A review of ecological impacts of global climate change on persistent organic pollutant and mercury pathways and exposures in arctic marine ecosystems

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Abstract   Bioaccumulative and biomagnifying contaminants, such as persistent organic pollutants (POPs) and mercury (Hg), have for decades been recognized as a health concern in arctic marine biota. In recent years, global climate change (GCC) and related loss of arctic sea ice have been observed to be driving substantial change in arctic ecosystems. This review summarizes findings documenting empirical links between GCC-induced ecological changes and alterations in POP and Hg exposures and pathways in arctic marine ecosystems. Most of the studies have reported changes in POP or Hg concentrations in tissue in relation to GCC-induced changes in species trophic interactions. These studies have typically focused on the role of changes in abundance, habitat range or accessibility of prey species, particularly in relation to sea ice changes. Yet, the ecological change that resulted in contaminant trend changes has often been unclear or assumed. Other studies have successfully used chemical tracers, such as stable nitrogen and carbon isotope ratios and fatty acid signatures to link such ecological changes to contaminant level variations or trends. Lower sea ice linked-diet changes/variation were associated with higher contaminant levels in some populations of polar bears, ringed seals, and thick-billed murres, but the influence of changing trophic interactions on POP levels and trends varied widely in both magnitude and direction. We suggest that future research in this new area of GCC-linked ecotoxicology should focus on routine analysis of ancillary ecological metrics with POP and Hg studies, simultaneous consideration of the multiple mechanisms by which GCC and contaminant interactions can occur, and targeted research on changing exposures and toxicological effects in species known to be sensitive to both GCC and contaminants [*Current Zoology* 61 (4): 617–628, 2015].

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1   Introduction

Exposure to bioaccumulative contaminants has been recognized as a health concern in arctic marine fish and wildlife for several decades (Dietz et al., 2013b; Letcher et al., 2010). More recently, global climate change (GCC) and the subsequent loss of vast amounts of arctic sea ice have led to substantial changes in arctic ecosystems (Post et al., 2009; Post et al., 2013; Wassmann et al., 2011). Given the extreme difficulty of effectively mitigating and managing GCC, research on the interaction between climate-linked ecological change and other dynamic anthropogenic stressors, such as contaminant exposures, is paramount for developing adaptation and conservation measures (Parmesan et al., 2011). Contaminants, such as persistent organic pollutants (POPs) and mercury (Hg), bioaccumulate and biomagnify through food webs, and are thus subject to altered pathways and exposures as a function of changing ecosystems (Jenssen et al., 2015; Macdonald et al., 2003). The Arctic is an excellent region for research of this nature, given that GCC-linked abiotic and ecological changes

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in the Arctic surpass what has been reported in other regions and may herald future changes to take place in temperate latitudes (Post et al., 2009).

The Arctic is warming at more than twice the global average (Serreze and Barry, 2011). This phenomenon is known as arctic amplification and is thought to be largely driven by sea ice loss (Pistone et al., 2014; Screen and Simmonds, 2010). Indeed, the decline in arctic sea ice over the past three decades is considered to be one of the most impactful indicators of GCC that has so-far occurred (IPCC, 2013). Arctic sea ice is a key habitat for many arctic marine species, and declines in its extent and thickness, later freeze up and earlier break-up, in addition to warmer temperatures, less snow, and altered nutrient availability, have all led to major disturbances in arctic marine ecosystems (Kovacs et al., 2011; Post et al., 2009; Post et al., 2013; Wassmann et al., 2011).

Within these ecosystems, nonetheless, the number of studies are remarkable limited in showing significant biological changes that can be ascribed to GCC. Wassmann et al. (2011) found just 51 studies on arctic marine biota that met the Intergovernmental Panel on Climate Change (IPCC) criteria for effects in biota. The criteria specify that the study (1) finished in 1990 or later, (2) covered a 20 year period or more, and (3) demonstrated a significant biological change in either direction. Broadly speaking, patterns of northward range expansion and/or increased seasonal presence have been observed for subarctic and even temperate species, such as subarctic capelin *Mallotus villosus*, harp seals *Pagophilus groenlandicus* and ice-avoiding killer whales *Orcinus orca* (Ferguson et al., 2010; Higdon and Ferguson, 2009; Kovacs et al., 2011). In contrast, arctic marine species have less capability to shift their ranges further north (Kovacs et al., 2011). In certain regions, arctic species including ringed seals *Pusa hispida*, polar bears *Ursus maritimus*, Pacific walruses *Odobenus rosmarus divergens* and/or various seabirds have shown changes in habitat use, condition, reproductive success, abundance and/or feeding ecology attributed to sea ice change (Jay and Fischbach, 2008; McKinney et al., 2009; McKinney et al., 2013; Provencher et al., 2012; Regehr et al., 2010; Stirling et al., 1999; Stirling, 2005; Stirling and Parkinson, 2006). Changes in the lower food web, including the amount and timing of primary production and zooplankton and benthic invertebrate species composition have also been shown concurrent with sea ice change (Post et al., 2013; Wassmann et al., 2011).

