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1 Review

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3 Arctic Methylmercury Cycling

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27 Abstract

28

29 Anthropogenic mercury (Hg) undergoes long-range transport to the Arctic where it is transformed into
30 methylmercury (MeHg), potentially leading to high exposure in some Arctic inhabitants and wildlife.
31 The environmental exposure of Hg is determined not just by the amount of Hg entering the Arctic, but
32 also by biogeochemical and ecological processes occurring in the Arctic. These processes affect MeHg
33 uptake in biota by regulating the bioavailability, methylation and demethylation, bioaccumulation and
34 biomagnification of MeHg in Arctic ecosystems. Here, we present a new budget for pools and fluxes
35 of MeHg in the Arctic and review the scientific advances made in the last decade on processes leading
36 to environmental exposure of Hg. Methylation and demethylation are key processes controlling the
37 pool of MeHg available for bioaccumulation. Methylation of Hg occurs in diverse Arctic

38 environments including permafrost, sediments and the ocean water column, and is primarily a process
39 carried out by microorganisms. While microorganisms carrying the *hgcAB* gene pair (responsible for
40 Hg methylation) have been identified in Arctic soils and thawing permafrost, the formation pathway of
41 MeHg in oxic marine waters remains less clear. Hotspots for methylation of Hg in terrestrial
42 environments include thermokarst wetlands, ponds, and lakes. The shallow sub-surface enrichment of
43 MeHg in the Arctic Ocean, in comparison to other marine systems, is a possible explanation for high
44 MeHg concentrations in some Arctic biota. Bioconcentration of aqueous MeHg in bacteria and algae
45 is a critical step in the transfer of Hg to top predators, which may be dampened or enhanced by the
46 presence of organic matter. Variable trophic position has an important influence on MeHg
47 concentrations among populations of top predator species such as ringed seal and polar bear
48 distributed across the circumpolar Arctic. These scientific advances highlight key processes that affect
49 the fate of anthropogenic Hg deposited in Arctic environments.

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54 **1. Introduction**

55 Mercury (Hg) is a bioaccumulative neurotoxin that is released from the earth's crust through natural
56 processes and anthropogenic activities (Streets 2019). Since the industrial revolution, direct
57 anthropogenic emissions have exceeded natural emissions by an order of magnitude and concentration
58 of Hg in the atmosphere has increased 3-5 times (Driscoll et al. 2013, Li et al 2020). Since Hg is well
59 mixed globally in Earth's atmosphere these emissions have also reached the Arctic environment. Due
60 to a variety of physical, chemical, and biological processes, some of which are unique to environments
61 in cold regions, high levels of Hg exposure have been detected among Arctic communities and
62 wildlife (AMAP 2021).

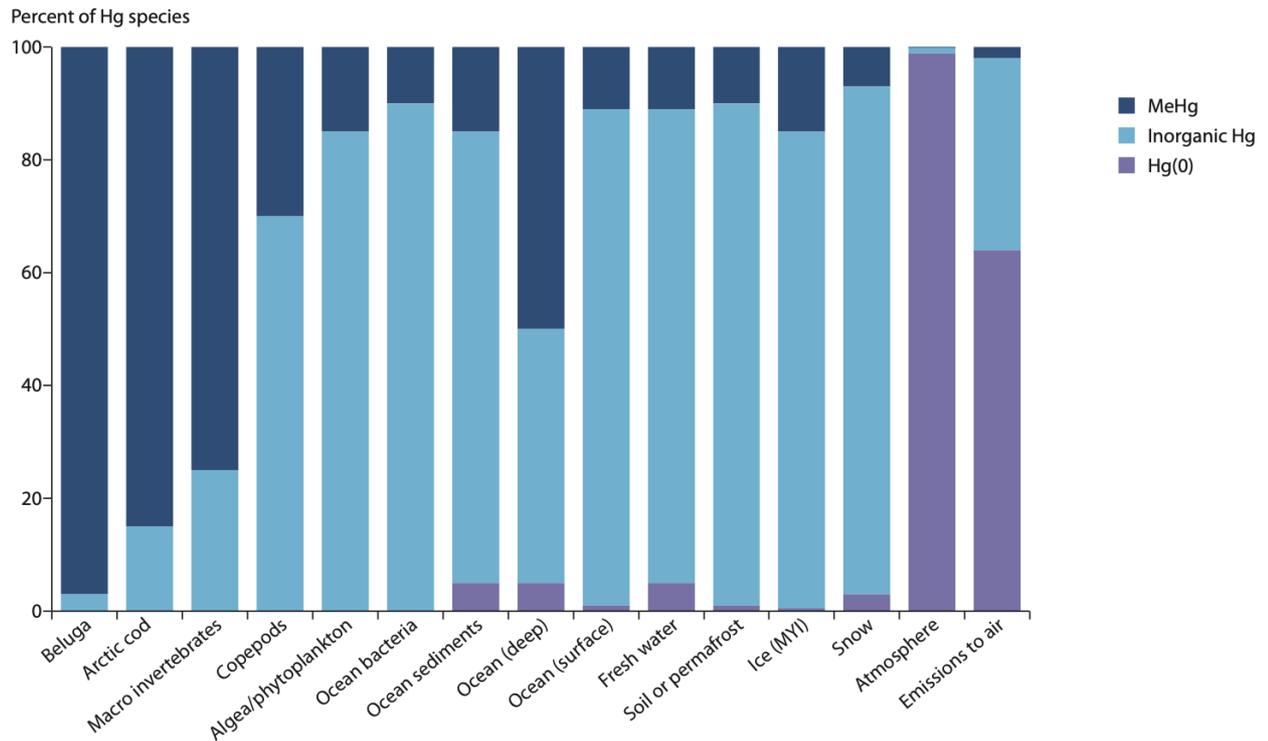
63 The biogeochemical cycle that connects Hg emissions to Hg in Arctic food webs involves inorganic
64 and organic forms of Hg that undergo a wide range of transformation processes in air, water, and land.
65 There are three main chemical forms of Hg in Arctic environmental compartments and biota: gaseous
66 elemental mercury (Hg(0)), inorganic divalent mercury (Hg(II)) and MeHg. Figure 1 shows the
67 partition of each in the major Arctic environmental compartments and biota.

68 Industrial activities (e.g., artisanal and small-scale gold mining, coal-fired power stations, waste
69 incineration, cement manufacture, and others) release both Hg(0) and Hg(II). The main natural Hg
70 sources to the atmosphere are volcanoes and forest fires. While Hg(II) is typically deposited closer to
71 the emission source, Hg(0) has a residence time ranging from several months to over a year in the
72 atmosphere and can be transported from the emission source to remote polar regions within a few days
73 to weeks (Durnford et al, 2010, Dastoor et al. 2015). Atmospheric mercury depletion events, during
74 polar sunrise occur in both Arctic and Antarctic regions and lead to the rapid oxidation of Hg(0) to the
75 more easily deposited Hg(II) species. A large fraction of Hg(0) also dissolves in natural waters where
76 it is referred to as dissolved gaseous mercury (DGM) (Jiskra et al. 2021). On land, foliar (plant) uptake
77 of Hg(0) drives the atmospheric deposition of Hg in the Arctic tundra (Obrist et al., 2017; Jiskra et al.
78 2018; Douglas and Blum, 2019). Through this process, large amounts of Hg have accumulated in
79 northern permafrost soils from the atmosphere over thousands of years. Today, concerns are raised

80 about the potential remobilization of this permafrost Hg pool as climate warming leads to increased
81 soil temperatures, thawing permafrost, and changes in the hydrological cycle (Schaefer et al., 2020,
82 Lim et al. 2020). In natural waters, soils and sediments, Hg is primarily found as Hg(II) (Skylberg,
83 2012). Due to a high affinity for particles (e.g. mineral surfaces and organic matter), most Hg(II) in
84 sediments and soils is found in the solid phase and these systems commonly act as important sinks for
85 Hg. At the same time, these systems are known to be important sites for Hg methylation. The main
86 form of MeHg existing in natural environments is monomethylmercury (MMHg), although, in some
87 environments, dimethylmercury (DMHg) may become the dominant species, especially in seawater.
88 Since most analytical data on methylated Hg do not differentiate MMHg and DMHg, we will use
89 MeHg to refer to methylated Hg in general from hereon, unless otherwise specified. While both
90 MMHg and DMHg are toxic and lipophilic, only MMHg has been shown to biomagnify in aquatic
91 food webs to concentrations of concern for wildlife and human health (Lavoie et al., 2013). Because of
92 this, the potential risk Hg pollution poses to Arctic ecosystems is not only controlled by the amount of
93 inorganic Hg transported into the system and cycling within the Arctic environment, but also by the
94 extent of the Hg pool that is methylated and accumulated in aquatic food webs.

95 In this review and contribution to the Virtual Special Issue on Arctic mercury, we present a synthesis
96 of work conducted for the AMAP Assessment 2021: Mercury in the Arctic on the processes affecting
97 mercury transformation and biotic uptake in the Arctic (AMAP 2021, Chapter 4). Given the high
98 complexity of the Hg biogeochemical cycle, it is not surprising that significant advances are still being
99 made despite over 50 years of active research. Here, we focus on scientific progress made in the last
100 decade on processes leading to environmental MeHg exposure by presenting (1) a new mass balance
101 for MeHg in the Arctic, and new information on (2) our mechanistic understanding of Hg
102 (de)methylation processes, (3) where hotspots of MeHg are found in the Arctic and, finally, (4)
103 processes affecting Hg accumulating in Arctic biota.

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107 **Figure 1.** Partition of different Hg species (Hg(0), inorganic divalent Hg(II) and MeHg) in
 108 emissions and in various Arctic environmental compartments and biota, expressed as
 109 percentages of total Hg (adopted from AMAP 2011, Douglas et al., 2012; Cossa et al., 2014;
 110 Schartup et al., 2020).

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112 **2. How much methylmercury is circulating in the Arctic**

113 **environment?**

114

115 A summary of MeHg concentrations, fluxes and estimated MeHg pools in different environmental
 116 compartments used for this mass balance is presented in Table 1, Table 2 and Figure 2. The MeHg
 117 budget for the Arctic Ocean was first estimated by Soerensen et al. (2016b) and later revised by
 118 Petrova et al. For the AMAP 2021 Hg assessment, THg (Dastoor et al. 2022) and MeHg (this review)

119 budgets were calculated for land areas north of 60°N and the Arctic Ocean, including the central basin,
120 and the Barents, Kara, Laptev, East Siberian, Chuckhi and Beauford Seas.

121 To estimate the stock for the 0-20 m, 20-200 m, 200-500 m and <500 m water masses (Table 2)
122 average concentrations of THg and MeHg in seawater calculated from available data (Wang et al.,
123 2012b, 2018; Heimbürger et al., 2015; Agather et al., 2019; Charette et al., 2020; Kim et al., 2020b;
124 Petrova et al., 2020; Jonsson et al., 2022, Kohler et al., 2022) were used. For biota, we used the
125 estimate previously provided by Soerensen et al., 2016b. Remaining reservoirs (soil, snow, ice and air)
126 were estimated from the stocks of Hg (Dastoor et al. 2022; Table 3.8.1 in AMAP (2021), key data for
127 this review are also shown in Table S1) and the best estimates of %MeHg for each environmental
128 compartment (Table 1 and S1). The MeHg pool in surface soils was estimated by multiplying the total
129 Hg pool by the median %MeHg in surface soils (St. Pierre et al., 2015: 0.82%) and wetlands (Tarbier
130 et al., 2021: 1.7%, not impacted by recent permafrost thaw) and assuming a 25% coverage of wetlands
131 for the Arctic Region (Kåresdotter et al., 2021). Methylmercury reservoirs in snowpack and glaciers
132 were estimated using %MeHg measurements for snow (6.1%, range: 0.11-11%; St. Pierre et al., 2019,
133 Varty et al., 2021 and Jonsson et al., 2022) and median %MeHg (5.5 %) reported by Zdanowicz et al.,
134 (2016) for glaciers, respectively. For sea ice, the mean %MeHg reported by Dimento et al., (2019) was
135 used, resulting in an estimated MeHg pool of 0.32 Mg. This value is close to the estimate of MeHg
136 (0.33 Mg) previously estimated by Schartup et al. (2020). For the atmosphere, %MeHg were
137 calculated from the concentrations of MMHg and DMHg (Table 1) previously reported by Baya et al.
138 (2015) and total Hg concentration reported by Dastoor et al. (2022).

139 Using geochemical modelling, Soerensen et al., (2016b) estimated MeHg reservoirs in the Arctic
140 Ocean (also including the Canadian Arctic Archipelago and the Baffin Bay), sediments and plankton
141 resulting in a total of 576 Mg. In the updated MeHg budget presented by Petrova et al. (2020), the total
142 pool of MeHg in the Arctic Ocean was estimated to 800 Mg MeHg. Our budget for the Arctic Ocean
143 water sums up to 590 ±341 Mg MeHg and is thus in the same range as earlier estimates.

144 Petrova et al. (2020) found a MeHg maxima in the subsurface waters (100–300 m) of the Arctic Ocean
145 (in line with observations from Wang et al. (2012b, 2018), Heimbürger et al. (2015), Agather et al.

