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# Climate change and mercury in the Arctic: Abiotic interactions

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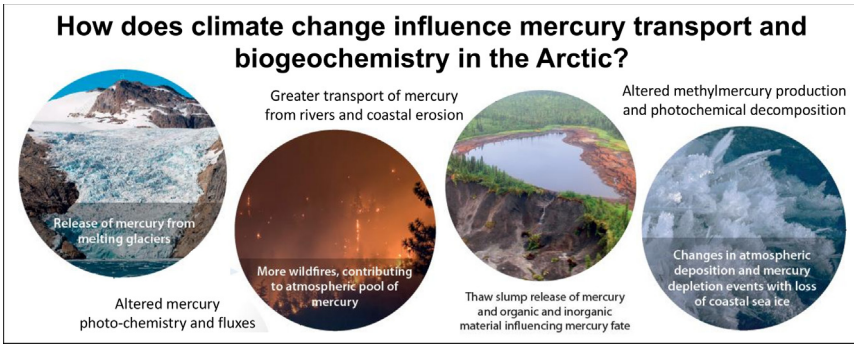
## HIGHLIGHTS

- Current evidence indicates climate change is influencing Hg cycling in the Arctic.
- Permafrost thaw, glacier melt, and coastal erosion are increasing Hg export.
- Arctic permafrost is a global Hg reservoir, vulnerable to degradation and release.
- Experiments show warmer temperatures increase MeHg in Arctic sediments and soils.
- Net effects of climate change on Arctic Hg contamination remain poorly resolved.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Dramatic environmental shifts are occuring throughout the Arctic from climate change, with consequences for the cycling of mercury (Hg). This review summarizes the latest science on how climate change is influencing Hg transport and biogeochemical cycling in Arctic terrestrial, freshwater and marine ecosystems. As environmental changes in the Arctic continue to accelerate, a clearer picture is emerging of the profound shifts in the climate and cryosphere, and their connections to Hg cycling. Modeling results suggest climate influences seasonal and interannual variability of atmospheric Hg deposition. The clearest evidence of current climate change effects is for Hg transport from terrestrial catchments, where widespread permafrost thaw, glacier melt and coastal erosion are increasing the export of Hg to downstream environments. Recent estimates suggest Arctic permafrost is a large global reservoir of Hg, which is vulnerable to degradation with climate warming, although the fate of permafrost soil Hg is unclear. The increasing

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development of thermokarst features, the formation and expansion of thaw lakes, and increased soil erosion in terrestrial landscapes are increasing river transport of particulate-bound Hg and altering conditions for aquatic Hg transformations. Greater organic matter transport may also be influencing the downstream transport and fate of Hg. More severe and frequent wildfires within the Arctic and across boreal regions may be contributing to the atmospheric pool of Hg. Climate change influences on Hg biogeochemical cycling remain poorly understood. Seasonal evasion and retention of inorganic Hg may be altered by reduced sea-ice cover and higher chloride content in snow. Experimental evidence indicates warmer temperatures enhance methylmercury production in ocean and lake sediments as well as in tundra soils. Improved geographic coverage of measurements and modeling approaches are needed to better evaluate net effects of climate change and long-term implications for Hg contamination in the Arctic.

## 1. Introduction

Global climate change is most pronounced in the Arctic, where surface air temperatures have risen at more than twice the rates elsewhere due to Arctic amplification (Serreze and Barry, 2011; Meredith et al., 2019). Arctic warming is also projected to continue to increase faster than the global mean under future scenarios (IPCC, 2013). A recent assessment by the Arctic Monitoring and Assessment Programme (AMAP) concluded that the climate in the Arctic is moving towards a new state, and that although enhanced efforts to control greenhouse gas emissions would reduce further loss, the system will not return to earlier conditions during the course of this century (AMAP, 2017). Physical changes to the Arctic environment are marked: reductions in sea-ice extent and thickness, dramatic loss of multi-year ice, decreased ice-season length, retreating mountain and tidewater glaciers, permafrost thaw and thermokarst development, reduced seasonal snow cover, increased river runoff, and altered nutrient availability (Post et al., 2009; Perovich and Richter-Menge, 2009; Box et al., 2019). These physical changes to Arctic ecosystems are likely to have consequences for the long-term cycling and biogeochemistry of mercury (Hg) (Stern et al., 2012).

The effects of climate change on Hg cycling in the Arctic are complex and interactive because of potential alterations to multiple processes including Hg transport (St. Pierre et al., 2018; Zdanowicz et al., 2018) and Hg biogeochemical transformations such as methylmercury (MeHg) production (MacMillan et al., 2015; Yang et al., 2016). Recent indications of climate change vary across the circumpolar Arctic, including regional differences in warming, sea-ice loss, and altered snow cover (AMAP, 2017). As the environmental change currently underway in the Arctic may differ across both spatial and temporal scales, consideration of effects specific to marine, freshwater, and terrestrial ecosystems is warranted, as well as seasonal, annual, and decadal variation, to assess climate change impacts on Hg cycling.

This review is a contribution to the Virtual Special Issue on the AMAP Assessment 2021: Mercury in the Arctic (AMAP, 2021). It presents a comprehensive synthesis of current evidence for climate change influences on abiotic Hg cycling in the Arctic environment, which was conducted for the assessment. A review of climate change influences on biological uptake and food web exposure is presented separately in this special issue (McKinney et al., 2022, this issue). Whereas the science on climate-Hg interactions in the Arctic was largely hypothetical in earlier reviews (Stern et al., 2012; Macdonald et al., 2005), substantial new empirical, experimental, and modeling evidence has emerged over the last decade. Further, environmental change in the Arctic has continued to accelerate, and a clearer picture is emerging of the profound shifts in the climate and the cryospheric processes (AMAP, 2019; Box et al., 2019; Saros et al., 2019). In this review, a brief summary is provided of what is known about climate change effects on physical and biogeochemical processes within Arctic marine, freshwater, and terrestrial ecosystems. Then, the science of connections between physical changes and the environmental fate of Arctic Hg is integrated by examining effects on Hg transport and biogeochemical transformations of Hg (Fig. 1). Some aspects of the Hg cycle (such as the fate of dissolved inorganic Hg in aquatic ecosystems, or elemental Hg fluxes between air, water, and soil) are not addressed when information on climate change effects was absent. Geographic disparities are also not addressed due to limited information despite probable variation with latitude and among Arctic regions. Finally, conclusions and recommendations are provided for future evaluation of climate change impacts on Hg transport and cycling in the Arctic.

## 2. How has climate change affected the physical and biogeochemical characteristics of Arctic environments?

### 2.1. Atmosphere

Warmer air temperature is arguably the greatest agent of change for physical and biogeochemical alterations in the Arctic environment. Recent temperature increases are more than two times greater in the Arctic than at lower latitudes (Serreze and Barry, 2011; Meredith et al., 2019). The enhanced susceptibility to warming in this region is referred to as Arctic amplification, a phenomenon thought to largely be due to a positive feedback with sea-ice loss (see Fig. 2); decreased ice cover means more open water, which better absorbs solar radiation, which in turn, leads to higher water temperatures and further reductions in ice cover (Serreze and Barry, 2011; Pistone et al., 2014; Screen and Simmonds, 2010). Air temperature increases are reflected in annual averages for the Arctic, seasonal temperatures (particularly in winter), and extreme warm temperatures (Meredith et al., 2019). Total annual precipitation is also increasing, such as in coastal Greenland where meteorological observations are available for the period since 1890 (Mernild et al., 2015) and more broadly across the Arctic in recent decades (Box et al., 2019). Currently, much of the Arctic's annual precipitation falls as snow, which is released from terrestrial environments during spring melt. Climate modeling projects a long-term shift towards precipitation predominantly in the form of rain instead of snow during the 21st century (Bintanja and Andry, 2017). Extreme events are occurring more frequently, such as freezing rain and rain-on-snow events due to warmer temperatures in autumn and winter (Liston and Hiemstra, 2011; Hartmann et al., 2013; IPCC, 2013; Hansen et al., 2014; AMAP, 2017). Other changes to the Arctic atmosphere include shifts in air pressure at sea level, aerosol optical properties, and wind speeds (IPCC, 2013). Furthermore, there are regional disparities in recent atmospheric climate trends observed across the Arctic (AMAP, 2017). These broad changes to the atmospheric environment may have implications for long-range transport of Hg to the Arctic and exchange with marine and terrestrial environments.

### 2.2. Terrestrial environments

Arctic amplification of global climate warming is leading to profound physical changes in Arctic terrestrial environments, including a longer summer growing season (Tagesson et al., 2012), changing precipitation patterns with an increase in summer-wet precipitation (Zhang et al., 2013; Bintanja and Andry, 2017), altered seasonality (Vihma, 2014), and increasing instability of the cryosphere, particularly permafrost thaw (AMAP, 2017, 2019). The major seasonal transitions of fall to winter and spring melt have shifted two to three weeks later and earlier, respectively, shortening the winter season. This means spring melt runoff, the largest hydrologic event in many Arctic locations, is occurring earlier. In addition, there are an increasing number of days without snow cover and with soil and vegetation above freezing temperatures (more growing degree days).