Extensive and on-going studies of POP and Hg exposures in arctic marine biota and food webs have been reported over the past three decades (Dietz et al., 2013b; Letcher et al., 2010). In general, POPs are persistent, lipophilic, bioaccumulative, toxic, and subject to long-range transport, resulting in contamination of even remote environments such as the Arctic (Letcher et al., 2010). A number of halogenated organic chemicals are categorized as POPs and regulated under the internationally binding Stockholm Convention, including organochlorine pesticides like dichlorodiphenyltrichloroethane (DDT), hexachlorocyclohexanes (HCHs) and endosulfan and industrial chemicals and by-products like polychlorinated biphenyls (PCBs) and certain polybrominated diphenyl ethers (PBDEs) (chm.pops.int). The levels of many older and internationally regulated "legacy" POPs, like DDT and PCBs, have declined in arctic marine species due to historic regulations (Dietz et al., 2013a; Rigét et al., 2010a). Yet, POP trends in some populations recently appear to have leveled off and at concentrations that continue to be above thresholds for toxic effects in laboratory animals (Dietz et al., 2013a; Letcher et al., 2010; Rigét et al., 2010a). In addition, the list of chemicals found in arctic biota continues to grow, and increasing temporal trends have been reported for some current-use chemicals (e.g., NCP, 2013).

Although Hg is a naturally occurring element, emissions from anthropogenic activity have led to elevated environmental Hg levels, including some currently increasing trends particularly in western arctic marine biota (Braune et al., 2015a; Rigét et al., 2011). Mercury accumulates in biota in its lipophilic monomethyl form, MeHg, which is a particular concern due to its potency as a neurotoxin (Basu et al., 2009; Dietz et al., 2013b; Scheuhammer et al., 2015). An international treaty on Hg, the Minamata Convention on Mercury, was recently agreed upon and is pending ratification (mercuryconvention.org).

A basic toxicological principle states that the biological risk posed by contaminants is a function of not only their inherent toxicity, but also the levels to which biota are exposed (i.e., the dose). Levels of exposure to POPs and Hg in arctic ecosystems are driven by physical and chemical factors that result in them reaching remote arctic regions and chemical and ecological factors that result in them concentrating within biota (bioaccumulating) and through food webs (biomagnifying) (Macdonald et al., 2000). It was hypothesized over a decade ago with respect to GCC-contaminant interactions that, although physical and chemical changes are easier to model and understand, ecological changes have a higher

potential to alter contaminant pathways and exposures (Macdonald et al., 2003). At that time, however, no empirical evidence of such relationships existed. Since then, this new research area in ecotoxicology has transformed from prediction and speculation to documented empirical evidence.

The objective of this review is to summarize recent reports of climate-associated ecological changes that have altered POP and Hg pathways and exposures in arctic marine biota. Studies based on models, predictions, speculation or lab experiments of changes that *could* occur are not covered here. Nonetheless, we would be reviewing a small number of studies indeed, if we only focused on studies documenting these linkages that met the three IPCC criteria for GCC impacts in biota. Thus, we relax the second and third criteria to also include studies that cover periods shorter than twenty years and not just those that show an increasing or decreasing trend, but also those that report variation in contaminant pathways and exposures and possible links to climate-ecological variation. The documented ecological change or variation may be correlatively linked or simply suggested by the study's authors to be related to climate change, with other explanations possible and/or proposed (as per Wassmann et al., 2011). In addition, climate metrics, such as sea ice extent, the North Atlantic oscillation (NAO) and Arctic oscillation (AO), include those reported to have varying degrees of support for associations with anthropogenic climate change and natural oscillations in climate may play a role. That being said, it is *extremely likely* that anthropogenic forcing is dominant cause of global temperature increases since the 1950s and *very likely* that anthropogenic forcing has contributed to arctic sea ice loss since 1979 (IPCC, 2013). Thus, arctic marine ecosystems are highly relevant for examining GCC-ecological-contaminant linkages, including through examination of sea ice changes.