146 (2019), Jonsson et al. (2022)), leading to a net export of MeHg from the Arctic Ocean to the Nordic
147 Seas and the North Atlantic of 7.5 Mg/yr. A part of the MeHg produced in the subsurface ocean layer
148 (20–200 m; 17 Mg/yr) is converted to DMHg, which diffuses up to the polar mixed layer (PML; 0–20
149 m), where some of it evades to the atmosphere. Here DMHg is proposed to rapidly photochemically
150 degrade back to MMHg and deposit back to the PML. For the AMAP 2021 report, MeHg (MMHg +
151 DMHg) evasion was estimated to 4.5 (2.8–6.3) Mg/yr using a DMHg to Hg⁰ evasion ratio of 0.16
152 (Soerensen et al, 2016b) and the new Hg⁰ evasion estimate (32 (20–45) Mg/yr (Table 3.8.1 in AMAP
153 (2021)). The MeHg deposition was then calculated to 2.6 Mg/yr assuming the evasion to deposition
154 ratio (0.57) presented by Soerensen et al. (2016b). In addition, DMHg may be photochemically
155 degraded in surface waters to MMHg (West et al. 2022). However, as our budget deals with the total
156 stock of MeHg, transformation reactions between methylated Hg species within compartments are not
157 considered here. Riverine MeHg inputs (0.8 Mg/yr) were estimated from Sonke et al., (2018) and
158 equal inputs by coastal erosion was assumed (as done for Hg, Table 3.8.1 in AMAP (2021)). The
159 benthic flux of MeHg to subsurface waters was estimated to 0.9–4.6 Mg/yr (Soerensen et al., 2016b,
160 Kim et al., 2020b). The *in situ* net water column methylation (5.1 Mg/yr) was estimated by calculating
161 the difference between estimated inputs (total input: 18.4 Mg/yr; rivers: 0.8 Mg/yr; coastal erosion: 0.8
162 Mg/yr; atmospheric deposition: 2.6 Mg/yr; oceans: 9 Mg/yr; sediment: 4.7 Mg/yr, sea ice: 0.5 Mg/yr)
163 and outputs (total output: 23.5 Mg/yr; atmospheric evasion: 4.5 Mg/yr; ocean inputs: 16.5 Mg/yr;
164 sediment: 1.4 Mg/yr, sea ice: 0.4 Mg/yr; plankton uptake 0.7 Mg/yr). Our *in situ* net water column
165 methylation for the Arctic Ocean is close to the *in situ* net water column methylation estimates
166 provided from earlier models (Kim et al., 2020: 10 Mg/yr; Soerensen et al., 2022: 15 Mg/yr for the
167 upper 200 m). Our estimated *in situ* production corresponds to a daily net Hg methylation potential of
168 $7.5 \cdot 10^{-6} \text{ d}^{-1}$ (calculated using the water column total Hg budget estimate (1,870 Mg Hg) from Dastoor
169 et al., 2022). This calculated net Hg methylation potential (net result of *in situ* Hg methylation and *in*
170 *situ* MeHg demethylation processes) is three orders of magnitude lower than the potential Hg
171 methylation rate constants ($0.6\text{--}13 \cdot 10^{-3} \text{ d}^{-1}$) reported from incubation studies by Lehnherr et al., (2011).

172 The Arctic MeHg budget highlights the importance of several processes that control MeHg reservoirs.
173 Specifically, MeHg production in the water column and in shelf sediments are dominant inputs to the
174 Arctic Ocean (Figure 2). Atmospheric deposition and marine evasion also represent large fluxes of
175 MeHg, with a net deposition flux. Similarly, fluxes of MeHg via oceanic currents are large, with an
176 estimated net export from the Arctic Ocean to the North Atlantic. The modelling of estimates for pools
177 and fluxes is based on existing data, which have varying degrees of uncertainty. In particular, our *in*
178 *situ* net Hg methylation potential is associated with high uncertainty, although the much lower *in situ*
179 net methylation used in our model (in comparison to potential methylation rates (k_m) values available)
180 suggests a rapid turnover of MeHg in Arctic waters (Lehnherr et al., 2011 reported potential
181 demethylation rates of 0.2-0.6 d⁻¹). Some budget estimates are based on a "best estimate" approach
182 (when no data was available) or a very limited set of observations. Therefore, improved spatial and
183 temporal coverage of measurements is important to improve estimates of MeHg fluxes, including from
184 meltwater and erosion.

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Table 1. Total MeHg concentrations, fluxes and pools in the Arctic environment (excluding aquatic biota larger than plankton). The total MeHg concentrations reported cover both freshwater and the marine compartments, while the fluxes focus on those relevant for the Arctic Ocean.

| Environmental Compartment | MeHg Concentration | %MeHg | Description | Region | References |
|--|--|-----------|---|--------------------|-------------------------|
| Atmosphere | 2.9±3.6 pg/m ³ MMHg 3.8±3.1 pg/m ³ DMHg | <1% | | Hudson Bay and CAA | Baya et al., 2015 |
| Soil | 0.02–2.11 ng/g (d.w) | 0.82% | | Canada | St. Pierre et al., 2015 |
| | 0.5 and 3.4 ng/g (d.w) | 1% | Tundra (organic horizon) (n=2) | Alaska | Olson et al., 2018 |
| | 0.1 and 1.0 ng/g (d.w) | 3% | Tundra (B-horizon) (n=2) | Alaska | Olson et al., 2018 |
| | 0.17±0.55 ng/g (d.w) | 0.48±1.7% | Peat plateaus underlain by permafrost (upper 50 cm, n=4) | Subarctic Sweden | Tarbier et al., 2021 |
| | 1.8±4.7 ng/g (d.w) | 6.1±6.5% | Wetlands forming due to permafrost thaw (upper 50 cm, n=5) | Subarctic Sweden | Tarbier et al., 2021 |
| | 0.78±1.2 ng/g (d.w) | 1.7±2.4% | Wetlands not impacted by permafrost thaw (upper 50 cm, n=5) | Subarctic Sweden | Tarbier et al., 2021 |
| | 0.11–6.71 ng/g (d.w) | 0.3–17% | Wetland soils | Canada | Varty et al., 2021 |
| | 0.08±0.01 ng/g (d.w) | 0.1% | Organic layer | Alaska | Yang et al., 2016 |
| | 0.07±0.01 ng/g (d.w) | 0.1% | Mineral layer | Alaska | Yang et al., 2016 |
| Soil pore water/wetland and stream water | 0.43 ng/L | 11 | Poor fens (recent permafrost thaw) | | Gordon et al., 2016 |
| | 0.01–0.05 ng/L | | Palsas underlain by permafrost | Subarctic Sweden | Fahnestock et al., 2019 |
| | 1.0–2.0 ng/L | | Wetlands formed due to permafrost thaw | Subarctic Sweden | Fahnestock et al., 2019 |
| | 0.23 and 0.51 ng/L | Up to 20% | Wetlands | Canada | Varty et al., 2021 |
| | 0.216 ± 0.020 ng/L to 3.45 ± 1.45 ng/L | 0.47–9.9% | Downstream permafrost thaw slump | Canada | St Pierre et al., 2018 |
| Rivers | 0.04–0.18 ng/L | | | | Table S2 |
| Lakes | 0.02–0.65 ng/L | | | | Table S2 |
| Ponds | <0.02–0.53 ng/L | | | | Table S2 |

| | | | | | |
|----------------------|-----------------------------------|---------------------------|---------------------------|---|---|
| Glaciar ice and firn | 0.1-0.7 ng/L | 1-17% (median 5.5%) | Firn and ice Core Samples | Canada | Zdanowicz et al., 2016 |
| Glacial meltwater | <0.010–0.235 ng/L | | Unfiltered | | Table S3 |
| | <0.010–0.075 ng/L | | Filtered | | Table S3 |
| Lichen | 1.41–17.1 ng/g | | High Arctic coast | Canada | St. Pierre et al., 2015 |
| Seawater | 0.018±0.016 ng/L | 12 ± 6% | | Western Arctic Ocean | Agather et al., 2019 |
| | MMHg: 0.018±0.010 ng/L (n=162) | | | | |
| | DMHg: 0.008±0.006 ng/L | | | | |
| | (MMHg:DMHg ~ 1.38) | | | | |
| | 0.036 ±0.024 ng/L | | | Beaufort Sea | Wang et al., 2012b (only stations included for this review) |
| | 0.048 ±0.028 ng/L | 18±12% | | Canadian Arctic | Wang et al., 2018 (only stations included for this review) |
| | 0.045±0.017 ng/L | | | Western Arctic Ocean | Heimbüger et al., 2015 |
| | 0.034±0.022 ng/L | 30-33% | | Eastern Arctic Ocean | Heimbüger et al., 2015 |
| | 0.027±0.019 ng/L | 41±38% | | Western and eastern Arctic Ocean | Jonsson et al., 2022 |
| | MMHg 0.018±0.016 ng/L | | | | |
| | DMHg 0.006±0.007 ng/L | | | | |
| | (MMHg:DMHg ~ 1:2) | | | | |
| | 0.013±0.004 ng/L | | | Barents Sea | Petrova et al., 2020 |
| | 0.049±0.03 ng/L | | | Opening/shelf | |
| | MMHg 0.019±0.013 ng/L | 24±14% | | Fram Strait | Petrova et al., 2020 |
| | DMHg 0.039±0.028 ng/L | | | | |
| | (MMHg:DMHg ~ 2:1) | | | | |
| | 0.075±0.019 ng/L | | | Fram Strait outflow (East Greenland Current) | Petrova et al., 2020 |
| | 0.020±0.012 ng/L (n=122) | 9% | 0-20 m | Arctic Ocean (this review) | |

| | | | | |
|--------------------------|-----|-----------|--|---|
| 0.040±0.026 ng/L (n=439) | 21% | 20-200 m | | |
| 0.046±0.028 ng/L (n=177) | 32% | 200-500 m | | |
| 0.042±0.021 ng/L (n=288) | 30% | >500 m | | Wang et al., 2012, 2018; Heimbürger et al., 2015; Agather et al., 2019; Charette et al., 2020; Kim et al., 2020b; Petrova et al., 2020, Jonsson et al., 2022; Kohler et al., (2022) |

| | | | | | |
|------------------------------------|------------------|--------------|--|--|---|
| Shelf sediment | 0.15–0.37 ng/g | 0.43±0.17% | | Beaufort Sea and Chukchi Sea | Fox et al., 2014 |
| | 0.2 ng/g | | | Western and eastern Arctic Ocean (<i>data synthesis</i>) | Soerensen et al., 2016b |
| | 0.019-2.4 ng/g | 0.06%–2.6% | High MeHg observed close to the Lena river delta | Laptev Sea | Liem-Nguyen et al., 2022 |
| | 0.002-0.168 ng/g | 0.003%–0.27% | | Eastern Siberian Sea | Kim et al., (2020) |
| Basin sediment | 0.05 ng/g | | | Western Arctic Ocean | Soerensen et al., 2016b |
| Plankton (freshwater) | 0.03 ng/g | | Phytoplankton | Lake Melville, Canada | Schartup et al., 2015a |
| | <0.4–30 ng/g | | Water column seston | | Chételat et al., 2018 |
| | 10-99 ng/g | | Zooplankton | Skeleton Lake, Canada | Varty et al. (2021) |
| Plankton (marine) | <1.5 ng/g | | Phytoplankton | Chukchi Sea | Fox et al., 2017 |
| | 8–96 ng/g | | | Circumpolar Arctic | Foster et al., 2012; Pućko et al., 2014; Pomerleau et al., 2016 |
| Biofilms/microbial mats/Periphyton | 2–12 ng/g | | Shoreline rock biofilm, lakes | Lakes in East Canadian Arctic | Chételat et al., 2018 |
| | 0.3–1.3 ng/g | | Microbial mats, lakes | Lakes in East Canadian Arctic | Camacho et al., 2015 |
| | 2–56 ng/g | | Periphyton, filamentous algae | Lakes in East Canadian Arctic | van der Velden et al., 2013a |
| Snow | 0.02–0.07 ng/L | 0.2-10% | Surface snow | <i>data synthesis</i> | Soerensen et al., 2016b |
| | <0.01–0.074 ng/L | 21-31% | Snowpack | <i>data synthesis</i> | Soerensen et al., 2016b |

| | | | | |
|-------------------|-----------|--|----------------------|-------------------------|
| 0.14-9.1 ng/L | 1.1-0.74% | Snow (unfiltered) | Lake Hazen, Canada | St. Pierre et al., 2019 |
| 0.039-1.2 ng/L | 13-42% | Snow (filtered) | | |
| 0.002±0.0002 ng/L | 0.41±49% | Surface snow from sea ice | Arctic Ocean | DiMento et al., 2019 |
| 0.19-0.67 ng/L | 8-11% | | Canada (High Arctic) | Varty et al., 2021 |
| 0.005-0.008 ng/L | 1.5-5.1% | Surface snow from sea ice (unfiltered) | Arctic Ocean | Jonsson et al., 2022 |

| | | | | | |
|---------|-----------------------------------|---|------------|---|-----------------------|
| Sea ice | 0–0.5 ng/L | 0 - 40% | multi-year | Eastern Beaufort Sea and McClure Strait | Beattie et al., 2014 |
| | 0.017±0.024 ng/L | 3.5 ± 4.3% | | Central Arctic Ocean | DiMento et al., 2019 |
| | 0.006±0.004 ng/L | 1.0-2.3% | first-year | Central Arctic Ocean | Schartup et al., 2020 |
| | 0.022±0.015 ng/L | 2.2-5.0% | multi-year | Central Arctic Ocean | Schartup et al., 2020 |
| | 0.014±0.011 ng/L | 8.2% (media) | | Central Arctic Ocean | Jonsson et al., 2022 |
| Air | MMHg: 2.9 ± 3.6 pg/m ³ | range: <0.5 (LOQ) to 12.7 pg/m ³ | | Hudson Bay, Canadian Arctic Archipelago | Baya et al. 2015 |
| | DMHg: 3.8 ± 3.1 pg/m ³ | range: <0.5 (LOQ) to 11.3 pg/m ³ | | | |

189 **Table 2.** Total MeHg fluxes and pools in the Arctic environment (excluding aquatic biota larger than
 190 plankton). While the MeHg concentrations reported (Table 1) cover both freshwater and the marine
 191 compartments, the fluxes focus on those relevant for the Arctic Ocean. Details on how the MeHg
 192 pools (and their uncertainty) were calculated is provided in Table S1.

