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Mercury and persistent organic pollutants in native and invading forage species of the Canadian Arctic: Consequences for food web dynamics☆

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ABSTRACT

Contaminant dynamics within Arctic marine food webs may be altered through the climate-driven northward invasions of temperate/boreal species. Here, we compare tissue concentrations of total mercury (THg) and legacy and emerging persistent organic pollutants (POPs) in native versus invading forage species sampled from 2012 to 2014 near Arviat, Clyde River, and Resolute Bay, NU, representing, low, mid- and high eastern Canadian Arctic regions, respectively. Concentrations of THg, legacy Σ-polychlorinated biphenyls (ΣPCB) and Σ-organochlorine (ΣOC) pesticides were detected in all forage species, whereas emerging halogenated flame retardants were detected in only a few individuals. Concentrations of major contaminant groups among regions did not vary for Arctic cod (*Boreogadus saida*), while for sculpin (Cottoidea) there was no clear latitudinal trend. Thus, considering interspecific variation, native sculpin and northern shrimp (*Pandalus borealis*) had the highest overall concentrations of THg (0.17 ± 0.02 and 0.21 ± 0.01 μg g^{-1} wet weight, respectively), ΣPCB (322 ± 35 and 245 ± 25 ng g^{-1} lipid weight (lw), respectively), and ΣOC (413 ± 38 and 734 ± 64 ng g^{-1} lw, respectively). Comparing the keystone native species, Arctic cod, to its 'replacement' species, capelin (*Mallotus villosus*) and sandlance (*Ammodytes* spp.), THg concentrations were higher in Arctic cod compared to capelin ($p < 0.001$), which was partly explained by differences in fish length. Conversely, capelin and sandlance had higher concentrations of most POPs than Arctic cod ($p < 0.02$). Neither feeding habitat (based on $\delta^{13}C$), trophic position (based on $\delta^{15}N$), nor fish length significantly explained these differences in POPs between Arctic cod, capelin and sandlance. Higher POPs concentrations, as well as variation in congener/compound patterns, in capelin and sandlance relative to Arctic cod seem, therefore, more likely related to a more "temperate"-type contaminant signature in the invaders. Nevertheless, the relatively small (up to two-fold) magnitude of these differences suggested limited effects of these ecological changes on contaminant uptake by Arctic piscivores.

1. Introduction

Surface air temperatures in the Arctic have increased at two times the global average in the past two decades (Screen and Simmonds, 2010). Ensuing warmer seas and sea ice loss have led to changes in the species composition of Arctic marine food webs over the same time period (Post et al., 2013). Such alterations to the Arctic food web have the potential to influence the exposures of sensitive native species to bioaccumulative environmental contaminants, such as persistent organic pollutants (POPs) and methylmercury (MeHg) (Macdonald et al., 2005), which are already a concern for wildlife and human health (Binnington et al., 2016; Dietz et al., 2013; Letcher et al., 2010). These contaminants

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originate mainly from anthropogenic activities in temperate regions and are transported to the Arctic through atmospheric, oceanic and fluvial pathways (Macdonald et al., 2000). Transient and non-native species also act as biological vectors of contaminants into Arctic marine food webs (Blais et al., 2005; Krümmel et al., 2003; McKinney et al., 2012). Several recent studies have reported altered temporal trends in tissue contaminant levels of top predators linked to shifts in prey species distribution. These shifts included invasions of temperate/boreal species associated with climate change (Braune et al., 2014b, 2015b; McKinney et al., 2009, 2015).

Arctic cod (*Boreogadus saida*) is a primary food source for whales, seals, and piscivorous fish in the Arctic (Craig et al., 1982). The large amount of Arctic cod biomass consumed by Arctic predators makes it a critical element of energy transfer between lower and upper trophic levels (Crawford and Jorgenson, 1996; Welch et al., 1993). Yet, Arctic cod appears to have declined in abundance in recent decades in the eastern Canadian Arctic, while sub-Arctic sandlance (*Ammodytes* spp.) and capelin (*Mallotus villosus*), suggested to be replacing Arctic cod in some regions, have become more common (Gaston et al., 2003, 2005; Hop and Gjøsæter, 2013; Provencher et al., 2012). These changes have occurred to the greatest extent in the low Arctic region, where capelin now dominates, and have become more regularly observed in the mid-Arctic (low, mid- and high Arctic regions defined by Salomonsen (1965) and Provencher et al. (2012) based on a gradient of water composition of a mixture of boreal and polar water to unmixed polar water; Provencher et al., 2012). Shifts in capelin distribution have frequently occurred in the past and have been strongly linked to climatic changes (e.g., temperature variation; Rose, 2005). In the Beaufort Sea, a decrease in Arctic cod abundance since the 1970s led to changes in Arctic char (*Salvelinus alpinus*) diets from mainly Arctic cod to capelin and sandlance in 2013–2014 (Harwood et al., 2015). In Hudson Bay, ringed seals (*Pusa hispida*) have been found to consume about ten times more sandlance than Arctic cod (Chambellant et al., 2013). In contrast to the declines in Arctic cod, the abundance of other Arctic native fish, such as sculpin (Cottoidea), does not appear to have changed over the same time period (Provencher et al., 2012).

Sandlance and capelin may exhibit differences in contaminant levels compared to Arctic cod due to variation in trophic position, size, lipid content, feeding habits and habitats, or migration patterns (McKinney et al., 2012). For instance, a dietary shift from Arctic cod to capelin may have affected total mercury (THg) trends in thick-billed murres (*Uria lomvia*) from Hudson Bay from 1993 to 2013. Concentrations of THg would have been increasing, but because of a concomitant decline in trophic position, THg concentrations actually did not significantly change over the time period (Braune et al., 2014b). Conversely, higher concentrations of some legacy POPs and current-use pesticides (e.g. chlorothalonil, Σendosulfan, highly-chlorinated PCBs, *p,p'*-DDE, *trans*-nonachlor), have been reported in capelin from Cumberland Sound compared to some Arctic native species, possibly related to capelin migration to more temperate regions, where this species may pick up a more "temperate"-type contaminant signature (i.e., higher concentrations and/or proportions of less volatile contaminants which tend to accumulate closer to source regions; McKinney et al., 2012; Morris et al., 2016). It is thus important to better characterize contaminant levels in these changing forage communities among regions, to understand how the increasing presence of sub-Arctic forage fish may impact contaminant exposures in upper trophic levels, including people living in the North.

The main objective of this study was to compare contaminant concentrations in invading temperate/boreal species versus native prey fish and invertebrate species in eastern Canadian Arctic regions. We measured and compared the current (2012–2014) concentrations and relative proportions of THg, legacy PCBs and organochlorine (OC) pesticides, polybrominated diphenyl ethers (PBDEs) and newer halogenated flame retardants, in four native and two invading prey species/groups for which limited contaminant data have previously been published. We focused in particular on Arctic cod versus their 'replacements' capelin and sandlance. We further sought to determine the extent to which location (when possible), biological factors (fish length) and ecological factors (trophic position and feeding habitat) accounted for variation in contaminant concentrations among prey species.