2 GCC-Ecological Change-Contaminant Linkages in Arctic Marine Ecosystems

Over the past decade, a limited number of studies have been published examining GCC-ecological variation/change-POP/Hg linkages in arctic marine ecosystems. Studies have been carried out in the Canadian Arctic, Greenland, and Norway (mainly Svalbard), but not in the US (Alaska) or the Russian Arctic (Fig. 1). These reports have largely focused on upper trophic level spe-

cies, such as polar bear, ringed seal, beluga whale *Delphinapterus leucas* and certain seabirds. It is worth noting that perhaps an even smaller number of similar studies have been reported in other arctic ecosystems, such as arctic lakes (Outridge et al., 2007; Rigét et al., 2010b) and rivers (Carrie et al., 2010). Many of the arctic marine studies reviewed here have used chemical tracers, such as stable isotope ratios of nitrogen (δ^{15}N) and carbon (δ^{13}C) or fatty acid profiles, as indicators of diet or food web changes/variation. As δ^{15}N is predictably enriched through food webs, it has been used to infer trophic position and to quantify POP and Hg biomagnification through food webs (Campbell et al., 2005; Fisk et al., 2001) As δ^{13}C enrichment is minimal through food webs, it is used to infer carbon sources, such as benthic versus pelagic, or marine versus terrestrial, and the relationship of habitat or feeding location to POP and Hg bioaccumulation (Choy et al., 2009). Fatty acids consumed by monogastric species are deposited in storage tissues in similar proportions as in their diets, and thus predator fatty acid profiles can be used to infer dietary composition (Budge et al., 2006). Such chemical tracer profiles characterized in arctic marine organisms have been linked to POP and Hg level changes or variation and/or metrics of climate. Other studies have examined relationships between climate metrics and contaminant change or variation directly, but implicated ecological change as the mechanism.

2.1 Polar bears and arctic foxes

McKinney et al. (2009) first reported on climate-related feeding changes in western Hudson Bay polar bears, which altered contaminant time trends (Table 1). Lower δ^{13}C values and changes in fatty acid profiles in years of earlier summer sea ice break-up were suggested to be related to higher dietary proportions of subarctic seal species, namely harbor seals *Phoca vitulina* and harp seals, relative to arctic seals, namely bearded seals *Erignathus barbatus*. However, tracer profiles did not suggest a change in ringed seals, their main prey (Thiemann et al., 2008). By comparing actual trends to those adjusted for the influence of dietary tracers, the authors showed that this shift in diet generally resulted in slower rates of decrease of legacy POPs and faster rates of increase of newer POPs.

A similar but longer term and more detailed study of East Greenland polar bears showed a substantial diet shift over the past thirty years from one of mainly arctic-type ringed seals to mainly subarctic-type harp and hooded seals *Cystophora cristata* using a statistical approach known as quantitative fatty acid signature analy-

sis (McKinney et al., 2013). This study showed that higher consumption of subarctic seals occurred in years of warmer temperatures and lower ice extent, as demonstrated by correlations between prey consumption and the annual NAO index. This suggested that climate changes may influence the abundance, distribution

Fig. 1 Map of the circumpolar Arctic showing locations of studies reporting documented climate-ecological-POP/Hg linkages, with symbols designating polar bear/arctic fox, marine mammal, seabird or food web study

Green symbols indicate studies on POPs. Red symbols indicate studies on Hg. Numbers indicate a the cited study: (1) McKinney et al., 2009 (2) McKinney et al., 2013 (3) Andersen et al., 2015 (4) Gaden et al., 2009 (5) Gaden et al., 2012 (6) Gaden and Stern 2010 (7) Rigét et al., 2012 (8) Rigét et al., 2013 (9) Loseto et al., 2015 (10) Bustnes et al., 2010 (11) Bustnes et al., 2012 (12) Øverjordet et al., 2015 (13) Braune et al., 2014 (14) Braune et al., 2015b (15) Hallanger et al., 2011 (16) McKinney et al., 2012.

Table 1 Summary of studies on polar bears and arctic foxes examining GCC-ecological-POP/Hg linkages

Study species, year and location	Climate metric	Ecological change/variation	Influence on POPs or Hg		Reference
			Compound	POP/Hg variation/ change	
Polar bear, 1991–2007, western Hudson Bay	Sea ice break-up date	Diet (sub-arctic versus arctic seals)	α-HCH	↔	McKinney et al., 2009
			ΣPCB, ΣCHL	↑ instead of ↓ trend	
			ΣPBDE, β-HCH	Faster rate of ↑ trend	
			ΣDDT	Faster rate of ↓ trend	
Polar bear, 1984–2011, East Greenland	NAO	Diet (sub-arctic versus arctic seals)	ΣPCB, OCS, *t*-nonachlor, oxychlordane, CB170/190, CB180, CB153	Not significantly slower rate of ↓ trend	McKinney et al., 2013
			HCB, ΣDDT, *p,p′*-DDE	↔	
			BDE153, ΣPBDE	Non-significantly faster rate of ↑ trend	
Arctic fox, 1997–2013, Svalbard	Sea ice extent	Diet (marine versus terrestrial)	ΣPCB, ΣCHL, mirex, *p,p′*-DDE, ΣPBDE	↔	Andersen et al., 2015
			HCB	↓ in years with ↑ reindeer mortality (terrestrial diet)	
			β-HCH	↑ in years with ↑ sea ice cover (marine diet)	