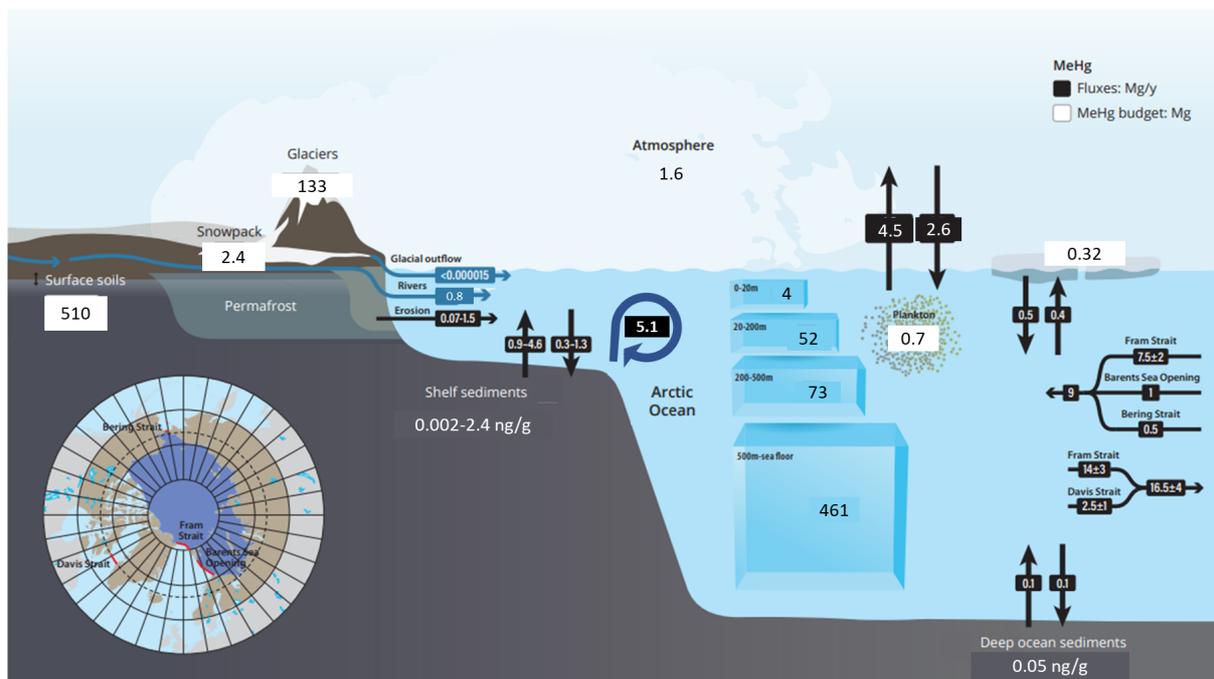
| Environmental Compartment | MeHg Fluxes | References |
|--|---|---|
| Atmospheric deposition | 8.5 (8–10) Mg/yr 2.6 Mg/yr | Soerensen et al., 2016b this review |
| Atmospheric evasion | 14 Mg/yr 4.5 (2.8–6.3) Mg/yr | Soerensen et al., 2016b this review |
| Sea ice uptake | 0.4 Mg/yr | Petrova et al., 2020 |
| Sea ice discharge | 0.5 Mg/yr | Soerensen et al., 2016b |
| Coastal erosion | 0.07–0.46 Mg/yr 1.5 Mg/yr | Soerensen et al., 2016b Petrova et al., 2020 |
| Wetland ponds | 1.8–40 ng/m ² , day | Lehnherr et al., 2012a |
| Glacial meltwater (unfiltered), Ellesmere Island | 0.03–14.9 g/y 0.49–11.7 mg/km ² | St. Pierre et al., 2019 |
| Riverine inputs | 0.8 Mg/yr | Sonke et al. 2018 |
| Inflow to Arctic Ocean | 3–13 Mg/yr | Soerensen et al., 2016b |
| —of Atlantic deep and mid- depth waters | 6.8 Mg/yr | Soerensen et al., 2016b |
| Inflow to Arctic Ocean | 9±1.5 Mg/yr | Petrova et al., 2020 |
| —from Fram Strait | 7.5±2 Mg/yr | Petrova et al., 2020 |
| —from Barents Sea Opening | 1 Mg/yr | Petrova et al., 2020 |
| —from Bering Strait | 0.5 Mg/yr | Petrova et al., 2020 |
| Outflow to North Atlantic | 8–27 Mg/yr | Soerensen et al., 2016b |
| Outflow to North Atlantic | 16.5±2 Mg/yr | Petrova et al., 2020 |
| —via Fram Strait | 14±3 Mg/yr | Petrova et al., 2020 |
| —via Davis Strait | 2.5±1 Mg/yr | Petrova et al., 2020 |
| Sedimentation load (shelf sediment) | 1.3 Mg/yr 0.3 Mg/yr | Soerensen et al., 2016b Petrova et al., 2020 |
| Sedimentation load (basin sediment) | 0.1 Mg/yr | Soerensen et al., 2016b |
| Sediment diffusion to water (shelf sediment) | 0.9 Mg/yr 4.6±0.2 Mg/yr | Soerensen et al., 2016b Kim et al., 2020b |
| Sediment diffusion to water (basin sediment) | 0.1 Mg/yr | Soerensen et al., 2016b |

| Environmental Compartment | MeHg Pool | References |
|--------------------------------------|-------------------|---|
| Atmosphere | 1.6 (0–3.2) Mg | Baya et al., 2015; Table 3.8.1 in AMAP (2021) |
| Surface soils | 510 (270–749) Mg | St. Pierre et al., 2015; Tarbier et al., 2021 |
| Snowpack | 2.4 (0.43–4.3) Mg | St. Pierre et al., 2019, Varty et al., 2021; Jonsson et al., 2022 |

| | | |
|----------------------|----------------------|---|
| Glaciers | 133 (± 1.2) Mg | Zdanowicz et al., 2016 |
| Sea ice | 0.32 (0.04–1.0) Mg | Dimento et al., 2019; Schartup et al., 2020 |
| Shelf sediments | 0.002–2.4 ng/g | Fox et al., 2014; Soerensen et al., 2016b; Liem-Nguyen et al., 2022; Kim et al., (2020) |
| Deep ocean sediments | 0.05 ng/g | Soerensen et al., 2016b |
| Water column | | Wang et al., 2012, 2018; Heimbürger et al., 2015; |
| 0–20 m depth | 4 (± 2) Mg | Agather et al., 2019; |
| 20–200 m depth | 52 (± 34) Mg | Charette et al., 2020; Kim et al., 2020b; Petrova et al., 2020, Jonsson et al., 2022; |
| 200–500 m depth | 73 (± 44) Mg | Kohler et al., 2022 |
| >500 m depth–bottom) | 461 (± 232) Mg | Soerensen et al., 2016b |
| Plankton | 0.7 Mg | |

193

194



195

196 **Figure 2.** MeHg fluxes and budget in the Arctic environment. Updated from Soerensen et al. (2016b)
197 and Petrova et al. (2020) with data shown in Table 1 and Table 2.

198

199 3. Mechanistic understanding of Hg (de)methylation processes

200

201 **3.1. Formation and degradation pathways of MeHg**

202 Given that MeHg is the bioaccumulative form of Hg, understanding the rates of both methylation and
203 demethylation as well as Hg(II) reduction, which might limit the availability of this species for
204 methylation, have implications for biotic uptake and exposure of Hg. While photochemical processes
205 might govern Hg chemistry at the surface of the waters, biological processes are the key to understand
206 Hg cycling in freshwater, seawater and in sediments.

207 Methylation of Hg is primarily thought to be a biotic process carried out by various microorganisms,
208 though evidence for abiotic methylation exists. In 2013, the gene pair involved in biological Hg
209 methylation (*hgcAB* genes) were discovered (Parks et al., 2013). This opened up new possibilities to
210 search for Hg methylators (Gilmour et al., 2013, Jones et al., 2019; Capo et al., 2020; Gionfriddo et
211 al., 2016; McDaniel et al., 2020) and uncover the intracellular processes involved in Hg methylation
212 (Smith et al., 2015). Carriers of the *hgcAB* genes that are known to methylate Hg include a wide range
213 of sulfate reducing bacteria, iron reducing bacteria, methanogens, and some fermentative
214 microorganisms (Gilmour et al., 2013, 2018). A global analysis of metagenomes was first to reveal the
215 presence of *hgcAB* in the Arctic environment by identifying the genes in *Deltaproteobacteria*,
216 *Firmicutes*, and particularly in methanogenic archaea from thawing permafrost soil (Podar et al.,
217 2015). Fahnestock et al. (2019) also showed that concentrations of MeHg in porewater increased
218 across a thaw gradient along with the abundance and diversity of potential Hg methylators. It is
219 important to note that not all Hg methylating microorganisms have the same capacity to methylate Hg
220 and that this appears to depend on individual strains rather than taxonomic ranking (Ma et al., 2019).
221 Marine microorganisms with a greater oxygen tolerance and broader habitat range might be important
222 Hg methylators in the seawater (Villar et al. 2020; Lin et al., 2021).

223 In the last decade, several studies have highlighted the importance of Hg methylation in suboxic and
224 even oxic waters for MeHg bioaccumulation and biomagnification in aquatic food webs (Sunderland
225 et al., 2009; Heimbürger et al. 2010; Lehnher et al., 2011; Mason et al., 2012; Wang et al., 2012b,
226 2018; Blum et al., 2013; Heimbürger et al., 2015; Schartup et al., 2015a; Soerensen et al., 2016a;
227 Gascón Díez et al., 2016; Rosati et al. 2018; Agather et al., 2019; Petrova et al., 2020). While

228 anaerobic biological Hg methylation is mediated by the *hgcAB* genes that are abundant in marine
229 sediments (Podar et al., 2015), the aerobic pathway, if it exists, remains unclear. *HgcA* or *hgcAB*-like
230 genes have recently been identified in global ocean oxic seawater (Villar et al., 2020) as well as in
231 Antarctic sea ice (Gionfriddo et al. 2020), though not in the Arctic Ocean (Bowman et al., 2020a).
232 Further studies are needed to identify key players for Hg methylation in polar marine waters and sea
233 ice.

234 It is generally agreed that primarily dissolved complexes of Hg(II) and MMHg are taken up by Hg-
235 methylating bacteria at the base of aquatic food webs (Drott et al., 2007; Schaefer and Morel, 2009;
236 Chiasson-Gould et al., 2014; Mangal et al., 2019). Adsorption of Hg(II) and MMHg to the solid phase
237 are thus important processes limiting their biological uptake (Schartup et al., 2015a; Mazrui et al.,
238 2016). However, as shown in previous work from other marine or brackish water systems, different
239 dissolved and solid chemical forms of Hg differ in their bioavailability (Jonsson et al., 2012, 2014;
240 Mazrui et al., 2016). Although recent terrestrial inputs of Hg(II) and MeHg did not contribute much to
241 the stock of MeHg in the estuarine system, 40 to 70% of the bioaccumulated Hg pool originated from
242 these recent terrestrial Hg inputs. In these studies, refractory pools of Hg(II) and MMHg were not fully
243 accounted for.

244 MeHg demethylation can be biotically or abiotically mediated. Abiotic demethylation is mostly
245 photochemical degradation in surface waters by different mechanisms including direct photolysis and
246 indirect photolysis via reactions with reactive oxygen species such as hydroxyl radicals. Research in
247 the Arctic, subarctic, and temperate areas has shown photochemical demethylation from ultraviolet
248 (UV) radiation is an important aspect of freshwater Hg budgets (Seller et al., 1996; Lehnerr et al.,
249 2012a; Poste et al., 2015; Klapstein and O'Driscoll, 2018). Because organic matter compounds in
250 surface waters readily absorb UV radiation, they mediate the photodemethylation of aqueous MeHg
251 (Klapstein et al., 2017). A recent study on ponds with high DOC concentrations resulting from
252 permafrost thaw slumps found higher rates of photodemethylation when compared to an oligotrophic
253 lake in the polar desert with low DOC concentrations (Girard et al., 2016). Photolytically produced
254 reactive oxygen species from organic matter have been shown to mediate MeHg demethylation in

255 Alaskan lacustrine systems (Hammerschmidt and Fitzgerald, 2010). In the Arctic, photodemethylation
256 rates are likely regulated by prolonged ice coverage (Poste et al., 2015; Klapstein and O'Driscoll
257 2018). Non-photochemical abiotic demethylation may also occur via reactions with selenoamino acids
258 (Khan and Wang, 2010), but the importance of such reactions remains to be further examined outside
259 of the laboratory.

260 Bacterial resistance to MeHg (and Hg(II)) is one of the most widely observed phenotypes in eubacteria
261 (Barkay et al., 2003) and is mediated by the *mer* operon, which has been found in a diverse range of
262 habitats including soils, sediment and water, both freshwater and marine systems (Barkay et al., 2003
263 and references therein). Within the operon (which contains different types of genes), attention has
264 been focused on the *merA* gene, which catalyzes reaction transforming Hg(II) into Hg(0) and *merB*,
265 which codifies for the alkaline mercury lyase, a protein that demethylates MeHg into Hg(II). Little is
266 known about the occurrence, diversity and distribution of *these genes* in the Arctic region. Of potential
267 importance for Hg demethylation, a recent study, however, unveiled the presence of *merB* in deep
268 waters of the Arctic Ocean (Bowman et al., 2020a). Also, a study performed in lake sediment cores
269 showed the presence of *hgcA* in Arctic lakes (Ruuskanen et al., 2020). The diversity of both Hg
270 methylators and MeHg demethylators in the Arctic needs to be further explored.

271

272 **3.2. Formation and degradation pathways of DMHg**

273 The main formation and degradation pathways of the DMHg remain unknown in marine waters,
274 including the Arctic Ocean (Agather et al., 2019; Petrova et al., 2020; Bowman et al., 2020b, Jonsson
275 et al. 2022). Although early studies identified synergies between the activity of organisms and
276 production of DMHg (Baldi et al., 1993; Sommar et al., 1999; Pongratz and Heumann, 1998), there is
277 currently no experimental data to support that it is a biological product. Baldi et al. (1993) suggested
278 the formation of DMHg in bacterial cultures was the result of an abiotic reaction between dissolved
279 sulfide and MeHg (via the formation of dimethylmercurysulfide). Recent work has also shown abiotic
280 formation of DMHg from MMHg when MMHg is adsorbed onto organic or inorganic surfaces

281 containing reduced sulfur sites (Jonsson et al., 2016). Other known pathways of DMHg formation of
282 potential relevance in natural environments include the reaction of MeHg with selenoamino acids
283 (Khan and Wang, 2010) and methylation with methylcobalamin (Imura et al., 1971).

