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Review

The distribution and trends of persistent organic pollutants and mercury in marine mammals from Canada's Eastern Arctic

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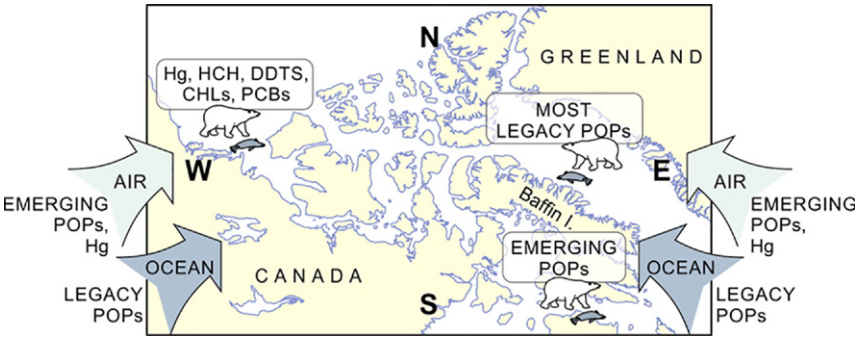
HIGHLIGHTS

- Mercury and legacy HCH in marine mammals are highest in the Western Arctic.
- Most legacy POPs in marine mammals are highest in the East and the Beaufort Sea.
- More recent POPs are highest in seals and polar bears at lower latitudes.
- Food web structure is shaping spatial trends of mercury and some legacy POPs.
- Water is driving legacy POPs whereas atmospheric deposition is driving recent POPs.

GRAPHICAL ABSTRACT



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ABSTRACT

Arctic contaminant research in the marine environment has focused on organohalogen compounds and mercury mainly because they are bioaccumulative, persistent and toxic. This review summarizes and discusses the patterns and trends of persistent organic pollutants (POPs) and mercury in ringed seals (*Pusa hispida*) and polar bears (*Ursus maritimus*) in the Eastern Canadian Arctic relative to the rest of the Canadian Arctic. The review provides explanations for these trends and looks at the implications of climate-related changes on contaminants in these marine mammals in a region that has been reviewed little. Presently, the highest levels of total mercury (THg) and the legacy pesticide HCH in ringed seals and polar bears are found in the Western Canadian Arctic relative to other locations. Whereas, highest levels of some legacy contaminants, including ∑PCBs, PCB 153, ∑DDTs, *p,p′*-DDE, ∑CHLs, ClBz are found in the east (i.e., Ungava Bay and Labrador) and in the Beaufort Sea relative to other locations. The highest levels of recent contaminants, including PBDEs and PFOS are found at lower latitudes. Feeding ecology (e.g., feeding at a higher trophic position) is shaping the elevated levels of THg and some legacy contaminants in the west compared to the east. Spatial and temporal trends for POPs and THg are underpinned by historical loadings of surface ocean reservoirs including the Western Arctic Ocean and the North Atlantic Ocean. Trends set up by the distribution of water masses across the Canadian Arctic Archipelago are then acted upon locally by on-going atmospheric deposition, which is the dominant contributor for more recent contaminants. Warming and continued decline in sea ice are likely to result in further shifts in food web structure, which are likely to increase contaminant burdens in marine mammals. Monitoring of seawater and a range of trophic levels would provide a better basis to inform communities about contaminants in traditionally

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harvested foods, allow us to understand the causes of contaminant trends in marine ecosystems, and to track environmental response to source controls instituted under international conventions.

Contents

1. Regional contaminant overview

Contaminants have entered the Arctic predominantly by atmospheric and oceanic long-range transport (Fig. 1) and secondarily from local sources. Long-range contaminants having the potential to produce consequential exposures in arctic biota exhibit a set of key characteristics. To transport long distances in the atmosphere or oceans, they must be volatile or semi-volatile, or partition favourably into water, but they must also be sufficiently involatile and chemically stable to deposit from the atmosphere within the Arctic once arriving there (Wania, 2003). Furthermore, to produce substantial risk at distant locations like the Arctic these contaminants must have been released in large quantities (e.g., 100 s of kt to >Mt (see, e.g., Li and Macdonald, 2005; Macdonald et al., 2000; Streets et al., 2011)), and persist in air or water for the time periods required to arrive in the Arctic or move within the Arctic (>5 days for winds, >1–10 years for ocean currents). Finally, these contaminants must readily enter biological food webs and be concentrated there to concentrations that exceed thresholds of known or potential adverse effects. Persistent, biologically accumulative, toxic (PBT) characteristics apply particularly to a wide range of organohalogen compounds (OHCs) released either intentionally (e.g., pesticides) or collaterally with industrial activities (e.g., PCBs, PBDEs and PFOS), and mercury (Hg), which has volatile and toxic forms (elemental and methyl mercury).

The OHCs comprise thousands of compounds that exhibit a wide range in physical chemical properties such as volatility and partitioning between media like air, water and particulates (Wania, 2003). Even within a single compound class (e.g., PCBs, PBDEs, DDTs, HCHs) there may be a wide range in these properties, with highly chlorinated compounds usually having higher affinity for particulates. These chemical properties ultimately decide much of the fate of OHCs: compounds with high affinity for particles like the highly chlorinated PCBs (high K_{OW}) tend to attach to particles. In the ocean some of these particles rapidly sink (days – weeks) from surface water into the deeper ocean or become buried in shallow-ocean sediments. Other OHCs, like the HCHs, tend to partition strongly from air into water, but not so much onto particles. Accordingly, the transport pathways of the OHCs into and through the Arctic Ocean are largely controlled by these physical chemical properties (Wania, 2003), and by the time scales of degradation (e.g., photolysis, hydroloyis, metabolism) and sedimentation (particle scavenging), which ultimately remove OHCs from the biosphere.

Hg can be buried in soils and sediments, which removes it from the biosphere, but unlike the OCHs, Hg cannot be destroyed. Transfer of Hg to long-term sequestration (sediments, permafrost and the deep oceans) takes centuries or longer during which the contaminant Hg has sufficient time to redistribute widely within air and ocean through volatilization, deposition and transport (e.g., see Driscoll et al., 2013). As a result of the historical loadings from human activities (e.g., mining, coal burning, industry), the ocean today is estimated to hold ~56 ± 16 kt of contaminant Hg in part manifested as an approximate tripling of the total Hg (THg) concentration in surface water above the pre-industrial, natural background (Lamborg et al., 2014).

Unlike the OHCs, Hg occurs naturally and thus anthropogenic Hg has loaded onto this natural background (Driscoll et al., 2013; Lamborg et al., 2014; Streets et al., 2011). Of great importance to fate and toxicity, Hg exhibits several forms in the environment including inorganic (elemental (Hg^0), dicationic Hg (Hg^{2+})) and organic (monomethylmercury (MMHg), dimethylmercury (DMHg)) forms (AMAP, 2011). Of these forms, MMHg presents by far the greatest concern for environmental toxicity, which implies that the inorganic forms of Hg, which dominate emissions and transport, must undergo methylation within the environment to convert them to the PBT form (Macdonald and Loseto, 2010). The potential for Hg to transfer between chemical forms presents a challenge to predicting Hg trends and toxicity within ecosystems because processes of oxidation, reduction and methylation are strongly controlled by environmental conditions like solar radiation and intensity of organic carbon processing. In this review we summarize and discuss the distribution and trends of POPs (and specifically OHCs) and mercury burdens in ringed seals and polar bears in the Eastern Canadian Arctic relative to the rest of the Canadian Arctic. Implications of climate-related changes on marine mammal contaminant levels are also discussed.

2. Contaminants in the Arctic

The risks to the Arctic from OHCs and Hg, which have long been recognized (AMAP, 1998; Barrie et al., 1992), provided much of the

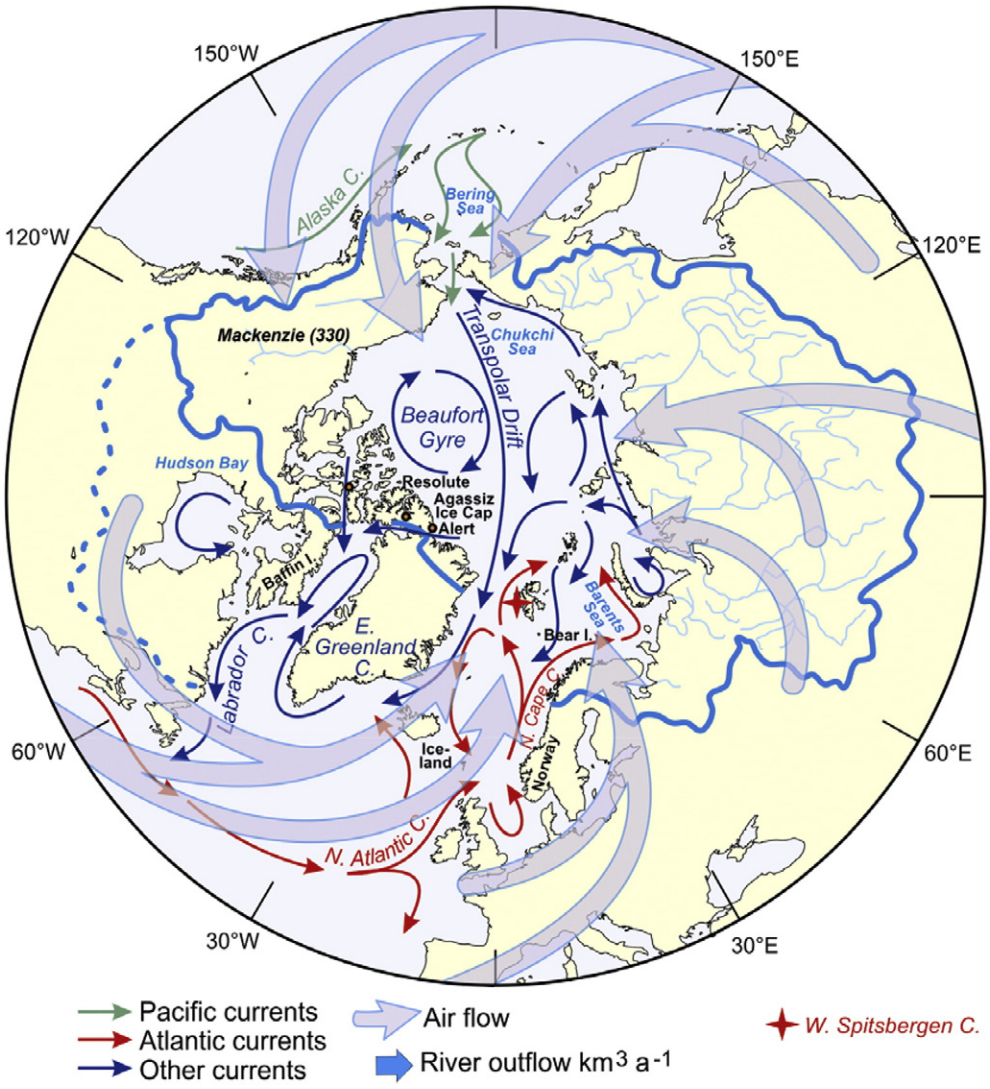

Fig. 1. A schematic diagram showing the major surface currents in the Arctic Ocean and the predominant atmospheric transport pathways.

motivation to reduce or eliminate industrial and agricultural sources in temperate regions through international conventions (e.g., Minimata, 2013; Stockholm, 2004). Accordingly, these contaminants all exhibit transient emission histories with globally significant quantities of the OHCs released predominantly after WWII. Mercury has been released for a much longer period dating back even to Roman times (Martínez-Cortizas et al., 1999) but, more pertinent to the present, Hg has been released in large quantities from multiple sources during the past 60 years with coal burning presently contributing a major fraction (AMAP, 2011; Streets et al., 2011). Streets et al. (2011) estimated that human activities all told have released 350 kt of Hg to the environment, 61% of that after 1850.

The primary historical loadings of OHCs and Hg to the environment have led to the present situation: persistent contaminants are distributed globally in air, soil, sediment, vegetation, other biota, water and ice. These environmental reservoirs may, in some cases, sequester contaminants for long periods (e.g., burial in marine and lake sediments, entry into abyssal ocean water), sequester them reversibly depending on climate and other conditions (e.g., glacial ice and snow, vegetation, surface water, food webs), or maintain them in active exchange cycles between reservoirs (e.g., air, surface water, vegetation, soil, and food webs). With the passage of time the contaminants *tend* to redistribute themselves away from locations of high concentration (technically, high fugacity; e.g., near industry or agriculture sources) toward locations of low concentration (e.g., regions remote from usage). Other environmental factors complicate the process of transport and re-deposition by providing the means to concentrate particular compounds but these processes require the input of energy (e.g., see Macdonald et al., 2002). International controls during the past few decades have reduced primary emissions of classical contaminants. As a result, these environmental reservoirs (i.e., secondary sources) now play a greater proportional role within the Arctic. Secondary sources are strongly driven by climate conditions like temperature, precipitation, ice cover, and vegetation (Macdonald et al., 2005; Wang et al., 2010).