2. Material and methods

2.1. Sampling

Fish and invertebrates that are important as food sources for Arctic top predators were targeted for collection. Five to 20 individuals of Arctic cod, capelin, sandlance, sculpin spp. and northern shrimp (*Pandalus borealis*), as well as 1–2 pools of amphipod spp., were collected near Arviat (low Arctic, 61° N, 94° W), Clyde River (mid-Arctic, 70° N, 69° W), and/or Resolute Bay (high Arctic, 75° N, 95° W), Nunavut, Canada (Fig. 1, Table S1) from 2012 to 2014. The fish and invertebrate species were collected by community members in Arviat and Resolute Bay using traditional methods, while Arctic cod in Resolute Bay was fished using rod and reel. In Clyde River, trawl fishing was used for all fish and invertebrate species. It should be noted that not all species were collected in all locations, despite repeated collection attempts. A small number of other miscellaneous fish were collected (cisco (*Coregonus artedi*), Greenland cod (*Gadus ogac*), goiter blacksmelt (*Bathylagus*

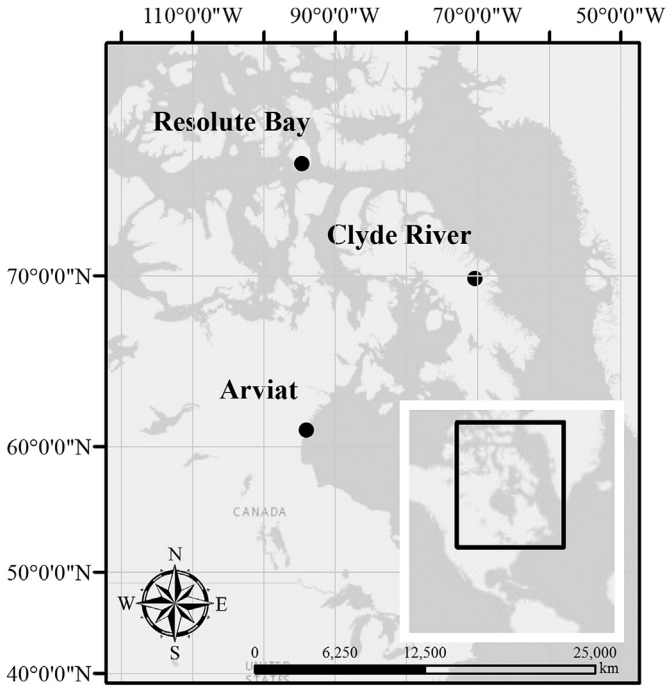

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Fig. 1. Sampling locations of marine forage fish and invertebrates from 2012 to 2014 in the eastern Canadian Arctic. Sampling was near Arviat, Clyde River, and Resolute Bay, Nunavut, representing low Arctic, mid-Arctic, and high Arctic locations, respectively.

euryops)) and analyzed, but are not thought to be major Arctic prey fish, so these results are provided only in the Supporting Information (Table S2). Collected samples were wrapped in solvent-rinsed foil and stored at −20 °C until shipped to the laboratory, where they were kept at −80 °C. Species were identified visually and/or by DNA barcoding at the Canadian Center for DNA Barcoding, Canada. Standard lengths (total length excluding the caudal fin) of all individual fish were recorded.

2.2. POP analysis

Prior to analysis, muscle tissues of individual fish and northern shrimp, and whole amphipod pools, were homogenized. For certain fish species, smaller individuals were pooled by combining equivalent mass aliquots of each homogenate (Table S3). Samples were analyzed for Σ_{40}polychlorinated biphenyl (ΣPCB) and OC pesticides Σdichlorodiphenyltrichloroethanes (ΣDDT), mirex, dieldrin, octachlorostyrene (OCS), Σchlordanes (ΣCHL), Σchlorobenzenes (ΣClBz) and Σhexachlorocyclohexanes (ΣHCH). Flame retardants (FRs) included ΣDechlorane Plus (ΣDP), hexabromobenzene (HBBz), pentabromoethylbenzene (PBEB) and Σ_{19}polybrominated diphenyl ether (ΣPBDE). A full list of individual compounds of each contaminant group is detailed in Table S4.

Chemical extraction and clean-up were performed as previously described by Fisk et al. (2001) at the Great Lakes Institute for Environmental Research (GLIER; University of Windsor, Canada). Briefly, an aliquot of 2.5 g of pooled or individual samples was homogenized with anhydrous sodium sulfate, spiked with the internal standards CB34 and BDE71 and eluted with 1:1 dichloromethane:hexane. Lipid content was determined gravimetrically. Samples were subject to gel permeation chromatography to remove lipids, followed by fractionation on a Florisil column. Fraction 1 contained *ortho*-PCBs and some OCs, fraction 2 contained the FRs and remaining OCs and PCBs, and fraction 3 contained heptachlor epoxide and dieldrin. PCBs and OCs were separated and quantified by gas chromatography with micro-electron capture detection on an Agilent 7890A GC with Agilent 7683 ^{63}Ni-μECD (60 m DB-5 column of 0.25 mm I.D. and 1.0 μm film thickness (J&W Scientific, USA)). FRs were analyzed at the Tomy Lab (University of Manitoba, Canada) by high resolution gas chromatography electron capture negative ion mass spectrometry on an Agilent 5973 GC-MSD fitted with a 10 m × 0.25 mm i.d. DB-5 capillary column (0.25 μm film thickness, J&W Scientific, CA), as previously described (Tomy et al., 2008). Reagent blanks, recovery standards and calibration standards were run at start and every six samples. An in-house carp reference material was extracted with each batch.

The GLIER and the Tomy labs routinely participate in the annual Northern Contaminants Program Inter-Laboratory Quality Assurance Program. Overall, the performance of both labs was classified as excellent, that is, the average results for PCBs, OCs, and PBDEs were within ±20% of the median values (see SI Methods).

The internal standards used for PCB, OC and FR analyses had recoveries of 97 ± 10% (CB34) and 99 ± 10% (BDE71). For PCB and OC, all blanks were below the detection limit and the method limit of quantification (MLOQ) was set to 10x the method detection limit (MDL), ranging from 0.004 to 0.05 ng g^{-1} (Table S5). For FRs, BDE28, 47, and 77 were the only congeners routinely detected in all method blanks (n = 5). The absolute amounts of these congeners in the samples were typically 1.2 to 2 times greater than in the method blanks. The MLOQ was set to the blank average plus 3x the standard deviation, ranging from 0.006 to 0.04 ng g^{-1}. All values were recovery corrected and blank subtracted for FRs. The values were within 9 ± 7% for ΣPCB, 15 ± 9% for ΣOC, and 38 ± 17% for ΣPBDE, of the repeated values measured for the in-house carp reference material. No reference was available for other FRs.

2.3. Mercury analysis

THg analyses were performed at the Center for Environmental Science and Engineering (University of Connecticut, USA). Aliquots of 0.07–0.10 g of each homogenized sample were digested in a hot block with potassium persulfate, sulfuric and nitric acids. Concentrations of THg were determined by flow-injection cold vapor atomic absorption spectrometry using a Perkin Elmer FIMS (adapted from EPA 1630 (Environmental Protection Agency, 1998)). DORM-3 (NRCC, fish protein, reference value of 0.38 μg g^{-1}), DOLT-4 (NRCC, dog-fish liver, reference value of 2.58 μg g^{-1}) and NIST 1946 (reference value of 0.43 μg g^{-1}) were used as standard reference materials and laboratory control samples, duplicates and acid blanks were run every 20 samples. Moisture content was determined gravimetrically.

All blanks were below the detection limit of 0.003 μg g^{-1} (details in SI Methods). The reference material concentrations were within 93 ± 9% (DORM-3), 95 ± 6% (DOLT-4) and 90 ± 2% (NIST, 1946) of the consensus values. The relative standard deviation of duplicate samples was between 2 and 12% and the recoveries of matrix spikes ranged from 75 to 103%.