284 Volatilization of DMHg from the marine boundary layer and the subsequent atmospheric
285 demethylation of DMHg to MMHg has been suggested as a source of MeHg in surface snow (Kirk et
286 al., 2008; St. Louis et al., 2007), marine waters (Baya et al., 2015) and coastal terrestrial environments
287 (St. Pierre et al., 2015). Baya et al. (2015), who were first to quantify DMHg in ambient air, also
288 suggested the evasion of DMHg from marine waters in Hudson Bay and the Canadian Arctic
289 Archipelago was enough to sustain the atmospheric concentrations of MMHg observed. In addition to
290 degradation of DMHg in air, DMHg could potentially also be degraded in marine waters, although
291 there have been contradictory claims in the literature on whether the rates are high enough for the
292 process to play a role in the cycling of DMHg (Mason and Sullivan, 1999; Black et al., 2009). West et
293 al. (2022), however, recently demonstrated that DMHg in arctic seawater readily degrades under
294 natural sunlight. The photodecomposition rates observed in tested waters were in the same range as the
295 rate observed for MMHg, suggesting that a significant fraction of the DMHg that is transported to
296 surface waters may be degraded to MMHg directly in surface waters rather than in the atmosphere. It
297 is also possible that DMHg is degraded through other pathways which do not involve photochemical
298 reactions. DMHg has recently been shown to degrade in aqueous solutions to MeHg in the presence of
299 reduced sulfur as dissolved sulfide or as the metastable iron sulfide mackinawite (West et al., 2020).

300

301 4. Where are methylmercury hotspots in the Arctic?

302

303 4.1. Terrestrial Environments

304

305 4.1.1 Arctic Tundra

306 Mercury has a strong affinity to organic substances, and organic soils therefore play an important role
307 in the mobilization and transportation of Hg from soils to water bodies. As such, surface vegetation,
308 soils, and organic matter are likely locations where elevated Hg is present and available for
309 transformation. St. Pierre et al. (2015) reported that soil MeHg concentrations varied by two orders of
310 magnitude (0.02–2.11 ng/g) across several sites in the Canadian Arctic Archipelago, with a median of
311 0.12 ng/g. A tundra study (Olson et al., 2018) reported concentrations of MeHg between 0.5 and 3.4
312 ng/g in organic horizons and 0.1 to 1.0 ng/g in B horizons, with MeHg on average accounting for 1%
313 of THg (THg with a maximum of 3% in the lower B horizon). A study from Fennoscandia further
314 found MeHg accounting for $0.48 \pm 1.7\%$ of THg in peat plateau soils, respectively (Tarbier et al.
315 2021). As discussed further below, higher %MeHg values has been reported from subarctic and Arctic
316 soils saturated with water (Tarbier et al. 2021: $6.1 \pm 6.5\%$, Varty et al. 2021: $>10\%$).

317 In addition to driving the deposition of Hg to Arctic tundra by foliar uptake (Obrist et al., 2017), plants
318 also bioaccumulate Hg from the soil via the roots (Zhou and Obrist, 2021). Generally, very few data
319 are available on MeHg concentrations in Arctic vegetation. Lichens in some coastal areas of the
320 Canadian High Arctic (MeHg: 1.41–17.1 ng/g) were more enriched in MeHg (and THg) relative to
321 underlying soils than at inland sites. This suggests greater uptake/deposition of MeHg may originate
322 from the evasion of DMHg and MMHg from open marine waters in coastal areas (St. Pierre et al.,
323 2015). Further research is needed to identify sources and pathways of MeHg uptake in terrestrial
324 vegetation.

325 A major consequence of climate warming in the Arctic and Subarctic has been the thawing and
326 degradation of permafrost (Jorgenson et al., 2001, 2006; Lantz and Kokelj, 2008; Walvoord et al.,
327 2019). Given the large amounts of Hg that potentially could be released from Arctic soils in a warmer
328 climate (Schaefer et al., 2020, Lim et al. 2020), methylation rates in the Arctic terrestrial environment
329 have gained more attention. Permafrost degrades through two dominant mechanisms; first is the top
330 down thaw of near-surface permafrost through the downward expansion of the seasonally thawed
331 active layer (Douglas et al., 2021). This mechanism, which typically encompasses a broad areal extent
332 but is not vertically extensive, generates wetter and more anoxic conditions that are potentially
333 favorable for Hg methylating bacteria (Yang et al., 2016). While increased Hg methylation under such
334 conditions has been demonstrated in temperate and tropical zones (Hsu-Kim et al., 2018), few studies
335 have covered Hg methylation in zones with thawing permafrost (Fahnestock et al., 2019; Tarbier et al.,
336 2021; Varty et al., 2021).

337 The second mechanism for permafrost degradation is the development of thermokarst features through
338 thaw subsidence of ice-rich frozen ground (Vonk et al., 2015; Olefeldt et al., 2016; Olefeldt et al.,
339 2021). This typically leads to creation of a variety of features including scarps, pits, thaw slumps, and
340 ponding of surface water that do not encompass large regions but are hotspots for geomorphologic
341 processes. Thermokarst wetlands, ponds and lakes have been repeatedly identified as Hg methylation
342 hotspots across the Arctic due to the concurrent mobilization of carbon and nutrients, creating
343 conditions suitable for methylation. In previously formed thaw ponds, Lehnherr et al. (2012a, 2012b)
344 measured potential Hg methylation rates (1.8 to 40 ng/m² /day) similar to rates in already identified Hg
345 methylation hotspots. High concentrations of dissolved MeHg (ranging up to 22 ng L⁻¹), likely from
346 either increased export of MeHg from peat soils, or *in situ* methylation in pond sediments, have also
347 been found in eastern Canadian thaw ponds (MacMillan et al., 2015). Gordon et al. (2016) observed
348 up to an order of magnitude higher MeHg concentrations in the poor fens associated with recent
349 permafrost thaw (0.43 ng/L, filtered) than in the hydrologically isolated bogs (0.04 to 0.1 ng/L,
350 filtered) in the Scotty Creek catchment of the discontinuous permafrost zone (in Canada's Northwest
351 Territories). In the Stordalen Mire in Subarctic Sweden, MeHg concentrations increased by two orders

352 of magnitude along the permafrost thaw gradient, from 0.01–0.05 ng/L in palsas underlain by
353 permafrost to 1.0–2.0 ng/L in thawed fens (Fahnestock et al., 2019). The thawed fens were associated
354 with heightened Hg accumulation, reducing conditions, higher sulfur concentrations and a greater
355 proportional representation of putative Hg methylators (i.e. microbial strains carrying the Hg
356 methylation gene pair but have not yet have been confirmed to methylate Hg in laboratory studies)
357 within the microbial communities (from 0.5% to 7.8%), compared to the dry frozen palsas and bogs at
358 intermediate stages of thaw (Fahnestock et al., 2019). In another Fennoscandian permafrost peatland
359 study, an order of magnitude higher concentration of MeHg (and fraction of THg as MeHg) was found
360 in topsoil (upper 50 cm) from collapse fens (edge of the fen receiving peat from the collapsing peat
361 plateau; $1.8 \pm 4.7 \text{ ng g}^{-1}$; $6.1 \pm 6.5\%$ of THg) compared to the peat plateau soils ($0.17 \pm 0.55 \text{ ng g}^{-1}$;
362 $0.48 \pm 1.7\%$ of THg; Tarbier et al. 2021). Median MeHg and %MeHg in the soil from the collapse fen
363 ($1.8 \pm 4.7 \text{ ng g}^{-1}$; $1.7 \pm 2.4\%$ of THg) was also 3.6 times higher than in the nearby fens assumed to
364 have no recent history of permafrost, suggesting higher methylation of Hg in the recently formed fens.
365 In line with these studies, Varty et al. (2021) observed higher %MeHg in soils saturated with water in
366 comparison to drier soils collected on Ellesmere Island (Canada).

367 Permafrost thaw has also been shown to influence microbial processes (Mackelprang et al., 2011) and
368 biogeochemical cycling (Frey and McClelland, 2009) in downstream freshwater systems. Several
369 recent studies have reported sites with permafrost slumps (i.e. slope failures or mass-wasting features
370 resulting from the thawing of ice-rich permafrost) are abundant with both organic matter compounds
371 and Hg. In fact, the highest concentrations of THg and MeHg (1200 and 7 ng/L unfiltered,
372 respectively) ever recorded in natural flowing freshwater were measured downstream of a permafrost
373 thaw slump in the Northwest Territories of Canada (St. Pierre et al., 2018). Other studies show
374 similarly elevated levels of both THg and MeHg in water and sediments from areas influenced by
375 permafrost degradation (MacMillan et al., 2015; Gordon et al., 2016; Houben et al., 2016). In stratified
376 thaw ponds, elevated MeHg concentrations and percentages in bottom waters suggest *in situ*
377 methylation is occurring in anoxic waters (MacMillan et al. 2015). In addition to slump age, many
378 factors may be contributing to the reported differences in organic matter and Hg levels downstream of

379 permafrost slumps, highlighting the need for further study of how permafrost degradation will affect
380 interactions between organic matter and Hg across various Arctic and Subarctic landscapes. Factors
381 such as the depth of the permafrost active layer, peatland buffering and interferences as well as ground
382 water influences may affect organic matter-Hg dynamics downstream of thermokarst processes (Frey
383 and McClelland, 2009; Lim et al., 2019). Furthermore, thaw slumps are likely affecting a wide range
384 of other biogeochemical processes that will either directly or indirectly alter Hg methylation or
385 bioaccumulation in receiving water bodies (Pokrovsky et al., 2018).

386 **4.1.2. Freshwater systems**

387 Arctic freshwater systems support several species of fish, which can be important in the traditional diet
388 of some Indigenous peoples (Lockhart et al, 2005). Mercury enters lakes and streams primarily via
389 terrestrial runoff and atmospheric deposition, but it can also originate from groundwater passing
390 through unfrozen parts of permafrost below the water body (i.e. taliks). Inorganic Hg(II) entering
391 Arctic freshwater systems can either be photolytically reduced to Hg(0) and evade back to the
392 atmosphere, be methylated to MeHg or bind to particles and sink to the bottom sediments. Where
393 methylation of Hg dominates in freshwater systems has been shown to vary between different Arctic
394 freshwater bodies (see Table S2; Chételat et al., 2015).

395 Mercury and MeHg dynamics in freshwater systems change with the seasons, having low or no
396 mobilization during freezing conditions (November–May) which increases with higher river flows and
397 flooding during the melting season from May to June. Methylmercury in terrestrial snow, for example,
398 enters High Arctic lakes during snowmelt via melt streams, having higher concentrations during spring
399 melt than during summer (Loseto et al., 2004). Methylmercury concentrations in Canadian Arctic
400 freshwater systems are generally low ranging from 0.04 to 0.18 ng/L in unfiltered samples in rivers,
401 0.02 to 0.10 ng/L in lakes and greater than 0.02 to 0.53 ng/L in ponds (see Table 1). Compared to
402 deeper lakes, shallow water bodies generally have higher MeHg due to warmer temperatures and
403 higher organic matter available for microbial metabolism and are probable methylation hotspots
404 (Lehnherr et al., 2012a; Lehnherr, 2014; MacMillan et al., 2015). Varty et al. (2021) showed that
405 small, shallow Arctic lakes can reach higher concentrations of water column MeHg (up to 0.72 ng/L)

406 in winter when ice covered. In addition, snow MeHg concentrations in spring (0.19-0.67 ng/L) suggest
407 that snowpack may be an important MeHg source during the melt period. Water residence time may
408 also play a role, with less within-lake processing of aqueous MeHg in smaller lakes (Richardson et al.,
409 2021). In Alaskan lakes, sediment methylation has been shown to be the major source of MeHg to
410 those ecosystems (Hammerschmidt and Fitzgerald, 2006). Varty et. al. (2021) also suggested sediment
411 MeHg production is important for the annual MeHg budget of Skeleton Lake and can explain the
412 elevated concentrations of MeHg observed during the ice-covered season.

413 Generally, in freshwater ecosystems, the main carrier for MeHg is DOC (Tsui and Finlay, 2011),
414 which has the double role of promoting the solubility and transport of MeHg in surface waters and
415 attenuating dissolved MeHg uptake by aquatic food webs by limiting transport across cell membranes.
416 High Arctic lakes typically have low biological productivity, limiting Hg uptake by algae and
417 suspended detrital organic matter. At the same time, many Arctic lakes have low DOC concentrations,
418 often below the suggested threshold of 8 to 11 mg/L DOC, above which DOC is hypothesized in some
419 studies to counteract MeHg bioaccumulation (French et al., 2014; Braaten et al., 2018). For MeHg
420 production, methylation of Hg in High Arctic wetland ponds was suggested to be negatively correlated
421 with DOC levels by decreasing Hg(II) availability to methylating microorganisms (Leinherr et al.,
422 2012a). While some studies suggest DOC concentrations may be declining in Arctic lakes (Saros et
423 al., 2015), others show increases in DOC because of climate change, land use practices and other
424 water chemistry interactions (Macdonald et al., 2005; Isles et al., 2020).

425 During melting, most transported Hg is bound to particles. A total flux of (dissolved) MeHg flux from
426 all Arctic rivers was estimated by Sonke et al. (2018) to be 584 kg/y. The larger variability
427 among different flux estimates for rivers, e.g. the Mackenzie River in various studies (72 kg/yr of
428 dissolved MeHg (Sonke et al. 2018), 14 kg/yr MeHg (Mu et al. 2019) and 13 and 15 kg/y MeHg
429 (unfiltered) (Emmertson et al, 2013)) stresses the need for additional studies to better constrain these
430 fluxes. The total Hg flux from larger Arctic rivers estimated by Sonke et al. (2018) was 27.6 Mg/yr
431 (13.4 Mg/yr as dissolved Hg and 13.9 as particulate Hg). The fate of Hg exported to the Arctic Ocean
432 is less clear. Coastal sediments and oligotrophic lake sediments in a High Arctic region of the

433 Canadian Arctic Archipelago were found to methylate Hg, though at a low rate (St. Pierre et al., 2014;
434 Hudelson et al., 2020).