↑= Increase. ↓ = Decrease. ↔ no influence of ecological variation or change. NAO = North Atlantic oscillation. HCH = hexachlorocyclohexane. HCB = hexachlorobenzene. CHL = chlordane. PBDE = polybrominated diphenyl ether. DDT = dichlorodiphenyltrichoroethane. *p,p′*-DDE = *para,para′*-dichlorodiphenyldichloroethylene. PCB = polychlorinated biphenyl. OCS = octachlorostyrene.

and/or accessibility of these seal species, and thus alter the composition of the polar bear diet. Legacy POP declines were generally more rapid than in western Hudson Bay polar bears, and adjusting for the proportion of ringed seal in the East Greenland polar bear diet showed that these rates of POP declines were not strongly influenced by the diet shift to date.

Andersen et al. (2015) examined POP trends in Svalbard arctic foxes in relation to sea ice cover, reindeer *Rangifer tarandus platyrhynchus* mortality and marine/terrestrial diets (as indicated by $\delta^{13}C$). There was a decreasing temporal trend in concentrations of all POPs after adjusting for sex, age, body condition, diet and food availability. In general, $\delta^{13}C$ was an important variable, suggesting that higher POP levels were associated with higher feeding on marine prey. Significantly lower levels of HCB were associated with higher reindeer mortality, indicating lower HCB intake when foxes fed more on terrestrial prey. An association between higher levels of β-HCH with greater sea ice-cover also pointed to higher β-HCH contamination in marine prey. Thus, GCC-linked reduced accessibility to marine prey and higher reindeer mortality could result in lower levels of some POPs in arctic fox through an increasingly terrestrial diet. Yet, these authors suggested a more complicated picture, including the possible role of feeding on seabirds. As well, the authors did not compare actual trends with those adjusted for the influence of diet.

2.2   Marine mammals

Total Hg (THg) trends in ringed seals were investigated for associations with the length of ice-free season in the western Canadian Arctic (community of Ulukhaktok, NWT) (Gaden et al., 2009) (Table 2). Although there was no temporal trend in Hg levels, years following both long and short ice-free seasons were characterized by higher THg levels in the ringed seals. No relationship between $\delta^{13}C$ or $\delta^{15}N$ with ice-free season was found, however, possibly due to differences in turnover rates between THg and stable isotopes. The authors suggested that environmental conditions in the previous ice-free season influenced ringed seal prey, particularly the abundance and age classes of arctic cod *Boreogadus saida*, and consequently THg exposures.

In the same region, Gaden et al. (2012) also reported associations between ringed seal POP trends and sea ice break-up date. The levels of *p,p'*-DDE and several PCB congeners, but not other legacy POPs, were higher in years of earlier sea ice break-up. Although no data on ecological change in relation to sea ice change were presented, the authors suggested that early ice break-up

influenced ringed seal prey availability type and/or quantity of prey consumed, resulting in higher blubber levels of certain POPs. Interestingly, ringed seals in this region have shown clear declines in body condition over the past two decades, as indicated by changes in blubber thickness (Harwood et al., 2012). This body condition decline could also be a GCC-linked ecological change. Indeed, declines in blubber thickness were inversely correlated with the levels of many of the monitored legacy POPs, and reported concentrations were adjusted for blubber thickness (Gaden et al., 2012).

A study conducted by Rigét et al. (2012) examined temporal trends of THg from the early 1980s to 2010 in Greenland ringed seal populations in a warming climate. The influences of climate indices such as ice coverage, water temperature and the AO Index on THg concentrations were evaluated. Increasing levels of THg in seals were found in Ittoqqortoormiit, central East Greenland, and Avanersuaq, Northwest Greenland, with an annual increase of +10.3 and +2%, respectively. Age was an important co-variate for all three regions and trophic position for two regions. The AO was an important explanatory variable for all three regions and was positively associated with THg concentrations in seals indicating the importance of global climatic processes on ringed seal populations in Greenland. In a related study, legacy POP levels in West Greenland ringed seal were higher in years of less sea ice (Rigét et al., 2013). The authors again suggested a relationship to availability or type of prey. The authors also found relationships of some POPs to AO and salinity, but in these cases, suggested that these relationships were related to variable abiotic inputs into the region.

