Supplementary information

The influence of global climate change on accumulation and toxicity of persistent organic pollutants and chemicals of emerging Arctic concern in Arctic food webs

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Table S1 Summary of predicted and empirical evidence, conclusions, and knowledge gaps for global climate change-related effects on contaminant exposure and bioaccumulation in Arctic food webs. Arrows signify evidence of increased (↑), decreased (↓), mixed (↑↓), or no influence (↔) of environmental or ecological change.

Climate-related change	Effect on contaminant exposure and bioaccumulation in Arctic biota and food webs	
	Predicted	Empirical/Correlational
Abiotic changes		
Increased temperature ↑↓ Chemical fate and behavior	↑ LRT ↑ Mobilization of POPs from primary and secondary sources ↔ Contaminant uptake and elimination ratesin fish (modeled) ↔ Bioaccumulation in Barents Sea food web(modeled)	↑ LRT ↑ Mobilization of POPs from primary and secondarysources
Altered climate patterns ↑↓ Atmospheric circulation ↑↓Ocean circulation ↑↓ Wind and precipitation patterns	None identified	POPs ↑ in gulls, ringed seals and landlocked Arctic charwith AO+/NAO+ in preceding year POPs ↓in gulls with AO+ in current winter POPs and/or OCPs ↑ or ↓ in murres and/or fulmars 4-9years after high rainfall and/or NAO+ or NAO- Declining trends of some POPs in ringed seals slowed inyears with greater influx of Atlantic currents
Reduced sea ice ↓ Sea ice cover ↓ Sea ice season ↓ Multi-year ice ↑ First-year ice	None identified	Pesticides ↑↓ in zooplankton and ice fauna in areas withmore sea ice in the Barents Sea Lipophilic POPs ↑ in ringed seals in years with early seaice break up in the Beaufort Sea PCB-153 ↑ in ringed seals in years with shorterduration of sea ice in West Greenland Lipophilic POPs ↑ in ringed seals with high total or multi-year sea ice in the Canadian Arctic Lipophilic POPs ↑ in polar bears in areas and seasonswith low sea ice extent around Svalbard Lipophilic POPs and PFAS ↑ in polar bears with high quality sea ice habitat β-HCH and PFAS ↑ in Arctic foxes with increasing seaice cover
Increased terrestrial runoff to surface waters ↑ Glacial and snow meltwater discharge ↑ Precipitation and associated runoff	↑ Mobilization of POPs from secondary sources ↑ Release of POPs to lakes and coastal regions ↑ Darkening and browning of lakes and rivers and coastal waters leading to ↑↓ changes to trophic structure and transfer ofPOPs	POPs ↔ in Arctic char in waters with glacial dischargeor increased turbidity POPs ↓ in littoral amphipods with increasing run-offduring melt season

References	Conclusions	Knowledge Gaps
MacLeod et al., this issue Halsall et al., this issue	Changes in sources and emissions could impact exposure in biota and will be important for interpreting observed changes	
Gouin et al., 2013	Climate-related temperature increases may have minimal impacts on bioaccumulation, due to species-specific biotransformation capacities and thermal tolerances	
Borgå et al., 2010	Seasonal variations in temperature could influence bioaccumulation on smaller time scales	
Bustnes et al., 2010 Rigét et al., 2013 Houde et al. 2019 Cabrerizo et al., 2018bFoster et al., 2019	Higher concentrations of POPs in Arctic biota observed following NAO+/AO+ states and influx ofAtlantic oceanic currents suggest higher exposure following transport of air and water masses from North America and Europe	Unknown underlying mechanisms of statistical relationship between AO/NAO and POPs in biota No data for terrestrial species Effects of intermittent extreme weather events unknown Little understanding of the variance allocation to confounding factors for large regional differences
Carlsson et al., 2014 Borgå et al., 2002a,b Gaden et al., 2012 Rigét et al., 2013a Houde et al., 2019 Tartu et al., 2017b Tartu et al., 2017a Routti et al., 2017 Tartu et al., 2018 Blévin et al., 2020 Andersen et al., 2015a Routti et al., 2017	Melting of multi-year sea ice may release stored contaminants into the Arctic marine food web or may dilute and reduce contaminants at base of thefood web Separating the direct effects of declining sea ice cover on biota POP levels from the indirect effects from changes in food web structure and function aredifficult	Contrasting results indicate sea ice may be a source of POPs/CEACs or dilute POP/CEACs Little is known regarding the match/mismatched of icemelt and biological production and the effect on POP/ CEAC accumulation Differences between POPs and CEACs Processes largely unknown apart from biotic processes
Cabrerizo et al., 2018b	Findings are based on the effects of glacial sources on Arctic freshwater biota and food webs are limited	Effect of terrestrial-deposited snow and melting glaciers onPOP accumulation in receiving freshwater, coastal marine, and terrestrial food webs
Skogsberg, 2019	Increased runoff may dilute and reduce bioavailable fraction of contaminants at base of food web	Net result of terrestrial runoff on contaminant accumulation in Arctic biota still unknown due to largeseasonal variations