435 **4.1.3 Glaciers**

436 Methylmercury has been shown to be released in glacier-fed streams of the Canadian High Arctic
437 (Ellesmere Island; St. Louis et al., 2005; St. Pierre et al., 2019), Yukon (Halm and Dornblaser, 2007)
438 and Alaska (Nagorski et al., 2014; Vermilyea et al., 2017). Reported MeHg concentrations in
439 unfiltered glacial meltwater vary from <0.010 to 0.26 ng/L and account for 1 to 26% of THg, while
440 concentrations of MeHg in filtered meltwater vary from <0.010 to 0.075 ng/L and account for 4 to
441 32% of THg (see Table S3). Estimates of unfiltered MeHg fluxes in glacier-fed streams of Ellesmere
442 Island are between 0.03 and 14.9 g/y, which translate to annual yields ranging from 0.49 to 11.7
443 mg/km²/y in basins with 21 to 82% glacier coverage (St. Pierre et al., 2019; see Table S3).

444 An increase in the glacial sediment Hg flux in response to enhanced glacier melt rates might lead to
445 higher exposure risks to inorganic Hg or MeHg in downstream aquatic ecosystems in some
446 catchments. On land, changes might occur with the development of seasonally wet areas in newly
447 deglaciaded forelands where methylation of sediment-bound Hg could occur. In southeastern Alaska,
448 for example, watersheds with greater wetland abundance have higher Hg concentrations, in both water
449 and aquatic fauna, than glacierized basins (Nagorski et al., 2014). An increased supply of labile and
450 bioavailable DOM from terrestrial sources such as wetlands or newly formed organic soils (e.g.,
451 Wietrzyk-Pełka et al., 2020) might also increase the risk of riverine Hg uptake by biota in these
452 watersheds. It could also promote Hg methylation by sulfur-reducing bacteria in catchments where
453 subglacial oxidation of sulfides causes enhanced sulfidization of ponds, wetlands and lakes (Graham et
454 al., 2012). Cryoconite holes (dirt-filled melt cavities on glacier surfaces) are abundant over glacier
455 margins or stagnant ice and can accumulate atmospherically deposited impurities released by ice melt,
456 as well as microbes (Cook et al., 2016). These micro-environments could potentially act as local
457 hotspots of Hg methylation in glacial environments, but direct evidence supporting this is presently
458 limited to Tibetan glaciers (Zhang et al., 2020).

459 In the marine environment, meltwater plumes from tidewater glaciers carrying both dissolved and
460 particulate Hg may be injected into zones of locally high primary productivity in coastal surface
461 waters, where subsequent methylation and biological uptake can occur (Schartup et al., 2015a). Some
462 glaciers of southern Alaska are sources of old (up to ~3900 y) and highly labile DOM, of possible
463 subglacial bacterial origin, which is discharged into coastal waters (Hood et al., 2009, 2015).
464 Likewise, retreating glaciers in fjords in southern Spitsbergen, Svalbard, discharge sediments
465 containing up to 2.8% weight of OC (Kim et al., 2020a). Seafloor sediments in these fjords have THg
466 concentrations of up to 84 ng/g. Hawking et al. (2021) recently reported dissolved MeHg
467 concentrations from Greenland glacial catchments that were similar to values previously reported from
468 wetlands. As discussed elsewhere (Dastoor et al., 2022) for THg, further studies are warranted to
469 confirm the high levels of MeHg in these glacial catchments, particularly since the source(s) and
470 processes leading to high MeHg yield remain unknown. Hg concentrations correlate closely with OC
471 and are thought to be largely derived from glacially eroded sedimentary rocks (e.g., shales) in the
472 glacierized basin. The discharge of Hg-bearing glacial sediments containing organic matter might
473 promote Hg methylation and uptake by marine biota within the water column, which has not been well
474 studied. However, as a recent and exhaustive review highlighted, the local effect of glacial runoff on
475 coastal marine primary productivity is likely limited to a few kilometers beyond glacier termini, and
476 whether runoff promotes or limits productivity varies between land- or marine-terminating glaciers
477 (Hopwood et al., 2020). For example, observations in fjords in northwestern Svalbard suggest that
478 glacial meltwater plumes could reduce the phytoplankton biomass (Piquet et al., 2014), and this might
479 effectively limit MeHg production and uptake in the marine food chain. Furthermore, the high
480 deposition rates of inorganic matter near tidewater glaciers, or at the mouth of proglacial channels,
481 dilute the organic matter content in the seafloor sediments of Svalbard fjords (Liu et al., 2015), and
482 this might limit benthic Hg methylation. On the whole, therefore, the environmental impact of Hg
483 releases from melting glaciers and ice caps is likely to be local, rather than widespread across the
484 Arctic. A study on the East Greenland shelf found no enhanced Hg or MeHg levels near the 79°N
485 glacier, precluding large inputs or locally enhanced MeHg production (Petrova et al. 2020).

486 4.2. Marine systems

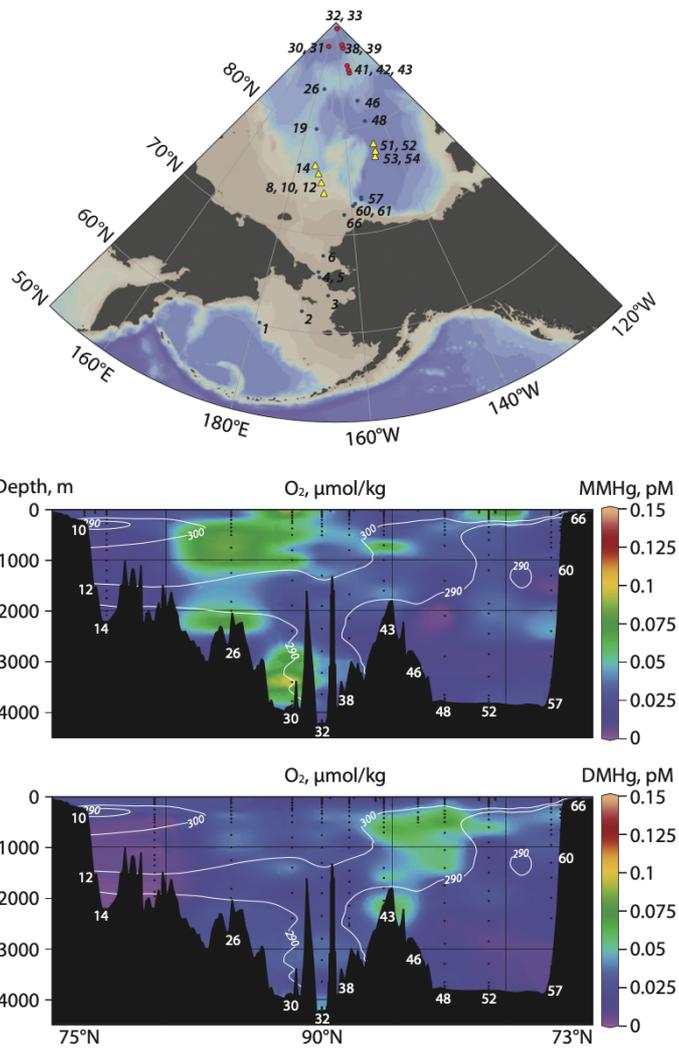
487 4.2.1 Seawater

488 MeHg concentrations in the western Arctic Ocean have been reported to be 0.011 ± 0.010 ng/L in
489 filtered seawater (Agather et al., 2019, Figure 3) and 0.046 ± 0.024 ng/L in unfiltered seawater (Wang
490 et al., 2018). Heimbürger et al. (2015) found similar levels of 0.045 ± 0.017 ng/L and 0.034 ± 0.022 ng/L
491 in unfiltered seawater in the Eastern Arctic and Central Ocean. Methylmercury (MeHg) inflow to the
492 Arctic Ocean totalled 9 ± 2 Mg/yr, most of which enters with deep and mid-depth waters of the Atlantic
493 Ocean (7.5 ± 2 Mg/yr; Petrova et al., 2020). Similar to THg, more MeHg flows out of the Arctic Ocean
494 into the North Atlantic (16.5 ± 3 Mg/yr, 14 ± 2 Mg/yr via the Fram Strait, 2.5 ± 1 Mg/yr via the Davis
495 Strait; Petrova et al. 2020) than flows in. The MeHg outflow via the Fram Strait conserves the shallow
496 MeHg peak found throughout the Arctic Ocean, and an average DMHg:MMHg ratio of 2.5:1 (Petrova
497 et al., 2020). *In situ* MeHg formation in the Arctic Ocean water column (Lehnherr et al., 2011; Wang
498 et al., 2012b, 2018; Heimbürger et al., 2015; Soerensen et al., 2016b; Petrova et al., 2020) results in a
499 net MeHg outflow of 7.5 ± 2 Mg/yr (Petrova et al. 2020).

500 A subsurface enrichment of MeHg in seawater has been observed in almost all major ocean basins
501 (AMAP/UNEP 2019), including the Atlantic Ocean (Bowman et al., 2015; Bratkič et al., 2016),
502 Pacific Ocean (Sunderland et al., 2009; Hammerschmidt and Bowman, 2012; Munson et al., 2015;
503 Bowman et al., 2016; Kim et al., 2017), Southern Ocean (Cossa et al., 2011; Gionfriddo et al., 2016);
504 Mediterranean Sea (Cossa et al., 2009, 2012, 2017; Heimbürger et al., 2010), Baltic Sea (Soerensen et
505 al., 2016b) and the Black Sea (Rosati et al., 2018) basins. However, the Arctic Ocean basin differs
506 from the others because the MeHg enrichment occurs at a much shallower depth (100–300 m depth)
507 where dissolved oxygen saturation is well above 75% (Wang et al., 2012b, 2018; Heimbürger et al.,
508 2015; see Figure 4). The Arctic subsurface enrichment of MMHg and DMHg is also detected in water
509 exiting the Arctic Ocean to the North Atlantic Ocean (Petrova et al. 2020). Heimbürger et al. (2015)
510 first hypothesized that the characteristic shallow MeHg peaks lead to enhanced biomagnification and
511 may be an alternative explanation for high arctic biota Hg levels, rather than anthropogenic Hg inputs.
512 As this subsurface MeHg maximum lies within the habitat of zooplankton and other lower trophic

513 level biota it has been suggested that biological uptake of the subsurface MeHg and subsequent
514 biomagnification explain a long-standing mystery in the Canadian Arctic on why marine animals in
515 the western Arctic have higher Hg levels than those in the east (see Figure 4; Wang et al., 2018).
516 Increasing amounts of bioavailable Hg in surface waters could have an effect on Hg accumulation at
517 the bottom of Arctic marine food webs. THg levels of up to 242 ng/g have been reported in
518 zooplankton from Hudson Bay (Foster et al., 2012). As discussed further below, regional differences
519 in food web length (and thus biomagnification) have also been provided as a contributing factor
520 explaining the higher Hg levels marine animals in the western Arctic (St. Louis et al., 2011; Brown et
521 al., 2016).

522 The mechanisms of subsurface MeHg formation in the Arctic Ocean and other ocean basins in the
523 world is under debate. Most of the studies have attributed subsurface MeHg to *in situ* Hg methylation
524 in the water column associated with organic matter remineralization (Sunderland et al., 2009; Cossa et
525 al., 2009; Wang et al., 2012b; Heimbürger et al., 2010, 2015), yet the biotic or abiotic processes that
526 are responsible for methylation remain unidentified (Bowman et al., 2020a). Recent work in the
527 Canadian Arctic (Wang et al., 2018) and the Siberian Shelf (Kim et al., 2020) suggest that advection
528 of MeHg produced in shelf sediments also could contribute to the subsurface peak of MeHg found in
529 the water column. This debate calls for further studies on MeHg sources and dynamics in the Arctic
530 and global oceans.



531

532

533 **Figure 3.** Monomethylmercury (MMHg) and dimethylmercury (DMHg) and dissolved oxygen (O₂) in
 534 filtered seawater along a looped latitudinal section (~75°N to the North Pole and then back to ~73°N)
 535 in the western Arctic Ocean. The top panel shows sampling station locations along the section.

536 Adapted from Agather et al., 2019.

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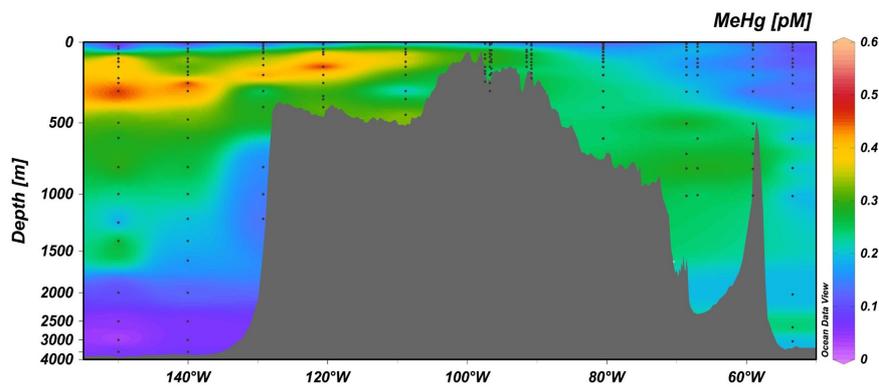
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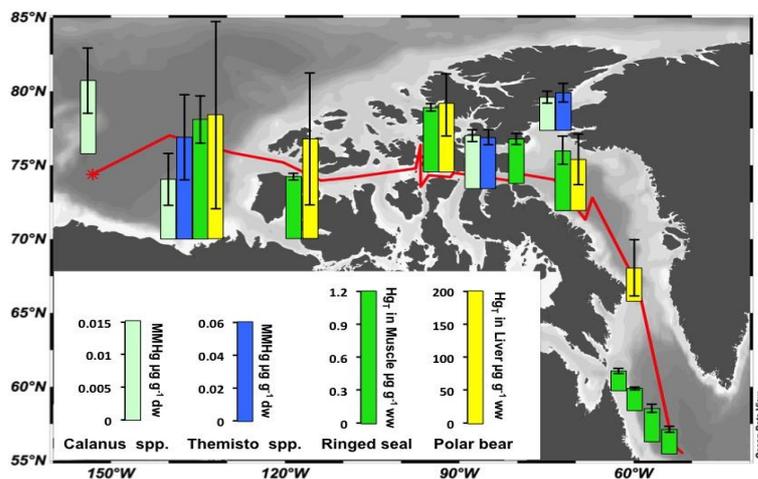
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Figure 4. Methylmercury (MeHg) concentrations in unfiltered seawater (top panel) and the marine food web (bottom panel) along a longitudinal (west-to-east) section (the red line in the bottom panel) across the Canadian Arctic and Labrador Sea. Bar charts in the bottom panel show mean concentrations of monomethylmercury (MMHg) in *Calanus* spp. and *Themisto* spp. collected from 1998 to 2012 ($\pm 1\text{SD}$), THg in muscle of adult ringed seals collected in 2007 and 2011 and THg in liver of polar bears (*Ursus maritimus*) collected from 2005 to 2008. Adapted from Wang et al., 2018.