In other marine mammals from Hudson Bay and/or Foxe Basin, female beluga whales showed decreasing THg trends, but male belugas, Atlantic walrus *Odobenus rosmarus rosmarus* and narwhal *Monodon monocerus* did not (Gaden and Stern, 2010). Declining $\delta^{13}C$ ratios concurrent with THg declines were suggested to indicate a shift in female beluga whale foraging habitat or prey type contributing to the THg declines. However, only weak associations between $\delta^{13}C$, but not $\delta^{15}N$ nor THg, and the NAO were found.

In Beaufort Sea belugas, Loseto et al. (2015) investigated possible climate and diet factors influencing THg trends. There were increasing trends in THg levels until 2002, but in recent years THg levels declined in larger/older belugas and stabilized in medium-sized/younger belugas. These variable trends were better explained by climate variables, such as Pacific Decadal

Table 2 Summary of studies on Arctic marine mammals examining GCC-ecological-POP/Hg linkages

Study species, year and location	Climate metric	Ecological change/variation	Compound	Influence on POPs or Hg POP/Hg variation/change	Reference
Ringed seal, 1973-2007, eastern Amundsen Gulf	Ice-free season length	Diet (arctic cod amount and age classes)	THg	↑ in both long and short ice-free seasons	Gaden et al., 2009
Ringed seal, 1993-2008, eastern Amundsen Gulf	Sea ice break-up date	Prey availability or type	p,p'-DDT, p,p'-DDE, ΣCHB, oxychlordane, CB31, CB52, CB101, CB118, CB138, CB153, CB180	↑ in years of earlier break-up	Gaden et al., 2012
			HCB, HCHs, c-chlordane, t-chlordane, dieldrin, mirex, CB28, CB105, CB156	↔	
Beluga whale, 1984-2008, Hudson Bay	NAO	Foraging region or diet	THg	↓ parallels δ^{13}C ↓ in females	Gaden and Stern 2010
Atlantic walrus, 1982-2008, Foxe Basin	NAO	None reported	THg	↔	
Narwhal, 1993-2001, Foxe Basin	NAO	None reported	THg	↔	
Ringed seal, 1994-2010, Central West, North West, and East Greenland	AO, ocean temperature, salinity, sea ice cover	Prey availability or type	THg	↑ in years of ↓ ice and/or ↑ AO (also possibly related to abiotic inputs)	Rigét et al., 2012
Ringed seal, 1994-2010, West Greenland	AO, ocean temperature, salinity, sea ice cover	Prey availability or type	p,p'-DDE, CB153	↑ in years of ↓ ice	Rigét et al., 2013
			α-HCH, β-HCH, CB52, CB153 and/or p,p'-DDE	↑ with ↑ AO or ↑ salinity (related to abiotic inputs)	
Beluga whale, 1981-2012, Beaufort Sea	AO, PDO, sea ice minimum	Unclear (possibly food web structure)	THg	Variable, parallels PDO with 8 yr time-lag	Loseto et al., 2015

↑= Increase. ↓ = Decrease. ↔ = no influence of ecological variation or change. AO= Arctic oscillation; NAO = North Atlantic oscillation; PDO = Pacific decadal oscillation. THg = total mercury. CHB = chlorobornane. HCH = hexachlorocyclohexane. HCB = hexachlorobenzene. CHL = chlordane. PBDE = polybrominated diphenyl ether. p,p'-DDT = $para,para'$-dichlorodiphenyltrichoroethane. p,p'-DDE = $para,para'$-dichlorodiphenyldichloroethylene. PCB = polychlorinated biphenyl.

Oscillation (PDO) with an 8 yr time-lag, rather than by stable isotopes. Oceanic changes were therefore suggested to have impacted long-term Hg intake in this species possibly by altering habitats and food web structures, as well as Hg methylation processes.

2.3 Seabirds

Glaucous gulls *Larus hyperboreus* were investigated for POP trends in relation to climate variation in the Norwegian Arctic (Bustnes et al., 2010) (Table 3). Concentrations of ΣPCB, HCB and oxychlordane declined between 16% and 60% over the period (1997–2006). After controlling for potentially confounding variables, there was a correlation between POP levels and the AO index. The current winter AO was negatively correlated with POP levels, whereas the preceding summer and winter AO were positively correlated with POP levels. The authors speculated that dietary variation, potentially through altered migratory patterns, or physiological (e.g., condition) variation may have led to higher POPs in colder winters. In addition, greater air transport in warmer conditions the previous year may have led to higher POPs the next winter.