Table S1. continued

Climate-related change	Effect on contaminant exposure and bioaccumulation in Arctic biota and food webs	
	Predicted	Empirical/Correlational
Freshwater hydrology and permafrost thaw ↓ Ice cover ↑ Permafrost thaw ↑ Suspended sediment and water turbidity ↑ Primary productivity	None identified	POPs ↑ in sedimentary organic matter of slump-affected lakes POPs ↑ in amphipods from slump-affected lakes
Seasonality changes ↑ Earlier ice melt ↑ Open water season ↓ Later ice freeze up	None identified	PCBs ↑ in later hatching geese eggs POPs ↑↓ in zooplankton, fish and seabirds withseasonal changes in an Atlantic fjord
Ecological changes		
Increased primary production in marine and freshwater	↓ bioaccumulation due to less bioavailabilityof dissolved POPs Systematic ↓ bioaccumulation due to lower bioconcentration in phytoplankton	
Changing species interactions: Marine-to-terrestrial prey shifts	None identified	↓ 30% in CHL for Southern Beaufort Sea polar bears; decrease not reaching statistical significance for PCB, HCH, ClBz and increase not reaching statistical significance for DDT ↓ PCBs, OCPs, PBDEs, PFAS in Arctic foxes from Svalbard, but long term changes in diet did not affect temporal trends of POPs ↓ PCBs, OCPs, PBDEs, PFAS in polar bears from Svalbard, but long term changes in diet did not affecttemporal trends of POPs
Changing species interactions: Arctic to sub-Arctic prey shifts	None identified	↑ instead of ↓ trend for PCBs and CHLs in Western Hudson Bay polar bears, faster rate of ↑ for PBDEs andβ-HCH, faster rate of ↓ for DDTs Non-significant slower rate of ↓ for PCBs and CHLs inEast Greenland polar bears, non-significant faster rate of ↑ of PBDEs. ↔ effect for HCB and DDTs ↑ biomagnification when transient/sub-Arctic species included in the food web
Changes in energy intake/ expenditure: ↑ Energy needs ↓ Body condition	None identified	↑ POPs during incubation in eiders with low body condition ↑ POPs in polar bears and Arctic foxes from Svalbard, but long-term changes in body condition do not affecttemporal trends of POPs ↑ POPs in polar bears in the Barents Sea ↓ Sea ice led to ↑ energy expenditure and ↑ Hg in seabirds due to more time flying, less time underwater,and deeper and longer dives
Changes in behavior and migration patterns	↑ Exposure expected due to migration and higher trophic level diet (i.e. elevated POP levels in migratory seabirds)	Delayed migration timing and routes affect contaminant transfer to offspring/eggs: heavier PCBs ↑

AO: Arctic oscillation; CC: climate change; CHL: chlordane; ClBz: chlorobenzene; CEAC: chemical of emerging Arctic concern; DDT: dichlorodiphenyltrichloroethane; HCH: hexachlorocyclohexane; LRT: long-range transport; NAO: North Atlantic oscillation; OCP: organochlorine pesticide; PBDE: polybrominated diphenyl ether; PCB: polychlorinated biphenyl; PFAS: per- and polyfluoroalkyl substances; POP: persistent organic pollutant