For soil and terrestrial environments, the largest change in the Arctic attributable to rising temperatures is an increase in permafrost thaw

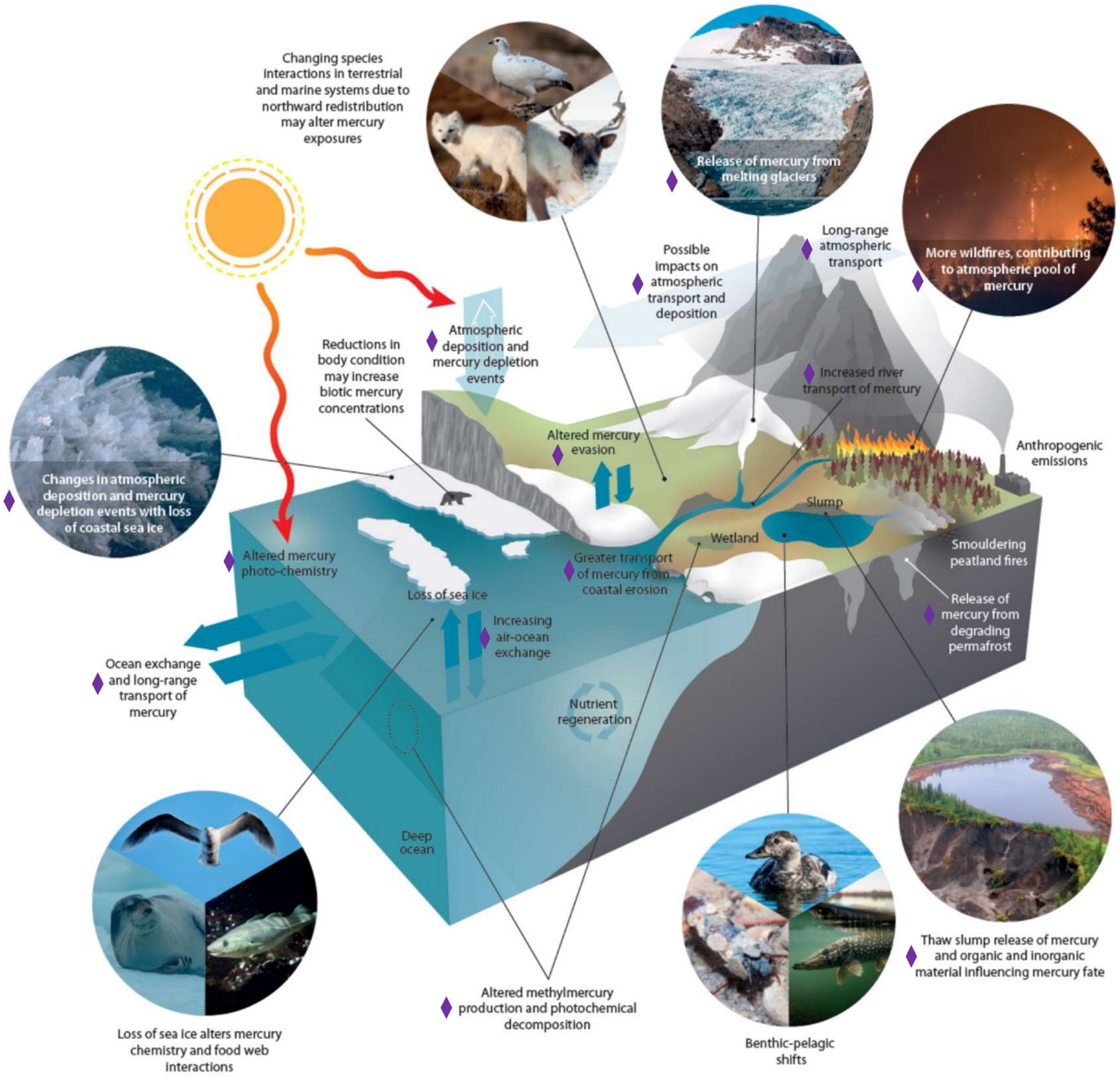


**Fig. 1.** Conceptual diagram highlighting key physical and ecological changes occurring in the Arctic and links to the environmental and biological fate of Hg. Abiotic processes are highlighted with a purple diamond symbol. For more detail on ecological changes and links to the biological fate of Hg, see McKinney et al. (2022), this issue.

degradation and the melting of land ice (Liljedahl et al., 2016). Permafrost is warming around the circumpolar Arctic (Biskaborn et al., 2019), which is leading to the formation of thermokarst features on the landscape such as sinkholes and thermokarst lakes (AMAP, 2017). The destabilization and slumping of permafrost is releasing sediment to downstream lakes and waterways as well as to the Arctic Ocean from coastal erosion (Lantuit and Pollard, 2008; Kokelj et al., 2015; Lewkowicz and Way, 2019). Thermokarst responses to climate warming are not uniform, however, and vary regionally within the circumpolar Arctic in relation to local landscape factors (AMAP, 2017; Loranty et al., 2018).

The greatest stores of land ice (e.g., glaciers, ice sheets, ice caps, ice fields) are in Greenland, the Russian Arctic, the northern Canadian Arctic, and Alaska, and all regions are losing ice mass at an accelerating rate due to anthropogenic climate warming (AMAP, 2017, 2019; Box et al., 2019). Melting Arctic land ice is contributing to sea-level rise, and transporting nutrients, particulates and contaminants to downstream freshwater and marine environments (Søndergaard et al., 2015; Zdanowicz et al., 2018). Together, these large-scale changes in the terrestrial cryosphere have important implications for Hg cycling and transport in the Arctic.

Wildfire has always been a critical process in the life cycle of boreal forests while tundra ecosystems, particularly in the High Arctic, have traditionally been characterized by low wildfire occurrence because of low temperatures, short snow-free seasons, and a lack of flammable biomass (Wein, 1976; Hu et al., 2015). Lake sediment charcoal records reveal that wildfires used to occur only in the driest and warmest regions of the tundra, such as in Western Alaska and Northeastern Siberia. Since 2007, however, some fires have begun to occur in areas where fires have been absent for the last 6500 to 35,000 years (Chipman et al., 2015). The susceptibility of tundra ecosystems to fire largely depends on the crossing of certain temperature and precipitation thresholds predicted to occur more often with climate change (Hu et al., 2015; Young et al., 2017). Climate change also has the potential to exacerbate the likelihood and intensity of wildfires on the tundra by impacting vegetation cover, lightning activity, fire-season length, precipitation patterns, and watershed connectivity

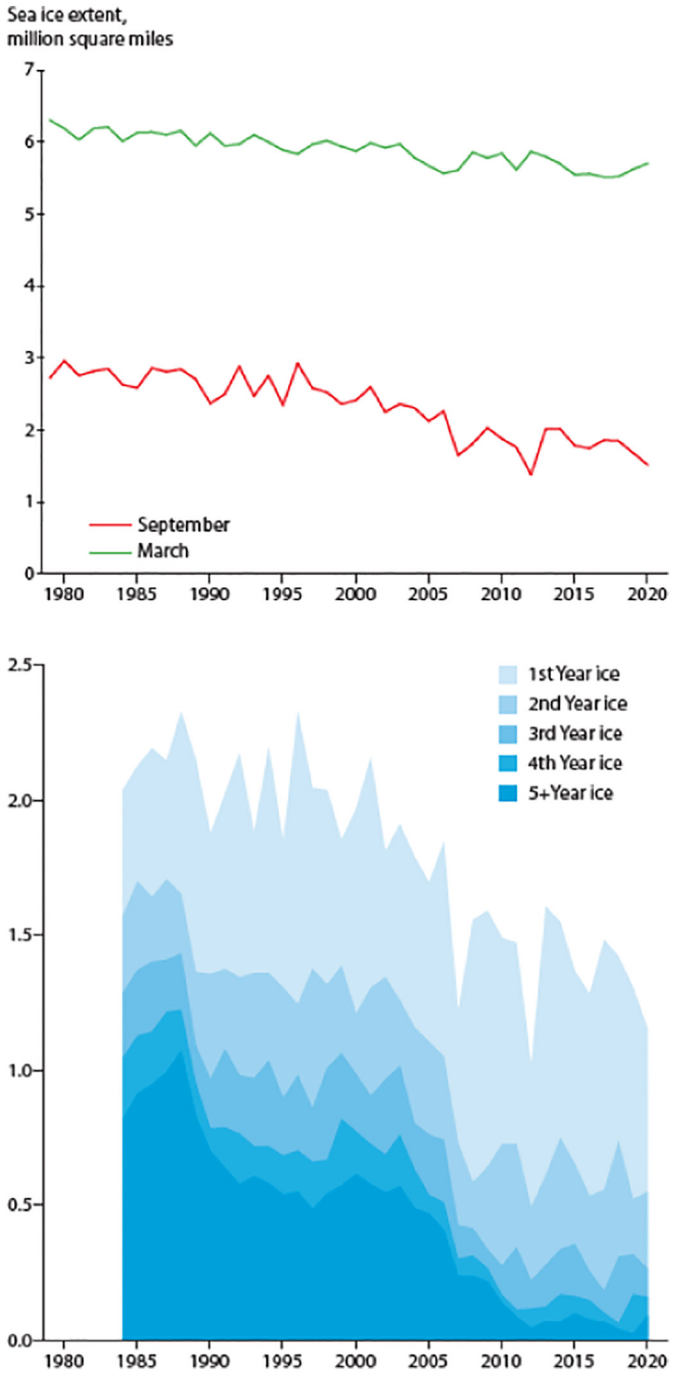


**Fig. 2.** Multi-decadal decline in sea-ice extent in the Arctic Ocean (upper panel) and change in sea-ice age during the period of minimum extent. Source: National Snow and Ice Data Center.

(Smith et al., 2005; Riordan et al., 2006; Hu et al., 2015; Coogan et al., 2019). These climate-driven changes in the terrestrial environment may have complex effects on Hg cycling and transport to downstream environments.

### 2.3. Freshwater environments

Changes to the hydrological cycle in response to Arctic warming have had important implications for the number of lakes and ponds dotting Arctic tundra landscapes. While new lakes and ponds may be forming downstream of retreating glaciers (Stokes et al., 2007; Milner et al., 2017), increasing evaporation and thermokarst development have led to dramatic reductions in the number and surface area of lakes and ponds in non-glacierized regions (Carroll et al., 2011; Finger Higgens et al., 2019; Roach and Griffith, 2015). Watershed geomorphology can play a role in recent shrinking and expanding of water surface area, which may be regional in nature within the Arctic (Carroll and Loboda, 2018). Water surface area has increased in parts of Alaska with warming lowland permafrost (Pastick et al., 2019), while it has decreased in lakes of the Northwest Territories that are situated on bedrock and likely isolated from groundwater (Carroll and Loboda, 2018).