The Western Arctic Ocean, which includes the Chukchi and Beaufort Seas, lies upstream of the Canadian Arctic Archipelago (CAA) and supplies almost all of the ocean water transiting its passages (Figs. 1 and 2). Therefore, concentrations of Hg and the OHCs in surface water of the Western Arctic Ocean provide the starting point for water passing through the CAA and eventually into Baffin Bay. Surface seawater concentrations in the Arctic Ocean, which are available for HCH, PCB, and other organics like the perfluoroalkyl acids (PFAAs) (e.g., Benskin et al., 2012a, 2012b; Jantunen et al., 2015; Li et al., 2002; Sobek and Gustafsson, 2014) and THg (Wang et al., 2012), imply that a reservoir has built up over time in the Arctic Ocean. Li et al. (2004) estimated that at its peak accumulation in 1982 6670 t of α-HCH was held by the Arctic Ocean, and by 2000 the total amount that had entered into the Arctic Ocean exceeded 27,000 t, accounting for about 0.6% of the

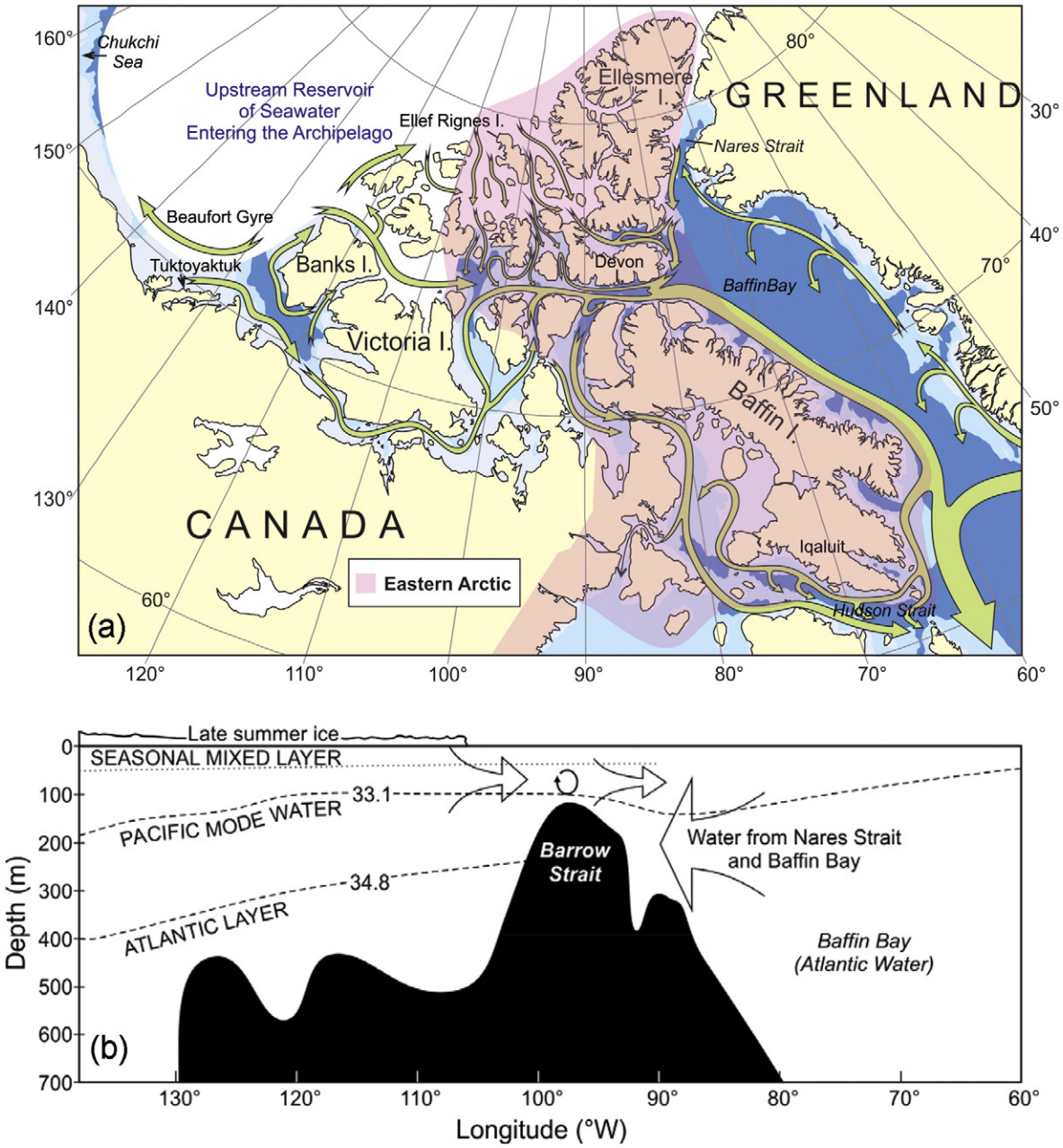

Fig. 2. A schematic diagram showing a) the major currents associated with the Canadian Arctic Archipelago, and b) the water structure and bathymetry within the Archipelago. After Bidleman et al., 2007.

global emission from land to atmosphere. Sobek and Gustafsson (2014) estimated the total inventory of $\sum_7$PCB to be 182 ± 40 t, the major contributors being river discharge (115 ± 11 t), ocean currents (52 ± 17 t) and atmospheric deposition (30 ± 28 t). Outridge et al. (2008) estimated that the Arctic Ocean contains 7920 t of THg, with 945 t of that in the upper ocean, where it would be most accessible to biota. Atmospheric deposition is thought to be an important ongoing source of inorganic Hg to the Arctic Ocean (~100 t yr^{-1}), partly because of atmospheric mercury depletion events (AMDEs) that occur with polar sunrise, but ocean currents from the Pacific and Atlantic also supply considerable amounts of dissolved Hg (48 t yr^{-1}). It is likely that rivers and coastal erosion (60 t yr^{-1}) also play important roles, possibly dominant regionally, although much of this Hg may enter on particulates and end up being buried on the shelves (see, e.g., Dastoor and Durnford, 2014; Fisher et al., 2012; Shang et al., 2015) rather than entering surface water passing through the Arctic Ocean.

3. The Eastern Canadian Arctic as a contaminant receptor region

The CAA is an important pathway for Arctic Ocean surface water to pass into Baffin Bay and the North Atlantic Ocean (Fig. 2a). Surface ocean water enters the western Arctic Ocean through Bering Strait. This water is then modified during its ~10 year residence time in the surface water (top 200 m) of the Canada Basin. A substantial proportion of this water then enters the CAA where it takes but a few years or less to transit into Baffin Bay. Shallow sill depths, 125 m at Barrow Strait and 220 m in Nares Strait, together with strong density stratification, constrain withdrawal of water from the Arctic Ocean to shallow depths (Fig. 2b). As a result, waters within the CAA are relatively rapidly replenished by Arctic Ocean surface seawater, and it is from this source that they inherit legacy contaminants. The Arctic Ocean surface water has a residence time of about 10 years, which means that the past few decades are the most important in terms of contaminant accumulations.

Within the CAA, water properties and contaminant burdens are further modified by atmospheric exchange (deposition and evasion), the addition of river water (~220 km^3 yr^{-1}), and processes like sea-ice formation and melting, primary production and degradation. Runoff within the CAA includes glacial melt, which under the present circumstances of warming in the Arctic provides a means to release legacy OHCs and Hg deposited during decades like the 1950s and '60s when usage was much higher (Blais et al., 2001; Macdonald et al., 2005).

The HCHs provide a clear example of how up-stream ocean reservoirs may affect OHC burdens in the CAA. The emission histories for HCHs are relatively well known, and there are adequate water and air data to infer trends with time in surface waters of the Arctic Ocean (e.g., Bidleman et al., 2007; Harner et al., 1999; Jantunen et al., 2015; Li et al., 2002, 2004; Macdonald et al., 2000; Shen et al., 2004; Wania and Mackay, 1999). Because the HCHs have a strong affinity for water, especially cold water, a large portion of the HCH transported by air ended up in northern surface waters producing concentrations of α-HCH that exceeded 6 ng L^{-1} in some places during the 1980s. β-HCH also loaded into Arctic Ocean surface waters, but because it had an even higher affinity for cold water the Asian source was rained out or exchanged into surface water in the Bering Sea, before it could get to the Arctic thence to enter the Chukchi and Beaufort Seas about 10 years later via water currents though Bering Strait (sill depth of ~50 m) (Li et al., 2002). This large-scale regional separation of HCH compounds due to differing physical properties is more than of casual interest: it has biological significance as elegantly exemplified by bowhead whales, whose tissues exhibited a seasonal variation in HCH α/β ratio due to altered contaminant exposure along their annual foraging migration between Bering and Beaufort Seas (Hoekstra et al., 2002).

Degradation rates for both α- and β-HCH are slow enough to permit buildup of the inventories in Arctic Ocean surface water over time, with a major removal pathway being outflow into the CAA channels. The Western CAA, therefore, reflects the upstream loading of surface waters in the Canada Basin (Fig. 2b). Once past the sill at Barrow Strait, the Eastern CAA surface waters reflect a mixture of water from the Beaufort Sea (historically higher concentrations) with water of lower HCH concentration from the Atlantic Ocean (Fig. 2b), which sets up a longitudinal gradient across the CAA (Bidleman et al., 2007). Presently, CAA waters are shedding some of this HCH by evasion to the atmosphere either within the CAA itself (Jantunen et al., 2007) or into the Labrador Sea (Shen et al., 2004), which would further augment concentration declines from west to east. Microbial degradation and export of HCH from the Canada Basin has, subsequently, led to the decline in HCH in the Arctic Ocean's surface waters such that HCH will become undetectable by ~2020 (Pućko et al., 2013). At that point the Beaufort Sea reservoir will play little role in supplying HCH to the CAA. Although we have fewer data for β-HCH, it seems clear that this compound will follow a similar temporal pattern to α-HCH but with a delay of ~10 yrs (Li et al., 2002).

Perfluoroalkyl acids (PFAAs) are highly chemically stable and "terminal" degradation products resulting from the transformation of precursor perfluorinated alkyl substances (PFASs). Chemicals categorized as PFASs have been manufactured in large quantities for over 60 years and used in a wide range of applications including aqueous firefighting foams, grease-proofing paper, textile stain and soil repellents, processing aids in fluoropolymer manufacturing. The use of PFASs in various commercial products has ultimately led to a release of large quantities to municipal waste waters and directly to the atmosphere, leading to the degradation of these precursors and with the formation of PFAAs which are essentially non-degradable in the environment (Li et al., 2017). In particular, perfluorooctane sulphonate (PFOS) would be widely recognized as a degradation product of the side-chain fluorinated polymers found in the pre-2002 fabric stain protector product called Scotchgard™ (Chu and Letcher, 2014). Because PFAAs are strong acids and that highly favour aqueous media in their conjugate base anion form (e.g., Gouin and Wania, 2007), they provide an interesting comparison with the HCHs. Based on surface water samples collected between 2005 and 2009, Benskin et al. (2012b) constructed a transect showing the spatial distributions of two dominant compounds, PFOA (perfluorooctanoic acid) and PFOS, in surface water across the CAA (Fig. 3). Like the HCHs, these PFAAs and PFASs have accumulated in Canada Basin surface waters, entering in part through Bering Strait. Although it is likely that the CAA inherits these upstream concentrations as a starting point, the higher concentrations observed in the Central and Eastern CAA suggest further atmospheric deposition of PFAAs and PFOS and/or their PFAS precursors. For waters east of Barrow Strait we would expect that transport within Atlantic surface currents has played a dominating role.

PFAA chemical compositions provide direct insight into manufacturing sources and timescales for these PFASs. Historical production using electrochemical fluorination, which produces a mixture of branched and linear compounds, was supplanted in the 2000s by telomerization, which produces only linear compounds. Examination of spatial data (Benskin et al., 2012a) indicates that surface water in the North Atlantic Ocean is strongly tagged by historical electrochemical production products, whereas the North Pacific and Western Arctic contains more recent telomerization products likely from Asian sources. These authors inferred from these data that the PFAAs endure for long periods within seawater, and that important sources to the CAA include Atlantic and Pacific oceanic pathways plus atmospheric deposition within the CAA.