560 **4.2.2. Marine sediment**

561 Methylmercury concentration measurements in deep Arctic Ocean sediments are sparse. Based on a
562 range of means, MeHg concentrations in Beaufort Sea and Chukchi Sea shelf sediments were
563 estimated to range from 0.15 to 0.37 ng/g (dry weight, Fox et al., 2014). A recent study in the East
564 Siberian Sea shows that MeHg concentrations range from 0.09 to 0.17 ng/g in shelf sediments, with
565 much lower concentrations (~0.003 ng/g dry weight) in the slope sediments (Kim et al., 2020b). Liem-
566 Nguyen et al (2022) reported MeHg concentrations ranging from 0.19 to 2.6 ng/g in Laptev Sea
567 sediments along a 330 km long cross-shelf transect. The higher concentrations of MeHg (and
568 MeHg/OC ratio) observed close to the Lena river delta than further out on the shelf was explained by
569 trapping of terrestrial MeHg close to the river delta. Based on estimated MeHg concentrations of 0.2
570 ng/g (dry weight) for shelf sediments and 0.05 ng/g (dry weight) for the deep basin, Soerensen et al.
571 (2016b) estimated a MeHg sedimentation flux of 1.3 Mg/yr and 0.1 Mg/yr to shelf and deep basin,
572 respectively. Resuspension was not estimated due to an absence of data. Soerensen et al., (2016b)
573 estimated MeHg diffusion to the overlying water column at 0.9 Mg/yr and 0.1 Mg/yr from shelf and
574 deep basin sediments, respectively, for a total flux of 1.0 Mg/yr. However, a recent study in the East
575 Siberian Sea suggests that the MeHg diffusion flux from sediments to the water column could be much
576 larger, amounting to 4.6 ± 0.2 Mg/yr for the Arctic shelf system alone (Kim et al., 2020b). There is a
577 need to better constrain the benthic MeHg flux.

578

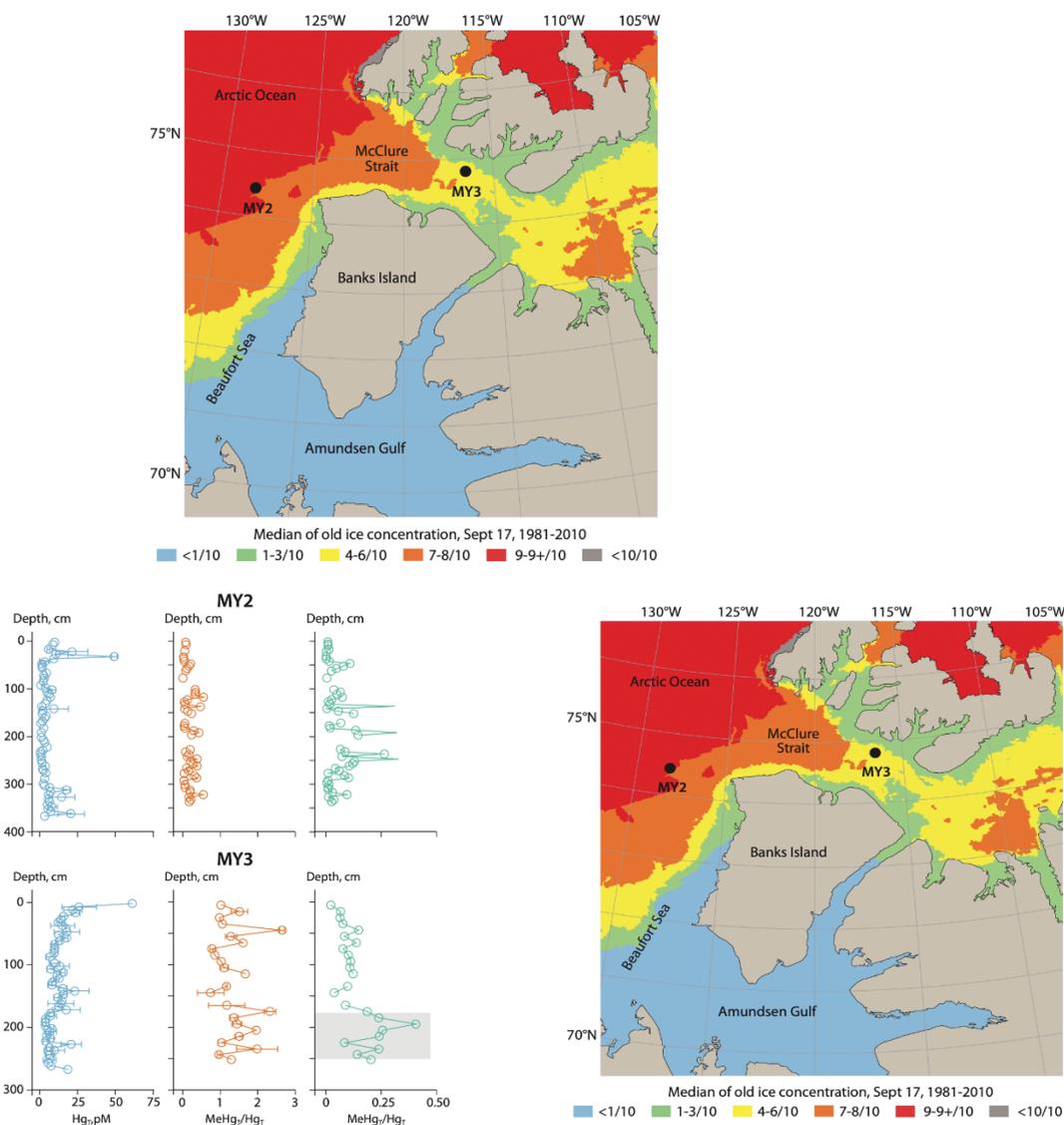
579 **4.2.3. Sea ice and snow**

580 The sea ice environment, including first-year ice, multi-year ice and the overlying snow cover,
581 provides a spatially and temporally dynamic interface between the ocean and the atmosphere in the
582 Arctic Ocean where physical transport, chemical reactions, and biological uptake occur (Wang et al.,
583 2017). While sea ice is semi-permeable (e.g., Loose et al. 2010), elevated Hg(0) concentrations under
584 sea ice compared with ice-free waters suggest that ice is a barrier for gas exchange (Dimento et al.
585 2019). The sea ice environment is also an important habitat and feeding ground for many organisms,

586 including bacteria, algae, zooplankton and larger mammals such as polar bears and whales. Beattie et
587 al. (2014) found elevated MeHg concentrations in multi-year Arctic sea ice of up to 0.5 ng/L
588 (unfiltered) in the lower part of the ice (see Figure 5). Recent work has shown concentrations of both
589 MMHg and DMHg in sea ice, especially within first-year ice (4.4% MMHg vs. 1.5% in multi-year ice)
590 from across the central Arctic Ocean (Schartup et al., 2020). Snow, which covers large parts of the
591 Arctic landscape as well as sea icescape has a large interstitial surface area for air-snow exchange of
592 Hg and plays a major role in atmospheric mercury depletion events (Douglas et al., 2012, 2017;
593 Steffen et al., 2014). However, Douglas et al. (2012) report that the small amounts of MeHg found in
594 snowpack before spring snowmelt does not add significantly to MeHg levels in aquatic systems.

595 As the high concentrations of MeHg in the bottom layer of sea ice (Beattie et al., 2014) cannot be
596 explained by downward transport of MeHg from the overlying snow (Gionfriddo et al., 2016;
597 Soerensen et al., 2016b), it has been postulated that Hg methylation could take place in sea ice.
598 Discrete peaks in MMHg and DMHg concentrations within sea ice cores (Schartup et al., 2020) also
599 suggest that they may be produced by microorganisms within the sea ice. This is supported by the fact
600 that microbes present in snow and ice are metabolically active even under freezing temperatures
601 (Douglas et al., 2012). Methylation of Hg has earlier been suggested to be strictly anaerobic and to
602 occur in the anaerobic brine environment (droplets, channels or pockets of salt solution in sea ice), or
603 in the lower part of the ice, due to the involvement of sulfate-reducing bacteria (Chaulk et al., 2011;
604 Gilmour et al., 2013; Beattie et al., 2014). During spring melt, heterotrophic activity could lead to
605 oxygen depletion in the bottom of the ice column and denitrification processes that favor anoxic
606 environments could then be important in the lower parts of the ice (Rysgaard and Glud, 2004). A study
607 in the Antarctic has suggested methylation of Hg in sea ice might be carried out by aerophilic bacteria,
608 such as *Nitrospina*, or by sea ice phytoplankton (Gionfriddo et al., 2016), but this has yet to be
609 confirmed in the Arctic. Gionfriddo et al. (2016) suggest that sea ice phytoplankton play a role in the
610 stimulation of methylation of oxidized Hg that may occur within brine pockets and periphytic biofilms
611 where organic matter is trapped and decaying. Due to the seasonality of primary production, MeHg
612 formation could show seasonal variations. Chlorophyll a (Chla) showed a covariation with MeHg,

613 which also has been shown in other studies (Antarctica: Cossa et al., 2011; Arctic: Beattie et al.,
 614 2014), suggesting a microbial mercury methylation and the involvement of phytoplankton and
 615 phototrophs. Gionfriddo et al. (2016) also identify Proteobacteria, which possesses the *mer* operon, as
 616 a potentially important Hg reductive phylum. It should be noted that direct support for the production
 617 of MeHg in Arctic snow and ice (from e.g. Hg methylation assays) is still limited. From terrestrial
 618 sites, where Hg methylation and demethylation assays have been conducted on intact snow or melted
 619 snow, low or non-detectable potential Hg methylation rates were reported (Varty et al. 2021; Willis et
 620 al., 2018). In addition, both Varty et al. (2021) and Willis et al. (2018) found low methylation to
 621 demethylation ratios and concluded that *in situ* MeHg production likely did not contribute
 622 significantly to the pool of MeHg observed in the snowpack.



624 **Figure 5** Profiles of total Hg (Hg_T) methylmercury ($MeHg_T$) concentrations and the ratio of MeHg to
625 THg ($MeHg:THg$) in multi-year sea ice sampled at two sites (MY2 and MY3) in the eastern Beaufort
626 Sea and the M'Clure Strait. Adapted from Beattie et al., 2014.

627

628 The uptake and transport of Hg and subsequent formation of MeHg in sea ice is determined by its
629 thickness and permeability, which varies dramatically with temperature and season. During winter, the
630 ice is cold and less permeable, which slows the transport of Hg species through the ice. However,
631 exchange continues with the seawater as the bottom of the ice is warmer (greater permeability) and
632 with the atmosphere as brine exclusion expels Hg to the overlying snowpack. In spring, the ice warms
633 up and the brine volume increases. Contaminants such as Hg then percolate from snow to ice and
634 could lead to their flushing into the underlying seawater. Summer sea ice melt leads not only to
635 dissolved Hg being released from the ice but also particle bound Hg and MeHg being incorporated in
636 the bulk ice (Wang et al., 2017). The presence of sea ice also decreases photodegradation of MeHg in
637 underlying water (Ebinghaus et al., 2002, Point et al. 2012). The dynamic motion of sea ice further
638 makes it possible to transport Hg and MeHg over time and space as the ice can drift over large areas in
639 the Arctic Ocean, depending on winds and underwater currents. The export of MeHg via drifting sea
640 ice has been calculated by Wang et al. (2017) at less than 4% of the total Hg export from the Arctic
641 Ocean. Petrova et al. (2020) estimated a net export of MeHg in sea ice of 0.4 Mg/yr, considering 10%
642 of the total Hg export is in the form of MeHg.

643 The total MeHg pool in sea ice in the Arctic Ocean was estimated by Schartup et al. (2020) to be 0.33
644 Mg, which is consistent with estimates of 0.3 to 2.6 Mg, based on studies performed in the M'Clure
645 Strait and Beaufort Sea (Beattie et al., 2014; Soerensen et al., 2016b).

646

647 **5. Processes affecting mercury accumulation in Arctic biota**

648 Methylmercury concentrations in fish and wildlife vary within and across geographic regions in the
649 circumpolar Arctic, reflecting the variability in Hg loading to the ecosystem and environmental
650 conditions that control *in situ* production of MeHg, both of which are discussed above. It also reflects
651 on differences in biological uptake at the base of the food web, and ecological characteristics that
652 control trophic transfer of MeHg within the food web (see Dietz et al., this issue). All of these
653 conditions can change over time resulting in temporal variation of MeHg concentrations in animals
654 (see Morris et al., this issue). The delineation of specific conditions promoting MeHg bioaccumulation
655 is important for identifying Arctic ecosystems at greater risk of MeHg exposure (Munthe et al., 2007).
656 Recent research has identified key environmental characteristics that enhance MeHg bioaccumulation
657 in the Arctic.

658 Mercury speciation determines (a) how Hg is transported between different environmental
659 compartments, (b) organismal exposure, (c) the potential toxicity of the exposure to the organism, (d)
660 the extent to which Hg is distributed between tissues within the organism, (e) the toxicity of the Hg
661 compound (e.g., MMHg, DMHg, Hg(II)), (f) organism assimilation and excretion, and (g) its
662 propensity to bioaccumulate and biomagnify through the food web. In this section, the uptake of Hg at
663 the base of Arctic aquatic and terrestrial food webs is described, with an emphasis on the biomagnified
664 chemical form MeHg.

665

666 **5.1 Mercury uptake at the base of Arctic food chains**

667 The bioconcentration of aqueous MeHg in bacteria and algae, either suspended in the water column
668 (plankton) or growing in biofilms on surfaces such as sediment, ice and rocks, is understood to be a
669 critical step in the transfer of Hg to top predators in freshwater and marine environments. Aquatic food
670 webs with greater MeHg uptake by microorganisms also have higher concentrations in subsequent
671 trophic levels (Wu et al., 2019), and greater uptake at the base of terrestrial food webs likely has the
672 same consequence (Tsz-Ki Tsui et al., 2019).