In the Arctic, ice up to two meters thick can cover lakes for as much as ten months of the year and is a critical feature regulating the physics, chemistry, and biology of these ecosystems. During winter, microbial metabolism can result in the build-up of gases such as methane under ice (AMAP, 2017). With warming, lake-ice melt has been happening earlier in the summer (Surdu et al., 2016), and the onset of lake-ice formation has been delayed (Lehnherr et al., 2018), trends which are predicted to continue in the future (Brown and Duguay, 2011). These shifts in ice phenology have occurred in concert with the warming of surface waters during the summer months (O'Reilly et al., 2015). The loss of lake ice has the potential to increase the seasonal light regime in the water column, as well as atmospheric deposition to surface waters, with implications for biological productivity, carbon processing (Cory et al., 2014), and contaminant cycling (Outridge et al., 2007).

Many Arctic lakes remain well-mixed or weakly stratified during the open-water season (Vincent et al., 2008; Priet-Mahéo et al., 2019). With rising temperatures, warmer waters and a longer ice-free period, thermal stratification patterns in lakes are expected to change by strengthening the temperature gradient in the water column and lengthening the stratification period in deep lakes (Prowse et al., 2006; Gebre et al., 2014). Little information is available, however, on the extent to which thermal stratification patterns are changing in Arctic lakes, likely because few long-term, high-frequency measurements are available for lake water columns. Recent studies in Greenland indicate that lake warming and the processes that affect water transparency (e.g., turbidity, browning, eutrophication) are likely to affect lake stratification (Saros et al., 2016; Cadieux et al., 2017). These shifts in water-column mixing are important because thermal stratification has a major influence on the biogeochemistry of lakes.

Warmer temperatures and shifting precipitation patterns are contributing to altered hydrology of Arctic catchments, including streamflow, hydrological connectivity, and water storage (Bring et al., 2016). In the western Canadian Arctic, the timing of stream discharge is changing, with greater flow in winter (Déry et al., 2009; Spence et al., 2014) and this greater winter flow is affecting the water concentrations and catchment export of solutes (Spence et al., 2015). The discharge of large Arctic rivers has also been increasing in recent decades in North America and Eurasia (Box et al., 2019) due to complex processes, though ultimately from greater delivery of atmospheric moisture (Bring et al., 2016). Increasing baseflows in Arctic rivers may also be related to permafrost thaw and associated changes to groundwater storage and/or circulation (Evans et al., 2020). These shifts in the water cycle are significant for Hg cycling given the critical role of streams and rivers in the transport of organic matter and Hg within the Arctic.

Increased leaching of dissolved organic matter (DOM) from terrestrial catchments to rivers and lakes, often estimated by dissolved organic carbon (DOC) concentrations, has been detected from boreal to Arctic regions (Monteith et al., 2007; Garmo et al., 2014; de Wit et al., 2016; Räike et al., 2016; Wauthy et al., 2018), resulting in widespread coloring of lake and river waters with chromophoric DOM (a process termed “browning”). A trend assessment from almost 500 lakes, rivers and streams in Norway, Sweden and Finland showed the largest trends in boreal regions. Significant long-term increases in DOC concentrations were also common in Subarctic fresh waters (de Wit et al., 2016). Browning of fresh waters in recent decades has coincided with improved air chemistry (i.e., lower sulphate deposition and altered sea-salt deposition), changing organic matter chemistry, and solubility in catchment soils (de Wit et al., 2007; Monteith et al., 2007; Evans et al., 2012; Oulehle et al., 2013; Valinia et al., 2015).

Recent changes in environmental conditions, such as greater precipitation and discharge, are increasing DOC concentrations in surface waters

(Räike et al., 2012; de Wit et al., 2016; Zwart et al., 2017). Increased vegetation from greening of catchments, intensive land use (e.g., forestry activity and peat mining) and elevated runoff will jointly increase DOC in Subarctic Fennoscandian watercourses (Finstad et al., 2016; Räike et al., 2016). In addition to the direct effect of changes in hydrological regimes, climate change has been observed to brown lakes in Arctic and Subarctic regions through the thawing of permafrost and deepening of the soil active layer, introducing more DOM to freshwaters (Vonk et al., 2015; Wauthy et al., 2018) and increasing organic carbon (OC) export in large Arctic rivers (Mu et al., 2018). On the other hand, declining concentrations of DOC throughout the 2000s have been observed in Arctic lakes in Greenland, but this decline is related to drivers other than changing air temperature or discharge (Saros et al., 2015). Many studies have addressed various aspects of DOM in Arctic rivers and lakes (Cory et al., 2014; O'Donnell et al., 2016; Kaiser et al., 2017; Osburn et al., 2017; Jiang et al., 2020), reporting that DOM derived from terrestrial environments is abundant and widely distributed in Arctic surface waters, and the quality of this terrestrial DOM is highly different compared to DOM produced within the lakes. Climate-driven changes to carbon cycling have important implications for particulate-sorbed Hg, photochemical processes, and river export.

### 2.4. Marine environments

Sea-ice loss in the marine environment, as documented by satellite records over the past four decades, has been identified as one of the most striking indications of climate change on a global basis (Meredith et al., 2019). There is much interannual variability in sea-ice extent, but an overall multi-decadal declining trend has been found and is projected to continue (Fig. 2). Declines in extent are significant in all months of the year and estimated at $-0.4$ and $-0.8$ million km$^2$ per decade in winter (March 1979–2019) and summer (September 1979–2018), respectively, with some evidence of small recent accelerations in ice loss (Meredith et al., 2019). The declines are also found across nearly all regions but show regional variability in magnitude, with the Beaufort and Chukchi seas showing the greatest reductions (AMAP, 2017). The Arctic Ocean is expected to become seasonally ice free in the coming decades, with some estimates suggesting as early as the next decade (2030s; Wang and Overland, 2012; AMAP, 2017).

In addition to changes in sea-ice extent, there have also been changes in sea-ice thickness, multi-year ice, ice timing, distribution, mobility, and snow depth over the ice. Sea-ice thickness and volume over the Arctic basin have declined by two-thirds from the 1980s to the 2010s (Overland and Wang, 2013; Lindsay and Schweiger, 2015; Fig. 2). After 2005, Arctic sea ice switched from predominantly multi-year cover to coverage dominated by annual ice (AMAP, 2017). The ice-covered season is now one to three months shorter than in the 1970s, with earlier melt onset and later freeze-up (Barber et al., 2015; AMAP, 2017). Likely due to later formation of sea ice in the fall, snow depth on the sea ice has shown consistent declines (Webster et al., 2014), with possible connections to rising proportions of first-year ice.

Shifts in the marine environment have been recorded related to ocean temperatures and circulation, freshwater river flux input, sea levels, and ocean acidification. Along with warming surface air temperatures in the Arctic, sea-surface temperatures have generally increased (Barents, Chukchi, Kara, East Siberian, and Laptev seas; Timmermans and Labe, 2021; Lind et al., 2018) and so have water temperatures at lower depths, due to warmer waters entering from the North Atlantic and North Pacific (AMAP, 2017; Lind et al., 2018). Increased river discharge and melting glaciers have led to a 50% increase in freshwater flux in less than two decades off South Greenland (Bamber et al., 2012). Because salinity is an important driver of ocean currents, this increased freshwater flux has been suggested to have played a role in recently observed reductions in the Atlantic Meridional Overturning Circulation (Rahmstorf et al., 2015). Globally, sea levels have risen by 20 cm or more since 1900 (AMAP, 2017). The Arctic is a main player in sea-level rise, with the melting of Arctic glaciers and ice caps contributing to more than one-third of the global increase in sea levels (AMAP, 2017). As elsewhere, due to climate change, the Arctic Ocean is taking up more carbon dioxide from the air, forming carbonic acid and increasing hydrogen ions, thereby decreasing the pH of the Arctic Ocean. This process also consumes carbonate ions, which are needed by calciferous organisms. These processes are called ocean acidification. The Arctic Ocean is more sensitive to acidification than other oceans due to various processes (AMAP, 2018), and significant declines in pH have been reported in the Canadian Basin in the period from 1997 to 2008 and in the Nordic seas in the period from 1985 to 2009 (AMAP, 2018).

## 3. What influence has climate change had on mercury transport processes?

### 3.1. Atmospheric deposition

The concentrations of gaseous elemental mercury (GEM) in the atmosphere are controlled by emission source strength (primary anthropogenic, natural emissions and re-emissions, as well as secondary legacy emissions), atmospheric relaxation time, transport and the final fate of Hg moving into terrestrial and aquatic reservoirs (Skov et al., 2020; AMAP, 2021). Atmospheric relaxation time is the net effect of all removal and formation reactions of GEM, and it represents the time needed before a change, such as to emissions, affects the atmospheric concentration of GEM. All of these processes (to a lesser extent anthropogenic emissions) have a climate component that might affect future Hg dynamics. The transport from mid-latitude regions is dependent on the position of the major weather systems, which is predicted to change with changing climate (IPCC, 2019). The relaxation time of atmospheric Hg is dependent on the oxidation rate of GEM to gaseous oxidized mercury (GOM) and particulate-bound mercury (PBM), as GOM and PBM are fast removed by either wet or dry deposition. The final fate of Hg is also dependent on the reduction of oxidized Hg (e.g., in the water column) back to elemental Hg (Skov et al., 2020). The reaction kinetics of gaseous elemental mercury, Hg(0), in the atmosphere are temperature dependent (Goodsite et al., 2004, 2012; Donohoue et al., 2006; Jiao and Dibble, 2015), whereby oxidation decreases with increasing temperature, and thus GOM and PBM formation is expected to decrease in a future warmer climate. The deposition is dependent on the stability of the atmosphere, wind speed, and surface properties (Skov et al., 2006); thus, the Hg distribution between atmosphere and land surfaces is climate-dependent. However, the direction of this change is uncertain as reactions with bromine will be affected by the release of bromine from refreezing leads, snowpack or from marine aerosols, resulting in slower oxidation of Hg(0) in the atmosphere (Yang et al., 2020; Zhao et al., 2016; Blechschmidt et al., 2016). Deposition will be faster as the aerodynamic resistance is less over vegetation and open waters than over snow and ice. Nonetheless, the concurrent impacts of changes in climate, chemical composition, land use and primary and secondary Hg emissions on Hg temporal trends make it difficult to detect the influence of changes in anthropogenic emissions in observed temporal trends. Despite rising global anthropogenic emissions over the past several decades (Streets et al., 2011), concentrations of Hg in the Arctic atmospheric have generally decreased (Cole and Steffen, 2010; Berg et al., 2013; MacSween et al., 2022, this issue). As rising air temperatures (particularly in spring) and reduced sea-ice extent and thickness continue with climate warming, the implications for Hg levels in Arctic ecosystems may be complex and multidirectional (Bekryaev et al., 2010; Cavalieri and Parkinson, 2012; Stern et al., 2012).