The detailed case for HCH demonstrates the importance of contrasts in contaminant concentrations in Arctic Ocean and Baffin Bay surface waters in determining background spatial trends in CAA waters. Unfortunately, we have insufficient data or time series for other legacy OHCs, but we can infer that gradients between east and west Archipelago waters have likely been set in the same manner as for the HCHs. This broad background pattern is then altered within the CAA by air-sea exchange, degradation, sedimentation and river input of OHCs. For contaminants that have been introduced more recently into global cycles (e.g., PBDEs and PFOS), we can infer that atmospheric deposition within the CAA may presently be more important because surface ocean reservoirs have not had sufficient time to accumulate contaminants to their maximal concentrations. In the case of PFOS, where ocean transport is relatively important (e.g., see Armitage et al., 2009), the ocean pathways may already rival atmospheric pathways to the CAA.

Surface water in the Beaufort Sea also provides baseline concentration of THg entering the CAA. THg concentration is then modified by losses and gains within the CAA. Due to the technical difficulty of measuring Hg in sea water at pM (pico molar = 10^{-12} mol L^{-1}) concentrations, there are relatively few reliable ocean-section data. Recently, several studies have produced high-quality data that provide a broad view of THg and MMHg in the North Pacific, Arctic and North Atlantic Oceans (Table 1). These data show no significant THg gradient between the N Pacific and the N Atlantic Oceans, with concentrations usually found to be about 1.1 ± 0.6 pM. The most interesting feature of the data in Table 1 is the relatively high concentrations seen in some surface samples in the CAA and Hudson Bay. There is mounting evidence that rivers may play an important role in supplying Hg to surface seawater in the Arctic (Andersson et al., 2008; Fisher et al., 2012; Heimbuerger et al., 2015; Zhang et al., 2015). Accordingly, it seems likely that the Mackenzie River may be an important modulator of THg in surface water entering the CAA (e.g., see Leitch et al., 2007; Wang et al., 2010). Within the CAA the ~220 km^3 yr^{-1} of runoff (Alkire et al., 2017) could produce high THg concentrations locally, if not regionally, in surface water, but there are no measurements of Hg in these small rivers. Hudson Bay receives ~700 km^3 yr^{-1} of runoff from many rivers distributed around the Bay (Déry et al., 2005). These rivers reportedly carry 0.7–3.5 pM THg concentration (Hare et al., 2008; Kirk and St. Louis, 2009) and could, therefore, contribute to high THg concentrations observed in surface waters of Hudson Bay.

Using stable Hg isotopes, Lehnherr et al. (2011) showed that reactions producing or demethylating MMHg were rapid, which implies

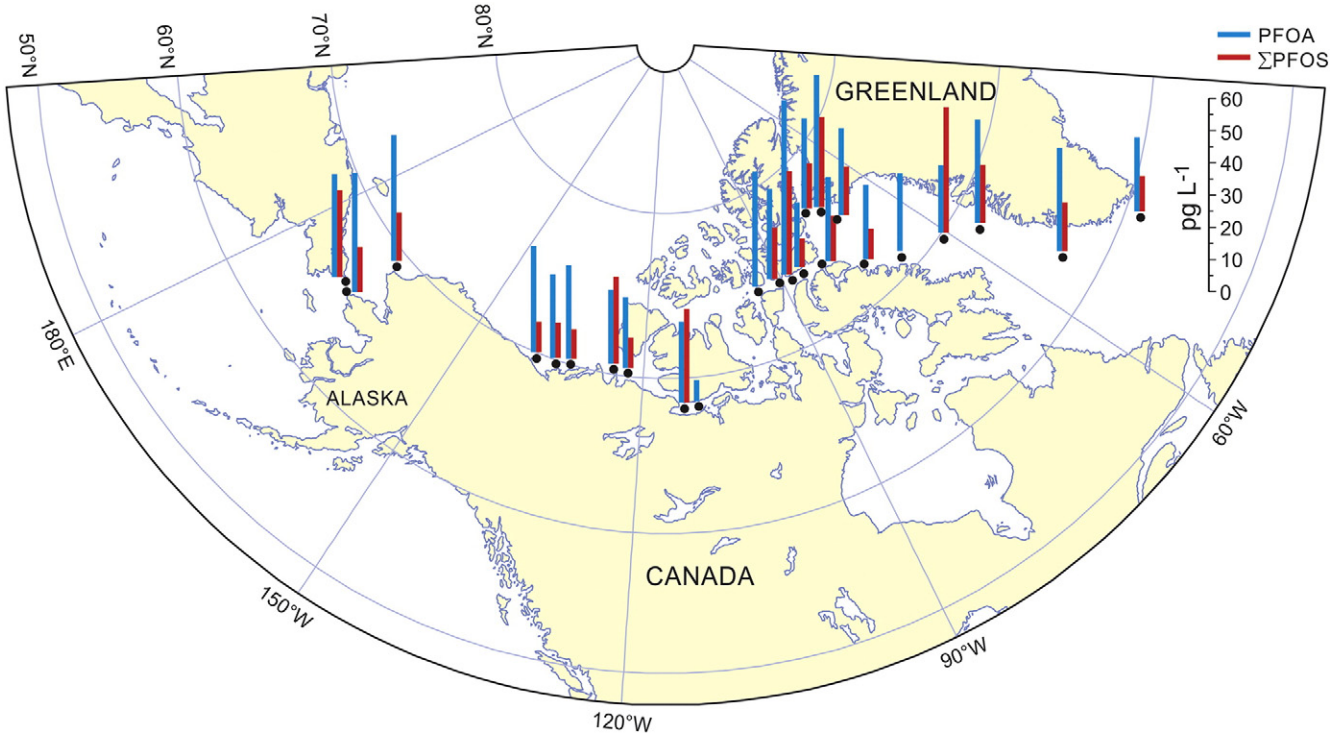

Fig. 3. The distribution of perfluorooctanoate (PFOA) and perfluorooctanesulfonate (PFOS) in surface waters of the CAA (2007 and 2009 data). Adapted from Benskin et al., 2012a.

that MMHg cannot be transported long distances in the water (Wang et al., 2012). Given that the residence time of water in the CAA is about three years, one would not expect MMHg entering the Archipelago from the Arctic Ocean to persist very far into the passages; instead, MMHg concentrations would reflect methylation processes occurring locally within the CAA (<a few 100 km). However, the life time of MMHg in natural seawater at ambient concentration remains unconfirmed. Further work is warranted given the potential importance of MMHg production over the Chukchi/Beaufort Shelves, the region where nutrient-rich waters entering the Western CAA are produced. The data in Table 1 suggest that production of MMHg is slow relative to destruction in the interior waters of the North Pacific, North Atlantic and at the North Pole, whereas the CAA and Baffin Bay (North Water), both of which have areas of higher primary production/regeneration, may produce more MMHg.

4. POPs in the marine ecosystem

4.1. Fish and invertebrates

Persistent organic pollutants (POPs), and more specifically organohalogen contaminants (OHCs), bioaccumulate in organisms and increase with each trophic level in the marine food web, biomagnifying to levels that can affect arctic wildlife and human health (Letcher et al., 2010; McKinney et al., 2012). Invertebrates and fish are excellent indicators of environmental contamination since these organisms can bioaccumulate POPs from the water, sediments, and their diet. Invertebrates and fish, especially Arctic cod (*Boreogadus saida*), are also major diet items of marine mammals in the Arctic. Measuring levels of contaminants in these organisms enables us to assess biomagnification in marine food webs and to better interpret the levels and trends of contaminants in marine mammals. Further, a number of fish species are important traditional food items for Inuit.

Very few studies have reported OHC concentrations in Arctic marine invertebrates and fish in the Eastern Canadian Arctic. Further, variations in POP concentrations and patterns can vary with trophic position (indicated by $\delta^{15}N$), body size, partition coefficients and bioaccumulation of individual compounds. Borgå et al. (2005) measured PCBs and organochlorine pesticide (OCP) concentrations in invertebrates and Arctic cod collected in 1998 from the North Water Polynya in northern Baffin Bay. In the copepod (*Calanus* spp.) $\sum$HCH (4.0 ± 3.2 ng/g ww; 47.6 ± 38.1 ng/g lw) was found in greatest concentration followed by $\sum$PCBs, $\sum$chlordanes (CHL), $\sum$dichlorodiphenyltrichloroethane (DDT) and hexachlorobenzene (HCB). The concentration profile

Table 1
A summary of dissolved total Hg and monomethyl Hg concentrations in the North Pacific, Arctic and North Atlantic Oceans.

Location	Reference	THg (pM)	MMHg (pM)	*n*
North Pacific	Sunderland et al., 2009	0.99 ± 0.32	0.095 ± 0.052	48
Beaufort Sea	Wang et al., 2012	1.1 ± 0.45	0.20 ± 0.15	90, 57
CAA	Kirk et al., 2008	2.3 ± 2.4	0.31 ± 0.22	15
Baffin Bay (North Water)	Kirk et al., 2008	1.5 ± 1.3	0.30 ± 0.17	15
Hudson Bay	Hare et al., 2008	3.8 ± 1.7		25
Hudson Bay	Kirk et al., 2008	2.1 ± 2.6	0.19 ± 0.12	29
North Pole	Heimbuerger et al., 2015	0.54 ± 0.09	0.053 ± 0.033	12
Makarov Basin (85°N)	Heimbuerger et al., 2015	1.0 ± 0.25	0.21 ± 0.08	27
Amundsen Basin (81°N)	Heimbuerger et al., 2015	1.3 ± 0.23	0.16 ± 0.10	22
North Atlantic	Bowman et al., 2015	0.89 ± 0.30	0.095 ± 0.098	605, 432

differed in the pelagic amphipod, *Themisto libellula* with ∑PCBs (2.9 ± 1.3 ng/g ww; 120.8 ± 54.2 ng/g lw) being found in greatest concentration, followed by ∑CHL, ∑DDT, ∑HCH and HCB. For Arctic cod, ∑CHL (2.7 ± 0.7 ng/g ww; 225 ± 58.3 ng/g lw) was found in greatest concentration, followed by ∑DDT, ∑PCB, ∑HCH and HCB. Fisk et al. (2003a) measured PCBs and OCP concentrations in seven species of zooplankton collected in 1998 in northern Baffin Bay. These included *C. hyperboreus* (herbaceous copepod); *Euchaeta glacialis* (omnivorous copepod), *Metridia longa* (omnivorous copepod), *Mysis occulata* (detritus feeding and predatory mysid), *Themisto libellula* (predatory amphipod), *Sagitta* sp. (predatory arrowworm), and *Pandalus* sp. (predatory shrimp). The ∑PCB and OCP concentrations varied between species, but in general ∑PCBs (5.1–33.7 ng/g ww; 81.0–1204 ng/g lw) had the greatest concentrations and ∑chlorobenzenes (CBz) (0.29–0.78 ng/g ww; 9.4–27.9 ng/g lw) had the lowest concentrations (Fisk et al., 2003a). HCB and HCH isomers in zooplankton are generally greater in the Western Canadian Arctic than the Eastern Arctic (Fisk et al., 2003b), which likely reflects the gradient set up by atmospheric and oceanic transport of these chemicals from Asian sources to the Western Arctic.

Benthic invertebrates tend to have a larger range in POP concentrations across species, with filter- and detritus-feeding invertebrates having some of the lowest concentrations among all biota (Fisk et al., 2003b). ∑PCBs are generally the contaminant found in highest concentration among most benthic invertebrates in the Canadian Arctic, with the greatest concentrations having been measured in the scavenging amphipod *Anonyx nugax* (Fisk et al., 2003b).

Studies in the Eastern Canadian Arctic have found that ∑polybrominated diphenyl ether (∑PBDE) concentrations are higher in zooplankton (e.g., 72.9 ± 10.1 ng/g lw) than upper trophic level organisms (e.g., 0.4 ± 0.2 ng/g lw in walrus) (Morris, 2015; Morris et al., 2007; Tomy et al., 2008). These results, however, diverge from reports of low concentrations found in invertebrates (range: 0.16–0.53 ng/g lw) from Svalbard (Sormo et al., 2006) including shrimp (0.39 ng/g lw) from the Beaufort Sea (Desforges et al., 2013). This may reflect large geographical variability in ∑PBDE concentrations in lower trophic level organisms across the circumpolar Arctic, which may be due to the more recent release of PBDEs compared to legacy OHCs whereby spatial distributions have not yet thoroughly mixed over space.

