ± 0.025 μg g^{-1} dw for Arctic cod ranging 42–48 mm in length sampled from the western North American Arctic (Chukchi Plateau-Canada Basin) during 1997–98, and 0.171 ± 0.074 μg g^{-1} dw for Arctic cod ranging 160–172 mm in length sampled during that same study. For northern Hudson Bay, the mean total Hg concentration for Arctic cod of 95–177 mm length was 0.11 μg g^{-1} dw with a maximum value of 0.17 μg g^{-1} dw which compares reasonably well with the values from the western Arctic. Arctic cod sampled from Lancaster Sound in 1988–1990 also averaged 0.19 μg g^{-1} dw of total Hg and two-horn sculpin (*Icelus bicornis*) averaged 0.24 μg g^{-1} dw total Hg (Atwell et al., 1998). Given that those data were for fish sampled about twenty years earlier from a different, more northerly ecosystem, the Hg levels are not that different from those found for northern Hudson Bay.

Total Hg was also measured in Arctic cod and sculpin sampled during the 1980s and 1990s from various locations around Greenland (Dietz et al., 1996; Rigét et al., 2000). However, more recent data from fish sampled in 2003–2004 from West Greenland indicated total Hg levels of 0.07 μg g^{-1} dw in 130–160 mm whole capelin and 0.34 μg g^{-1} dw in muscle of 300–400 mm shorthorn sculpin (Rigét et al., 2007) which compared well with our value of 0.06 μg g^{-1} dw in 81–134 mm capelin but was considerably higher than the values we found for 117–122 mm Arctic staghorn sculpin (0.10 μg g^{-1} dw) and 103–138 mm *Triglops* spp. (0.07 μg g^{-1} dw) from northern Hudson Bay. Differences in sculpin species and size analyzed could account for the differences. Total Hg measured in Arctic cod (177–204 mm) and capelin (126–151 mm) sampled in 2006 from Svalbard in the Norwegian Arctic ranged from nd-0.02 μg g^{-1} ww in cod muscle, nd-0.01 μg g^{-1} ww in cod liver, and was not detected (<0.01 μg g^{-1} ww) in whole capelin (Jaeger et al., 2009). Those values are lower than the concentrations we found in similarly-sized capelin (0.013–0.022 μg g^{-1} ww) and slightly smaller Arctic cod (0.009–0.039 μg g^{-1} ww) from northern Hudson Bay. This is not surprising given that Hg concentrations in marine biota from the Canadian Arc-

Table 4
Total Hg concentrations (μg g^{-1} dw) and $\delta^{15}N$ (‰) in forage fish sampled during 2007–2009 from Coats Island in northern Hudson Bay. Mean values are weighted to account for pooled samples comprised of 2–8 fish. The total number of fish analyzed (*n*) includes fish analyzed both individually and as pooled samples (see Table S1). Minimum and maximum values are given in brackets after the mean, where appropriate, and may include both individual fish and pooled samples.

Fish species	*n*	Length[a] (mm)	$\delta^{15}N$	% Moisture	Hg (μg g^{-1} dw)
Arctic cod	18	95–177	15.1 (13.3, 16.0)	77 (74, 80)	0.11 (0.04, 0.17)
Capelin	20	81–134	13.5 (12.2, 14.0)	73 (69, 79)	0.06 (0.05, 0.08)
Sand lance	16	125–156	12.9 (11.6, 13.7)	70 (65, 76)	0.09 (0.08, 0.12)
Benthic fish					
Atlantic poacher	2	142, 151	16.1	73	0.11
Arctic shanny	5	124–130	14.5 (14.1, 15.6)	71 (70, 74)	0.19 (0.18, 0.22)
Daubed shanny	4	117–220	14.2 (14.0, 14.2)	76 (76, 76)	0.13 (0.12, 0.15)
Banded gunnel	3	166–199	17.8 (17.7, 17.9)	62 (60, 66)	0.10 (0.10, 0.10)
Fish doctor	7	123–195	14.8 (13.7, 15.1)	71 (69, 76)	0.16 (0.10, 0.22)
Fourline snake blenny	6	113–132	15.7 (15.2, 15.9)	73 (68, 78)	0.30 (0.22, 0.49)
Arctic staghorn sculpin	2	117, 122	13.6 (12.9, 14.2)	78 (77, 79)	0.10 (0.09, 0.11)
Sculpin (*Triglops* spp.)	7	103–138	15.0 (14.9, 15.1)	73 (71, 77)	0.07 (0.07, 0.07)
Snailfish	1	124	14.5	80	0.16