The factors influencing Hg(0) trends in the Arctic from 1979 to 2008 were investigated by Fisher et al. (2013) using the GEOS-Chem (Goddard Earth Observing System - atmospheric chemistry) model. A small increasing trend in Hg(0) concentrations over 30 years was simulated by the model, mainly reflecting rising anthropogenic emissions (as per Streets et al., 2011). The model captured the springtime interannual variability in Hg(0) concentrations at Arctic sites and found it to be more significant compared to the temporal trend. The model reproduced the observed shift in minimum Hg(0) concentrations from May (1995–2001) to April (2002–2007) at Alert, Canada and attributed this shift to cooling in April

and warming in May, confirming Cole and Steffen (2010). However, a shift in minimum Hg(0) concentrations at Alert was not found to be a characteristic of the Arctic as a whole. Fisher et al. (2013) concluded that high temperatures and a low sea-ice fraction in spring decrease the importance of atmospheric mercury depletion events (AMDEs), while high solar radiation in spring enhances the photo-reduction and re-emission of Hg deposited to the snowpack. During summer, the same environmental changes increase photo-reduction of Hg(II) in the ocean and evasion of Hg(0) to the atmosphere. Thus, Fisher et al. (2013) suggested climate change may decrease Hg fluxes from the atmosphere to the cryosphere and Arctic Ocean.

Building on the work by Fisher et al. (2013), Chen et al. (2015) evaluated the influence of environmental conditions and anthropogenic emissions on temporal trends of atmospheric Hg in the Arctic. Anthropogenic emission inventories were used from AMAP/UNEP for the years 2000, 2005 and 2010. The model captured most of the seasonality in observed trends, especially the increases in spring and fall; however, it failed to reproduce increasing total gaseous Hg trends in July at Alert and in October at Ny-Ålesund, Svalbard. A main finding of the study was that lower Hg evasion from the Atlantic Ocean influenced atmospheric Hg at mid-latitudes and contributed to the decrease in Arctic Hg(0) concentrations from November to March.

The role of global anthropogenic emissions and meteorology in temporal trends of ambient Hg concentrations and deposition was assessed by Dastoor et al. (2015) for the Canadian Arctic from 1990 to 2005 using GEM-MACH-Hg (Global Environmental Multi-scale - Modeling Air quality and Chemistry - Mercury; see AMAP, 2021) and AMAP anthropogenic emissions (AMAP, 2011). Interannual variability in air concentration and deposition of Hg was found to be driven by interannual variability in meteorology. Modelled surface air Hg(0) concentrations in the Canadian Arctic declined by ~12% from 1990 to 2005, consistent with measurements at Alert (Cole and Steffen, 2010; Cole et al., 2013). The decreasing trend was influenced equally by changes in metereology and anthropogenic emissions. For net atmospheric deposition of Hg, the model simulated a 15% increase due to changes in meteorology and a 5% decrease in the High Arctic due to a decline in emissions in North America and Europe; this resulted in an overall 10% increase in Hg deposition between 1990 and 2005 (AMAP, 2021). Increasing snow-covered regions over first-year sea ice led to a decreasing trend in Hg re-emission fluxes from the snowpack, which resulted in increased net Hg deposition in the model. Halogen-enriched snowpacks over first-year sea ice suppressed re-emission of Hg from snow (Durnford and Dastoor, 2011). Additionally, a decrease in snow cover extent and a small increase in precipitation contributed to a small increase in deposition. For the marine environment, Fisher et al. (2013) and Chen et al. (2015) did not consider the impact of changing snow characteristics (i.e., halogen content in sea ice and snowpack) on Hg reduction and re-emission from snow, whereas Dastoor et al. (2015) neglected changes in ocean Hg evasion. Despite discrepancies between models, environmental factors associated with climate change played a dominant role in Arctic Hg trends in all three studies. The complexity of climate-Hg interactions are not fully represented in current Hg models.

### 3.2. Catchment transport

The impacts of climate change on the transport of Hg across Arctic catchments are complex, and catchment or watershed-scale studies are becoming increasingly common (e.g., Zdanowicz et al., 2018; St. Pierre et al., 2019) in recognition that ecosystem health is dependent on biogeochemical connections and processes within the catchment (e.g., Braaten et al., 2014). The following sections summarize how climate change factors, namely snowmelt and precipitation changes, permafrost degradation, forest fires, and glacial melt, have already affected and may continue to influence Hg transport across Arctic catchments. Lake sediments are also included in this section because this environmental archive is widely used to estimate Hg fluxes to lakes, which it can be strongly influenced by catchment transport and climate change processes.

#### 3.2.1. Snowmelt and rainfall

Snowmelt has traditionally been considered an important vector for the transfer of Hg to both terrestrial and aquatic ecosystems across the Arctic (e.g., Obrist et al., 2018; Dommergue et al., 2010). Across non-glacierized areas, the snowmelt period may account for the largest seasonal pulse of total mercury (THg) fluxes to downstream ecosystems (Semkin et al., 2005). Douglas et al. (2017) highlight the potential enhancement of Hg deposition to, and retention in, coastal Arctic snowpacks due to the combined deposition of Hg and halogens during AMDEs. However, the re-emission of Hg from snowpacks varies with latitude (Durnford et al., 2012), such that the impact of these changes may differ spatially. In inland regions, Hg in snowpacks is often associated with mineral dusts (Agnan et al., 2018). Extreme events, including low snowpack years and large wind events (St. Pierre et al., 2019), could enhance dust deposition to snowpacks and ecosystems across the Arctic.

Summertime rainfall events, which are becoming increasingly common (Bintanja, 2018), can also mobilize significant quantities of solutes and particulate matter from landscapes at the height of thaw. Already, in more southerly regions of the Arctic, like the Yukon Territory (Canada), areal rates of summertime wet deposition of Hg greatly exceed, by as much as six times, that of snowpack deposition (Zdanowicz et al., 2018). The impact that such shifts in precipitation patterns will have on the Hg budget of Arctic watersheds, especially as precipitation originates from and interacts with other changes across the region (e.g., reduced sea-ice cover), largely remains to be determined.

#### 3.2.2. Terrestrial organic matter transport

Arctic and Subarctic lakes are often characterized by clear water with low DOM concentrations of which DOC is a major component (Henriksen et al., 1997; Pienitz et al., 1997; Lim et al., 2001; Forsström et al., 2015), but many of these systems are browning (Macdonald et al., 2005; Wauthy et al., 2018). Browning of surface waters from increasing terrestrial DOC concentrations has important consequences for the biogeochemical cycling of Hg in Arctic and Subarctic ecosystems. DOM binds inorganic Hg and MeHg during transport from catchment soils to water bodies (Grigal, 2002). DOM plays a critical role in complexation, photochemical and microbial processing of Hg in aquatic environments (Lehnherr, 2014). Positive correlations between Hg and DOC concentrations in water are often observed; however, DOC quality and age may be important factors controlling these correlations (Forsström et al., 2015; Lescord et al., 2018; Poste et al., 2019; Richardson et al., 2021). Browning of lakes is also known to enhance thermal stratification (Snucins and Gunn, 2000) and thereby weaken aeration of the hypolimnion, promoting anoxia (Couture et al., 2015), thus potentially enhancing in-lake methylation of inorganic Hg and accumulation of MeHg in biota in small temperate and boreal lakes (Watras et al., 1995; Eckley et al., 2005; Rask et al., 2010; Verta et al., 2010). In addition to oxygen-related MeHg production, browning of lakes will shift primary production towards secondary bacterial production, which is linked to elevated Hg levels in boreal watercourses (Forsström et al., 2013; Lescord et al., 2018). Furthermore, increasing DOC will decrease the penetration of UV-radiation into the water column and reduce demethylation processes in surface water (DiMento and Mason, 2017; Williamson et al., 2019). Thus, browning processes may enhance MeHg exposure in lake systems.

#### 3.2.3. Permafrost degradation

With a loss of between 6% and 29% of high latitude permafrost projected for each 1 °C of warming (Koven et al., 2013), climate change-induced permafrost thaw could mobilize the vast amount of Hg currently stored in frozen soils. Soils in the Arctic and Subarctic permafrost contain substantial reservoirs of OC (Tarnocai et al., 2009; Schuur et al., 2015) and Hg is bound to this organic matter, although estimates of the amount of stored Hg remain poorly constrained (Schuster et al., 2018; Lim et al., 2020). The fate of this Hg will ultimately depend on the type of permafrost features created or affected (i.e., lake, wetland or hillslope thermokarst; Olefeldt et al., 2016) as well as on climatic factors controlling transport to downstream ecosystems.

Permafrost thaw across the Arctic results in the creation of small thermokarst lakes, ponds, and wetlands (Gordon et al., 2016; Olefeldt et al., 2016). These highly productive systems are shallow, have high inputs of organic matter and nutrients, and are microbially active, making them excellent environments for the production of MeHg (MacMillan et al., 2015; Gordon et al., 2016; Roth et al., 2021). When the ponds drain following slumping, further permafrost degradation, or erosion, they become an important source of MeHg to nearby rivers (e.g., Fortier et al., 2007). Changes to thermokarst along the edges of small Subarctic lakes have led to increased Hg deposition at depth (Rydberg et al., 2010) potentially enhancing MeHg production. Thermokarst wetlands show higher MeHg production than adjacent peat plateaus (Tarbier et al., 2021), though connectivity between wetlands and adjacent terrestrial and freshwater systems may modulate MeHg exports to downstream systems (Varty et al., 2021). Although MeHg photodemethylation is typically an important sink of MeHg in small pond systems (Lehnherr et al., 2012), browning as a result of large DOM inputs with permafrost thaw may actually reduce photodemethylation and increase the net production of MeHg.