Bustnes et al. (2012) examined changes in circulating POPs levels in fasting common eiders *Somateria mollissima* in the Norwegian high Arctic and Subarctic. The high arctic colony showed greater increases in circulating POP levels associated with higher lipid metabolism and lower body mass. Circulating POP increases were also higher in years when ambient temperatures were lower at both colonies, which may also be associated with higher energy expenditures.

Inter-year differences (2008–2009) in trophic position (using $\delta^{15}N$) and THg (and Cd) were examined in black-legged kittiwakes *Rissa tridactyla* and little auks *Alle alle* in two locations in Svalbard (Øverjordet et al., 2015). Lower THg was found in kittiwakes in years when they fed at a lower trophic position and had different feeding habits. The authors suggested that the lower trophic position years were years of less ice and consequently less access to arctic cod, a relatively high trophic position prey. In contrast, trophic position did not influence THg in little auks. The authors concluded that such differences can be attributed to the more specialized feeding behavior of auks on invertebrates versus the more opportunistic feeding behavior of kittiwakes on various fish and invertebrates.

Trends in THg levels in thick-billed murres *Uria lomvia* at both low and high Canadian Arctic colonies were recently reported in relation to changing diets and/or sea ice conditions (Braune et al., 2014). In the high Arctic, where increases in fish versus invertebrate prey have been documented (Provencher et al., 2012), THg levels increased at a faster rate than when adjusted for diet change (using $\delta^{15}N$). In the low Arctic, increases in lower trophic level capelin versus arctic cod have been recorded in thick-billed murre diets concurrent with sea ice declines. Levels of THg would have been increasing (as determined by adjusting for changes in $\delta^{15}N$), but instead showed non-significant trends possibly related to this concomitant shift to lower trophic level prey. Since trophic level changes did not seem to fully explain the THg trends, the authors suggested that other factors, such as changes in Hg cycles and bioavailability related to climate change, may also be influencing Hg trends.

Trends in legacy POPs were also examined in the same thick-billed murre colonies, considering $\delta^{15}N$ changes (Braune et al., 2015b). Legacy POPs generally declined in both colonies. Yet, the decline rate was generally slower in high arctic murres, as a result of the concomitant shift to higher trophic level prey. In contrast, POPs decline rates were faster in low arctic murres, due to the temporal shift to lower trophic level prey. Climate change associated shifts in diets may thus influence both Hg and POP trends in seabirds.

2.4 Arctic marine food webs

In a zooplankton-fish-seabird food web in Svalbard, biomagnification of several legacy POPs appeared to be higher in July relative to May and October (Hallanger et al., 2011) (Table 4). More subarctic-type zooplankton and fish species were sampled in July relative to other months. The authors concluded that this variation in calculated biomagnification was due to a higher presence of subarctic species in the food web in the summer relative to other seasons. In other words, the presence of subarctic species in arctic marine food webs may result in higher overall POP concentrations in the food web and lead to higher calculated rates of biomagnification.

In eastern Canadian Arctic marine ecosystems, documented changes in the presence of subarctic and transient species have occurred. Consistent with the Svalbard food web study, McKinney et al. (2012) showed that biomagnification of several legacy POPs through an eastern Canadian Arctic zooplankton-fish-marine mammal food web was apparently higher when subarctic/transient species were part of the food web relative to only arctic marine species. More specifically, it was the calculated biomagnification of temperate latitude-associated POPs that was higher in subarctic species (e.g., harp seal) versus arctic species (e.g., ringed seal).

Table 3 Summary of studies on Arctic seabirds examining GCC-ecological-POP/Hg linkages

Study species, year and location	Climate metric	Ecological change/variation	Compound	Influence on POPs or Hg POP/Hg variation/change	Reference
Glaucous gull, 1997–2006, Bear Island (Bjørnøya)	AO	Possibly foraging region, diet or condition	ΣPCB, HCB, oxychlordane	↑ in colder years (↓ AO), but ↑ if warmer the previous year (↑ AO), (possibly related to ↑ transport)	Bustnes et al., 2010
Common eider, 2005–2009, northern Norway and Svalbard	Air temperature	Body mass loss/lipid mobilization during fasting	CB153, p,p'-DDE, HCB	↑ in circulating levels in colder years and in colder region (Svalbard)	Bustnes et al., 2012
Black-legged kittiwake, 2008–2009, Svalbard	Sea ice cover	Diet change (subarctic versus arctic fish)	THg	↓ in years of ↓ ice	Øverjordet et al., 2015
Little auk, 2008-2009, Svalbard	None identified	None reported	THg	↔	
Thick-billed murre, 1975–2013, Canadian high Arctic	None identified	Diet change (fish versus invertebrates)	THg	Faster rate of ↑ trend	Braune et al., 2014
Thick-billed murre, 1993–2013, northern Hudson Bay	Sea ice conditions	Diet change (subarctic versus arctic fish)	THg	No trend instead of ↑ trend	
Thick-billed murre, 1975–2013, Canadian high Arctic	None identified	Diet change (fish versus invertebrates)	HCB, HE, dieldrin oxychlordane; p,p'-DDE, Σ_{69}PCB	Slower rate of ↓ trend; ↔	Braune et al., 2015b
Thick-billed murre, 1993–2013, northern Hudson Bay	Sea ice conditions	Diet change (subarctic versus arctic fish)	HCB, HE, dieldrin, oxychlordane, p,p'-DDE, Σ_{69}PCB	Faster rate of ↓ trend	