673 Despite the critical role of phytoplankton as the dominant energy source for food webs in the ocean,
674 few measurements of MeHg have been made of Arctic phytoplankton. In the Subarctic estuary of Lake
675 Melville, phytoplankton contained 0.03 ng/g dw of MeHg (Schartup et al., 2015a), which is low
676 relative to concentrations in marine phytoplankton at more temperate latitudes (Schartup et al., 2018).
677 Phytoplankton of the Chukchi Sea had MeHg concentrations below the study analytical detection limit
678 of 1.5 ng/g dw (Fox et al., 2017). Cellular uptake of MeHg by phytoplankton may occur by passive or
679 active transport across the cell membrane (Lee and Fisher, 2016; Skrobonja et al., 2019). Smaller
680 phytoplankton cells have larger surface to volume ratios and, consequently, bioconcentrate more
681 MeHg than larger cells (Lee and Fisher, 2016; Schartup et al., 2018). Smaller phytoplankton are also
682 known to stimulate MeHg production, because they have a higher surface to volume ratio and are
683 more effectively remineralized (Heimbürger et al., 2010). In the Arctic Ocean, water column MeHg
684 concentrations are often highest at subsurface depths of ~100–200 m, where the proximity to
685 phytoplankton may enhance uptake into pelagic food webs (Heimbürger et al., 2015; Wang et al.,
686 2018). Arctic estuaries may also be important zones for MeHg uptake by phytoplankton because of
687 salinity-driven water-column stratification and the stimulation of MeHg production by inputs of
688 terrestrial organic matter (Schartup et al., 2015b). Seasonal changes also affect Hg uptake by marine
689 phytoplankton, such as in the Baltic Sea, where reduced sea ice cover and eutrophication enhanced
690 bioaccumulation during warmer months (Bełdowska and Kobos, 2016; Soerensen et al., 2016a).

691 Sea ice is another habitat where microbial uptake of MeHg may be critical for basal uptake into food
692 webs. As stated before, MeHg production can occur in polar sea ice (Beattie et al., 2014; Gionfriddo et
693 al., 2016; Shartup et al., 2020) and in the marginal sea ice zone (Heimbürger et al., 2015). Algal
694 growth on the under-ice surface is a significant energy source for pelagic food webs in the Arctic
695 (Kohlbach et al., 2016). Burt et al. (2013) characterized uptake of inorganic Hg by ice algae during a
696 spring bloom associated with first-year ice and suggested that this could represent an important entry
697 point for marine invertebrates grazing either directly on the ice or on settling particulate matter
698 released from melting ice.

699 Methylmercury within the sea ice can be taken up by ice-associated biota, algae, under-ice amphipods
700 and ice-dependent animals such as fishes, polar bears and birds and can bioaccumulate in marine food
701 chains throughout the year. When sea ice melts, Hg and MeHg in the ice are released to the surface
702 ocean. Fast settling of sea ice algae upon melt (Boetius et al. 2013) may also be an important transport
703 route for MeHg into the deep ocean, but this needs further research.

704 With greater light penetration into the water column, the melting period is also a period of high
705 primary production in surface water, and the concurrent release of contaminants during this period
706 could lead to additional uptake of contaminants within the marine food web. Mercury attached to
707 particles is released from the ice during complete ice melt. Sinking particles may be remineralized in
708 the water column, releasing Hg for uptake by pelagic microbes, or reach the sediment and become a
709 source of Hg available for uptake by benthic organisms. Hg uptake by sea ice algae is only limited by
710 the amount of Hg available for uptake when spring blooms commence (Burt et al., 2013). This is
711 important since ice algae represent 10 to 60% of annual primary production in the Arctic, and they
712 may become even more abundant under mild climate conditions. Burt et al. (2013) estimate that with
713 the replacement of multi-year sea ice with first-year ice due to climate change, an extra 48 kg/y of
714 particle-bound Hg could enter the Beaufort Sea in the coming years.

715 Benthic algae and biofilms are a dominant primary energy source for food webs in many Arctic lakes
716 (Vadeboncoeur et al., 2003; Chételat et al., 2010; Rautio et al., 2011). In the eastern Canadian Arctic,
717 MeHg concentrations ranged from 2 to 12 ng/g dw for rocky shoreline biofilms (Chételat et al., 2018),
718 0.3 to 1.3 ng/g dw for microbial mats (Camacho et al., 2015), and 2 to 56 ng/g dw for periphyton and
719 filamentous algae (van der Velden et al., 2013a). In comparison, MeHg concentrations of water-
720 column seston measured in some of those same lakes ranged from less than 0.4 to 30 ng/g dw, and
721 bioconcentration factors of MeHg in biofilms and seston were higher in lakes with low aqueous DOC
722 (Chételat et al., 2018).

723 Invertebrates play a key role in the trophic transfer of MeHg entering food webs via microorganisms.
724 In the last decade, large amounts of data were compiled on MeHg concentrations in marine
725 zooplankton from across the circumpolar Arctic (Foster et al., 2012; Pućko et al., 2014; Pomerleau et

726 al., 2016). Zooplankton MeHg concentrations ranged from 8 to 96 ng/g dw and were highest in the
727 southern Beaufort Sea region compared to the Chukchi Sea, Canadian Arctic Archipelago, Hudson
728 Bay and northern Baffin Bay (Pomerleau et al., 2016; Wang et al., 2018). Larger taxa of marine
729 zooplankton such as *Themisto* and *Paraeuchaeta* predate on smaller zooplankton, and
730 biomagnification of MeHg was found between omnivorous and herbivorous taxa (Foster et al., 2012;
731 Pomerleau et al., 2016). Recent measurements of MeHg concentrations in zooplankton from Arctic
732 lakes (Lescord et al., 2015; Ch  telat et al., 2018, Varty et al., 2021) add to data from earlier studies
733 (Ch  telat and Amyot, 2009; Gantner et al., 2010), which showed that zooplankton in lakes can have
734 higher concentrations (>100 ng/g dw) than those in the marine environment. In a freshwater system on
735 Ellesmere Island (Canada), zooplankton MeHg concentrations were lowest under ice, highest at the
736 onset of ice-free conditions and then subsequently declined, indicating considerable seasonality in
737 bioaccumulation (Varty et al., 2021). While copepods tend to dominate in lacustrine zooplankton
738 communities in the Arctic, large-bodied grazers (Cladocera, *Daphnia*) are also present in some
739 systems and can have elevated MeHg concentrations (Ch  telat and Amyot, 2009; Lehnherr et al.,
740 2012a; Lescord et al. 2015; MacMillan, 2018). Modelling shows a potential for increase distribution of
741 daphnia across Arctic lakes due to climate-induced changes in lake productivity, a trend that could
742 enhance MeHg bioaccumulation patterns in pelagic food chains (Ch  telat and Amyot 2009). The
743 developmental stage can also influence the MeHg concentration of Arctic freshwater copepods, with
744 dilution occurring as adults accumulate lipid stores (Ch  telat et al., 2012).

745 A considerable number of studies of Arctic food webs have also characterized Hg concentrations in a
746 variety of benthic invertebrate taxa in marine environments (Bidleman et al., 2013; van der Velden et
747 al., 2013a; Fox et al., 2014; J  runds  ttir et al., 2014; Clayden et al., 2015; McMeans et al., 2015;
748 G  ngora et al., 2018; Pedro et al., 2019) as well as in lakes (van der Velden et al., 2013a; Ch  telat et
749 al., 2015, 2018; Kahilainen et al., 2016, 2017; Keva et al., 2017; Rohonczy et al., 2020). These studies
750 showed variation in Hg bioaccumulation among benthic invertebrate taxa, which was partly related to
751 trophic position.

752

753 **5.2. Processes affecting mercury bioaccumulation at higher trophic levels**

754 Mercury concentrations in fish and wildlife reflect ecological processes that influence dietary exposure
755 as well as animal physiology that affects internal accumulation of Hg (Bradley et al., 2017, Chételat et
756 al., 2020). Trophic position, shifts in diet, habitat-specific feeding, cross-ecosystem subsidies and
757 movement or migration can be important determinants of dietary Hg consumed by fish and wildlife.
758 Differences in the extent of assimilation and elimination of MeHg related to sex, age, body condition
759 and species also affect animal Hg burdens. Recent advances have identified Arctic-specific processes
760 affecting Hg bioaccumulation in fish and wildlife.

761 Arctic marine ecosystems have long food chains (~4–5 levels), and top predators (such as polar bear,
762 ringed seal, beluga whale, and Greenland shark) have elevated Hg concentrations due to feeding at a
763 higher trophic position (St. Louis et al., 2011; McMeans et al., 2015; Brown et al., 2016, 2018). These
764 slow-growing, long-lived top predators are prone to long-term Hg bioaccumulation with age (Masbou
765 et al., 2018; Ewald et al., 2019). Recent quantification of food-chain length using nitrogen stable
766 isotopes demonstrated that regional differences in Hg concentrations of top predators, such as between
767 western and eastern regions of the Canadian Arctic (St. Louis et al., 2011; Brown et al., 2016) is, in
768 part, due to food-chain length. There can also be spatial variation in the carbon sources which support
769 Arctic marine food webs, with potential consequences for food web bioaccumulation of Hg (Cyr et al.,
770 2019). Recent variation of Hg concentrations in beluga whales reflect complex influences of diet,
771 foraging habitat (e.g., benthic versus pelagic feeding), and climate drivers (Desforges et al., 2022;
772 Loseto et al., 2015, Loseto et al., 2008). Considerable research effort has also focused on measuring
773 the rate that Hg biomagnifies through food webs in Arctic ecosystems. A global meta-analysis showed
774 that biomagnification rates were higher at northern latitudes compared with the tropics (Lavoie et al.,
775 2013). Within the Arctic, Hg biomagnification rates can vary among marine and freshwater food webs
776 (Clayden et al., 2015; Lescord et al., 2015; McMeans et al., 2015; Ruus et al., 2015; Fox et al., 2017);
777 although in a study of co-located marine and lacustrine food webs of Arctic char, no difference was
778 found in biomagnification rates (van der Velden et al., 2013b). While some evidence suggests that

779 cold, low productivity ecosystems have higher Hg biomagnification (Lavoie et al., 2013), factors
780 controlling the biomagnification rate of Hg through Arctic food webs remain unclear.

781 Marine subsidies to the diet of terrestrial carnivores and freshwater fish can strongly affect Hg
782 bioaccumulation in the Arctic. Wolves and Arctic foxes that scavenged on carcasses of marine
783 mammals had higher Hg concentrations than those that had an exclusively terrestrial diet (Bocharova
784 et al., 2013; McGrew et al., 2014; Hallanger et al., 2019). In the case of Arctic fox at Svalbard,
785 interannual variation in their Hg concentration was positively related to the extent of sea ice cover,
786 which provides a platform to access marine carcasses (Hallanger et al., 2019). Sea-run Arctic char tend
787 to be larger and have faster growth rates than their lacustrine conspecifics, and therefore they have
788 lower muscle Hg concentrations (Swanson et al., 2011; van der Velden et al., 2013b). The lower Hg
789 bioaccumulation in sea-run char was also attributed to lower Hg exposure at the base of marine food
790 webs relative to lakes in the Arctic (van der Velden et al., 2013a).

791 Migration by wildlife such as seabirds and marine mammals can influence their dietary Hg exposure.
792 Little auks (*Alle alle*) that breed in Greenland during summer and overwinter off the coast of eastern
793 Canada were found to accumulate more Hg during the wintering period (Fort et al., 2014). Similarly,
794 yellow-billed loons (*Gavia adamsii*), which breed in lakes of Arctic North America and overwinter in
795 marine areas of eastern Asia, showed evidence of higher winter exposure (Evers et al., 2014). An
796 evaluation of seasonal variation in Hg contamination of nine species of Arctic seabirds showed, on
797 average, three times higher Hg concentrations in feathers grown at lower latitudes during the non-
798 breeding period than those grown while breeding in the Arctic (Albert et al., 2021). Greater Hg
799 bioaccumulation was found for seabirds overwintering in the western Atlantic relative to the eastern
800 Atlantic or the Pacific region (Albert et al., 2021). Thus, consideration of overwinter exposure at lower
801 latitudes is necessary for interpreting Hg bioaccumulation in migratory Arctic seabirds. Bowhead
802 whales (*Balaena mysticetus*) undergo long seasonal migrations within the Arctic in response to sea ice
803 conditions and prey availability, and examination of their baleen plates provided a record of temporal
804 variation in Hg exposure (Pomerleau et al., 2018). Annual cycles of Hg and $\delta^{13}\text{C}$ values (an isotopic

805 indicator of diet) in the baleen plates showed seasonal shifts in diet and Hg accumulation by bowhead
806 whales in both the eastern and western Arctic.

807 Biological transport is also a pathway for the mobility of contaminants in the Arctic environment such
808 as from migratory spawning fish and bird colonies (Michelutti et al. 2010, Choy et al. 2010,
809 Kristiansen et al., 2019). Migrating salmonids that return to remote freshwater ecosystems to spawn
810 act as important transport (bio)vectors of Hg from ocean to rivers (Blais et al., 2007). Similarly, high-
811 density nesting of seabirds and their deposition of guano can lead to inputs of marine-derived nutrients
812 and Hg to nearby ponds and tundra (Michelutti et al., 2010; Kristiansen et al., 2019, Choy et al., 2010).