References	Conclusions	Knowledge Gaps
Eickmeyer et al., 2016	Findings based on limited studies from small lakes	Broader suite of lakes need to be studied
D'Onofrio, 2014	Sedimentation of organic matter and particulatesthe facilitate the delivery of associated POPs/CEACs from water column to benthic communities	
Hitchcock et al., 2019a	Changes in seasonal timing may impact migration routes and/or relative time spent at more contaminated wintering grounds	Drivers of seasonal contaminant shifts in food webs (e.g. sea ice and meltwater) still unknown and are largeconfounding factors
Hallanger et al., 2011a,b,c	Seasonal effects may differ between POPs in relation to physiochemical properties (e.g. volatility, water solubility)	
Borgå et al., 2010	Current understanding of the net effect of increasing primary production on contaminant accumulation is limited	Effects of regional boosts in primary production and biodilution are unknown Little is know regarding the cascading effects on trophicinteractions in the food web
Atwood et al., 2017	Studies to date suggest a shift from marine to terrestrial prey is associated with decreasing POP levels	Knowledge of the mechanisms and processes underpinning observed correlations is limited
Andersen et al.,2015a Routti et al., 2017 Vorkamp et al., this issue		
Tartu et al., .2017a, b Lippold et al., 2018 Routti et al., 2017		
McKinney et al., 2009 McKinney et al., 2013	Difficult to conclude as effects appear to depend on the particular prey shift occurring and may be contaminant-specific	Effects of climate-related changes in food webs and species interactions on contaminant accumulation are largely unknown and would benefit from future application of modeling approaches
McKinney et al., 2012		
Bustnes et al., 2012 Baert et al., 2013	Difficult to conclude as most studies focus only on the relationship between body condition and POP levels (e.g. lipid depletion increasing POP concentrations)	Understanding of the effects of climate-related changes in behavior and migration on POP/CEAC accumulation is limited
Tartu et al., 2017b Andersen et al. 2015a Lippold et al., 2018 Routti et al., 2017		
Blévin et al. 2020		
Amélineau et al., 2019 Elliott and Fernie 2019		
Baert et al., 2013	Models suggest higher POP concentrations in migrating seabirds	
Hitchcock et al., 2019a	Behavior and migration changes lead to differences in diet and condition which affect POP/CEAC exposures	