One of the most striking consequences of permafrost degradation in parts of the Arctic subject to hillslope thermokarst is the development of retrogressive thaw slumps, a form of mass wasting characteristic of hilly regions underlain by ice-rich permafrost. These features, which can be up to 40 ha in area, can release large quantities of sediments and solutes into lakes, rivers, and coastal waters (Kokelj et al., 2013). Concentrations of Hg and MeHg in streams draining slumps in the western Canadian Arctic have been recorded as high as 1270 ng/L for THg and 7 ng/L for MeHg (St. Pierre et al., 2018). High sedimentation rates in slump-affected lakes have been hypothesized to dilute Hg deposition in these environments (Deison et al., 2012), but monitoring of these sites is needed to understand the long-term impact of these events. At present, Hg mobilization through the streams draining slump-affected areas is transport-limited (i.e., sediment supply exceeds water volume); however, during high-flow events, like the spring freshet, or if predictions of a wetter Arctic are realized (Bintanja and Andry, 2017), such conditions could enable the mobilization of vast quantities of Hg to downstream ecosystems. In the Mackenzie River (Canada), which drains 1.7 million $km^2$, river water concentrations of particulate and dissolved Hg were positively correlated with the age of water OC, suggesting catchment processes releasing older carbon (e.g., permaforst degradation) were important Hg sources (Campeau et al., 2022). Clearly more studies need to be done to establish whether permafrost Hg hotspots exist and to identify environmental processes that could promote the transport or accumulation of Hg. From this, a better projection can be made of the broader-scale impacts of Hg release from permafrost thaws.

### 3.2.4. Coastal erosion

There is substantial evidence that average coastal erosion rates are significantly increasing across many Arctic regions, and are now higher than at any time since observations began 50 to 60 years ago (Overduin et al., 2014; Irrgang et al., 2018). For example, erosion rates in rapidly eroding sections of the coastlines along the Laptev and Beaufort seas have doubled over the past 50 years (Jones et al., 2009; Günther et al., 2013). A number of interacting climatic, oceanographic, and onshore geomorphological processes have been suggested as contributing to this trend. These include: ongoing warming and destabilization of coastal ground ice, declining sea-ice extent, warmer summertime sea-surface temperatures and wind speeds, and rising sea levels. These changing oceanic conditions promote increases in storm frequency and intensity, and thus, the effects of wave action upon thawing, exposed shoreline permafrost (Jones et al., 2009; Overduin et al., 2014). Erosion of coastal soils is one of the major contributors of Hg to the Arctic Ocean (Outridge et al., 2008; Soerensen et al., 2016; AMAP, 2021), and this flux is likely to increase with increasing erosion rates during the 21st century.

### 3.2.5. Forest fires and soils

Arctic Hg cycling will be impacted by the anticipated increase in wildfire activity caused by climate change in three ways: (1) the appearance of tundra fires, which will release terrestrial Hg to the atmosphere and streams locally; (2) the transport of wildfire-derived Hg from lower latitudes via long-range atmospheric transport, and possibly rivers that flow northwards; and (3) by causing physical and biological changes in the local environment. Understanding the impacts of fire on Hg dynamics through Arctic ecosystems, particularly on watershed connectivity and stream/river transport to downstream systems, will be critical.

Mercury dynamics across the Arctic region are affected by fires occurring both locally and further afield. Gaseous elemental mercury, typically considered the dominant species emitted by fire, can be transported long distances from source regions (Fraser et al., 2018). Conversely, PBM has a shorter residence time in the atmosphere and is often deposited closer to the emission source (Fraser et al., 2018). Model estimates (GEOS-Chem) suggest ~10% of total annual Hg deposition (15 Mg/y) to the Arctic originates from forest fires, mainly from the large swaths of boreal forest in Eurasia (Kumar and Wu, 2019). At the same time, local wildfires are a minor deposition source of Hg to Subarctic Canadian lakes (Pelletier et al., 2020) and represent <5% of the total Hg deposition to the Canadian High Arctic during peak fire season (Fraser et al., 2018). The combination of increasing legacy contamination and greater fire frequency could lead to greater Hg release and deposition in the future (Biswas et al., 2007; Obrist et al., 2018). For example, the higher Hg concentrations in boreal forests could increase Hg emissions from wildfires in Eurasia by 41% between 2000 and 2050 (Kumar et al., 2018). More information is needed to better constrain estimates of Hg emissions from wildfires in boreal and Subarctic regions, including estimates of the propensity of boreal peatlands to burn and of the proportion of gaseous to particulate Hg produced by wildfires across these landscapes (Turetsky et al., 2006; Fraser et al., 2018).

Aside from the direct deposition of GEM and PBM to the landscape, local wildfires may also indirectly enhance the mobilization of Hg stored in soils through permafrost degradation, active layer deepening or warming and thermokarst feature development (Jones et al., 2015; Gibson et al., 2018). In particular, wildfires are estimated to be responsible for 2200 ± 1500 $km^2$ of thermokarst bog formation over a 400,000 $km^2$ area of sporadic and discontinuous permafrost zones in Subarctic Canada (Gibson et al., 2018). Thermokarst development may then promote the mobilization and production of MeHg (MacMillan et al., 2015). Recent work in forested watersheds suggests that wildfires may not affect exports of dissolved Hg, though stream concentrations of particulate Hg—an important byproduct of wildfire, especially close to the source (Fraser et al., 2018; Obrist et al., 2018)—may increase substantially for up to 8 months post-fire (Jensen et al., 2017). Greater erosional transport of Hg to a Subarctic montane lake continued for >20 years following two wildfires, which severely damaged catchment soils and vegetation (Pelletier et al., 2022). Furthermore, fires in permafrost zones typically result in the substantial loss of soil organic matter and increased active layer water storage and soil temperatures, leading to reductions in the permafrost, all of which can influence the biogeochemical cycling of Hg through northern ecosystems (Nossov et al., 2013).

### 3.2.6. Glacier melt

Within many glacierized catchments in the Arctic, glacier melt currently accounts for the largest source of both water and Hg to downstream ecosystems (Zdanowicz et al., 2018; St. Pierre et al., 2019). Glacial meltwaters integrate two principle sources of Hg: (1) legacy and modern Hg archived in glacial ice and snow; and (2) geogenic Hg transported by meltwaters as they flow across poorly consolidated proglacial landscapes (Zdanowicz et al., 2013). Hg deposition to glaciers has varied substantially over time in response to changes in both natural (e.g., volcanic eruptions) and anthropogenic sources (Beal et al., 2015). We would therefore expect the Hg contribution from ice and snow to decline as older ice begins to melt. In many cases, however, geogenic Hg contributions to meltwaters along glacier margins or across proglacial landscapes can be as important, if not more, than those from the glacier area (Zdanowicz et al., 2013). Large, periodic fluctuations in meltwater volume, including glacial lake outburst floods, can also mobilize substantial quantities of Hg from the surrounding

landscape. In the Zackenberg River of northeast Greenland, for example, glacial lake outburst floods are responsible for between 5% and 10% of the river discharge in years when they occur, but between 15% and 31% of the Hg export (Rigét et al., 2011; Søndergaard et al., 2015). These extreme meltwater discharge events may become increasingly common with climate change (Harrison et al., 2018; Nilsson et al., 2015), with the potential for substantial mobilization of Hg across landscapes.

Although glacial meltwaters typically contain very little MeHg (<0.1 ng/L; Zdanowicz et al., 2013; St. Pierre et al., 2019), little attention has been paid to Hg dynamics in subglacial channels or within cryoconite on glacier surfaces, both of which could support methylation. Subglacial meltwaters can become anoxic and contain enough bioavailable carbon to support significant production of methane, similar conditions required for Hg methylation (e.g., Lamarche-Gagnon et al., 2019). Higher concentrations of MeHg (1.0 ng/g) have been detected in cryoconite of the Tibetan Plateau, suggesting either the preferential accumulation of MeHg there or active methylation (Huang et al., 2019).

Deglaciation on land also has important consequences for Hg transport and processing within downstream coastal waters. In a survey of three fjords impacted by glacial melt in Svalbard, the highest THg concentrations were observed in Hornsund, which was experiencing the most dramatic glacial retreat of the three fjords at the time of sampling (Kim et al., 2020). This Hg was strongly associated with terrestrial (78 ± 17%) organic matter, suggesting that the Hg itself may have originated from the surrounding watersheds. Evidence from Hg stable isotopes indicated that the Hg transported to the fjords was primarily from bedrock erosion (Lee et al., 2021).

### 3.2.7. Lake sediment

Lake sediments are important environmental archives to assess both climate-related changes in catchment transport and atmospheric deposition of Hg in relation to watershed ecosystem function and disturbance over time (Fig. 3; Goodsite et al., 2013; Drevnick et al., 2016). However, given that these two processes overlap in time, disentangling the relative contribution of each can be challenging (Korosi et al., 2018). Whereas post-industrial increases in Hg deposition, largely due to anthropogenic activities, are reported across most northern lakes, the impacts of climate change on catchments are much more variable and depend on local characteristics, such as catchment-to-lake-area ratio (Drevnick et al., 2012), the presence of glaciers, or susceptibility to different thermokarst landform types (Burke et al., 2018). Numerous studies have noted widespread increases in sedimentation rates, particularly in recent sediment layers, a change attributed to increased catchment erosion (Fitzgerald et al., 2005; Muir et al., 2009; Cooke et al., 2010; Kirk et al., 2011). Mercury flux estimates are commonly adjusted using geological tracers (crustal elements) or changes in sedimentation rates to tease apart catchment and atmospheric Hg inputs so that these sedimentary records could be used to examine changes in atmospheric Hg deposition.