Mean ∑PBDE concentrations from the Hudson Bay region were 5.4 ng/g lw in the bivalve (*Mytilis edulis*), 9.8 ng/g lw in Arctic cod (*Boreogadus saida*) and 72.8 ng/g lw in sculpin (*Myoxocephalus scorpioides*) (Kelly et al., 2008). Hydroxylated (OH-) PBDEs were not detected in these three species, however methoxylated (MeO-) PBDEs were detected with OHBDE-47 representing 30–40% of ∑PBDEs (Kelly et al., 2008).

Arctic cod mean ∑PBDE concentrations collected from Davis Strait (23 ng/g lw, Tomy et al., 2008) were similar to Arctic cod PBDE concentrations collected from Barrow Strait (23 ng/g lw, Morris et al., 2007), Hudson Bay (9.8 ng/g lw, Kelly et al., 2008), and the Eastern Beaufort Sea (offshore cod, Mackenzie River: 0.69 ng/g lw; nearshore cod, Franklin Bay: 20.3 ng/g lw, Desforges et al., 2013) but much lower than those measured in cod (205 ng/g lw, Tomy et al., 2009) that were also from the Eastern Beaufort Sea (Tomy et al., 2009) and collected during a similar time period (2004–06) as Desforges et al. (2013). If we compare Tomy et al. (2009) Arctic cod PBDE concentrations to those reported in the east, then there appears to be considerable spatial variability across the Canadian Arctic which could be due to differences in feeding ecology or due to the later release of PBDEs into the environment resulting in less time to even out through mixing, transport and exchange through the marine food web. Variation in condition (lipid weight), age of fish samples, and different analytical laboratories may have contributed to the differences observed in the Eastern Beaufort Sea cod concentrations, although the reported concentration may be an accurate reflection of their feeding ecology. Further research and longer time series are needed to understand these geographical patterns.

4.2. Ringed seals and polar bears

The ringed seal (*Pusa hispida*) and the polar bear (*Ursus maritimus*) are ideal species for assessing the spatiotemporal trends of OHCs within arctic marine ecosystems. Both species have circumpolar distributions, but individuals of regional subpopulations have a relatively modest home range. Further, ongoing studies have secured samples from both species at various locations across the Canadian Arctic enabling a strong spatial OHCs dataset for comparing average levels between regions. OHCs including PCBs, PBDEs, PFOS, PFCAs, and pesticides (e.g., DDT, CHL, and HCH) can biomagnify to high levels in top predators (Muir et al., 2013). As the top trophic feeding predator of arctic and subarctic marine food webs, polar bears accumulate high levels of OHCs from their primary prey species, the ringed seal.

The ringed seal, which is the most abundant arctic pinniped species, is also considered a top predator in nearshore pelagic food webs. This seal species has a diet that consists of a wide variety of pelagic and benthic invertebrates and fish (e.g., Arctic cod, capelin (*Mallotus villosus*), sand lance (*Ammodytes sp.*), and sculpin (Cottidae)) (Gjertz and Lydersen, 1986; Labansen et al., 2007; McLaren, 1958). At higher latitudes in the Arctic, adult ringed seal have been reported to consume more forage fish (e.g., Arctic cod), than sub-adults which consume more pelagic zooplankton. In contrast to sub-adult and adult seals at lower latitudes which both consume more sub-Arctic fish species and pelagic zooplankton (Yurkowski et al., 2015). As mentioned above, ringed seals generally forage within limited areas (Thiemann et al., 2007), however, some regional movements, e.g., from the Western Canadian Arctic to the Bering Sea and East Cape of Siberia (Harwood et al., 2012, 2015; Smith, 1987), from the Labrador coast to Ungava Bay and western Baffin Island (Brown et al., 2014), and between Greenland and Canadian waters have been documented (Teilmann et al., 1999). Adult female ringed seals and polar bears typically have lower levels of OHCs than adult males, due to loss by reproductive females of contaminant load to offspring.

4.2.1. Spatial trends of organohalogen contaminants in ringed seals

Despite being banned for numerous decades, ∑PCBs remain the dominant legacy organochlorine (OC) contaminant in ringed seals across the Arctic, followed by ∑DDTs and ∑CHLs (Table 2). This trend is consistent with previous findings which show PCBs remaining the major POP in ringed seal blubber (Addison et al., 2014; Muir et al., 2013). One previous study, however, found ∑DDTs to be the major OC in the Western Canadian Arctic (Gaden et al., 2012). Legacy OC concentrations, which include PCBs and some OCPs, are generally greater in the east compared with the west, with the exception of toxaphene, ∑HCHs, α-HCH, and β-HCH (Table 2; Fig. 4). This is consistent with previous Canadian Arctic studies which have shown higher levels of most of the legacy OCs, except for toxaphene, and β-HCH, in ringed seals from the Eastern Canadian Arctic than in seals from the Western and Central Canadian Arctic (Fisk et al., 2003b). Elevated levels in the west likely reflect the greater use of β-HCH in Asia, the strong partitioning of HCHs and toxaphene into cold water, and efficient atmospheric/oceanic transport pathways to the Western Arctic (Fig. 1, Li et al., 2002). The reason for generally greater concentrations of PCBs and OCPs in the east is not clear. However, these greater concentrations may reflect global usage patterns: many of these chemicals were used earlier and in greater amounts in North America and Europe than in Asia (Breivik et al., 2002). Furthermore, these compounds do not partition as strongly into cold water as the HCHs and, therefore, the pathway involving atmospheric transport directly to the Arctic followed by air-sea exchange may have been less important than deposition in the ocean close to sources and subsequent transport northward in ocean surface currents (see, e.g., Stemmler and Lammel, 2009). The greater Eastern Arctic concentrations could also reflect, partially, the release of archived chemicals deposited onto permanent snowfields and small glaciers within the CAA, which are now receding. Some of the legacy

Table 2
Geometric means and 95% confidence intervals (ng/g lipid weight) of chlorinated, brominated and fluorinated contaminants in ringed seals collected from 2003 to 2010.
Data from Muir et al., 2013.

	Sachs Harbour	Ulukhaktok	Arctic Bay	Gjoa Haven	Grise Fjord	Resolute	Qikiqtarjuaq	Pangnirtung	Arviat	Inukjuak	Ungava	Nain
n	33	16	19	22	17	52	7	22	62	10	3	8
∑ PCBs[a]	488.7	328.1	334.2	415.0	509.3	358.1	248.9	275.4	338.1	247.2	539.5	912.0
	399.9–597.0	267.3–402.7	274.8–406.4	349.1–492.0	379.3–685.5	310.5–414.0	201.4–307.6	208.4–364.8	287.7–397.2	167.5–363.9	92.0–3169.6	653.1–1270.6
PCB153[a]	69.3	15.8	45.1	72.1	67.5	40.3	33.0	45.6	43.0	42.3	71.1	201.8
	49.2–97.7	9.1–27.2	34.3–59.2	59.0–88.1	47.6–95.5	33.5–48.4	20.8–52.2	33.5–61.9	33.4–55.3	26.7–66.8	11.5–439.5	142.6–285.8
∑ CHL[a]	225.4	121.3	125.3	120.8	183.7	193.6	152.1	179.1	121.3	101.2	131.5	283.1
	185.8–273.5	97.7–150.7	104.2–151.0	101.2–143.9	128.5–263.0	166.0–225.9	122.2–189.2	143.5–222.8	97.5–150.7	66.2–154.5	10.5–1648.2	196.8–407.4
∑ ClBz[a]	51.2	38.2	29.4	41.4	35.2	41.8	41.0	37.5	18.0	12.6	51.5	32.6
	42.3–61.8	33.1–44.1	24.2–35.6	36.4–47.1	30.3–40.9	37.8–46.2	32.9–51.3	32.4–43.5	15.9–20.4	10.5–15.2	25.1–105.7	23.5–45.1
∑ DDTs[a]	248.3	113.0	181.1	106.9	276.7	186.2	177.0	228.0	145.5	132.4	323.6	599.8
	189.2–325.8	84.3–151.4	137.1–239.3	88.7–128.5	203.7–376.7	155.6–222.3	127.1–246.6	180.7–287.7	119.9–177.0	84.7–206.5	39.5–2654.6	376.7–957.2
p,p′-DDT[a]	36.5	21.2	26.2	18.1	48.8	27.0	22.9	22.8	10.4	n.a.	n.a.	n.a.
	26.6–50.0	16.8–26.7	20.6–33.2	15.0–21.9	37.8–63.0	22.2–33.0	18.1–28.8	17.3–29.9	8.2–13.2			
p,p′-DDE[a]	196.8	91.6	140.0	78.2	205.1	141.6	134.3	194.5	128.8	116.7	298.5	540.8
	150.0–258.2	65.9–127.1	102.1–191.4	64.1–95.5	146.2–287.1	117.2–171.0	91.0–198.2	153.5–246.6	105.9–157.0	73.8–184.5	36.6–2426.6	351.6–829.9
Dieldrin[a]	42.1	34.5	36.7	37.8	46.2	50.9	45.0	41.0	25.1	27.1	48.4	61.5
	33.3–53.2	26.2–45.6	29.0–46.7	32.1–44.6	37.4–57.3	44.2–58.7	33.0–61.4	32.7–51.5	20.8–30.2	20.3–36.2	13.4–175.4	38.4–98.6
Toxaphene[a]	95.3	150.3	104.2	45.1	118.3	134.6	n.a.	101.6	52.6	84.5	73.1	n.a.
	70.8–128.2	94.2–239.9	78.5–138.0	36.3–56.0	76.0–184.1	106.7–169.4		71.3–144.9	43.3–64.0	54.1–131.8	9.4–564.9	
∑ HCHs[a]	147.9	99.1	78.2	61.0	61.7	100.5	85.7	91.8	50.9	53.8	75.5	102.1
	121.1–181.1	71.8–136.8	67.1–90.8	49.5–75.0	48.8–77.8	88.5–114.3	73.8-	77.4–108.6	41.9–62.1	42.4–68.5	11.2–509.3	80.2–130.3
α-HCHs[a]	100.0	72.9	49.4	42.5	35.6	64.9	56.5	51.5	40.0	42.8	27.2	67.8
	78.9–126.8	49.5–107.2	41.3–59.3	34.4–52.2	27.4–46.2	56.5–74.5	47.5–67.0	44.5–59.6	33.0–48.5	33.9–53.8	0.7–1013.9	53.7–85.3
β-HCH[a]	40.4	27.9	24.4	15.6	22.5	30.3	26.6	35.8	7.9	9.0	40.1	30.9
	34.3–47.5	23.7–32.8	20.5–29.0	12.1–20.0	18.2–28.0	26.2–34.8	22.3–31.7	28.2–45.4	6.1–10.3	6.4–12.5	15.2–105.7	21.4–44.7
∑ PBDEs[a]	5.1	4.9	2.8	11.1	5.0	5.1	5.7	8.1	10.0	16.1	13.5	28.8
	3.7–6.9	3.3–7.4	1.4–5.6	8.9–13.9	3.3–7.6	4.0–6.5	3.2–10.0	6.9–9.5	8.4–11.9	9.6–26.9	1.8–101.9	19.1–43.6
∑ PFCAs[b]	14.5	15.8	15.9	71.3	20.2	11.7	27.8	8.2	28.1	43.9	5.9	20.0
	11.0–19.1	10.8–23.3	10.9–23.2	52.1–97.7	13.8–29.7	10.0–13.7	23.7–32.7	5.6–12.1	24.5–32.2	35.0–55.1	1.3–26.7	14.5–27.7
PFOS[b]	8.3	8.3	10.4	32.4	21.5	5.6	22.1	5.6	17.5	36.5	5.1	21.9
	6.3–10.9	5.5–12.5	7.7–13.9	20.7–50.7	13.7–33.9	4.9–6.5	17.2–28.5	3.7–8.5	15.0–20.3	28.3–47.0	2.6–10.0	15.3–31.2

[a] Blubber tissue.
[b] Liver tissue.