2.4. Stable isotopes analysis

Stable nitrogen and carbon isotope ratios in muscle tissues (fish) and soft tissues (invertebrates) were determined at the Chemical Tracers Laboratory at the University of Windsor, as previously described (McMeans et al., 2009). All homogenized samples were freeze dried and ground, lipids were extracted with a 2:1 chloroform:methanol solution and lipid content was determined gravimetrically. A 400–600 μg portion of each sample was weighed, combusted in an elemental combustion system and analyzed by a coupled Delta V Advantage isotope ratio mass spectrometer (Thermo Scientific, USA). Stable isotopes were expressed as ratios of the sample relative to the standard (Kelly, 2000):

$$\delta = \left(\left(R_{sample} \Big/ R_{standard}\right) - 1\right) \times 1000‰$$

Precision of stable isotopes analysis was determined based on replicate analyses of four standards (NIST1577c, internal laboratory standard (tilapia muscle), USGS 40 and urea (n = 15 for all except n = 7 for USGS 40). Standard deviations measured ≤0.19‰ for δ^{15}N and ≤0.26‰ for δ^{13}C. Accuracy was determined by closeness to certified values of USGS 40 (n = 7), and means were within 0.04‰ for δ^{15}N and 0.07‰ for δ^{13}C. Instrumental accuracy was checked throughout sample analysis based on repeated runs of NIST standards 8573, 8547 and 8574 for δ^{15}N and 8542, 8573, 8574 for δ^{13}C (n = 10 for all). The mean differences from the certified values were 0.07, −0.14, −0.19‰ for δ^{15}N and 0.04, 0.06 and 0.07‰ for δ^{13}C, respectively.

Species trophic positions were calculated relative to amphipods in each region assuming that amphipods occupy a trophic position of 2.5, as previously demonstrated for gammarids and *Themisto spp.* (Clayden et al., 2015; Fisk et al., 2003; Foster et al., 2012). and using a constant trophic enrichment factor of 3.8‰ (Hobson and Welch, 1992):

$$TP = 2.5 + \left(\delta^{15}N_{fish/shrimp} - \delta^{15}N_{amphipod}\right) \Big/ 3.8‰$$

Primary consumers such as amphipods are more typical as a reference value for carbon source (Fisk et al., 2003) however, amphipods collected in the three locations here were of different genera and widely different δ^{13}C values (Table 1). Due to this variation, we used sculpin as the reference carbon source as it was the only species group collected in all locations exhibiting

Table 1
Biological and ecological characteristics of prey fish and invertebrate species in the eastern Canadian Arctic territory of Nunavut collected from 2012 to 2014. Results denoted as arithmetic mean (min-max).

	Arctic cod		Capelin	Sandlance	Sculpin			Northern shrimp	Amphipods		
	Clyde River	Resolute Bay	Arviat	Arviat	Arviat	Clyde River	Resolute Bay	Clyde River	Arviat	Clyde River	Resolute Bay
	Boreogadus saida		*Mallotus villosus*	*Ammodytes* spp.	*Myoxocephalus* spp.	*Cottuncullus microps*	*Myoxocephalus* spp.	*Pandalus borealis*	*Gammarus oceanicus*	*Themisto libellula*	*Gammarus setosus*
N[a]	20	20	11	13	10	10	10	10	1 pool	1 pool	2 pools
Feeding mode[b]	Predator, pelagic		Predator, pelagic	Predator, pelagic	Predator/Scavenger, benthic			Predator/detritivore supra-benthic	Grazer, pelagic	Predator, pelagic	Grazer, pelagic
Habitat range[c]	Arctic native		Sub-Arctic; migratory	Sub-Arctic	Arctic native			Arctic native	Arctic native	Arctic native	Arctic native
Length (mm)	142 (109–182)	148 (125–174)	103 (86–123)	96 (84–109)	197 (129–230)	127 (82–168)	148 (117–213)	–	–	–	–
Weight (g)	31.3 (12.3–60.7)	26.0 (15.0–35.0)	8.7 (4.0–15.0)	3.8 (2.7–5.5)	149 (25.2–228)	77.1 (19.0–163)	78.3 (27.0–224)	13.2 (10.6–15.7)	–	–	–
% lipid	1.4 (0.3–2.7)	2.6 (1.4–4.0)	1.8 (1.3–3.2)	1.5 (1.2–1.7)	0.8 (0.2–1.3)	0.3 (0.2–0.5)	0.7 (0.4–1.3)	0.3 (0.2–0.5)	0.75	11.0	1.7 (1.2–2.2)
% moisture	79.8 (76.9–82.5)	78.8 (76.2–82.1)	80.6 (79.3–83.3)	78.3 (73.5–81.5)	79.9 (78.0–82.2)	81.7 (78.6–85.3)	78.2 (77.3–80.0)	76.7 (75.4–79.7)	74.2	67.8	80.4 (78.3–82.5)
$\delta^{15}N$ ‰	14.9 (14.2–16.3)	15.2 (14.3–15.9)	14.7 (13.9–15.3)	14.2 (12.7–15.8)	15.7 (14.4–14.5)	18.0 (17.1–18.6)	16.1 (15.5–16.7)	15.8 (15.4–16.3)	10.0	13.1	13.0 (12.6–13.3)
$\delta^{13}C$ ‰	−20.7 (−21.1–−20.4)	−19.9 (−20.4–−19.5)	−21.1 (−23.1–−20.0)	−21.6 (−23.2–−19.6)	−16.6 (−18.9–−14.2)	−19.1 (−17.9–−19.8)	−17.7 (−18.5–−17.0)	−19.0 (−19.3–−18.7)	−16.5	−21.4	−17.3 (−18.3–−16.3)
Trophic position[d]	2.96 (2.78–3.34)	3.08 (2.86–3.27)	3.73 (3.52–3.90)	3.61 (3.20–4.02)	3.99 (3.66–4.45)	3.80 (3.56–3.94)	3.31 (3.18–3.49)	3.21 (3.10–3.33)	2.50	2.50	2.50
Relative carbon source[e]	1.08 (1.07–1.10)	1.12 (1.10–1.15)	1.27 (1.20–1.39)	1.30 (1.18–1.39)	1.00	1.00	1.00	0.99 (0.98–1.01)	0.99	1.12	0.98 (0.92–1.03)

[a] A number of individuals varying with species were pooled for persistent organic pollutant analyses. Details can be found in Table S3.
[b] Feeding mode based on Falardeau et al. (2014) for Arctic cod and sandlance, Rose (2005) for capelin, Giraldo et al. (2016) for sculpin, Fisk et al. (2003) for northern shrimp and *T. libellula* and Clayden et al. (2015) for gammarids.
[c] Habitat range based on Rose (2005) for capelin and Falardeau et al. (2014) for sandlance.
[d] Trophic position of forage fish and shrimp species in relation to the amphipods per region, assuming they occupy a trophic position of 2.5 (Fisk et al., 2003). 3.8‰ was considered to be the isotopic enrichment along the food web in each trophic level. The equation used was: $TP = 2.5 + (\delta^{15}N_{fish/invertebrate} - \delta^{15}N_{amphipods})/3.8$.
[e] Relative carbon source of forage fish and shrimp species was calculated in relation to sculpin per region based on the equation: $\delta^{13}C_{fish/invertebrate}/\delta^{13}C_{sculpin}$ (Fisk et al., 2003).

Table 2
Concentrations of total mercury (THg; μg g^{-1} wet weight) and persistent organic pollutants (ng g^{-1} lipid weight) in prey fish and invertebrate species in the eastern Canadian Arctic territory of Nunavut collected from 2012 to 2014. Results denoted as arithmetic mean (min-max) or ND (not detected) when <70% of the individuals had concentrations above the detection limit.