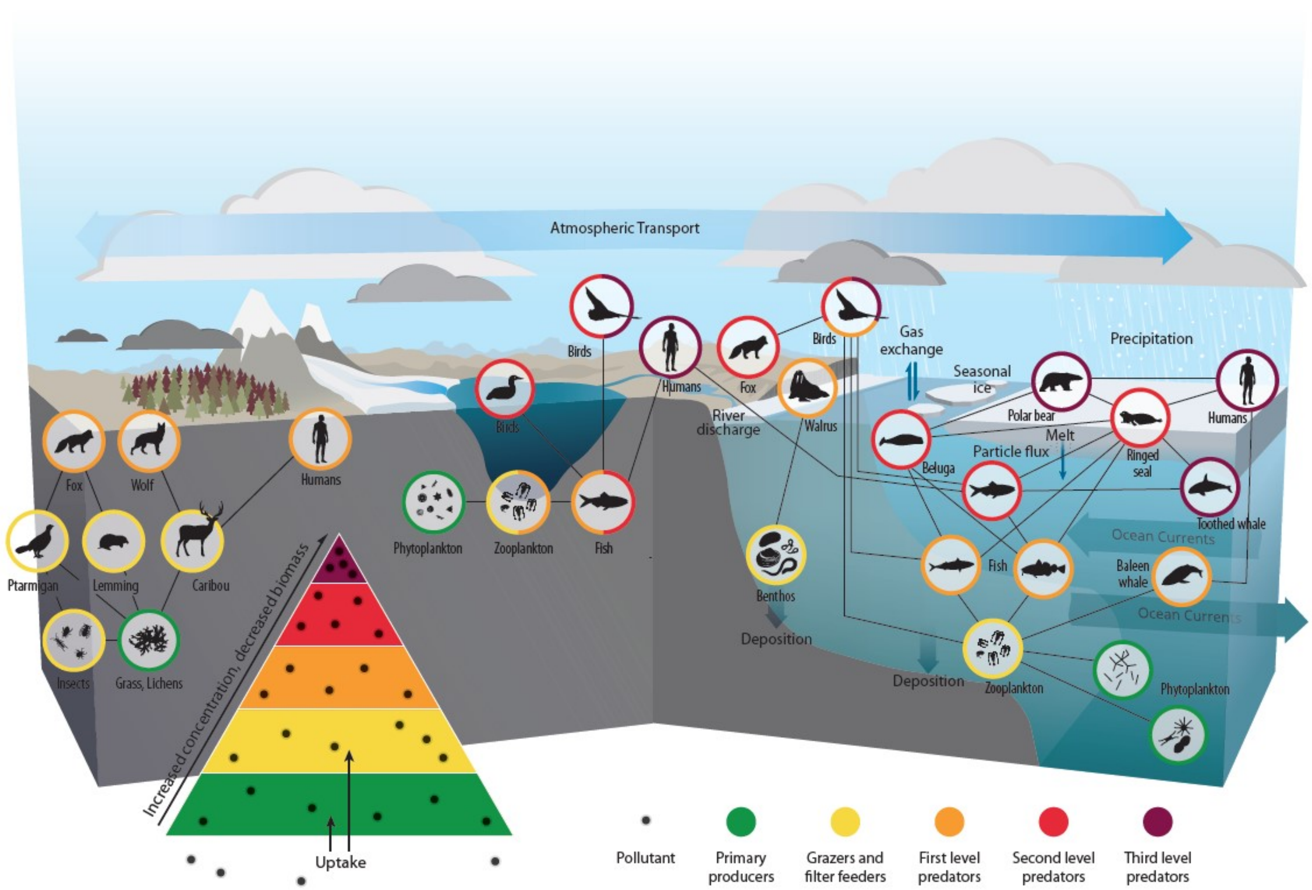

Figure S1 Pictorial representation of the physical and ecological pathways of persistent organic pollutants moving into and through the Arctic environment. Global climate change may influence the depicted pathways, and in turn, the dynamics and fate of Arctic contaminants. Adapted from Macdonald et al. (2005).

Case Study S1: Effects of permafrost thaw on Arctic char condition

Two lakes, East Lake and West Lake, on Melville Island in the central Canadian Arctic Archipelago have been the subjects of a long-term study examining the effect of permafrost thaw and degradation on lake water (Roberts et al., 2017). Since 2008, West Lake has been increasingly impacted by permafrost degradation and subaqueous slumps, which together increased turbidity 50- to 100-fold compared to the nearby East Lake by 2016 (Figure S2). Over this same time period, the physical condition of the resident Arctic char in both lakes changed, as determined using condition factors (K), a standard measure of fish health calculated using an individual's mass relative to its length. Whereas condition factors increased in char from East Lake, they declined in char from the permafrost thaw-impacted West Lake (Figure S2). With turbidity >100 times higher than the East Lake, visibility in

West Lake has been substantially reduced, limiting the ability of visual predators such as Arctic char to feed, which likely explains the lake's declining fish condition. Alternatively, the increased fish condition in East Lake may be due to warmer water temperatures and reduced ice cover, although this requires further investigation. Analysis of carbon stable isotopes ($\delta^{13}C$) and nitrogen stable isotopes ($\delta^{15}N$) showed that mean $\delta^{13}C$ values (± SD) in adult char from East Lake (-27.27±0.81 ‰; n=98) were depleted compared to those of West Lake char (-24.73±1.17 ‰; n=97), indicating greater terrestrial and benthic carbon inputs to West Lake (Muir, unpubl. data). Also, mean $\delta^{15}N$ values (± SD) were lower in West Lake char (10.1±0.98 ‰) compared to those in East Lake char (11.2±0.50 ‰), suggesting differences in food sources. By comparison, fish condition has not changed significantly in three of four Arctic char populations in lakes of similar size on Cornwallis Island in the central Canadian Arctic Archipelago (Figure S2) (Hudelson et al., 2019), despite similar warming temperatures, especially during the period 2008 to 2012.