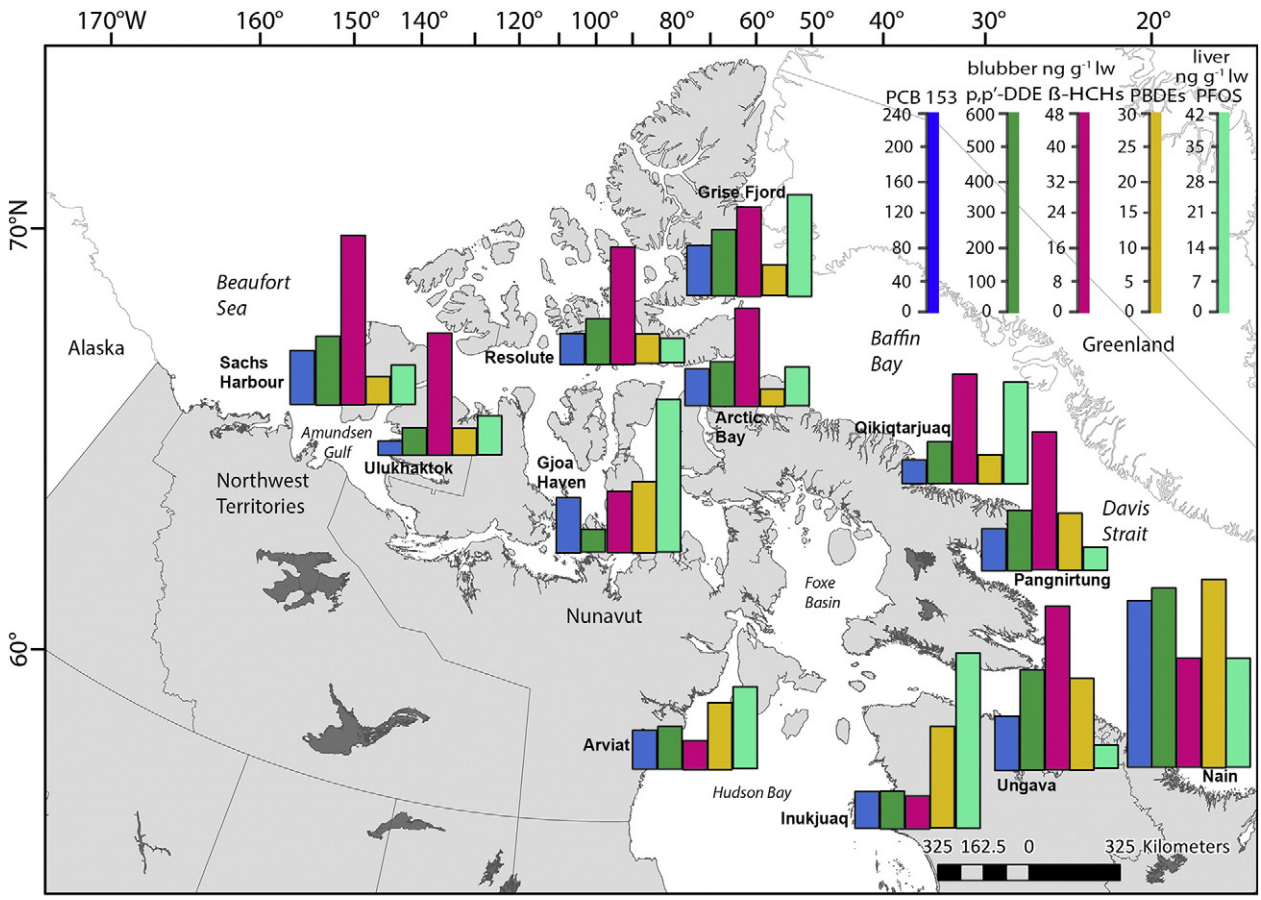

Fig. 4. Geometric means (ng/g lipid weight) of PCB 153, *p,p′*-DDE, β-HCHs, ∑PBDEs, and PFOS of ringed seals collected from 2003 to 2010. Data from Muir et al., 2013.

OCs (e.g., ∑PCBs, PCB 153, ∑CHLs, ∑DDTs, *p,p′*-DDE, and ClBzs) are relatively elevated in ringed seals from Sachs Harbour (Table 2, Fig. 4). In addition to assessing the trends for ∑PCBs and ∑DDTs in ringed seals and polar bears, we assessed the trends for PCB 153 and *p,p′*-DDE as these compounds have been shown to be recalcitrant and biomagnify in marine mammals. The greater relative OC concentrations from the Beaufort Sea region may be due to these seals feeding at a higher trophic position relative to seals from other regions (see Table 3 in Brown et al., 2016) or that there have been some continuing inputs of some of these compounds (e.g., ∑DDTs, *p,p′*-DDE) to the Western Arctic, despite restrictions on their use in North America during the 1970s (Addison et al., 2014).

PBDEs have been used since the 1980s, mainly as flame retardants in fabrics and other materials, and continue to be used in in-use products despite increasing restrictions and bans on use which were implemented in the early 2000s. Like other POPs, PBDEs accumulate to high levels in upper trophic level marine mammals. BDE-47 is the dominant congener in ringed seals and represents approximately 55–80% of ∑PBDEs (Ikonomou and Addison, 2008; Kelly et al., 2008). The most recent data show greater concentrations in the south, which decline northward in the Canadian Arctic; greatest average ∑PBDEs concentrations are found in ringed seals from southern locations in the Eastern Canadian Arctic, e.g., Nain, Inukjuak, and Ungava (Table 2; Fig. 4). This is consistent with a recent Canadian Arctic study which showed higher levels of PBDEs in ringed seals from the south than in seals from the northern Canadian Arctic (Houde et al., 2017). This broad south to north distribution likely reflects the relatively early stage of environmental loading of the PBDEs compared to the legacy contaminants. At this stage highly contaminated reservoirs (soils, vegetation, etc.) in temperate industrial regions supply atmospheric transport through seasonal volatilization, which spreads the contaminants northward (see, e.g., Gouin et al., 2004). Average ∑PBDE concentrations in Gjoa Haven were also relatively great, which may be due more to seals from this area feeding at the highest level of the food web (mean $\delta^{15}N$ value of 5.6) and having a depleted relative carbon source (RCS) (mean value of 0.8), whereby they are feeding more on pelagic prey, relative to seals from other areas (Brown et al., 2016), than it has to do with major sources or ∑PBDE transport routes.

PFASs were produced via two major synthesis processes: electrochemical fluorination (ECF) and telomerisation (Buck et al., 2011). The ECF process was used primarily by the 3 M Company from the 1950s to 2001 in the production of perfluorooctane sulfonyl fluoride (PFOSF) chemicals which include PFOS. In 2001, the 3 M Company announced the phase out of its PFOSF-based chemicals in favour of shorter chain-length compounds (Butt et al., 2010). However, PFOS has continued to be produced in China since 2003. In contrast, the telomerisation process has been used by various companies since the 1970s for the production of fluorotelomer alcohols (FTOHs), fluorotelomer olefins, fluorotelomer acrylates, fluorotelomer idodides, and PFCAs. FTOHs degrade to PFCAs (Hurley et al., 2004) and research suggest that the fluorotelomer olefins (Nakayama et al., 2007), fluorotelomer acrylates (Butt et al., 2009), and fluorotelomer idodides (Young et al., 2008) may form PFCAs via atmospheric oxidation. In contrast to legacy POPs which accumulate in lipid rich tissues, PFCAs and PFOS are associated with proteins and accumulate mainly in the liver, kidneys, and bile secretions (Butt et al., 2010). The sources and transport routes of PFASs to the Arctic are not well understood. However, two pathways have been suggested: atmospheric transport and oxidation of volatile precursors and direct transport of PFCAs and PFSAs via ocean currents (Butt et al., 2010). Local inputs are likely minimal, however a few studies indicate that both atmosphere

Table 3
Geometric means and 95% confidence intervals (ng/g lipid weight) of chlorinated, brominated and fluorinated contaminants of adult and sub-adult male and female polar bears collected from 2005 to 2008.
Data from McKinney et al., 2011a and Letcher et al., 2010.

	South Beaufort Sea	North Beaufort Sea	Gulf of Boothia	Lancaster/Jones Sound	Baffin Bay	Davis Strait	West Hudson Bay	South Hudson Bay
n	17	29	7	13	14	9	12	21
∑PCBs[a]	3688	5541	2445	2598	3211	4674	4634	5523
	2600–5232	4518–6797	1599–3739	2005–3366	2305–4472	2693–8114	3072–6992	4617–6608
PCB153[a]	1168	1934	864	970	1113	1176	1660	1937
	1158–1179	1923–1945	853–874	963–977	1002–1125	1158–1195	1639–1681	1927–1948
∑CHL[a]	1268	1982	1824	1130	2167	2135	3477	2166
	926–1736	1555–2525	1135–2930	788–1619	1523–3083	1383–3298	2386–5068	1604–2924
∑ClBz[a]	145	237	304	234	266	255	221	171
	106–197	199–282	231–400	194–283	205–344	149–435	174–279	150–194
∑DDTs[a]	81.8	93.7	31.5	64.0	179	104	88.1	152
	60.5–110	79.9–110	19.8–49.7	44.1–92.7	125–257	60.0–181	54.8–141	127–182
p,p′-DDT[a]	18.1	12.5	1.3	5.4	8.2	2.7	6.3	5.1
	17.9–18.3	12.4–12.7	1.3–1.3	5.3–5.5	8.0–8.3	2.6–2.7	6.2–6.4	5.0–5.2
p,p′-DDE[a]	29.9	53.0	21.1	44.0	124	46.4	56.1	113
	29.5–30.4	52.6–53.3	20.7–21.4	43.0–45.0	123–126	44.6–48.2	55.1–57.2	112–114
Dieldrin[a]	69.0	126	150	115	197	183	244	143
	47.5–100	104–153	111–202	90.8–146	161–241	103–323	185–322	111–185
∑Mirex[a]	32.9	50.0	22.6	22.5	33.1	33.3	50.4	51.7
	24.0–44.8	42.4–59.0	14.7–34.6	18.3–27.6	25.5–42.9	17.9–61.1	34.5–73.5	41.7–64.0
α-HCHs[a]	38.9	63.5	91.3	46.7	32.7	34.9	48.9	65.7
	28.4–53.1	56.7–71.1	70.3–119	34.3–63.4	27.8–38.3	23.4–51.9	38.1–62.8	53.7–80.3
β-HCH[a]	249	307	542	238	137	202	141	113
	190–326	250–379	361–815	174–324	101–186	139–294	102–196	87–147
∑PBDEs[a]	5.8	8.8	7.0	6.8	14.0	27.1	38.6	78.4
	4.3–7.9	7.7–10.0	4.5–10.7	5.1–9.0	11.4–17.0	16.7–43.4	27.5–54.0	65.6–93.6
∑PFCAs[b]	420	445	387	321	433	243	362	657
	418–422	443–447	384–391	320–323	429–437	242–244	359–364	653–661
PFOS[b]	723	639	515	638	1007	494	793	1551
	717–729	634–643	512–518	633–643	1000–1014	489–498	788–798	1543–1559

[a] Blubber tissue.
[b] Liver tissue.

and ocean provide transport pathways to the CAA (Benskin et al., 2012a, 2012b).

Similar to the spatial ∑PBDEs trends in ringed seals, ∑PFCAs and PFOS were greater in more southern locations in the Eastern Canadian Arctic, Hudson Bay and Labrador (e.g., Inukjuak, Arviat, Qikiqtarjuaq, and Nain) than in ringed seals from other locations further north (Table 2, Fig. 4). These observations are consistent with previous findings which also showed higher levels of PFCs in ringed seals from these locations (Butt et al., 2008). Consistent with ∑PBDE trends, ringed seals from Gjoa Haven had relatively greater average ∑PFCAs and PFOS concentrations. Similarly, these observations may be a result of these seals feeding at a high trophic level and having a depleted RCS than due to predominant transport routes of PFASs to the Arctic.