	Arctic cod		Capelin	Sandlance	Sculpin			Northern shrimp	Amphipods		
	Clyde River	Resolute Bay	Arviat	Arviat	Arviat	Clyde River	Resolute Bay	Clyde River	Arviat	Clyde River	Resolute Bay
	Boreogadus saida		*Mallotus villosus*	*Ammodytes* spp.	*Myoxocephalus spp.*	*Cottunculus microps*	*Myoxocephalus spp.*	*Pandalus borealis*	*Gammarus oceanicus*	*Themisto libellula*	*Gammarus setosus*
THg	0.03 (0.02–0.05)	0.03 (0.02–0.04)	0.01 (0.01–0.02)	0.02 (0.02–0.04)	0.13 (0.05–0.26)	0.27 (0.12–0.39)	0.12 (0.06–0.23)	0.21 (0.17–0.25)	0.02	0.01	0.01 (0.01–0.02)
ΣPCB	98 (55–210)	78 (57–106)	138 (83–218)	113 (70–158)	234 (117–437)	423 (176–903)	310 (163–611)	245 (132–339)	510	34.7	83.8 (81.4–86.2)
Dieldrin	21.1 (12.9–41.8)	16.8 (8.9–27.1)	39.3 (24.3–53.3)	16.5 (12.6–19.4)	19.7 (9.6–26.1)	20 (2–58)	48.7 (34.8–71.4)	79 (31–121)	72.7	15.9	25.2 (19.6–30.9)
Mirex	0.6 (0.2–1.4)	0.3 (0.1–0.6)	1.9 (0.7–3.7)	1.4 (0.7–2.1)	3.0 (1.0–6.4)	6.0 (2.8–17.7)	2.8 (0.8–7.5)	6.2 (3.3–8.5)	1.6	1.5	0.4 (0.3–0.4)
ΣHCH	15.4 (6.4–33.6)	15.7 (10.1–18.7)	11.7 (9.7–15.3)	11.8 (9.2–15.4)	16.6 (11.2–21.2)	64.1 (11.3–97.6)	27.8 (18.8–48.2)	35.7 (12.3–53.9)	14.4	15.2	24.4 (19.7–29.1)
ΣClBz	85.9 (37.8–267)	47.5 (32.3–73.6)	49.1 (39.1–63.9)	42.0 (36.6–52.3)	49.6 (19.2–81.6)	70.6 (24.5–170)	103 (65–170)	426 (170–569)	47.3	26.2	61.0 (41.1–81.0)
ΣDDT	14.4 (7.70–41.8)	9.55 (6.2–15.0)	33.6 (17.0–54.0)	23.0 (11.0–31.0)	108 (20–258)	118 (71.7–173)	171 (53–460)	19.2 (8.4–31.6)	105	8.9	19.0 (15.8–22.1)
OCS	0.5 (0.2–1.1)	0.3 (0.1–0.6)	1.85 (0.9–4.0)	1.0 (0.4–2.0)	1.4 (0.6–2.3)	2.1 (1.2–3.6)	1.5 (0.8–3.2)	1.8 (0.8–2.3)	1.2	0.3	1.6 (1.5–1.7)
ΣCHL	44.2 (15.4–69.0)	40.4 (27.9–56.0)	103 (55.4–167)	70.3 (33.8–158)	147 (42–323)	33.9 (19.5–52.7)	228 (129–360)	166 (79–281)	420	30.9	80.2 (65.2–95.2)
ΣOC	182 (109–435)	131 (90–174)	241 (158–317)	166 (106–216)	345 (131–695)	312 (179–491)	583 (334–978)	734 (305–937)	662	97.5	212 (179–245)
ΣPBDE	ND (<0.04–10.6)	ND (<0.04–63.6)	11.1 (0.02–17.4)	ND	ND (<0.04–329)	ND	ND (<0.04–68.8)	ND	303.3	ND	ND

Contaminants represented are Σ_{40}polychlorinated biphenyl (ΣPCB), Σhexachlorocyclohexanes (ΣHCH), Σchlorobenzenes (ΣClBz), Σdichlorodiphenyltrichloroethanes (ΣDDT), octachlorostyrene (OCS), Σchlordanes (ΣCHL), Σorganochlorinated pesticides (ΣOC) and Σ_{19}polybrominated diphenyl ether (ΣPBDE).

consistent (benthic) feeding habits (Gray, 2015; Hoekstra et al., 2003; Rigét et al., 2007). The equation used here was modified from Fisk et al. (2003):

$$\text{Relative carbon source} = \delta^{13}C_{\text{fish/shrimp}} \Big/ \delta^{13}C_{\text{sculpin}}$$

Carbon source of more than 1 thus represents organisms with more pelagic feeding habits and equal or lower represents benthic feeding habits (Fisk et al., 2003).

2.5. Statistical analyses

THg concentrations were reported in μg g^{-1} wet weight (ww). As there were significant correlations between ΣPCB, ΣCHL, ΣHCH, ΣClBz, mirex and dieldrin concentrations and lipid content ($R^2 > 0.23$, $p < 0.02$), POP concentrations were reported in ng g^{-1} lipid weight (lw) and all statistical analysis were performed with lipid corrected data. Contaminant concentrations were log-transformed and proportions of compounds within each POP group were arcsine-transformed to meet normality requirements of linear models.

Summary statistics were calculated only for contaminants detected in more than 70% of samples per species/location (details in Table 2). In those cases, the small number of individuals with non-detect values were set to a random value between 0 and 0.5 MDL. Although we show contaminant concentrations for amphipods, they were not included in statistical analysis due to the low sample size. To compare contaminant concentrations and proportions among species/locations, we used linear models (one-way analysis of variance) including *Species per Location* as the independent variable and each individual or sum (Σ) contaminant concentration as the dependent variable,

$$log/asin(contaminant) = \beta_0 + \beta_{Species} * Species/Location + \varepsilon$$

followed by *post-hoc* Tukey's honestly significant difference tests. We did not consider location separately from species as not all species were collected in all locations.

To evaluate the influence of biological and ecological factors on contaminant concentrations, we first used a Pearson correlation matrix to assess linear relationships between weight, length, relative carbon source and trophic position. Only weight and length were significantly correlated ($R^2 = 0.78$, $p < 0.0001$). Since length is related to age in fish species and consequently with bioaccumulation (Morrongiello et al., 2012), length and not weight was included in subsequent analyses. Next, the model selection was performed in a step-wise approach with linear mixed effects models using the package *lme4* (Bates et al., 2014). The explanatory variables included in the initial model were trophic position (TP), relative carbon source (C), length and their interactions as fixed effects, and species/location as random effects.

$$log(contaminant) = \beta_0 + \beta_{TP} * TP * \beta_{\mathbf{C}} * \mathbf{C} * \beta_{Length} * Length + (1|Species/Location) + \varepsilon$$