↑= Increase. ↓ = Decrease. ↔ no influence of ecological variation or change. AO = Arctic oscillation. THg = total mercury. HCB = hexachlorobenzene. HE = heptachlor epoxide. p,p'-DDE = *para,para'*-dichlorodiphenyltrichoroethane. PCB = polychlorinated biphenyl.

Table 4 Summary of studies on Arctic marine food webs examining GCC-ecological-POP/Hg linkages

Study species, year and location	Climate metric	Ecological change/variation	Influence on POPs or Hg		Reference
			Compound	POP/Hg variation/ change	
Zooplankton-fish-seabird food web, 2007, Svalbard	Climate change (in general)	Increase in sub-arctic species	CB28, CB101, CB118, CB138, CB149, CB153, HCB, *p,p'*-DDE, *c*-chlordane, oxychlordane, BDE47, BDE99, BDE100	↑ biomagnification in food web with subarc-tic species	Hallanger et al., 2011
			CB52, α-HCH, *c*-nonachlor, *t*-nonachlor, BDE28, BDE154	↔	
Zooplankton-fish-marine mammal food web, 2007–2008, eastern Canadian Arctic	Climate change (in general)	Increase in sub-arctic/transient species	Cl_4 to Cl_7-PCBs, *t*-nonachlor,, heptachlor epoxide, *p,p'*-DDE, dieldrin	↑ biomagnification in food web with subarc-tic/ transient species	McKinney et al., 2012
			Cl_3-PCBs, HCB, α-HCH	↔	

↑= Increase. ↓ = Decrease. ↔ no influence of ecological variation or change. AO = Arctic oscillation. THg = total mercury. HCB = hexachloroben-zene. HE = heptachlor epoxide. *p,p'*-DDE = *para,para'*-dichlorodiphenyltrichloroethane. PCB = polychlorinated biphenyl.

This suggests that at least some subarctic species are acting as contaminant biovectors, bringing their higher contaminant loads from feeding in subarctic or temperate regions to arctic food webs. These findings suggest exposure implications for arctic species that shift their diets from arctic to more subarctic type prey, in line with results from several of the individual-species studies already discussed.

3 Conclusions and Future Directions

Overall, these studies have provided empirical support for predictions that climate-linked ecological changes will influence contaminant levels and pathways within arctic ecosystems and beyond (e.g., Armitage et al., 2011; Macdonald et al., 2003; Noyes et al., 2009). From this small number of studies, though, it is difficult to speculate as to how extensive the impacts are and which ecological changes are having the greatest impact. Ecological changes could include changes in abundance, behavior/phenology, community structure, condition/growth, and/or habitat range (Wassmann et al., 2011). Certainly, the majority of the studies reported an effect on POPs/Hg of changing/varied species trophic interactions as a consequence of presumed climate-induced changes/variation in type, abundance, habitat range or accessibility of prey species, mainly in relation to sea ice changes. However, the particular ecological change resulting in POP/Hg changes was often unclear or not assessed. Lower sea ice linked-diet changes/variation were associated with higher contaminant levels in some populations of polar bears, ringed seals, and thick-billed murres, but the influence of these changing trophic interactions (whatever the cause) on POP and Hg levels and trends varied widely in both magnitude and direction. The possible influence of changes in body condition was also examined in a single study (Bustnes et al., 2012), and could be important, particularly with respect

to altered circulating levels of contaminants. Additional changes that have been predicted, but not empirically tested in arctic marine ecosystems, include altered primary production and food web lipid dynamics, in the case of lipophilic POPs (Kainz and Fisk 2009; Macdonald et al., 2003).