4.2.2. Spatial trends of organohalogen contaminants in polar bears

PCBs are among the dominant POPs in arctic marine mammals and levels in polar bears are among the highest reported in any species (Letcher et al., 2010, 2017a, 2017b; McKinney et al., 2011a; Norstrom et al., 1998). ∑PCBs are the major legacy OC in polar bears from all locations across the Canadian Arctic, followed by ∑CHLs, ∑ClBz and ∑DDTs (Table 3). ∑PCBs, PCB153, ∑DDTs, and *p,p′*-DDE concentrations were generally greater in polar bears from the Eastern Canadian Arctic compared with the west, except for ∑PCBs and PCB153 concentrations in bears from the Beaufort Sea which also showed relatively high levels (Table 3; Fig. 5). These observations are consistent with previous studies on polar bears from Alaska to East Greenland and Svalbard which showed ∑PCBs and ∑DDTs to be negatively correlated with longitude and that level in Beaufort Sea populations were also elevated (Dietz et al., 2015; McKinney et al., 2011a; Verreault et al., 2005). Previous studies have hypothesized that the elevated levels in the Beaufort Sea polar bears could be due a greater reliance on ice-associated food webs than pelagic food webs or that the higher levels of these particle-associated OCs are related to the large input of suspended particulate matter from the Mackenzie River (McKinney et al., 2011a; Norstrom et al., 1998). Perhaps, the higher levels of these OCs in the Beaufort Sea polar bears are due to the consumption of ringed seals from the Beaufort Sea region, which also have elevated levels of these contaminants relative to other locations in the Canadian Arctic. Average ∑PCBs and ∑DDTs in polar bears sampled across the Canadian Arctic are lowest in bears from the Gulf of Boothia and Lancaster/Jones Sound area (Table 3; Fig. 5). Average ∑CHLs concentrations in polar bears showed a similar spatial trend to PCBs and DDTs and were greater in the east than the west (Table 3). These results however are inconsistent with a previous study which found ∑CHLs in polar bear subpopulations from Alaska to Svalbard to be spatially uniform (McKinney et al., 2011a). The varied interpretation between the present study and McKinney et al. (2011a) may be due to the inherent challenges in detecting regional differences when looking across a much broader spatial area. Unlike ∑PCBs and ∑DDTs, mean β-HCH was positively correlated with longitude due to higher levels in the west compared with the east (Table 3). As was the case for invertebrates, fish, and ringed seal elevated concentrations of β-HCH in the west likely reflect the greater use of β-HCH in Asia and atmospheric/oceanic transport to the Western Arctic. OCS, Dieldrin and Mirex levels were relatively low in polar bears (<250 ng/g lw) and were spatially uniform across the Arctic. The lack of spatial trend observed in these legacy OCPs may result from a lower historical loading of the Beaufort Sea through gas exchange, resulting in a more even distribution produced by historical atmospheric transport and deposition.

∑PBDEs, ΣPFCAs, and PFOS concentrations in polar bears vary across the Arctic, with the highest levels found in Hudson Bay, intermediate levels in the Eastern Canadian Arctic (Baffin Bay and David Strait) and low levels in the Gulf of Boothia, Lancaster/Jones Sound, and the Western Canadian Arctic (Table 3; Fig. 5). Similar to ringed seals, the

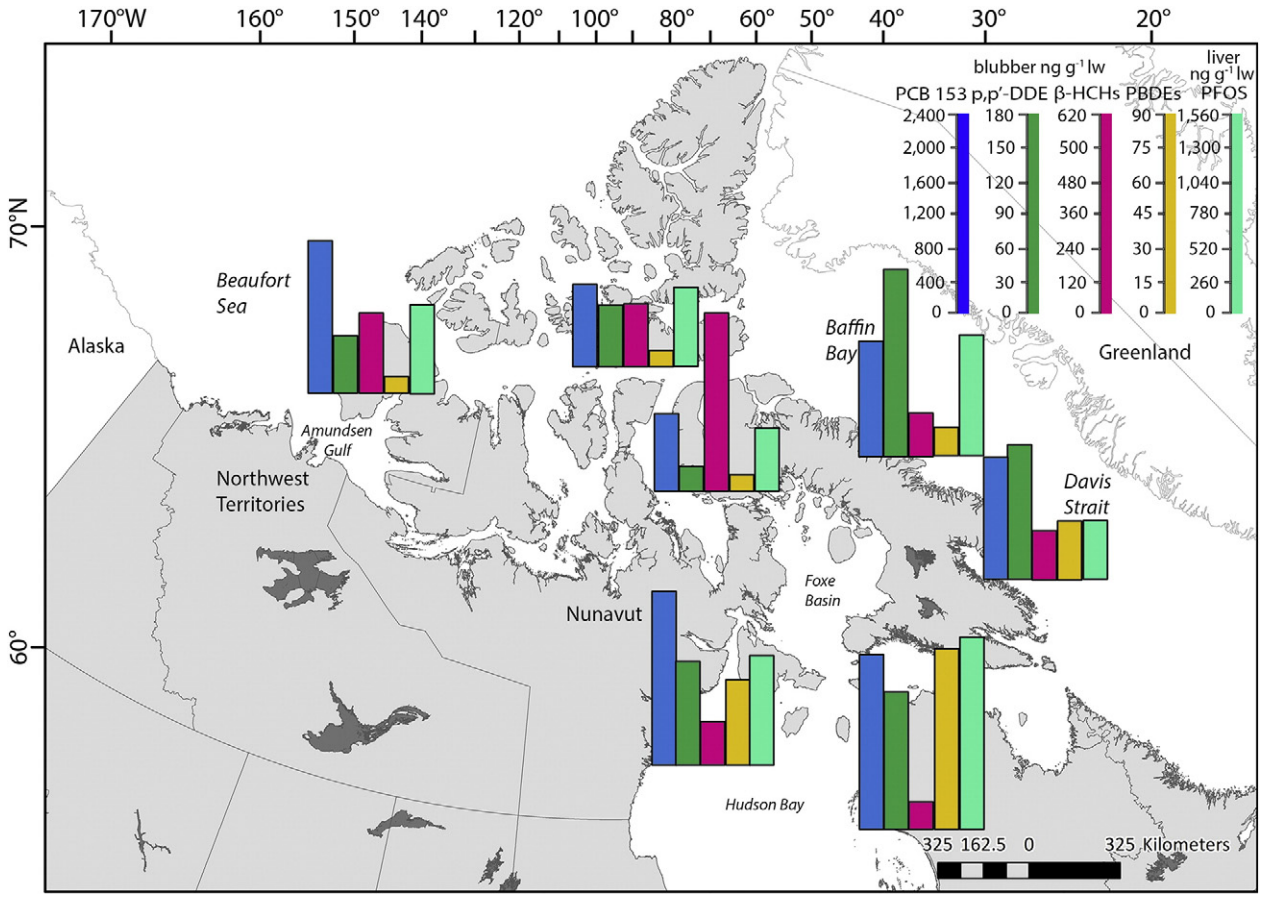

Fig. 5. Geometric means (ng/g lipid weight) of PCB 153, *p,p′*-DDE, β-HCHs, ∑PBDEs, and PFOS of polar bears collected from 2005 to 2008. Data from McKinney et al., 2011a,b and Letcher et al., 2010.

higher levels of these compounds observed in polar bears from southern latitudes (e.g., Hudson Bay) relative to higher latitude locations may reflect that these are newer POPs which have had a shorter time period to be transported to the Arctic and to work their way up through the Arctic food web.

4.2.3. Temporal trends of organohalogen contaminants in ringed seals and polar bears

Most of the legacy OCs appear to be declining in ringed seals and polar bears; however, there is strong evidence over the last decade of stalling declines in PCB and CHL levels (Letcher et al., 2017a,b), which may be related either to continued emissions from in-use materials and stored wastes despite the banning of these product in the 1970's (McKinney et al., 2011a) or the release of these compounds archived earlier in local or global environmental reservoirs (soils, vegetation, snow, ice) due to climate change (e.g., warming, permafrost melting, forest fires) and subsequent redistribution (Ma et al., 2016; Macdonald et al., 2005). ∑DDTs have declined significantly in ringed seals since 1998. Toxaphene concentrations in ringed seals have increased in Lancaster Sound and East Baffin over the period of 2008 to 2010. α-HCH has declined significantly in ringed seals in the east, whereas β-HCH increased in Resolute but not in East Baffin (Muir et al., 2013). Earlier studies reported that although global emissions of α- and β-HCH have declined since the early 1980's, there appears to be a lag in response time in ringed seal tissue concentrations (Addison et al., 2009), which is not surprising given the time required to load these compounds into polar seas and then release them through, for example, outflow through the CAA (Li et al., 2000; Pućko et al., 2013).

∑CHL and *p,p′*-DDE concentrations in polar bears have decreased from 1989 to 1996 and 1996 to 2002 and from 2005 to 2008 in most Canadian bear subpopulations. Higher PCB concentrations were reported in the period of 2005–2008 compared to 2001–2002 for all locations across the Arctic, except for Hudson Bay. Dieldrin levels show no overall declines. Overall PCB levels appear to have levelled off in the 2000s in most subpopulations, subsequent to the declines observed in the 1990s (Norstrom et al., 1998). Unfortunately, temporal comparisons of α-HCH and CBzs were confounded by erroneously low adipose levels in some of the bears and therefore were not reliable (McKinney et al., 2010). Longer-term trends (17-year period) in Hudson Bay show that ∑DDT decreased (11% per year), α-HCH decreased (12% per year), β-HCH increased (8.3% per year), and ∑PCBs and ∑CHL showed no distinct trends dating back to 1968.

∑PBDEs have generally increased from 1990s until early 2000s in ringed seals and/or polar bears and are now declining (2005–present). Ringed seals and polar bears achieved maximum ∑PBDEs concentrations during the period of 2000–2004, which were then followed by a decline from 2005 onwards which likely reflects the 2004 pentaBDE and octaBDE phase-outs (de Wit et al., 2010). This rapid response likely indicates that the upstream reservoir in the Beaufort Sea was not strongly loaded and therefore provided a source of low-concentration water to hasten the removal of chemicals deposited atmospherically into the CAA. Maximum levels of PFCs in ringed seal and polar bear livers were reached in the early 2000s in Hudson Bay and the Eastern Canadian Arctic and have continued to decline. As reported in Letcher et al. (2017a,b), in the liver and in samples from 2010 and including up to 2016, PFOS concentrations were consistently greater than for ΣPFCAs, and there were no obvious increasing or decreasing trends for ΣPFCAs and PFOS for both subpopulations of bears over the 2007–2014 period. In contrast, PFAA levels in the Beaufort Sea seals have continued to increase slowly to 2011 (Muir et al., 2013). The maximum

concentrations for PFOS in seals and polar bears achieved during the late 1990's and early 2000's was similar to the increased production of PFOSF-based products during the 1990s.

Modeling results suggest that the redistribution of more water soluble contaminants (e.g, PFOS and PFOA) from lower latitudes to the Arctic is ongoing and concentrations of these contaminants in the marine environment are expected to increase for the next 10 to 15 years (Stemmler and Lammel, 2010; Wania, 2007). As a result the distribution of more water soluble POPs in the Arctic will likely result in a more varied distribution due to oceanic transport into the Arctic. This pattern may therefore be distinct from long range atmospheric transport (LRAT) of these contaminants as well as others, e.g., PBDEs, which tend to result in more uniform deposition fluxes.

5. Mercury in the marine environment

Atmospheric, terrestrial, and oceanic pathways deliver Hg to Arctic marine waters, where they are taken up by algae and bacteria and transferred through the Arctic marine food web (Atwell et al., 1998; Campbell et al., 2005). Hg, mainly in the form of MMHg can bioaccumulate and biomagnify in marine food webs (Atwell et al., 1998). The biomagnification of Hg in marine food webs in the Canadian Arctic has been studied in the North Water Polynya (Campbell et al., 2005), Cumberland Sound (McMeans et al., 2015), Hudson Bay (Young et al., 2008), Queens Channel (Clayden et al., 2015) and in the Eastern Beaufort Sea and Amundsen Gulf (Lockhart et al., 2005; Loseto et al., 2008a, 2008b). In the Eastern Canadian Arctic the biomagnification factor (*C. hyperboreus* to seabird; 0.267) of MeHg in Queens Channel, located east of Bathurst Island was higher than in the North Water Polynya (bulk zooplankton to seal; 0.223). Further research is needed to determine whether the physical, chemical, or biological characteristics of these ecosystems are influencing Hg bioaccumulation or biomagnification.

5.1. Marine fish

Very few studies have measured Hg concentrations in marine fish species in the Eastern Canadian Arctic. Arctic char (*Salvelinus alpinus*) and Dolly Varden char (*S. malmo*) are members of the subfamily Salmonidae. Both char species inhabit freshwater throughout most of their lives with the anadromous (sea-run) forms migrating to sea in the summer for a few weeks of feeding before returning inland to spawn. Anadromous char typically have low Hg concentrations (<0.05 μg/g ww) in contrast to greater concentrations measured in freshwater fish species and marine mammals. Hg concentrations in anadromous char from the Eastern Canadian Arctic range from 0.04 ± 0.01 μg/g ww in Igloolik and Cape Dorset to 0.07 ± 0.03 μg/g ww in Pangnirtung, southern Baffin Island (Evans et al., 2015). Hg concentrations across the Arctic increased as a function of char size, decreasing condition factor and cooler springs (Evans et al., 2015). Temporal trends have shown that Hg concentrations in Cambridge Bay char have increased from 1977 to 2012. Trends of increasing Hg were also found at Pangnirtung (1990–2009) and Hall Beach west of Fox Basin (1978–2007). In contrast, Pond Inlet char have shown no trend over the time period of 2005 to 2012 and a decreasing trend was found at Iqaluit on southern Baffin Island (1992–2009) (Evans et al., 2015). Overall, temporal trends for Hg concentrations in sea-run char suggest a recent increase in levels across the Eastern Arctic.