Akaike information criterion for small sample sizes (AICc) was

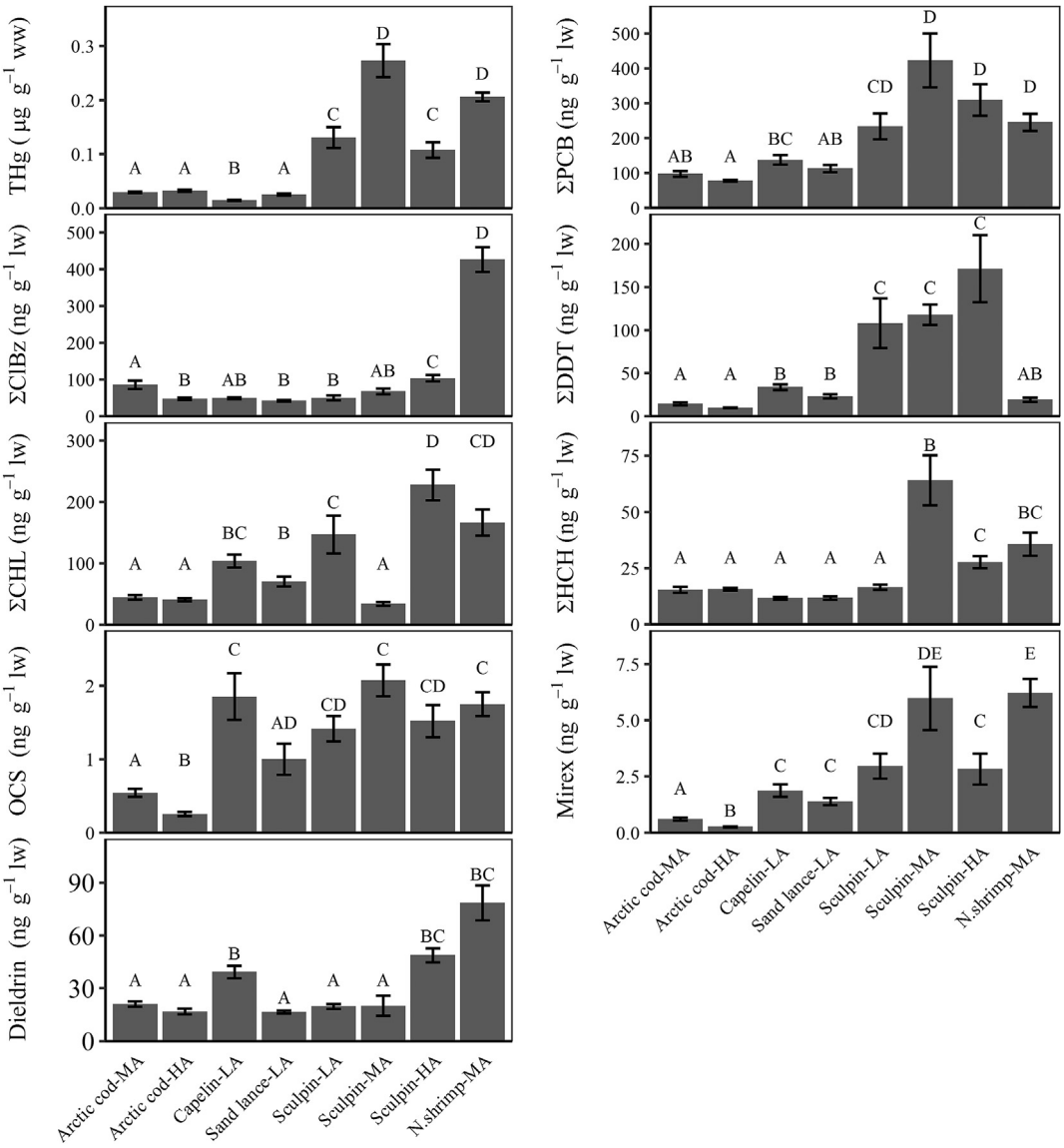

Fig. 2. Concentrations of total mercury (THg) in μg g^{-1} wet weight (ww) and persistent organic pollutants: Σpolychlorinated biphenyl (ΣPCB), Σchlorobenzenes (ΣClBz), dieldrin, Σhexachlorocyclohexanes (ΣHCH), octachlorostyrene (OCS), Σdichlorodiphenyltrichloroethanes (ΣDDT), mirex and Σchlordanes (ΣCHL) in ng g^{-1} lipid weight (lw) in major prey fish and invertebrates from the low (LA), mid- (MA) and high (HA) eastern Canadian Arctic from 2012 to 2014. Significant differences in contaminant concentrations among species are indicated by different letters above each bar. Note that the y-axis for each contaminant group vary in units and scales.

used to determine the best relative fit model with the package *AICcmodgav* (Mazerolle, 2016). The best model was selected based on the lowest AICc and the significance of fixed effects was determined by comparing the fit model with and without the term of interest using an ANOVA-type parametric bootstrap test with the package *pbkrtest* (Halekoh and Hojsgaard, 2014). The best fit model was inspected for normality of residuals and heteroscedasticity. All analyses were performed using R software version 3.2.0 (R Core Team, 2013) and statistical significance was considered at $p < 0.05$.

3. Results and discussion

3.1. THg and legacy POP concentrations and spatial variation

Concentrations of THg, ΣPCB and ΣOC varied among species/locations with up to four-times higher levels in sculpin and northern shrimp, compared to Arctic cod, capelin and sandlance (THg: $F_{7,96} = 130$, $p < 0.001$; ΣPCB: $F_{7,95} = 29.8$, $p < 0.001$; ΣOC: $F_{7,95} = 36.3$, $p < 0.001$; Table 2, Fig. 2). For THg, sculpin and northern shrimp exhibited significantly higher levels ($p < 0.001$) than other species regardless of sampling location. In fact, THg concentrations in sculpin and northern shrimp were close to the 0.5–1.2 μg g^{-1} ww range of muscle Hg toxicity thresholds for freshwater fish (Dietz et al., 2013; >73% in the form of MeHg for sculpin and northern shrimp; Pedro *et al*, unpublished data). Overall, POP concentrations were more variable than THg among species/locations. For example, sculpin from all three locations showed higher ΣDDT levels ($p < 0.02$) than Arctic cod, capelin, and sandlance, while northern shrimp showed similar ΣDDT levels to these species. For ΣPCB, ΣClBz, ΣHCH and mirex, northern shrimp showed higher levels ($p < 0.001$) than Arctic cod, capelin, and sandlance, while dieldrin and ΣCHL levels were higher ($p < 0.001$) only in northern shrimp compared to Arctic cod and sandlance. Sculpin occasionally showed higher levels of these contaminants compared to other species, depending on sampling location. These differences among species/locations may to some extent be explained by the more

benthic feeding habits of sculpin and northern shrimp compared to the other more pelagic fish (Giraldo et al., 2016; further discussed in *Influence of biological and ecological factors*). Notably, amphipods generally showed high levels of ΣPCB and OCs in the low and mid-Arctic compared to the high Arctic, possibly related to their relatively benthic scavenging habits (Tables 1 and 2). However, due to small sample sizes and previously reported inconsistent contaminant concentrations in these invertebrates relative to those in predators (Fisk et al., 2003), we did not include amphipods in the statistical analysis or attempt to draw any further comparisons with other focal species.

Spatial variation was assessed by comparing Arctic cod only successfully sampled from the mid- and high Arctic, and sculpin from all three locations, although the sculpin species sampled was not identical among locations (Table 2). Concentrations of THg, ΣPCB, and most OCs were similar between Arctic cod sampled in the mid- and high Arctic, except for small but significant differences in ΣClBz ($t = -4.0$, $p = 0.003$), OCS ($t = -5.1$, $p < 0.001$), and mirex ($t = -5.2$, $p < 0.001$; Fig. 2). Chlorobenzenes are highly volatile contaminants that tend to increase in concentration with latitude in water and in biota within the Arctic (Wania and Mackay, 1996). However, we observed the opposite pattern, i.e., higher concentrations in Arctic cod from the mid-Arctic compared to conspecifics in the high Arctic. Higher OCS and mirex in mid-Arctic cod relative to high Arctic may be in part related to their relatively low volatility and thus more pronounced latitudinal gradient relative to more volatile contaminants (Mackay et al., 1997, 1991).