Some studies have found strong positive correlations between Hg accumulation in sediments and proxies of lake primary production, such as total organic carbon (TOC) and S2 carbon (algal-derived kerogen), and suggested that this relationship was due to algal scavenging of Hg or the absorption/adsorption by algal biomass of available Hg in the water column followed by its sinking and sedimentation (Sanei et al., 2012; Brazeau et al., 2013). It has been estimated that 70% to 96% of Hg deposition recorded in sediments over the post-industrial period in Canadian and Norwegian Arctic lakes can be attributed to algal scavenging (Outridge et al., 2007; Stern et al., 2009; Rydberg et al., 2010; Jiang et al., 2011; Sanei et al., 2012; Outridge et al., 2019), which has implications for the use of sedimentary records to reconstruct Hg deposition. However, other studies have found strong relationships between Hg and proxies of primary productivity, such as S2 carbon and chlorophyll *a*, in only a subset of lakes and concluded that the effect of algal scavenging on the sedimentary Hg record was not a wide-spread phenomenon across the circumpolar Arctic (Kirk et al., 2011; Cooke et al., 2012; Deison et al., 2012; Korosi et al., 2018; Lehnherr et al., 2018). Outridge et al. (2019) suggest the absence of relationships between sediment organic matter and THg in some lakes may be due a limitation of forms of Hg and/or labile organic matter suitable for binding each other, such as complexation of Hg by humic acids (Le Faucheur et al., 2014; Schartup et al., 2015). They suggest the importance of algal scavenging may vary over time as the lake's climate and environmental characteristics change and algal scavenging may partially explain why recent Hg fluxes have increased in many Arctic lake sediments while atmospheric Hg concentrations have decreased or remained stable (Goodsite et al., 2013).

Increasingly, changes in catchment transport processes are being reflected in Hg deposition rates in lake sediment cores across the Arctic, especially in watersheds undergoing dramatic change as a result of permafrost thaw (Deison et al., 2012; Korosi et al., 2015), glacial melt (St. Pierre et al., 2019) and erosion by enhanced river flows. Deison et al. (2012) analyzed dated sediment cores from seven thermokarst-affected lakes in the Mackenzie Delta uplands (Northwest Territories, Canada) and compared

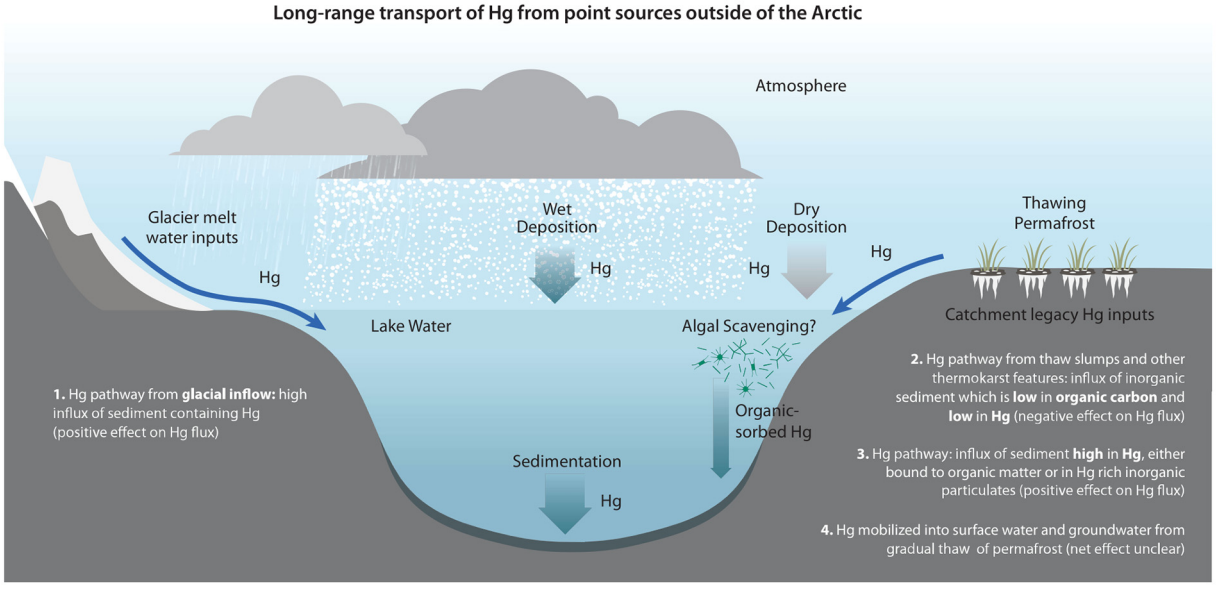


**Fig. 3.** Conceptual diagram highlighting connections between climate change and catchment Hg fluxes to Arctic lake sediments.

them to sediment cores of seven similar lakes where retrogressive thaw slumps were absent. Focus-corrected sedimentation rates were ~ two-fold higher in lakes with retrogressive thaw slumps on their shores (269 ± 66 SD g/m$^2$/y) compared to lakes with no thaw slumps (120 ± 37 g/m$^2$/y) due to higher influx of new material into slump-affected lakes. OC concentrations were higher in sediments of reference lakes than in slump lakes, and Hg concentrations were negatively related to sedimentation rate in the 14 study lakes, leading to the conclusion that retrogressive thaw slump development decreased concentrations of OC and Hg in the lake sediments due loading of inorganic materials from the slumps. Korosi et al. (2015) reported that the post-1970 subsidence of permafrost-associated peatlands resulted in changes to terrestrial organic matter transport to downstream lakes but not necessarily to Hg enrichment. In a meta-analysis of lakes across Arctic Alaska, lakes with catchments susceptible to thermokarst development were more likely to have high, but variable, Hg accumulation rates (Burke et al., 2018).

Lake Hazen on Ellesmere Island (Nunavut, Canada) provides an example of dramatic climate change impacts on Hg transport recorded in its lake sediments. The Lake Hazen watershed experienced a 1 °C increase in summer air temperature between 2001 and 2012 relative to the 1986 to 2000 baseline period (Lehnherr et al., 2018). This seemingly small change in temperature has translated to wholesale changes across the watershed, including the deepening of the soil active layer, extensive melting of the watershed's glaciers and a ~ ten-fold increase in the delivery of glacial meltwaters to the lake, an up to 70% decrease in lake water residence time, and regular ice-free conditions in summer. A number of Lake Hazen sediment cores have been analyzed for Hg, OC, carbon and nitrogen (C:N) ratios, stable N and C isotopes ($\delta^{15}$N and $\delta^{13}$C), and algal (diatom) assemblages (Kirk et al., 2011; Lehnherr et al., 2018; St. Pierre et al., 2019). Sedimentation rates in Lake Hazen have increased by eight-fold relative to the 1948 baseline since the post-2007 acceleration of glacial melt across the watershed, translating to an increase in Hg accumulation rates from 20 to 180 μg/m$^2$/y. These increases have occurred concomitantly with a 1000-fold increase in OC accumulation within the sediments (Lehnherr et al., 2018). Annual Hg sedimentation rates from cores collected in Lake Hazen matched well with a THg mass balance of the lake, suggesting that the sediment cores were a particularly good archive of interannual variability in catchment inputs (St. Pierre et al., 2019).

### 3.3. River transport

Glacier melt, permafrost thaw, and precipitation changes across the Arctic have important consequences for the transport of Hg by streams and rivers. Rivers integrate changes happening throughout their catchments, acting as conduits for previously archived Hg mobilized by increased precipitation, active-layer deepening (and other modes of permafrost thaw) and glacial melt to downstream freshwater and marine ecosystems. As rivers are one of the primary sources of Hg to the Arctic Ocean (Fisher et al., 2012), such changes have potentially important implications for the transformation and accumulation of Hg in marine food webs (Schartup et al., 2015).

The dramatic transformation of Arctic landscapes in response to climate change may lead to an increase in the transport of particulate-bound Hg. The transition from sporadic to discontinuous permafrost is associated with maximal particulate-bound Hg mobilization (Lim et al., 2019). Similarly, high temporal variability in the dynamics of glacial meltwater rivers across poorly consolidated High Arctic landscapes also results in Hg fluxes dominated by the particle-bound fraction (Søndergaard et al., 2015; St. Pierre et al., 2019). The anticipated climatic effects across regions may lead to substantial changes in landscape stability, especially in regions susceptible to hillslope thermokarst (Olefeldt et al., 2016). In these areas, we might expect to see enhanced mobilization and transport of particulate-bound Hg (St. Pierre et al., 2018).

Model estimates and in-situ measurements have begun to converge on a baseline pan-Arctic riverine THg flux to Arctic marine waters (AMAP, 2021). In particular, Zolkos et al. (2020) estimated an approximately 37,000 kg/y flux from samples collected in the six major Arctic rivers (the Kolyma, Lena, Mackenzie, Ob, Yenisey, and Yukon rivers) located across the Russian and Northern American Arctic, which is comparable to the 40,000 kg/y to 50,000 kg/y output of biogeochemical models (Zhang et al., 2015; Sonke et al., 2018). This was an important first step, against which future changes in riverine exports can be compared, though similar studies evaluating MeHg exports across the Arctic remain scarce (Jonsson et al., 2022, this issue).

The importance of rivers as a source of Hg to the Arctic Ocean may increase in the future. An increase over recent decades in the discharge of major Arctic rivers is well established (e.g., Serreze et al., 2006), with Eurasian rivers showing the greatest increase. Box et al. (2019) assessed the temporal trends in discharge for six of the largest Eurasian rivers (the Ob, Pechora, Severnaya Dvina, Yenisey, Lena, and Kolyma rivers) and for the two major North American Arctic rivers (the Mackenzie and Yukon rivers). For the composite Eurasian river dataset, river flows increased on average by 18.7 km$^3$ per decade between 1981 and 2011, whereas the two North American rivers exhibited an increase of 5.9 km$^3$ per decade between 1975 and 2015. These volumetric increases correspond to a 12% increase for Eurasian rivers and a 9% increase for North American rivers over approximately the past three decades.

### 3.4. Ocean currents

The largest oceanic fluxes occur at the Fram Strait, which is located between Greenland and Svalbard (Rudels, 2012), and although there is strong seasonal variability (Tsubouchi et al., 2018), these fluxes drive the largest oceanic Hg inflow and outflow (Soerensen et al., 2016). Only little long-term changes in those Hg fluxes are expected (Petrova et al., 2020). The authors used the first Hg observations at the Fram Strait to ascertain Hg species concentrations in different water masses. The largest Hg fluxes to the Arctic Ocean are associated with the West Spitzbergen Current, 43 Mg/y via the Fram Strait and 6 Mg/y via the Barents Sea Opening. The largest outflow occurs via the East Greenland Current, 54 Mg/y. Another important outflow occurs via the Davis Strait (between Greenland and Baffin Island), 19 Mg/y. Inflow volumes from the North Atlantic side have remained relatively stable between the mid-1990s and mid-2010s, although there is a slight, non-significant, increasing trend (Østerhus et al., 2019). However, the strong seasonal cycle in water mass transport (Tsubouchi et al., 2018) likely results in varying Hg fluxes throughout the year.