813 The body condition of fish and wildlife is sometimes correlated with their Hg concentration, although
814 reported observations have been both positive and negative. Trudel and Rasmussen (2006) showed
815 that Hg concentration may be either positively or negatively correlated to changes in growth and
816 growth efficiency, depending on how fish allocate energy between activity costs and growth rates.
817 Mercury concentrations of northern pike were negatively correlated with growth rate though also
818 influenced by water chemistry and characteristics of the subarctic catchments (Moslemi-Aqdam et al.,
819 2022). Tissue Hg concentrations were negatively correlated with body condition in sea-run char
820 (Evans et al., 2015), caribou (*Rangifer tarandus groenlandicus*; Gamberg et al., 2016) and little auks
821 (Amélineau et al., 2019), while positive correlations were observed for eider (Provencher et al.,
822 2016). For polar bear, both negative and positive correlations were found between tissue Hg
823 concentrations and body condition (Bechshoft et al., 2016; McKinney et al., 2017). Those contrasting
824 observations reflect several potential underlying physiological processes driving the effect of body
825 condition. Fasting or lower food availability and the subsequent loss of body mass can concentrate Hg
826 already in tissues (Keva et al., 2017; Seewagen et al., 2016; Peterson et al., 2018; Martyniuk et al.
827 2020). The reverse has also been suggested, that poorer body condition may be a negative effect of a
828 higher Hg burden (Provencher et al., 2016; Amélineau et al., 2019), although this has been discounted
829 for northern pike (*Esox lucius*; Sandheinrich and Drevnick, 2016). A positive association between
830 body condition and Hg concentration could result from greater Hg intake associated with increased
831 feeding (Bechshoft et al., 2016). Given these bioenergetic influences, seasonal and interannual

832 changes in food resource availability in the Arctic can have important consequences for Hg
833 concentrations in animals (Amélineau et al., 2019; Eckbo et al., 2019; Hallanger et al., 2019).

834

835 **5.3. Organic matter influences on mercury bioaccumulation**

836 While numerous water chemistry parameters have been shown to alter MeHg bioaccumulation in
837 Arctic systems, organic matter has complex linkages with the Hg cycle that can both dampen or
838 promote MeHg bioaccumulation. The complexity of these interactions is reflected in the varying
839 relationships reported between organic matter and Hg in the literature (Lavoie et al., 2019). A recent
840 global review found these relationships to be broadly influenced by geographic location and temporal
841 scale, as well by as human activities (Lavoie et al., 2019). The chemical composition of DOC also
842 plays a critical role in its effects on Hg biogeochemical cycling (Schartup et al., 2015b; Lescord et al.,
843 2018; Jiang et al., 2020), including bioaccumulation (see Section 4.4). For example, OC of terrestrial
844 origin is involved in the transport of MeHg to downstream freshwater and marine environments. In the
845 Subarctic Baltic Sea, interannual variation of seawater MeHg concentrations were related to DOC
846 inputs and composition (Soerensen et al., 2017). In Arctic fresh waters, DOC can promote or inhibit
847 the photodegradation of MeHg depending on the amount of DOC present (Girard et al., 2016) and
848 alter Hg(II) bioavailability to microbes (Chiasson-Gould et al., 2014) because of the multiple roles that
849 organic matter plays in those processes. Dissolved organic carbon can play a key role in promoting
850 MeHg bioaccumulation, although those interactions are complex, and consideration of site-specific
851 conditions are likely important.

852 Numerous studies on the effects of organic matter on Hg accumulation in Arctic biota have been
853 conducted in lakes and rivers across the Arctic over the past ten years. For example, Hudelson et al.
854 (2019) recently reported strong negative correlations between DOC and POC concentrations versus
855 total Hg (THg) levels in landlocked Arctic char (*Salvelinus alpinus*) in lakes around Resolute Bay in
856 Nunavut, Canada. In the same set of lakes, Lescord et al. (2015) reported negative correlations
857 between DOC (in combination with total nutrient levels) and MeHg concentrations in zooplankton. In

858 High Arctic lakes, waters with very low concentrations of DOC were associated with greater
859 bioaccumulation of MeHg at the base of the food web (e.g., chironomid larvae and zooplankton) even
860 when concentrations of MeHg in the water were low (Chételat et al., 2018). Overall, these results
861 suggest that Arctic aquatic systems with lower DOC concentrations, in combination with other
862 physicochemical variables, may have higher rates of Hg bioaccumulation in biota. However, variation
863 in the influence of DOC likely occurs. Furthermore, climate change is rapidly altering both carbon and
864 Hg cycling, resulting in a general increase in concentrations of organic matter in aquatic environments
865 across the Arctic (MacDonald et al., 2005; Isles et al., 2020; Chételat et al. 2022).

866 Some research in Arctic and Subarctic systems has also suggested that the relationship between biotic
867 Hg and organic matter concentrations may not be linear and may, in fact, be dependent on DOC
868 concentrations. More specifically, several studies have reported a curvilinear relationship between
869 DOC concentrations and THg or MeHg concentrations in aquatic biota, with an inflection point of ~8–
870 11 mg/L of DOC (French et al., 2014; Braaten et al., 2018). This relationship was first suggested based
871 on data from the Adirondack lakes and is dependent on system pH (Driscoll et al., 1994). However,
872 given that concentrations of DOC are often strongly correlated with aqueous concentrations of both
873 THg and MeHg, the curvilinear relationship between DOC and biotic Hg concentrations warrants
874 further investigation with consideration of other factors such as water chemistry and trophic position
875 (Chételat et al., 2018; Broadley et al., 2019). For example, Chételat et al. (2018) found that the ratio of
876 aqueous MeHg concentrations to DOC concentrations was a strong and linear predictor of invertebrate
877 MeHg concentrations across an array of Arctic lakes.

878 Recent studies, some related to this curvilinear Hg-DOC relationship, also consider the effect of
879 various measures of DOM quality rather than quantity (i.e., DOC concentrations) have on Hg
880 bioaccumulation. French et al. (2014) suggest that changes in DOM compounds and bioaccessibility
881 may account for changes in Hg bioavailability and bioaccumulation in Arctic freshwater invertebrates;
882 at lower DOC concentrations, Hg was found to be bound with smaller fulvic acids, while at higher
883 DOC concentrations Hg was bound with larger and less bioaccessible humic acids. In the Subarctic,
884 biotic Hg concentrations were similarly higher in systems with higher proportions of more

885 bioaccessible DOM compounds and lower proportions of large and aromatic humic substances
886 (Lescord et al., 2018). Conversely, other studies have reported that humic lakes are associated with
887 greater MeHg bioaccumulation in Subarctic boreal ecoregions (Braaten et al., 2018; Poste et al., 2019).
888 Future studies on Arctic Hg and DOM interactions may benefit from incorporating simple structural
889 measurements, such as SUVA₂₅₄, which provides an estimate of DOM aromaticity (e.g., Skjellberg et
890 al., 2009; Burns et al., 2013).

891 While several studies have shown changes in aqueous Hg levels in relation to permafrost degradation
892 and changing organic matter dynamics, none to our knowledge have examined their combined effect
893 on biotic Hg levels, Hg bioaccumulation potentials, or rates of biomagnification. In the retrogressive
894 thaw slump in the western Canadian Arctic, over 95% of the Hg was released in the particulate phase
895 (St. Pierre et al. 2018) and may not be readily bioavailable. Furthermore, a recent review by Tank et al.
896 (2020) found considerable variation in DOC concentrations and DOM quality among studies
897 examining the impact of permafrost thawing on biogeochemical cycling across aquatic networks.
898 Therefore, future studies assessing the impact of thawing permafrost on downstream bioaccumulation
899 patterns would benefit from considering the effect of DOM, including the structural or quality
900 measurements discussed above.

901

902

903

904 6. Conclusions and recommendations

905

906 Conclusions (in numbered bullets) are provided below under section headings followed by
907 recommendations in italics where appropriate.

908 **How much methylmercury is circulating in the Arctic environment?**

- 909 1. While no estimates of MeHg pools in the environment reservoirs, nor fluxes, were available in
910 the previous AMAP 2011 Mercury Assessment, measurements of MeHg in the Arctic
911 environment over the last decade have allowed for a model budget, as presented in Table 1,
912 Table 2 and Figure 1.
- 913 2. Seawater *in situ* Hg methylation is an important feature leading to the high MeHg portion
914 (32%) and a net export of MeHg (~5–11 Mg/y) from the Arctic Ocean to the North Atlantic.
- 915 3. Arctic sediments are a potential source of MeHg to the Arctic marine water column.

916 *Estimated and modeled MeHg budgets and fluxes are based on limited in situ measurements. Hence,*
917 *more field measurements are needed to fill important knowledge gaps and complete the total Arctic*
918 *MeHg budget. Geographically, the Siberian shelf seas and the central Arctic Ocean are important*
919 *regions where there are important data gaps to address. In addition, future models would benefit from*
920 *a better understanding of how the cycling of Hg varies over seasons and between years. Further work*
921 *is also warranted to better understand and improve the comparability of data between studies (e.g.*
922 *through inter laboratory comparisons and development of new certified reference materials) as future*
923 *synthesis and modelling work will continue to rely on data from multiple labs.*

924

925 *Given the biogeochemical cycling of Hg in the Arctic environment is sensitive to climate change, its*
926 *effects on MeHg production and breakdown in the Arctic, including the connection between climate*

927 *change and changes in DOM discharges, needs further study. Further studies should thus cover both*
928 *mercury speciation (i.e. quantification of methylated Hg forms) as well as ancillary parameters to link*
929 *Hg speciation with changes in water chemistry and biological factors (such as the composition of the*
930 *microbial community).*

931 **What controls the pool of mercury available for bioaccumulation?**

932 4. Methylation and demethylation are key processes controlling the pool of MeHg available for
933 bioaccumulation in Arctic food webs. Methylation of inorganic Hg in the environment is
934 primarily a microbial process while demethylation of MeHg occurs through both biotic and
935 abiotic pathways.

936 5. Organic matter quantity and quality vary greatly among Arctic environments and plays a key
937 role in both the availability of Hg for microbial methylation as well as the uptake of MeHg
938 into organisms.

939 6. Microbial Hg methylation occurs via organisms that carry the *hgcAB* gene pair. Micro-
940 organisms carrying the *hgcAB* gene pair have been identified in Arctic soils and thawing
941 permafrost, and those carrying HgcA or hgcAB-like genes have recently been identified in
942 global ocean seawater, though not in the Arctic Ocean. Organisms carrying genes that encode
943 the *merA* and *merB* operons (a biotic pathway for Hg reduction and demethylation) have also
944 been observed in the Arctic environment.

945
946 7. Evasion of DMHg from the marine boundary layer, and subsequent demethylation of DMHg
947 in the atmosphere, may be a source of MeHg deposition to Arctic snow, surface ocean water,
948 and coastal terrestrial ecosystems. In addition, DMHg may degrade to MeHg through
949 photochemical or dark abiotic pathways while still being dissolved in surface waters.

950

951 *Future mapping of microbial communities carrying the genes involved in methylation and*
952 *demethylation of Hg should be carried out as such mapping will provide critical information*
953 *on these processes across the Arctic, as well as reveal key linkages to the biogeochemical*

954 *cycle of other elements such as carbon (e.g. by understanding the involvement of methanogens*
955 *in Hg methylation).*

956 *The thawing of permafrost represents a potential source of both DOM and Hg to Arctic and*
957 *Subarctic aquatic environments; future work should examine the effects of permafrost-derived*
958 *organic matter on Hg bioaccumulation in Arctic biota, which may add to our understanding*
959 *of the complex Hg-DOM relationship as permafrost thawing increases in a changing climate.*

960 *Future research should also be directed towards a better understanding of the formation and*
961 *environmental fate of DMHg in the Arctic Ocean, and its role in controlling the pool of Hg*
962 *available for bioaccumulation in marine and coastal food webs.*

963

964 **Where are the hotspots of methylmercury in the Arctic?**

965 8. We previously knew that the production of MeHg in natural systems was controlled by
966 substrate availability (e.g., inorganic Hg and organic matter), the presence and abundance of
967 microbial Hg methylators, and MeHg degradation processes. Thus, anoxic systems, such as
968 organic-rich sediments and flooded soils, are understood to be typical hotspots for MeHg
969 production in the Arctic as elsewhere.

970 9. In the terrestrial environment, foliar uptake of Hg from the atmosphere, a process that could be
971 enhanced in recently or soon-to-be deglaciated foreland, is a key driver for sequestering
972 inorganic Hg that may be available for methylation.

973 10. Permafrost thaw in low-relief regions with ice-rich soils and sediments may result in the
974 formation of thermokarst wetlands, ponds and lakes that have been repeatedly identified as Hg
975 methylation hotspots across the Arctic due to the concurrent mobilization of carbon and
976 nutrients, creating conditions suitable for methylation.

977 11. Locally significant MeHg production may occur in newly-deglaciated forelands (especially
978 where wetlands develop), in cryoconite holes on the margins of glacier surfaces or margins,

979 and at the head of glacially-fed fjords and in the sediments within these fjords, though limited
980 evidence is available on the extent of occurrence.

981 12. In the Arctic Ocean., MeHg is found to be enriched in subsurface, oxic seawater at shallower
982 depths (100–300 m) than in the Global Ocean.

983

984 *Future work is needed to better identify sources and pathways of MeHg production and*
985 *storage in the Arctic terrestrial environment and how melting glaciers (and glacier sediments)*
986 *and thawing permafrost would contribute to MeHg loading of downstream environments.*

987 *The processes that are responsible for the enrichment of MeHg in Arctic subsurface seawater*
988 *remain poorly understood. Methylation and demethylation rates of Hg in Arctic seawater need*
989 *to be better quantified.*

990 *Future work should also aim to confirm the occurrence, or lack thereof, of Hg methylation in*
991 *Arctic sea ice and/or snow.*

992

993 **What processes are important for biological uptake of methylmercury?**

994 13. As we understood previously, bioconcentration of aqueous MeHg in bacteria and algae is a
995 critical step in the transfer of Hg to top predators in freshwater and marine environments.

996 Methylmercury is biomagnified through the food web. Thus, the concentration of Hg Arctic
997 animals at the top of the food web depends on the amounts of MeHg entering the food web,
998 the length of the food web as well as seasonal dietary changes, migration, and body condition.

999 14. Sea ice algae may be an important entry point for MeHg in marine ecosystems.

1000 15. Zooplankton in Arctic lakes can have higher concentrations than those in the marine
1001 environment.

1002 16. Recent nitrogen stable isotope measurements specific to Arctic biota have highlighted the
1003 importance of variable trophic position in controlling MeHg concentrations among species and
1004 among populations of the same species distributed across the circumpolar Arctic.

1005

1006

While some evidence suggests cold, low-productivity ecosystems have higher Hg

1007

biomagnification, factors controlling the biomagnification rate of Hg through Arctic food

1008

webs needs to be understood more clearly.

1009

The large magnification at the base of the food chain and its connection to DOM is a critical

1010

process that needs further study.

1011

1012

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