For sculpin, regional variation in contaminant concentrations was greater and not consistent with the patterns observed for Arctic cod (Fig. 2) or with a clear latitudinal trend. The only exception was an increase with latitude for ΣClBz ($p < 0.05$). Therefore, regional variation in sculpin contaminant concentrations was likely influenced by other intra- or inter-specific variation. Although sculpin feed on benthic amphipods, small fish and polychaetes (Evenset et al., 2016), their diets tend to be variable with no dominant prey species. Moreover, sculpin diet can vary among species and with the region and depth at which they feed (Giraldo et al., 2016; Gray, 2015). Thus, sculpin may not be well suited for use as a biomonitor of regional variation in contaminant levels. For example, higher levels of ΣCHL, ΣClBz and ΣHCH were previously found in sculpins of *Triglops spp.* compared to Arctic staghorn sculpin (*Gymnocanthus tricuspis*) sampled within the same region, and were related to differences in diet (Braune et al., 2014a). In summary, there was no consistent latitudinal gradient in contaminant concentrations based on our results for Arctic cod and for sculpin. Nonetheless, because we found spatial variation for

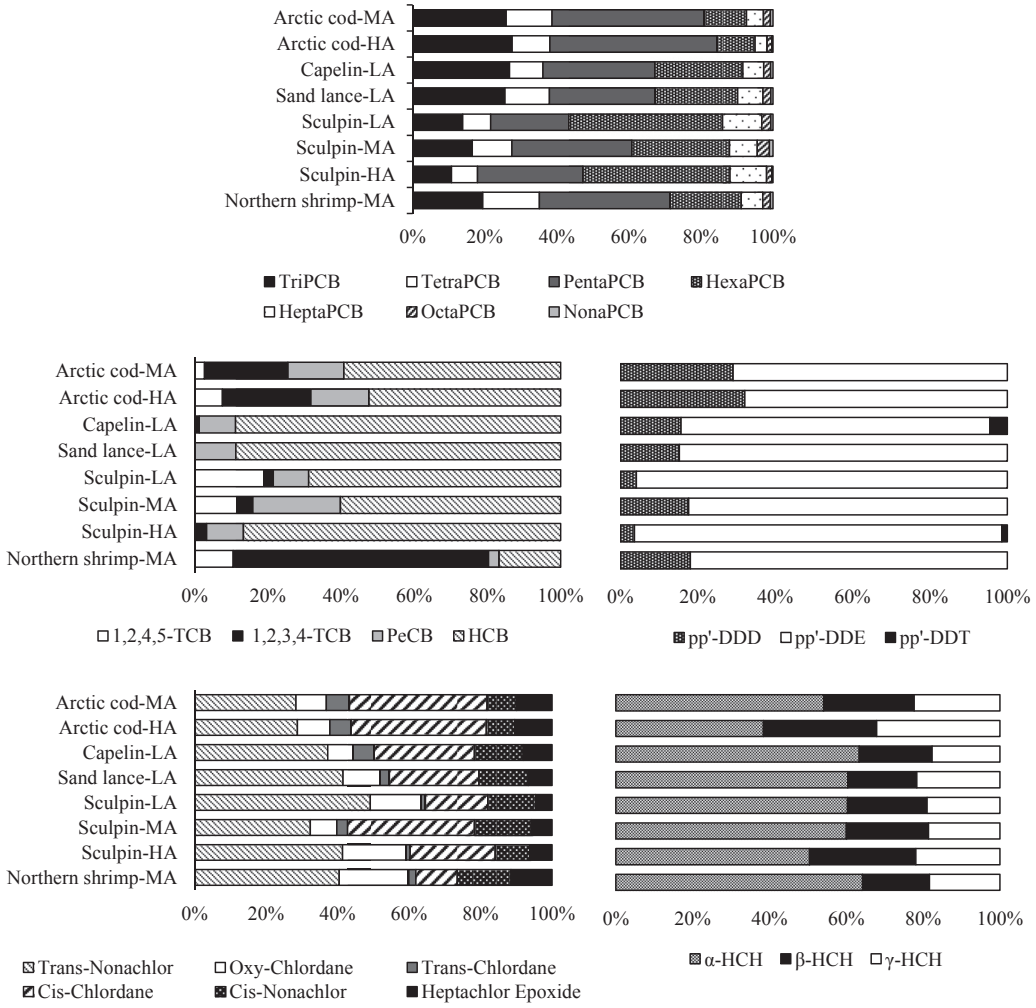

Fig. 3. Relative proportions (%) of polychlorinated biphenyl (PCB) homologue groups to ΣPCB, chlorobenzene compounds to ΣClBz, dichlorodiphenyltrichloroethanes compounds to ΣDDT, chlordane compounds to ΣCHL and hexachlorocyclohexane compounds to ΣHCH in marine prey fish and invertebrates sampled in the low Arctic (LA), mid-Arctic (MA) and high (HA) eastern Canadian Arctic from 2012 to 2014.

ΣClBz, OCS, and mirex in Arctic cod, we did not consider these specific contaminants in subsequent, more detailed comparisons among Arctic cod, capelin, and sandlance to avoid a possible confounding influence of spatial variation in environmental concentrations.

3.2. Legacy POP patterns and spatial variation

Contaminant patterns (proportions of individual compounds in each contaminant class) were generally similar among most species (Fig. 3) and were in agreement with previous studies (Fisk et al., 2003). However, we found some significant differences in contaminant patterns among species ($p < 0.001$). Higher proportions of the highly chlorinated hexa- and hepta-PCB and lower proportions of tri-PCB ($p < 0.001$) were commonly found in sculpins relative to Arctic cod, capelin and sandlance (except that no differences were found for hexa- and hepta-PCB in sculpin in the mid-Arctic compared to invading species). This finding may be related to the nearshore/benthic feeding habits of sculpin relative to these other more pelagic forage fish. Penta- and hexa-PCB congeners are more associated with sediments rich in organic matter at nearshore shallow waters (Ma et al., 2015), and consequently may tend to occur at higher concentrations where benthic/nearshore organisms commonly feed (Giraldo et al., 2016; Hoekstra et al., 2003). In addition to differences found for PCBs among species, we also found differences in PCB patterns within species among locations. Higher proportions of penta-PCBs and lower proportions of nona-PCBs ($p < 0.02$) in Arctic cod in the high Arctic compared to the mid-Arctic demonstrate the commonly observed latitudinal patterns seen for PCB profiles (Sobek et al., 2010; Wania and Mackay, 1996). For sculpin, lower proportions of hexa- and hepta-PCB ($p < 0.02$) in the mid-Arctic were found compared to the low and high Arctic. Sculpin in the low and high Arctic regions were of the same genera, but of a different genus than sculpin collected in the mid-Arctic. Diet differences among these sculpin species (Gray, 2015) could influence their contaminant patterns. We also found lower proportions of α-HCH and higher proportions of γ-HCH in Arctic cod from the high Arctic compared to Arctic cod and other fish in the mid- and low-Arctic, although the reason for these differences is not clear.

Higher proportions of HCB found in invading species ($p < 0.01$) relative to other species sampled (except for sculpin in the high Arctic) suggests a more "temperate"-type signature for at least the highly migratory capelin (Rose, 2005). By "temperate", we mean higher overall contaminant concentrations and/or proportions of the heavier, less volatile contaminants, such as HCB relative to the lighter tetra- and penta-ClBz, are to be expected at temperate latitudes relative to the Arctic. A more "temperate"-type signature in capelin relative to year-round resident Arctic forage species (McKinney et al., 2012; Morris et al., 2016; Rose, 2005) is consistent with capelin movements between North Atlantic and Arctic waters. Less is known about sandlance movements in the Arctic, although it was suggested that low temperatures and prey availability favor sandlance reproduction (Danielsen et al., 2016; Robards et al., 2002). Thus, it is possible that warming ocean waters could have favored the overall increase in sandlance abundance in Arctic regions (Falardeau et al., 2014; Provencher et al., 2012). Regardless, the intermediate contaminant signature and concentrations of sandlance relative to Arctic cod and capelin (further details in *Comparisons between Arctic cod and 'replacement' invasive species*), is not as clearly supportive of sandlance acting as a contaminant biovector from regions further south as per capelin. The ClBz signature for northern shrimp was markedly different compared to other species, with higher proportions of 1,2,3,4-TCB ($p < 0.001$) and lower of HCB ($p < 0.01$), possibly related to limited capacity to biotransform xenobiotics in invertebrates relative to teleost fish (Borgå et al., 2004). However, we could not explain this signature based on other data. In terms of spatial variation, we found higher proportions of HCB in the high Arctic sculpin compared to sculpin in other locations ($p < 0.01$) which may be related to spatial variation in sculpin feeding habits (Giraldo et al., 2016; Gray, 2015).