[a] Fish length reported as total length or fork length, as appropriate; i.e. fork length was reported for Arctic cod, capelin and sand lance.

tic have been found to be substantially higher than elsewhere in the circumpolar Arctic (Braune et al., 2005) likely reflecting the variable influence of air masses originating from different source regions (see AMAP, 2011).

Mean species trophic position ($\delta^{15}N$) was not significantly correlated with mean Hg concentrations in the fish from our study. This suggests that factors associated with bioaccumulation (e.g. size, age) as well as dietary differences may have been more important than biomagnification. Stern and Macdonald (2005) demonstrated a strong correlation between $\delta^{15}N$ and fish length in Arctic cod with concomitantly higher total Hg concentrations in the larger fish. Foster et al. (2012) suggested that conclusions as to whether total or methyl Hg biomagnify in zooplankton food webs, for example, differ depending on whether the computed relationships are based on known food web linkages. In other words, the assumption would have to be made that the higher trophic-position fish in our study were feeding on fish at a lower trophic position, which is not necessarily the case. However, the BMFs for Hg (Table 2) clearly indicated biomagnification from fish to thick-billed murre. Biomagnification from fish to seabirds and other upper trophic level species (e.g. marine mammals) has been well-established in other arctic food webs (Atwell et al., 1998; Campbell et al., 2005; Dehn et al., 2006; Jaeger et al., 2009; Rigét et al., 2007).

In summary, no one fish species had the highest contaminant levels across all contaminant groups. Of the two pelagic species, concentrations of the major OC groups (e.g. Σ_{21}PCB, ΣDDT, ΣCHL, ΣCBz), ΣPBDE, ΣPFCA and Hg were consistently higher in Arctic cod than in capelin delivered to thick-billed murre chicks at Coats Island during 2007–2009. Arctic cod, Atlantic poacher and fourline snake blenny had the highest mean ΣOC concentrations whereas Arctic shanny, Arctic staghorn sculpin and banded gunnel had the highest mean ΣPBDE concentrations. Arctic cod and the *Triglops* spp. of sculpins had the highest mean ΣPFCA, whereas daubed shanny had the highest mean PFOS concentration. Fourline snake blenny had the highest mean Hg concentration.

Acknowledgements

At NWRC, thanks to R. Lavoie and A. Shoji for organizing and documenting the samples, and J. Corriveau, A. Lekorchi, A. Idrissi, S. Chu, E. Pelletier and E. Neugebauer for the contaminant analyses. Stable-nitrogen isotope analyses for the 2007 samples were carried out at the University of Winnipeg Isotope Laboratory, Winnipeg, Manitoba, and for the 2008-2009 samples, at the University of Waterloo Environmental Isotope Laboratory, Waterloo, Ontario. Financial support was provided by Environment Canada, Fisheries and Oceans Canada, the Nunavut Wildlife Management Board, International Polar Year 2007–2009, Aboriginal Affairs and Northern Development Canada (Northern Contaminants Program). NSERC, Arctic Institute of North America and Northern Scientific Training Program support was provided for K. Elliott and J. Provencher. Support for J. Provencher was also provided by J. Dower at the University of Victoria. M. Chambellant received support from Manitoba Hydro, the ArcticNet Center of Excellence and the University of Manitoba. Logistical support was provided by Natural Resources Canada (Polar Continental Shelf Program).

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2013.11.003.

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