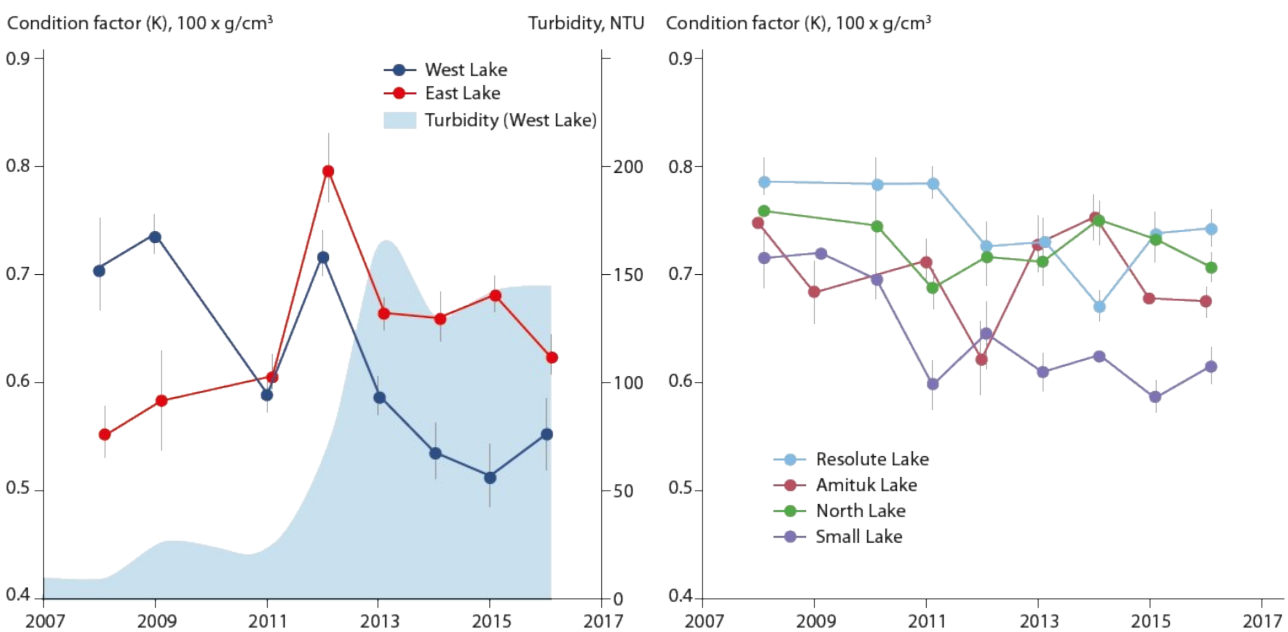

Figure S2 Condition of land-locked Arctic char in Canadian lakes. (Left) Condition factors (K) (100 x g/cm³) for landlocked Arctic char in the clear East Lake and turbid West Lake (2008–2016). Turbidity in West Lake is shown as a shaded background. Tubidity values in East Lake were < 5 NTU and not shown. Data from Roberts et al. (2017). (Right) Condition factors of adult Arctic char from four other lakes on Cornwallis Island in the central Canadian Arctic Archipelago for comparison. Data from Hudelson et al., (2019).

The physiological condition of Arctic char in Lake Hazen, the largest lake by volume north of the Arctic Circle, was also reported to have declined from 1981 to 2015 based on the overall trend in K values (Lehnherr et al., 2018). This trend also coincided with increased turbidity in the lake, arising from increased discharge of sediment-rich glacier-fed rivers, which was particularly apparent between 2008–2013. However, it should be noted the conclusion of Lehnherr et al. (2018) – that ecological changes have resulted in a significant decline in the condition of Lake Hazen char – has been challenged, with independent statistical analysis of the dataset suggesting no significant trend exists (Moore et al., 2018). Nevertheless, trends in the phytoplankton ecology of Lake Hazen have also been observed based on paleolimnological studies, with the planktonic diatom, *Cyclotella sensu lato,* increasing in relative abundance and supplanting benthic species by the late 1990s, reflecting longer ice-free periods in the lake. Thus, the food web of Lake Hazen is undergoing changes, but further monitoring is needed to establish the effects on Arctic char, the lake's top predator. Concentrations of lipid-normalized ΣPCBs and ΣDDTs in Arctic char in Lake Hazen declined over the period 2001–2015 but were positively correlated with NAO conditions (Cabrerizo et al., 2018b; Vorkamp et al., this issue). However, trends of POPs in Arctic char in Lake Hazen were not related to the increased glacial discharge or turbidity.