Variation in ΣDDT and ΣCHL patterns were likely related to specific differences in diet or biotransformation capacity (Borgå et al., 2001; Hoekstra et al., 2003; Hop et al., 2002). For example, higher proportions of *p,p'*-DDE found in sculpin of *Myoxocephalus* spp. (in the low and high Arctic) compared to other forage species ($p < 0.001$) has previously been found in the southern Beaufort-Chukchi Seas. The authors related the results to a high capacity of this sculpin or their prey to dehydrochlorinate DDT compounds (Hoekstra et al., 2003). Similarly, higher proportions of the metabolite oxychlordane in sculpin of *Myoxocephalus* spp. and northern shrimp ($p < 0.001$) compared to other species sampled may indicate increased biotransformation capacity in these species (Hoekstra et al., 2003).

3.3. PBDE and other FR concentrations

In contrast to THg and legacy POPs, and despite the temporal increases in some FR concentrations reported in Arctic biota (Braune et al., 2005), PBDEs were the only FR measured above the detection limits for >70% of individuals, and only in some species/locations. Specifically, in the low Arctic, BDE-47 was detected in sculpin, BDE-85 in capelin, and BDEs-28, 47, 99, and 100 in amphipods. For other species/locations, only a few individuals had detectable levels of certain PBDEs (Table S6). These findings of low to non-detectable levels in these prey species are not necessarily surprising, since FR are generally found at lower concentrations, and are not as persistent or bioaccumulative, relative to legacy POPs and THg (Braune et al., 2014a; de Wit et al., 2006; Kelly et al., 2008). Regardless, these results make it difficult to assess FR spatial trends and consequently, we did not further evaluate FR concentrations and patterns among species/locations.

3.4. Comparisons between Arctic cod and 'replacement' species

We found inter-specific differences in contaminant concentrations and patterns between native Arctic cod and the 'replacement' species capelin and to a lesser extent, sandlance (Fig. 2), which may be related to, among other factors, migratory behavior of capelin relative to Arctic cod. Levels of THg were significantly higher in Arctic cod than in capelin ($p < 0.001$), but were not different relative to sandlance. Conversely, ΣPCB, dieldrin, ΣDDT, and ΣCHL ($p < 0.02$) were significantly higher in capelin than in Arctic cod. Concentrations of ΣCHL and ΣDDT ($p < 0.02$) were also higher in sandlance compared to Arctic cod, while ΣPCB and dieldrin concentrations were similar between sandlance and Arctic cod. No differences were found among these three species for ΣHCH concentrations. Certain contaminants such as *p,p'*-DDT and ΣPBDE were not consistently detected in Arctic cod, whereas they were measured in all capelin analyzed. In addition, we found variation in contaminant patterns between Arctic cod and non-native species, i.e., higher proportions of α-HCH, hexa- and hepta-PCB, *cis*- and *trans*-nonachlor ($p < 0.001$) in capelin and sandlance relative to Arctic cod (Fig. 3). Conversely, Arctic cod had higher proportions of γ-HCH, penta-PCB and *cis*-chlordane compared to both invaders ($p < 0.001$).

The observed higher proportions of less volatile PCBs and differences in the fractions of compound species of CHL technical mixture in capelin and sandlance relative to Arctic cod, suggest a more "temperate"-type contaminant signature in the invaders,

especially capelin (Jantunen et al., 2015; Sobek et al., 2010; Su et al., 2008; Wania, 1998). This signature suggests that migration patterns of capelin may also have contributed to our finding of significantly higher concentrations of ΣPCB, dieldrin, ΣDDT, and ΣCHL in capelin versus Arctic cod and the opposite for THg. Most of these organic contaminants have been reported at higher concentrations in temperate relative to Arctic waters and food webs (Sobek et al., 2010; Wania and Mackay, 1993). For example, some PCB congeners were found at ten times higher concentrations in water and double the concentration in zooplankton in the temperate Baltic Sea compared to Arctic Barents Sea (Sobek et al., 2010). Concentrations of THg, on the other hand, tend to be higher in the Arctic Ocean relative to waters at lower latitudes due to increased inputs from rivers, reduced water-air exchanged prevented by sea-ice and mercury depletion events (Andersson et al., 2008; Emmerton et al., 2013; Zhang et al., 2015). Nonetheless, the detection of *p,p'*-DDT and PBDEs in capelin may also reflect regional variation in contaminant concentrations, since we detected these compounds in amphipods in the low Arctic, but not in the mid- and high Arctic. Possible influence of biological and ecological factors is discussed below.

3.5. Influence of biological and ecological factors

General linear models indicated that relative carbon source was an important variable explaining differences in contaminant concentrations among species/locations (Table 3). Relative carbon source was weakly but positively associated with ΣPCB (Fig. 4a), ΣCHL and ΣDDT concentrations and although not significant, was also included in the best fit models for mirex and THg. ΣPCB (especially penta- and hexa-PCB), ΣCHL, ΣDDT and mirex are very hydrophobic contaminants (log K_{ow} from 5.8 to 7.3; Mackay et al. (1997, 1991)) that tend to show higher concentrations in more benthic/shallow waters rich in organic matter relative to pelagic habitats (Ma et al., 2015). Likewise, MeHg, the bioaccumulative form of THg, is more abundant within highly productive nearshore environments (Barkay and Poulain, 2007; Kainz et al., 2003; Lehnherr et al., 2012). Thus, higher contaminant concentrations observed in sculpin relative to other fish species are likely explained, in part, by their foraging habitats (Giraldo et al., 2016). In support of this, relative carbon source of sculpin was significantly more benthic than for Arctic cod, capelin and sandlance ($p < 0.001$). Benthic organisms tend to have higher concentrations of most contaminants than pelagic organisms and to show more inter-individual variability in POP concentrations (Evenset et al., 2016; Fisk et al., 2003). Similarly, more benthic relative carbon sources found in Arctic cod relative to capelin and sandlance ($p < 0.001$) could partly explain higher THg concentrations in the former. However, carbon source did not explain the higher concentrations of ΣPCB, ΣCHL and ΣDDT measured in capelin and sandlance compared to Arctic cod because, unlike THg, Arctic cod had lower concentrations of these contaminants relative to capelin and sandlance.

Trophic position explained a significant amount of the variation for OCS concentrations, but not for other contaminants (Table 3). We found significantly higher TPs in sculpin compared to other species ($p < 0.001$) that would partially explain the higher OCS concentrations observed in sculpin (Fisk et al., 2001). The limited trophic range of the forage species examined may explain the lack of consistent relationships of other biomagnifying POPs with trophic position. Biomagnification is better assessed through more fulsome sampling of the food web, including top predators (Hop et al., 2002; Loseto et al., 2008; Ruus et al., 2015).

Fish length significantly explained variation in THg concentrations, but not other contaminants (Table 3, Fig. 4d). Increasing THg concentrations with fish length have been reported in previous studies (Braune et al., 2015a; Evans et al., 2005; Sackett et al., 2013). Length is positively associated with age in fish and due to bioaccumulation and/or differences in diet (e.g. Arctic cod switches to larger zooplankton prey species at different growing stages (Falardeau et al., 2014)), older fish and/or larger species may show higher THg levels (Schneider et al., 2000). Significantly larger Arctic cod ($p < 0.001$) compared to capelin and sandlance (Hop and Gjøsæter, 2013) could thus partly explain higher THg levels found in the former.

Our results for most contaminants, except ΣClBz and THg, differ from those recently found for Arctic cod, capelin, and sandlance in the low Arctic region of Northern Hudson Bay (Braune et al., 2014a). Although the authors did not undertake statistical comparisons among species, in that study Arctic cod showed apparently higher concentrations of POPs than capelin and sandlance in that region. However, Arctic cod in this previous study showed ΣOC concentrations averaging twice as much as those we found, while capelin and sandlance showed two times lower ΣOC concentrations than we measured. Considering the biological and ecological variables measured in both studies, our finding of higher trophic position (based on $\delta^{15}N$) of capelin and sandlance, but similar trophic position of Arctic cod, relative to Braune et al. (2014a) could partially explain this variation, between studies. Fish lengths and sampling years were similar between the two studies.