It is difficult to find evidence for change in seawater Hg species composition due to the lack of consistent temporal Hg observations. Wang et al. (2012) provided the first off-shore data on the Canadian Arctic Archipelago. Heimbürger et al. (2015) provided the first four profiles of Hg in the central Arctic Ocean (with data from a 2011 cruise). Both studies reported a surface enrichment in THg and a shallow MeHg peak at the lower halocline (200 m depth compared to 1000 m in the North Atlantic), which might be responsible for the high biota Hg levels in the Arctic (AMAP, 2021). The GEOTRACES program organized the first pan-Arctic survey with two Canadian cruises (2015 Canadian Arctic Archipelago and Labrador Sea; Wang et al., 2018), two German cruises (2015 central Arctic Ocean, 2016 Fram Strait; Petrova et al., 2020) and one American cruise (2015 central Arctic; Agather et al., 2019; DiMento et al., 2019). The new Hg species data are comparable to the 2011 observations and now include dimethylmercury (DMHg) in addition to monomethylmercury (MMHg). It is not surprising to see no difference five years later because the expected changes are minor (Soerensen et al., 2016) and require longer observation time series.

Evidence from a series of permanent in situ moorings indicates the seawater volume flowing through the Bering Strait has increased significantly over the last two decades at an average rate of 1.8% per year (Østerhus et al., 2019). There is considerable interannual variability to the northward Bering flow, but the difference between the minimum in 2001 and the maximum in 2014 represented an overall 70% increase (Woodgate, 2018). The most likely explanation for this trend is lower water pressure in the East Siberian Sea, which is caused by increasing westerly winds over the past two

decades; this drives surface water from the shelf into the deeper ocean basin, thereby creating a pressure differential between the Bering Sea and the Arctic Ocean (Peralta-Ferriz and Woodgate, 2017). Because the Pacific Ocean supplies only about 10% of the total seawater inflow to the Arctic Ocean (Woodgate, 2018; Østerhus et al., 2019), the impact on Hg delivery to the Arctic Ocean as a whole from increasing Pacific inflows is likely to be minor. However, the Hg delivered could affect Hg budgets in the Chukchi and Beaufort seas, where the effects of Pacific waters are noticeable on regional water chemistry, nutrient, and heat budgets (Woodgate, 2018; Torres-Valdés et al., 2013; Haine et al., 2015).

## 4. What are the impacts of climate change on mercury biogeochemical processes?

The main biogeochemical processes affecting Hg cycling in the Arctic are redox reactions between divalent and elemental Hg, and methylation/demethylation reactions. These reactions can occur to different extents in a variety of matrices, including fresh water, seawater, snow, sea ice, soils and sediments. They may be biotic or abiotic in nature and are driven by environmental variables, such as temperature, exposure to solar radiation, organic matter, nutrients and chloride concentrations. All of these variables are expected to be modified by climate change. Conceptual descriptions and predictions of how these changes may occur have been presented in the previous AMAP assessment on Hg (AMAP, 2011) and in the follow-up paper by Stern et al. (2012). In this section, we focus on non-atmospheric studies providing new data that can better constrain these predictions. These data are summarized in Table 1.

### 4.1. Inorganic mercury redox processes

Redox processes can occur through photochemical, abiotic and microbial processes (Møller et al., 2011). These processes greatly influence interfacial fluxes because of the high volatility of Hg(0) compared to Hg(II) species. Since the last AMAP Hg assessment (AMAP, 2011), there have been few studies on redox processes in fresh water, soils, and sediments in the Arctic. In contrast, recent advances have been made regarding Hg redox cycling in snow, seawater, sea ice, and the tundra, and some of these studies considered interactions with climate change.

In snow, approximately 75% of deposited Hg is revolatilized back to the atmosphere due to Hg(II) photoreduction (Wang et al., 2017a). However, precise predictions have been hampered by a lack of kinetic data (Mann et al., 2014). Recently, experiments have been conducted with High Arctic snow samples to assess photoreduction kinetics during melting, as a function of chloride content and UV intensity (Mann et al., 2015, 2018). Increasing UV intensity had a parabolic effect on reduction rate constants of Hg in frozen and melted snow. In contrast, total photoreduced Hg (resulting from a balance between photoreduction and photooxidation) increased linearly with UV intensity. Since the snowmelt period is characterized by relatively low UV intensity, less Hg(0) should be produced and evaded at that time. These studies have also shown that chloride can enhance Hg retention in melted snow by decreasing photoreduction of Hg(0). Considering future projections for higher chloride loading in coastal snowpacks due to enhanced sea-salt deposition from thinner first-year sea ice and more open water, these experimental studies suggest that more Hg could be retained in snowpack and delivered to Arctic aquatic systems during snowmelt (Mann et al., 2018).

With respect to the Arctic marine environment, Hg(0) has been shown to significantly accumulate under contiguous ice (DiMento et al., 2019), suggesting continual under-ice net Hg reduction by yet unidentified mechanisms. Mercury evasion is therefore likely altered by ongoing and anticipated changes in the extent of sea-ice cover, with pulses of Hg evasion predicted to occur over two to three weeks when ice cover disappears (DiMento et al., 2019). One redox process occurring in the marine water column is Hg reduction by microorganisms carrying the mercuric reductase (*merA*) gene (Bowman et al., 2020). Recent studies have identified *mer* genes in the Arctic basin and shown Hg(0) production in the presence of bacterioplankton possessing *merA* (Lee and Fischer, 2019). This process could be favored in a warmer ocean.

Furthermore, sea ice itself may modify fluxes through redox processes. Indeed, atmospheric Hg speciation measurements taken over sea ice and tundra have shown that Hg emissions are higher over tundra systems (Steffen et al., 2013). These trends are consistent with suppressed

**Table 1**
Anticipated effects of climate change on mercury biogeochemical transformations in Arctic environments, according to recent studies.

| Biogeochemical transformations | Matrices | Specific process(es) | Key variables considered | Effect(s) of climate change | References |
|---|---|---|---|---|---|
| Redox transformations | Snow and sea ice | Photoreduction and oxidation | Chloride and UV | Lower photoproduction of Hg(0)<br>Retention of Hg(II) | Mann et al. (2015, 2018); Steffen et al. (2013) |
| | Seawater | Hg(II) reduction under ice<br>Bacterial reduction (*merA* gene) | Sea-ice cover<br>Temperature | Pulses of Hg(0) evasion with sea-ice loss<br>Increased Hg(0) production | DiMento et al. (2019) |
| | Tundra | Hg(0) uptake by plants | Vegetation cover and length of growing season | Increased accumulation of atmospheric Hg by plant uptake in the tundra | Obrist et al. (2017) |
| Methylation and demethylation | Freshwater systems | Sediment methylation | $\delta^{201}Hg/\delta^{199}Hg$ ratios and changes in microbial communities<br>Temperature | Lower Hg methylation during past warming events<br>Higher net methylation in warmer sediments | Jackson (2019); Hudelson et al. (2020) |
| | | Methylation in hypolimnia and sediments | Change in thermocline depth | Change in volume of water and sediments where methylation occurs | Perron et al. (2014) |
| | | Photodemethylation | DOC | Increased demethylation in clear lakes/decreased demethylation in high DOC systems | Girard et al. (2016) |
| | Seawater (surface, subsurface) | Net methylation | Hg(II)<br>Organic matter mineralization<br>Nutrients from coastal rivers<br>Shift in plankton communities<br>Stratification | Increased net methylation | Wang et al. (2012, 2018); Heimbürger et al. (2015); Schartup et al., 2015) |
| | | Photodemethylation | UV penetration<br>Sea-ice cover | Increased photodemethylation | Point et al. (2011); DiMento and Mason (2017) |
| | Snow, first-year sea ice, multi-year sea ice | Net methylation | Loss of multi-year sea ice<br>Increased melting of sea ice and snow | Increased MeHg flux to the ocean | Beattie et al. (2014) |
| | Marine sediments | Net methylation | Temperature | Increased net methylation | St. Pierre et al. (2014) |
| | Arctic soils | | Temperature<br>Permafrost thaw | Increased net methylation | Yang et al. (2016) |

photoreduction in chloride-containing snow over sea ice. Climate change is also causing a decrease of multi-year ice, with saltier first-year ice being favored. This could result in more Hg being concentrated in sea ice from the underlying seawater due to freeze rejection and from the overlying snow if AMDEs are enhanced (Steffen et al., 2008; Chaulk et al., 2011; Beattie et al., 2014; Wang et al., 2017a). However, our understanding of redox and complexation processes in sea ice and sea-ice brines is too limited to make climate-related projections at this point (Wang et al., 2017a).

Ocean acidification could also influence Hg cycling in the Arctic, although no specific field study has addressed this question. In seawater, solubility, adsorption and redox processes of metals may be influenced by decreases in hydroxide ($OH^-$) and carbonate ($CO_3^{2-}$) concentrations (Millero et al., 2009). In the case of Hg(II), for which seawater speciation includes strong complexes with chloride, little change in speciation linked to acidification is anticipated, since the change in pH will not influence chloride concentrations (Millero et al., 2009). Recent laboratory studies have shown that ocean acidification could alleviate Hg toxicity in marine copepods not through change in Hg speciation, but rather through alteration of physiological processes (Li et al., 2017; Wang et al., 2017b).

For terrestrial systems, there is now evidence that Hg deposition on the Arctic tundra is controlled by the uptake of Hg(0) by plants (Obrist et al., 2017). From a mass balance analysis, it was determined that 70% of Hg in the interior tundra is derived from Hg(0), and that this Hg(0) deposition peaked during the summer. This important uptake by plants has been confirmed by Hg stable isotope analyses (Obrist et al., 2017). It follows that a climate-related increase in vegetation cover and the length of the growing season may lead to higher Hg deposition over the tundra (Zhou et al., 2021). Also, the production and evasion of Hg(0) in this landscape (for instance, in rivers, lakes, and ponds) could be redeposited nearby and taken up by vegetation.