Relatively high THg concentrations have been found in Cumberland Sound Arctic skate muscle (0.42 ± 0.18 μg/g ww) compared to Greenland halibut (*Reinhardtius hippoglossoides*) (0.17 ± 0.16), shorthorn sculpin (*Myoxocephalus scorpius*) (0.16 ± 0.07), Arctic char (0.04 ± 0.02), and capelin (*Mallotus villosus*) (0.02 ± 0.01) (McMeans et al., 2015). The reasons for the elevated levels in the Arctic skate remain unclear (McMeans et al., 2015). Greenland shark (*Somniosus microcephalus*) are the largest predatory fish in the Arctic and had the highest THg concentrations in muscle (1.62 ± 0.52 μg/g ww) (McMeans et al., 2015). These levels were within the range reported for beluga whale muscle and were consistent with their upper trophic level position in the marine food web.

5.2. Marine mammals

Elevated THg was first reported in marine mammals in the Canadian Arctic in the 1970s (Muir et al., 1992; Smith and Armstrong, 1975; Wagemann and Muir, 1984). Previous studies have observed that THg concentrations were greater in marine mammals from the Western Canadian Arctic compared with the Eastern Canadian Arctic (Muir et al., 1992; Rigét et al., 2005). These observations are likely due to a combination of local geological influences, atmospheric and riverine sources, dietary preferences, and methylation and biomagnification processes (Cardona-Marek et al., 2009; Gaden et al., 2009; Horton et al., 2009; Leitch et al., 2007; Loseto et al., 2008a; Loseto et al., 2008b; St. Louis et al., 2011).

5.2.1. Ringed seals

Studies have found that THg concentrations in ringed seals from the Eastern Canadian Arctic are two- to three-fold lower than ringed seals from the southern Beaufort Sea (Sachs Harbour and Ulukhaktok) (Muir et al., 1992; Wagemann and Muir, 1984). Braune et al. (2015) also reported greater THg concentrations in liver of adult ringed seals (≥5 years) in the Western Arctic (Sachs Harbour and Ulukhaktok) compared with other locations across the Canadian Arctic. Similar to liver, THg concentrations in muscle of adult seals were greater in the Western Arctic and in two sites in the Eastern Canadian Arctic (Resolute and Arviat). A recent investigation of THg in sub adult and adult ringed seals from across the Canadian Arctic confirmed previous reports of greater concentrations in muscle and liver tissue of seals from the Western Arctic and from Resolute and Arviat (Brown et al., 2016) (Table 4, Fig. 6). The higher levels observed at these locations in the east were attributed to seals feeding at a higher trophic level, whereas the higher levels in the west were attributed to both natural geological differences and to seals feeding higher in the food chain (Brown et al., 2016). The higher levels of THg in the Beaufort Sea seals may be due to active conversion of THg to MMHg in the Western Arctic Halocline where oxygen concentrations are low and organic matter is decomposing (Wang et al., 2012), or to the regional addition of Hg to Beaufort Sea surface waters from the Mackenzie River (e.g., Andersson et al., 2008; Stern and Macdonald, 2005).

Samples collected over time suggest that THg concentrations in ringed seals increased from the early-1970s to mid-1980s, and continued to increase to the mid- and late-1990s (AMAP, 2005; Wagemann et al., 1996). While present day THg levels in ringed seals exceed historical concentrations, no significant changes have been reported from 1999 to 2009 (Braune et al., 2015).

5.2.2. Polar bears

The polar bear has some of the highest THg concentrations in the Canadian Arctic (Rigét et al., 2011). Concentrations of THg in polar bear liver from the Western Arctic (northern and southern Beaufort Sea) were significantly greater than other areas across the Arctic, except for bears from the Gulf of Boothia and Lancaster Jones Sound which also had relatively higher levels compared with the rest of the Arctic (Routti et al., 2011) (Table 5, Fig. 6). These results are consistent with the spatial THg trends reported for ringed seal, which are the dominant prey of polar bears. A number of polynyas are within and/or adjacent to these two areas and have higher primary productivity relative to other areas in the Arctic (Hannah et al., 2009). Lancaster Sound in particular is one of the Arctic's most biologically productive marine areas and is characterised by two polynyas, one is along the northern coast of Lancaster Sound and the other at the Eastern outflow (Barber and Massom, 2007). A consequence of increased productivity could be

Table 4
Geometric means and 95% confidence intervals (μg/g wet weight) of total Hg in muscle and liver of sub-adult and adult male and female ringed seals collected from 2007 to 2011. Data from Brown et al., 2016.

	THg muscle μg/g ww				THg liver μg/g ww	
	n	Sub adult	*n*	Adult	Sub adult	Adult
Sachs Harbour	31	0.411 0.409-0.414	9	0.890 0.878-0.902	2.26 2.22-2.29	34.6 33.2-36.1
Ulukhaktok	4	0.387 0.385-0.389	16	0.543 0.541-0.545	11.7 11.6-11.9	20.1 19.9-20.3
Arctic Bay	9	0.190 0.188-0.191	9	0.377 0.373-0.381	1.08 1.05-1.10	6.39 6.27-6.50
Gjoa Haven	12	0.171 0.135-0.206	9	0.325 0.317-0.332	1.52 1.48-1.56	5.40 5.23-5.56
Grise Fiord	13	0.102 0.100-0.104	7	0.093 0.092-0.094	1.35 1.32-1.38	16.7 16.5-16.9
Resolute	61	0.380 0.378-0.382	33	0.543 0.541-0.544	4.43 4.40-4.45	11.3 11.2-11.5
Pond Inlet	12	0.180 0.179-0.182	4	0.487 0.480-0.494	2.13 2.06-2.20	8.07 7.89-8.26
Pangnirtung	14	0.235 0.234-0.237	0	n.a.	4.23 4.17-4.29	n.a.
Arviat	58	0.245 0243-0.246	66	0.270 0.261-0.273	4.76 4.71-4.81	16.2 16.1-16.3
Inukjuaq	13	0.102 0.098-0.107	5	0.139 0.137-0.141	0.99 0.98-1.01	4.05 3.92-4.17
Nachvak Fjord	6	0.141 0.136-0.146	19	0.150 0.146-0.153	n.a.	n.a.
Saglek Fjord	16	0.136 0.127-0.145	12	0.187 0.186-0.188	n.a.	n.a.
Okak Bay	14	0.125 0.124-0.126	14	0.262 0.260-0.264	n.a.	n.a.
Anaktalak Bay	7	0.096 0.095-0.097	4	0.210 0.208-0.212	n.a.	n.a.

enhanced methylation of inorganic Hg during regeneration of the labile organic matter as it sinks below the surface. Western and Southern Hudson Bay bears had some of the lowest average Hg levels in the Canadian Arctic, followed by bears from Davis Strait (Table 5, Fig. 6).

THg concentrations measured in 2007–2008 in polar bears from the Eastern Canadian Arctic were similar to the concentrations reported in the 1980s (Routti et al., 2011). Similar temporal observations have also been observed in ringed seals from several locations in the Canadian Arctic (Gaden et al., 2009). In a very recent report, THg concentrations in the liver collected in 2016 from bears from Western and Southern Hudson Bay were retrospectively comparable to concentrations in samples from years going back before 2000, i.e. 5 to 25 μg/g (wet weight). Similar to previous years, in 2016 liver samples THg concentrations in Western Hudson Bay bears were slightly greater than for the southern Hudson Bay bears. However, the THg concentrations were variable from year to year, and so there is a need to continue annual monitoring of THg in Hudson Bay bears.

6. Summary and outlook

There appear to be two dominant factors that contribute to spatial trends in Hg and legacy OHCs observed in the CAA. First, the contaminant concentrations in the water column are strongly affected by deposition and transport of contaminants by air and water, and by the historical loading of upstream oceanic reservoirs in the Western Arctic Ocean. Second, the concentrations of contaminants in higher trophic levels (e.g., bears, seals) are strongly mediated by local or regional food web structure.

For the first factor, the clearest example is the effect of HCHs loading into the upstream ocean reservoir in the Beaufort Sea leading to a significant west to east decreasing concentration across the CAA. However, direct atmospheric deposition and river input are also important in the CAA, especially for contaminants more recently released like the PBDEs and PFOS. These compounds presently exhibit higher concentrations in the south indicating that northward spread from more highly contaminated locations in temperate regions of the Northern Hemisphere is important. The spatial and temporal patterns of historical usage are also important, with Europe and North America having released PCBs and other compounds in greater amounts and earlier than Asia, and Asian sources favouring compounds like HCHs or telomeric (linear) PFOS. Hg is also strongly affected by sources, with coal burning being presently among the most important. During the past two decades, controls have reduced Hg emissions in Europe and North America whereas continued dependency on coal has resulted in Asia becoming the most important source (AMAP, 2011). As was the case for the HCHs, the Western Arctic Ocean reservoir is likely to become more heavily loaded with Hg from the Asian sources whereas the Atlantic Ocean contaminant Hg burden will decline. Although contaminant inorganic Hg is important, toxic risk and biological accumulation depend crucially on methylation within aquatic systems. This step, which accompanies organic decomposition, is controlled by regional factors including dissolved oxygen concentration.

Food web structure plays an important role in shaping spatial trends of MMHg and some legacy contaminants, including $\sum$PCBs, PCB 153, $\sum$DDTs, *p,p′*-DDE: Concentrations of these contaminants in ringed seals and polar bears from the Beaufort Sea are elevated relative to other locations across the central and eastern Canadian Arctic, which for ringed seals is consistent with differences in trophic position (Brown et al., 2016). The consumption of larger, older cod by ringed seals was thought to be associated with elevated Hg levels during poor ice years in the Western Arctic relative to other years (Gaden et al., 2009). The observation that ringed seals from the Amundsen Gulf feed more benthically, relative to other locations in the Arctic (Whitehouse et al., 2014) points to inter-regional differences in feeding ecology of the ringed seal that cannot be ignored when interpreting contaminants trends. Lastly, since ringed seals in the west were sampled at a relatively high latitude compared to a number of sites in the east, the elevated contaminant levels could also be partly attributed to the consumption of more forage fish (e.g., Arctic cod), than sub-adult and adult seals at lower latitudes which have been reported to consume