Table 3
Akaike's Information Criteria for small sample sizes (AICc) and linear mixed effects model results assessing the influence of fish length, trophic position and relative carbon source on THg and POPs concentrations in marine forage fish and invertebrates sampled in the eastern Canadian Arctic from 2012 to 2014.

Contaminant	Best relative fit model[a]	Degrees of freedom	parameter estimates bootstrap (p < 0.0001 '***' p < 0.001 '**' p < 0.01 '*')				
			Conditional R^2	Species/Location	TP	Carbon source	Length
THg	Length + carbon source + (1 | Species/Location)	1	0.90	<0.0001***		0.2475	0.0099 **
Dieldrin	(1 | Species/Location)	–	0.56	<0.0001***			
Mirex	carbon source + (1 | Species/Location)	1	0.77	<0.0001***		0.1485	
ΣPCB	carbon source + (1 | Species/Location)	1	0.67	<0.0001***		0.0099 **	
ΣHCH	(1 | Species/Location)	–	0.53	<0.0001***			
ΣClBz	(1 | Species/Location)	–	0.73	<0.0001***			
ΣDDT	carbon source + (1 | Species/Location)	1	0.86	<0.0001***		0.0099 **	
OCS	TP + (1 | Species/Location)	1	0.65	<0.0001***	0.0099 **		
ΣCHL	carbon source + (1 | Species/Location)	1	0.78	<0.0001***		0.0198 *	

Contaminants represented are total mercury (THg), Σ_{40}polychlorinated biphenyl (ΣPCB), Σhexachlorocyclohexanes (ΣHCH), Σchlorobenzenes (ΣClBz), Σdichlorodiphenyltrichloroethanes (ΣDDT), octachlorostyrene (OCS), Σchlordanes (ΣCHL), Σorganochlorinated pesticides (ΣOC) and Σ_{19}polybrominated diphenyl ether (ΣPBDE).

[a] Mixed effects models with log transformed contaminant concentrations. PCB and OCs are in ng g^{-1} lipid weight and THg is in μg g^{-1} wet weight. Trophic position (TP) and carbon source were calculated relative to amphipod $\delta^{15}N$ and sculpin $\delta^{13}C$ values in each location (see Table 1).

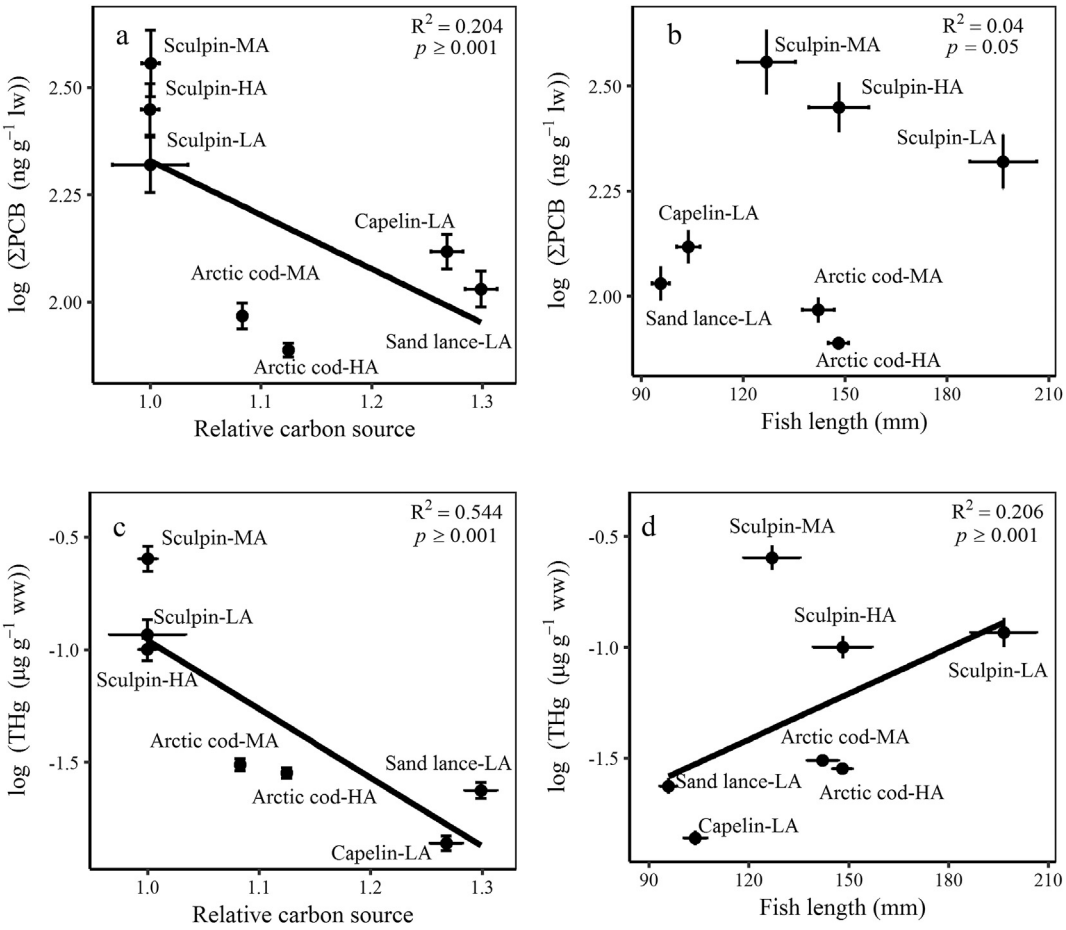

Fig. 4. Linear relationships between a) relative carbon source and b) fish length and log transformed polychlorinated biphenyl (ΣPCB) concentrations (on a lipid weight basis) and c) relative carbon source and d) fish length and log transformed total mercury (THg) concentrations (on a wet weight basis) in fish sampled in the low Arctic (LA), mid-Arctic (MA) and high (HA) eastern Canadian Arctic from 2012 to 2014. Points show mean (+/− standard error) for each fish species. Similar relationships to those found for ΣPCB were found between relative carbon source and dichlorodiphenyltrichloroethanes (ΣDDT) and chlordanes (ΣCHL).

Except for THg, it was not possible to explain the differences in contaminant concentrations between Arctic cod, capelin, and sandlance based on the biological and ecological variables we considered. Thus, our findings of higher concentrations of PCBs and most OCs, as well as different contaminant patterns in capelin and to some extent in sandlance, relative to Arctic cod, are consistent with the hypothesis that invading forage species can act as vectors for at least, certain organic contaminants into Arctic marine food webs. Nonetheless, native northern shrimp and sculpin showed higher concentrations of all contaminants compared to capelin and sandlance. Furthermore, POP concentrations in capelin and/or sandlance relative to Arctic cod were generally higher by only up to two-fold or less, and THg and ΣClBz concentrations were actually higher in Arctic cod than in capelin and sandlance. Taken together, our findings indicate that changes in Arctic native piscivore diets from Arctic cod to capelin and/or sandlance may only have a weak influence on their exposures to both THg and legacy POPs. Future work will focus on the potential impacts of these changes in the forage community for predators beyond contaminant concentrations, e.g., including concentrations of important micronutrients, such as selenium and essential fatty acids. If these dietary shifts are occurring towards prey of lower nutrient content, coupled with higher levels of certain contaminants, then the impact of these prey fish community changes may be a greater concern for Arctic seabirds, predatory fish, and marine mammals.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2017.05.085.

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