### 4.2. Methylmercury production and decomposition

Mercury methylation occurs via microbial processes. In anoxic environments (or in anoxic niches in oxic environments, such as marine particles), this methylation is controlled by the *hgcAB* gene cluster (Parks et al., 2013) which is present, among others, in some iron and sulfate reducing bacteria and in methanogens (Gilmour et al., 2018). However, the methylation capacity of *hgc*-like genes in the ocean is uncertain (Bowman et al., 2020). Mercury demethylation can result from photochemical or microbial pathways. Microbial demethylation can occur through either an oxidative or a reductive mechanism (Lehnherr, 2014). The main recent climate-related advances on Arctic methylation and demethylation have focused on marine and freshwater systems.

In fresh waters, microbial Hg methylation and demethylation occur mostly in anoxic hypolimnia and sediments, whereas photodemethylation is confined to the upper portion of the water column (Lehnherr, 2014). Climate-related changes in thermal regimes could alter the temperature-dependent methylation and demethylation rates in sediments. A paleolimnological study of Arctic sediment cores used $\delta^{201}Hg/\delta^{199}Hg$ ratios to estimate past MeHg production in lakes during the last 100 years (Jackson, 2019). According to this research, done on one core from one Arctic lake, the isotope ratio decreased during warming phases of climate change. Therefore, during warming events, net sediment methylation would be lower, presumably because of changes in the microbial community structure linked to changes in the phytoplanktonic communities providing nutrients. More research is warranted to see if such a trend holds for different types of freshwater systems especially along environmental gradients. With respect to the direct effect of temperature on the balance between methylation and demethylation, it has been recently experimentally shown that net methylation is increased when sediment cores are warmed (Hudelson et al., 2020). Thermal shifts may also alter the depth of the thermocline of stratified systems, and therefore the volume of sediments and hypolimnetic waters where Hg methylation usually takes place. The effect of thermocline deepening has been assessed through whole-ecosystem experiments on lower latitude systems (Perron et al., 2014); such studies have shown that deeper thermoclines are associated with a smaller volume of MeHg-rich hypolimnion and an important decrease in MeHg levels along the food web.

In the water column, changes in DOC loadings have been predicted to increase epilimnetic MeHg levels by impeding photodemethylation, based on lower latitude studies (Poste et al., 2015; Klapstein et al., 2018). In contrast, field experiments conducted in Arctic freshwater systems with contrasting DOC levels suggest a non-linear, unimodal relationship between DOC and photodemethylation rates (Girard et al., 2016), similar to the bell-shaped relationship previously described between DOC and Hg bioaccumulation (French et al., 2014; Braaten et al., 2018). As a result, increase in DOC inputs from thawing permafrost and other mechanisms could increase photodemethylation in clear lakes and decrease it in browner systems. Climate change is also promoting the formation of shallow thaw ponds that can become transient hotspots of MeHg production in the landscape (MacMillan et al., 2015; Tarbier et al., 2021).

In marine systems, the primary local methylation site appears to be in the water column rather than the sediments, except near coastal areas (Jonsson et al., 2022, this issue). MeHg production in Arctic seawater is thought to be tightly linked to mineralization of organic matter (Wang et al., 2012; Heimbürger et al., 2015), although the methylation mechanism remains unknown. Hence, climate change-induced increases in primary productivity in the Arctic Ocean, which would stimulate organic matter remineralization, will likely lead to higher net MeHg production. Such an increase in MeHg production is also predicted for shallow marginal sea-ice zones because of intensified stratification and shifts in plankton dynamics (Heimbürger et al., 2015). Coastal MeHg production may also be exacerbated by increased nutrient inputs from coastal rivers (Schartup et al., 2015). However, large uncertainties remain regarding production and loss mechanisms (e.g., changes in demethylation rates) (Wang et al., 2018, 2020). In particular, MeHg photodemethylation could be promoted by loss of sea ice and increased irradiation of surface waters (Point et al., 2011), especially in the open ocean where UV penetration is maximal (DiMento and Mason, 2017).

In addition to the water column, MeHg production may also occur in other marine matrices. It can be produced in snow, first- and multi-year sea ice, and accelerated melting of sea ice may release significant fluxes of MeHg into the Arctic Ocean (Beattie et al., 2014; Schartup et al., 2020). Recent measurements suggest first year sea ice may also be a source of DMHg (Schartup et al., 2020). Production of MeHg in coastal sediments can be significant, and the effect of increased temperature in shallow sediments could lead to increased net methylation rates as reported for lakes (Hudelson et al., 2020), with methylation being more sensitive to these temperature changes than demethylation according to experiments with slurries of marine sediments (St. Pierre et al., 2014).

Experimental warming studies have also been conducted on homogenized Arctic soil, and they have similarly concluded that permafrost thaw from climate warming could enhance MeHg production by an order of magnitude (Yang et al., 2016).

## 5. Conclusions and recommendations

Conclusions (in numbered bullets) are organized under section headings followed by recommendations in italics where appropriate.

### 5.1. How has climate change affected the physical and biogeochemical characteristics of Arctic environments?

1. Climate change, at local and regional scales, is profoundly affecting physical and biogeochemical characteristics that interact with the Hg cycle across all Arctic environmental compartments.
2. Atmospheric conditions are changing with warmer air temperatures in winter and summer, greater precipitation, and a shift from snow to rain.
3. Ocean circulation and particularly Arctic sea-ice conditions are changing dramatically, including reduced extent, thickness, and snow depth

over ice, altered timing, increased mobility, loss of multi-year ice, and an increase in the extent of halogen-rich first-year ice.
4. In the terrestrial environment, warmer and wetter conditions are leading to permafrost thaw and glacier melt, as well as changes in soil and hydrological processes; greater evaporation and drought conditions are resulting in an increase in the severity and extent of wildfires.
5. Earlier ice and snow melt and warmer temperatures are altering lake stratification, while altered hydrology and catchment characteristics are increasing the transport of organic matter to downstream water bodies.

*5.2. What influence has climate change had on mercury transport processes?*

6. Modeling results suggest that climate change may be impacting atmospheric Hg deposition in the Arctic. However, process-focused information on interactions between climate and Hg deposition is lacking, and current Hg models may not represent the complexity of those climate-sensitive processes.
7. Arguably the clearest evidence of current climate change effects on Hg cycling in the Arctic is for Hg transport from terrestrial catchments. Widespread permafrost thaw, glacier melt and coastal erosion are altering the sources and export of Hg from Arctic catchments. Changes in catchment transport processes are being reflected in Hg accumulation rates in lake sediment cores, especially in watersheds undergoing dramatic change as a result of permafrost thaw and glacial melt. Greater organic matter transport, particularly dissolved organic carbon, may also be influencing the downstream transport and fate of Hg in Arctic freshwater ecosystems.
8. Recent estimates suggest that Arctic permafrost is a large global reservoir of Hg, which is vulnerable to degradation under a warming climate during the 21st century. The fate of permafrost soil Hg is not well understood.
9. Alterations to the terrestrial landscape including the development of thermokarst features, the formation and expansion of thaw lakes, and increased soil erosion will result in greater transport of particulate-bound Hg to Arctic rivers. In-stream processing and deposition processes will ultimately determine whether particulate Hg travels downstream to estuaries or is stored more locally in river and stream sediments.
10. Increasing severity and incidence of wildfires both locally within the Arctic and across boreal regions may be contributing to the atmospheric pool of Hg. Long-term implications of climate change interactions with wildfires are not well understood.
11. Limited information is available on recent climate change-driven alterations in transport of Hg by ocean currents.

*Atmospheric deposition of Hg may be affected by climate change and further research to identify key climate influences on this flux is a priority. Changing winter and spring conditions and an increase in the presence of halogens in the lower atmosphere and on snow and ice surfaces are important factors to include in future modeling.*

*Long-term monitoring of Hg in Arctic terrestrial environments at the catchment scale would improve the detection of changes in Hg transport due to climate warming. Quantification of Hg sources and downstream fate (e.g., lake sediments) at the catchment scale should be combined with Hg transport in large rivers, which reflects processes occurring across Arctic catchments.*

*Improved geographic coverage for measurements and modeling of changes in Hg transport associated with thawing permafrost, melting glaciers, and wildfires is a priority to better constrain implications for the Arctic Hg cycle. Interactions between climate and Hg transport in the Arctic Ocean remains a major knowledge gap that warrants further research.*

*5.3. What are the impacts of climate change on mercury biogeochemical processes?*

12. Interactions with climate change remain poorly understood for many aspects of Hg biogeochemical cycling in the Arctic.
13. Redox transformations of inorganic Hg affect fluxes between the atmosphere and terrestrial or aquatic environments. Changes to the cryosphere, particularly reduced sea-ice cover and higher chloride in snow, are likely to alter the seasonal evasion or retention of inorganic Hg.
14. Mercury deposition on the Arctic tundra is controlled by uptake of Hg (0) by plants. Greater vegetation-cover and length of the growing season on the tundra may increase the accumulation of atmospheric Hg by plant uptake.
15. Experimental evidence indicates warmer temperatures can enhance MeHg production in freshwater and marine sediments as well as in tundra soils.
16. Evidence from Hg stable isotopes in the marine environment and lake experiments suggests the photochemical breakdown of MeHg will be enhanced in seawater due to sea-ice loss and altered in fresh waters due to changes in dissolved organic carbon loading.

*Further investigation of climate change impacts on redox transformations of inorganic Hg and the production of MeHg is a priority to better constrain long-term changes in Hg bioavailability to food webs. In situ process measurements and the development of mechanistic/stochastic models that couple Hg transport and transformations with climate models would allow projections under various climate change scenarios in the Arctic. The effects of increasing vegetation cover and length of the growing season on plant uptake of atmospheric Hg(0) are a priority for research because of the potential for enhanced Hg accumulation in the Arctic tundra.*

## CRediT authorship contribution statement

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## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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