These studies have relied on correlational analyses between GCC metrics and/or ecological parameters and tissue POP or Hg concentrations. We would be remiss if we did not acknowledge that, by their nature, such studies are unable to address cause-effect linkages. Modeling work involving altered ecological input parameters (e.g., diet) that showed impacts on contaminant levels or time trends would lend additional support to these empirical studies (IPCC, 2013). For instance, physiologically-based pharmacokinetic modeling has been successful in supporting correlative studies of tissue contaminant level-effects studies in arctic wildlife (Dietz et al., 2015; Sonne, 2010), though bringing effects evidence to the population level still remains. In addition, the possible GCC-contaminant interactions are numerous and complex (Jenssen, 2006; Jenssen et al., 2015). For example, although we assumed here that feeding habits change as a consequence of environmental change, contaminants that exhibit endocrine disrupting potential may alter foraging behavior, which could similarly result in altered contaminant intake (Tartu et al., 2014).

The reports reviewed here have provided preliminary insight into ecological aspects of GCC-contaminant interactions. We would like to recommend possible improvements and future directions for this new area of GCC-linked ecotoxicology, namely (1) routine analysis of ancillary ecological metrics with POP and Hg studies, (2) simultaneous consideration of the multiple mechanisms by which GCC and contaminant interactions can occur, (3) targeted research on species known to be sen-

sitive to both GCC and contaminants, and (4) studies linking these changes to changes in major impact parameters like immune and reproductive function and development, particularly at the population level (as described above).

A number of the reviewed studies reported ancillary ecological metric data, such as $\delta^{15}N$, $\delta^{13}C$, fatty acid profiles, body mass/condition and prey mortality rates. Other studies suggested ecological change without actually collecting such corroborating data. In remote and aquatic environments, it can be exceedingly difficult to directly assess ecosystem change and particularly changing species trophic relationships (Bowen and Iverson 2013). The analysis of ecological tracers like stable isotopes and fatty acids is inexpensive relative to contaminant analysis and can often be done without or with minimal additional sample collection requirements. Ecological tracer data can also be generated for individuals with paired contaminant data and can often result in a better understanding of the ecological change(s) that may be driving changes in contaminant levels in a population, species or food web. Developing and refining additional tissue-derived tracers of habitat use and trophic interactions (e.g., compound-specific isotopes analysis; McKinney et al., 2013) or other ecological information (e.g., lipid content as a body condition metric; McKinney et al., 2014) may also be useful in understanding changing contaminant levels and pathways.

Although ecological changes are hypothesized to be a major driver of altered GCC-induced changes in contaminant exposures and pathways, these changes are ultimately a function of physical, chemical and biological factors, as well as source emissions (Macdonald et al., 2003). It is no simple exercise to isolate which are the key factors impacting contaminant time trends, however. This is particularly true for legacy POPs for which secondary emissions and other environmental drivers are likely playing a more important role. Nonetheless, these time trends are used to assess the effectiveness of international controls on emissions (de Wit et al., 2004). Thus, failure to consider other changes to a system that may contribute to increasing or decreasing exposures could lead to erroneous attribution of causation. There has been success in examining the impacts of changing physical and chemical factors (Ma et al., 2011) and ecological factors (reviewed in the current paper). Moving forward, atmospheric environmental chemists and eco(toxico)logists should work together to consider the combined impacts of these changes on contaminant exposures in arctic marine biota.

Laidre et al., (2008) concluded that species most likely to be affected by climate change in the Arctic are those that depend on sea ice habitat. The authors highlighted ivory gull *Pagophila eburnea*, pacific walrus *Odobenus rosmarus divergens*, ringed seal, hooded seal, narwhal, and polar bear as species particularly sensitive in this regard. Some of these species are also top trophic feeders within arctic marine food webs, and thus tend to have relatively elevated contaminant levels (Letcher et al., 2010). As summarized in the current review, altered contaminant exposures linked to GCC have been documented in a limited number of polar bear and ringed seal populations. These studies are the result of successful long term contaminant monitoring projects and should remain a priority, especially with trends of sea ice loss projected to continue in the future (Perovich and Richter-Menge, 2009). A variety of long term climate data is also available, generally for at least as long or longer periods of time (e.g., nsidc.org). Ecological change data, however, is less available (Wassmann et al., 2011). As discussed, developing new, and refining existing, ecological tracer tools that can be measured in archived tissues with paired contaminant measurements will be an important part of these on-going studies. In addition, pacific walrus, hooded seal, and narwhal could be targeted for future research as species that may be sensitive to the interactive effects of climate-ecological-contaminant stressors.

In the most recent, comprehensive review of POP exposures and effects in arctic biota, Letcher et al (2010) identified a need to measure the interactive effects of multiple stressors, given that POPs, Hg, climate change, habitat alteration, resource development and exploitation, pathogens and disease do not act in isolation. Much work remains to even begin to address knowledge gaps regarding multiple stressor interactions and impacts on arctic wildlife and ecosystems. These recent studies documenting climate-ecological-contaminant linkages and changes are a step in that direction.

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