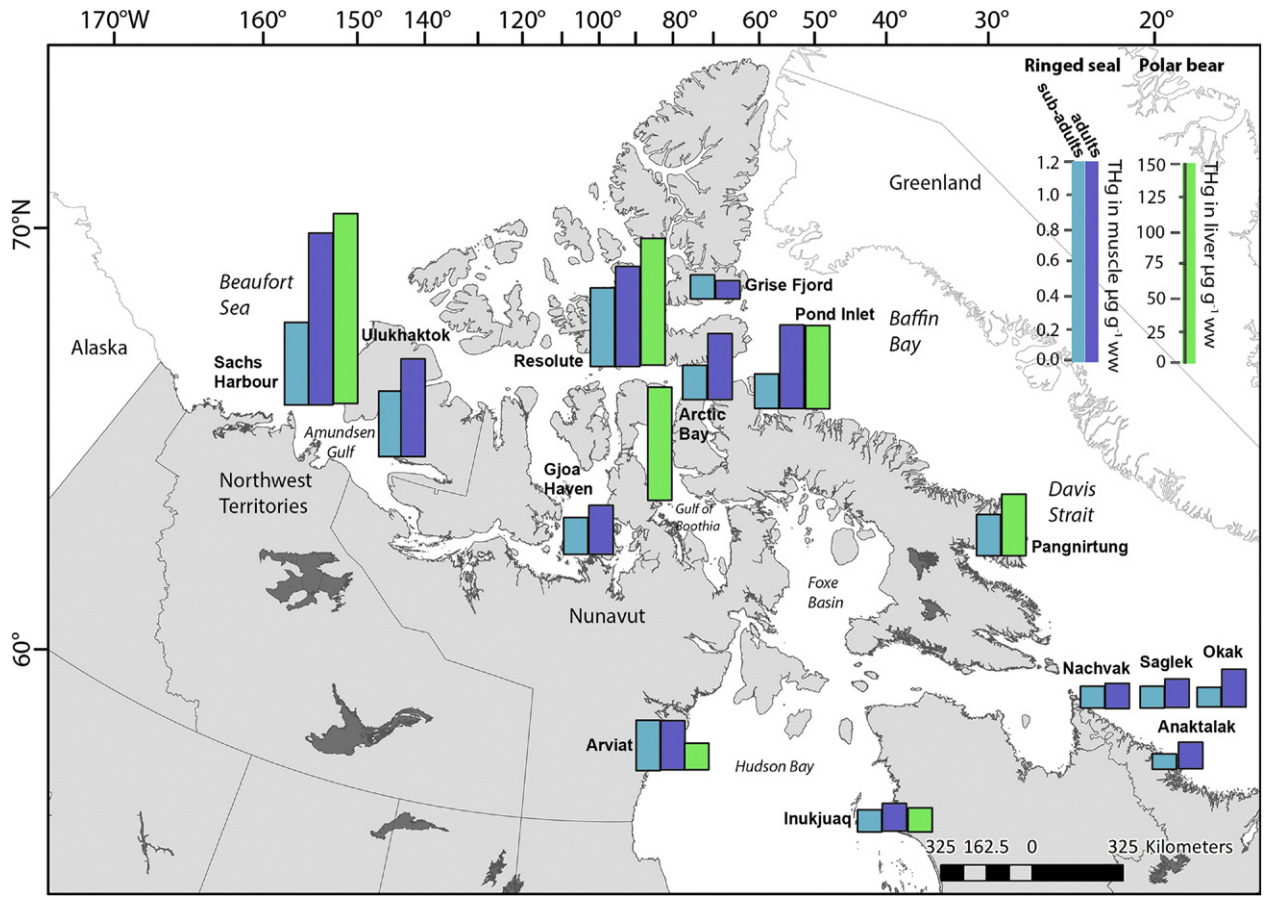

Fig. 6. Geometric means (µg/g wet weight) of THg in sub-adult and adult male and female ringed seals collected from 2007 and 2011 and polar bears collected from 2005 to 2008. Data from Brown et al., 2016 and Routti et al., 2011.

more sub-Arctic fish species and pelagic zooplankton (Yurkowski et al., 2015). The higher relative concentrations of these contaminants in polar bears in the Western Arctic may also reflect differences in contaminants in their primary prey, whereby the consumption of more contaminated western ringed seals compared to seals in the east. For more recent contaminants like PBDEs and PFOS, the rapid atmospheric delivery northward will contaminate the more southern locations in the near-term, but these emerging contaminants will become more evenly and widely distributed once primary sources are controlled.

Like the rest of the Arctic, the CAA is undergoing climate change associated with increasing temperatures. Increased warming of 4 to 8 °C by 2050 has been forecasted in fall and winter seasons for the Eastern Canadian Arctic (Brown et al., 2017). The current trend of decreased summer sea ice extent and earlier sea ice breakup in the Arctic Ocean is projected to continue (Brown et al. 2017). Within the CAA itself, summer sea ice extent has been declining, temperatures have been rising, and precipitation has been increasing (Fig. 7). These changes will likely enhance air-sea exchange, wet deposition, and transfers from land to rivers. Additionally, change in sea-ice cover strongly affects food web structure, and seasonal timing of primary production is expected to change (Tremblay et al., 2017), ultimately resulting in an early spring in the CAA. Although continued warming, further loss of sea ice and greater precipitation have been projected by a number of authors, only a few studies have shown the effects of such changes on exposure to pollutants within arctic ecosystems. For example, McKinney et al. (2009) found that changes in feeding ecology of polar bears from Western Hudson Bay, which was largely caused by earlier sea ice breakup, resulted in increased tissue concentrations of several chlorinated and brominated contaminants. Using food web tracer patterns (e.g., quantitative fatty acid signature analysis and fatty acid carbon isotope) McKinney et al. (2013) reported a shift in diet structure for East Greenland polar bears over 28-years which showed a decrease in ringed seal consumption and an increase in hooded seal consumption. The authors concluded that such a shift could result in slower temporal declines of legacy POP contaminants since subarctic seals have higher contaminant burdens of these pollutants than arctic seals. Gaden et al. (2009) found that both long and short ice-free seasons resulted in increased THg concentrations in ringed seals from the Beaufort Sea. The increased THg levels were attributed to changes in cohort composition of the Arctic cod population, which is the dominant prey of ringed seals. In shorter ice-free seasons a large percentage of young, less contaminated Arctic cod may not survive, leaving older more highly contaminated Arctic cod as the dominant prey available for predators (Gaden et al., 2009). Brown et al. (2014) reported a shift to feeding at

Table 5
Geometric means and 95% confidence intervals (µg/g wet weight) of total Hg in liver of sub-adult and adult male and female polar bears collected from 2005 to 2008. Data from Routti et al., 2011.

	n	THg liver µg/g ww
South Beaufort Sea	11	137 (135–138)
North Beaufort Sea	26	153 (152–155)
Gulf of Boothia	6	85.7 (85.1–86.3)
Lancaster/Jones Sound	13	94.1 (93.4–94.9)
Baffin Bay	14	59.2 (58.3–60.2)
Davis Strait	6	46.1 (45.4–46.7)
West Hudson Bay	11	19.5 (19.2–19.7)
South Hudson Bay	14	18.2 (18.1–18.4)

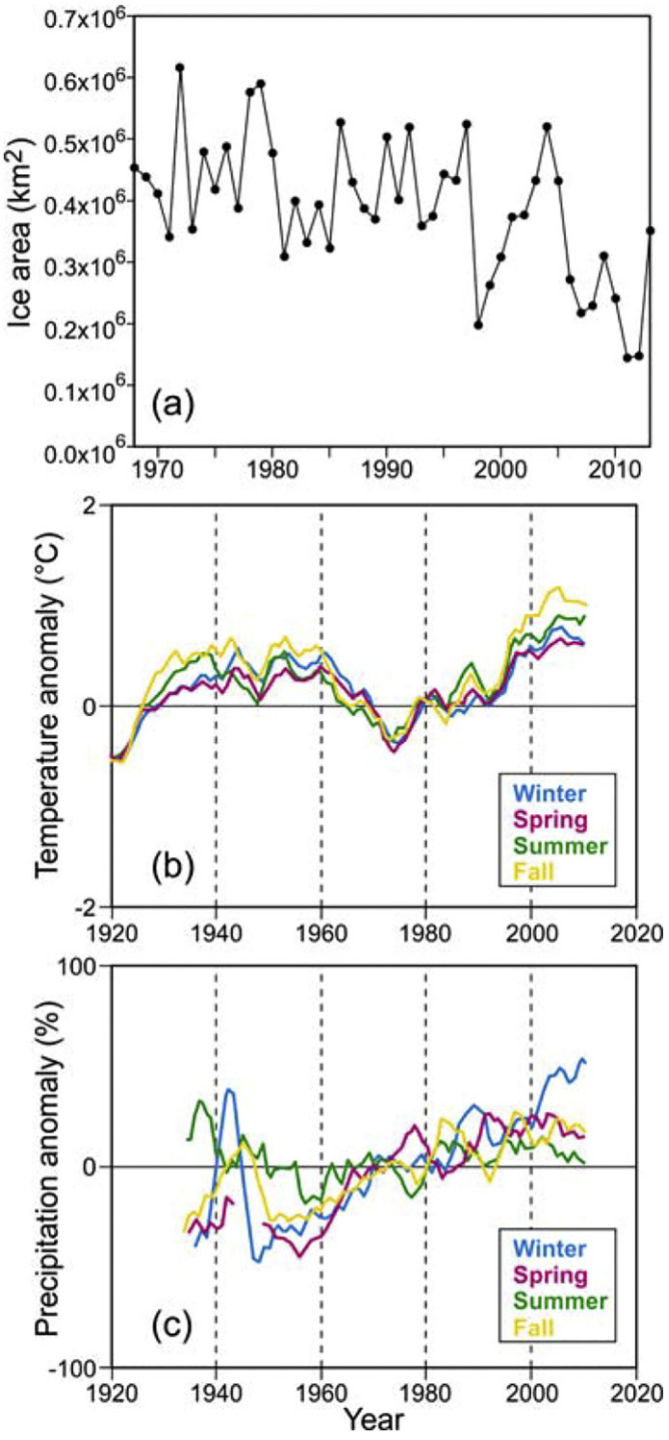

Fig. 7. Recent trends in a) summer sea-ice extent, b) temperature anomaly and c) precipitation anomaly for the CAA.
Modified from Howell et al., 2015, Steiner et al., 2015.

a higher trophic level in Labrador ringed seals sampled in 2010, which corresponded to a poor ice year along the coast (Colbourne et al., 2011). This shift in diet appears to have resulted in increased THg concentrations in these seals (Brown et al., 2017).

Recent studies suggest that onshore habitat use for some subpopulations of polar bears (e.g., Southern Beaufort Sea) is increasing with annual reduced sea ice duration (Atwood et al., 2016) and that bears annual dietary proportions are shifting (McKinney et al., 2017). For example, during poor ice years the diet of polar bears from the Southern Beaufort Sea shows a reduced consumption of the omnivorous ringed seal, bearded seal, and beluga whale and an increased consumption of the planktivorous-feeding bowhead whale. The bowhead whale occupies a lower trophic feeding position and thus is generally less contaminated compared with ringed seal and beluga whale, which feed at higher trophic levels (Hoekstra et al., 2003). The increased consumption of harvested and beach-cast bowhead whales on land over free-ranging ringed seal or beluga whale has the potential to therefore alter contaminant exposure in polar bears.

Yurkowski et al. (2015) reported interannual variation in ringed seal diet and an increased adult isotopic niche size over time, which suggests these seals have adaptability with respect to their diet and to a changing climate. The results also suggested that high-Arctic ringed seals may be at increased risk, due to their more restricted diet, than lower-Arctic populations from ecosystem perturbations, such as climate change. How these changes will affect contaminant concentrations and trends in ringed seals across the Arctic is yet to be determined.

Another possible change for individual ringed seals and polar bears is a variation in body condition which could alter contaminant burdens in both species. For example, reduced body condition may result in increased lipid and/or protein catabolism, which may subsequently mobilize and release contaminants into circulation (Polischuk et al., 2002), which can increase health risks to the affected individuals. Fasting periods and/or decreased body mass has been found to result in increased circulating concentrations of lipophilic POPs (Christiansen et al., 2007; Helgason et al., 2013; Polischuk et al., 2002) and THg (Seewagen et al., 2016). Circulating POPs can induce xenobiotic metabolizing enzymes in the liver (Wolkers et al., 2008), which lead to the toxic form of hydroxyl (OH) metabolites of POPs (Letcher et al., 2000). For example, OH-PCBs and OH-PBDE metabolites/contaminants have been reported mainly in the liver and/or plasma of East Greenland ringed seals and/or polar bears, but not in the adipose tissue (Gebbink et al., 2008; Letcher et al., 2009). Therefore, the health risks to individual animals or populations with reduced body condition and/or periods of fasting may be increased due to the mobilization of hydroxylated POPs.

The loss of summer sea ice is likely to favour pelagic primary production which would extend deeper into the water column due to greater penetration by light. Regeneration of this organic matter may enhance Hg methylation below the euphotic zone, but this may be offset by photodecomposition of MMHg in surface water (e.g., see Point et al., 2011). However, greater amounts of freshwater and sea-ice melt will lead to greater summer stratification, which may oppose the increase in productivity that should occur under reduced-ice conditions (Tremblay et al., 2017). This poorly-buffered freshwater is more vulnerable to ocean acidification, which may dramatically affect lower trophic food-web structure (AMAP, 2013). The concentration of MMHg in the water column is maintained as a balance between dynamic (i.e., rapid) processes of production and degradation. While it seems clear that projected changes in climate will lead to increases in the rates of both of these processes, it is exceptionally difficult to project how the balance point will be affected, regionally or locally.

Expansion of species distribution and invasion of southern species further north due to a warming climate has the potential to alter food web structure and subsequently contaminant accumulation in marine mammals across the Canadian Arctic. For example, a poor ice year can influence the abundance and/or availability of key prey species for marine mammals which may force them to feed on alternate prey that may be more or less contaminated than their preferred prey.

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