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## Arctic methylmercury cycling

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► **To cite this version:**

Sofi Jonsson, Michelle Nerentorp Mastromonaco, Feiyue Wang, Andrea G Bravo, Warren R.L. Cairns, et al.. Arctic methylmercury cycling. *Science of the Total Environment*, 2022, pp.157445. 10.1016/j.scitotenv.2022.157445 . hal-03737703

**HAL Id: hal-03737703**

**<https://hal.science/hal-03737703v1>**

Submitted on 15 Nov 2022

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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).

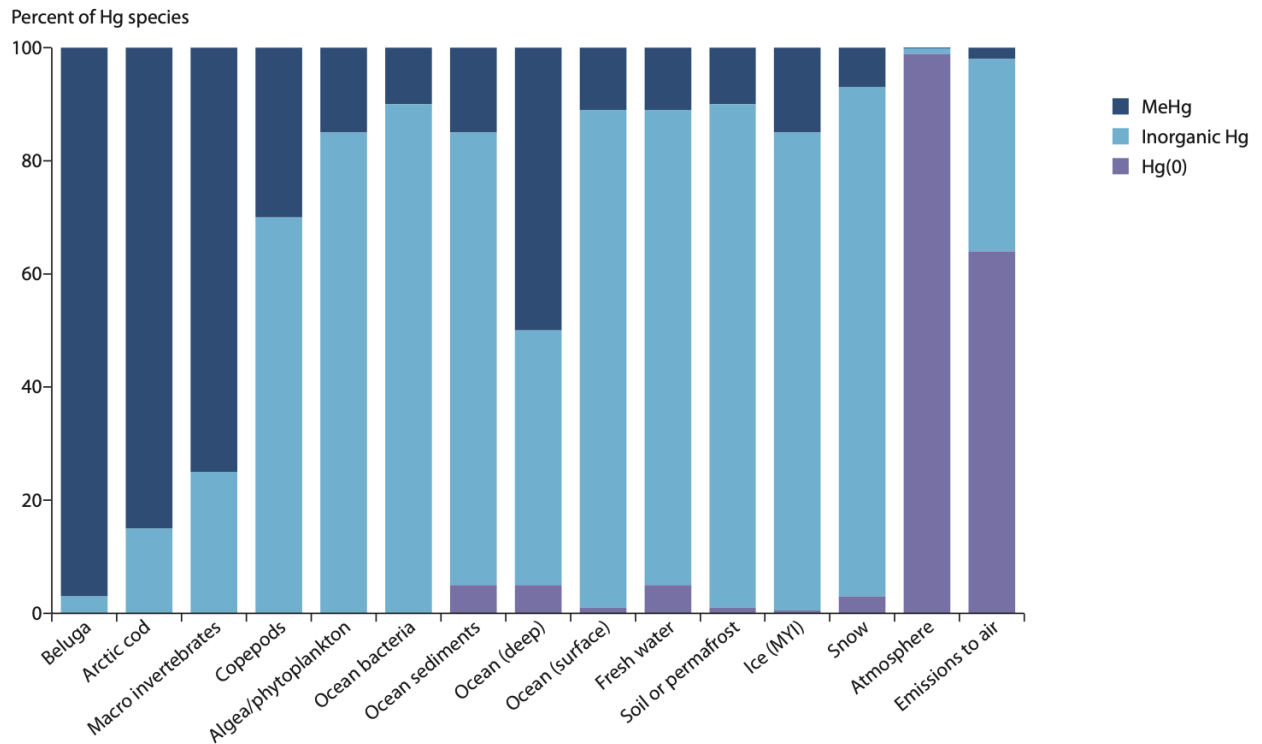
63 The biogeochemical cycle that connects Hg emissions to Hg in Arctic food webs involves inorganic  
64 and organic forms of Hg that undergo a wide range of transformation processes in air, water, and land.  
65 There are three main chemical forms of Hg in Arctic environmental compartments and biota: 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).

111

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 Hg<sup>0</sup> evasion ratio of 0.16  
152 (Soerensen et al, 2016b) and the new Hg<sup>0</sup> 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<sup>-1</sup>). 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 <sup>3</sup> MMHg 3.8±3.1 pg/m <sup>3</sup> 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 <sup>3</sup>	range: <0.5 (LOQ) to 12.7 pg/m <sup>3</sup>		Hudson Bay, Canadian Arctic Archipelago	Baya et al. 2015
	DMHg: 3.8 ± 3.1 pg/m <sup>3</sup>	range: <0.5 (LOQ) to 11.3 pg/m <sup>3</sup>			

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.

<b>Environmental Compartment</b>	<b>MeHg Fluxes</b>	<b>References</b>
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 <sup>2</sup> , day	Lehnherr et al., 2012a
Glacial meltwater (unfiltered), Ellesmere Island	0.03–14.9 g/y 0.49–11.7 mg/km <sup>2</sup>	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

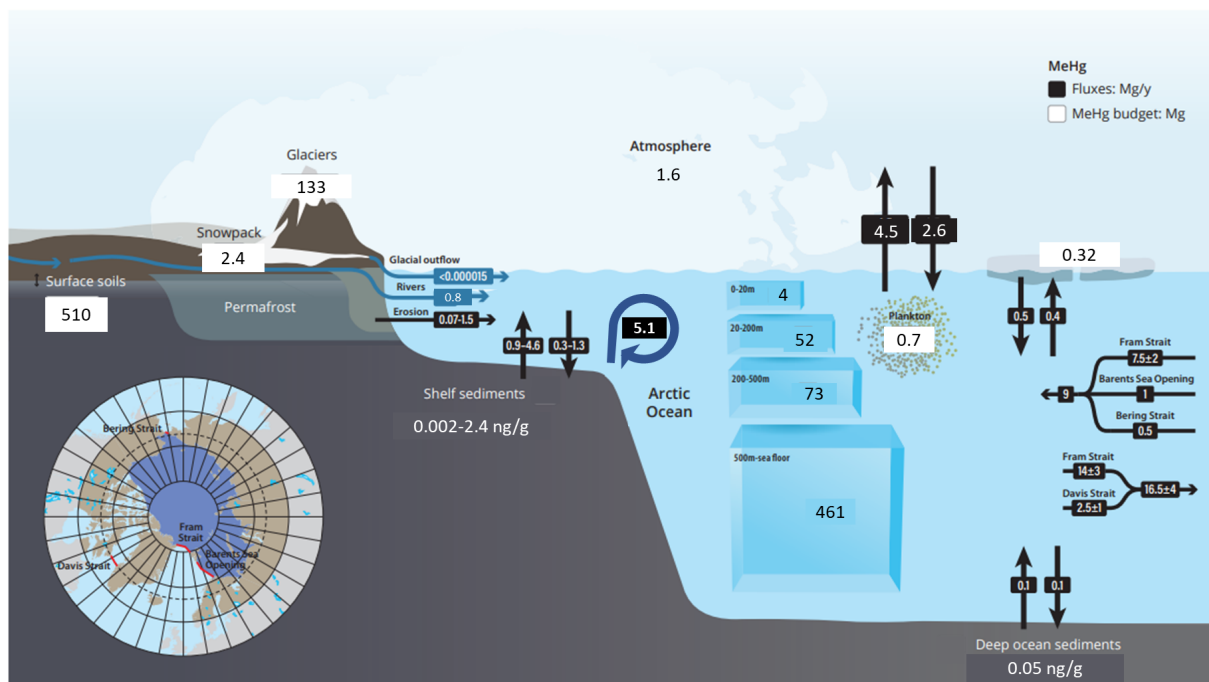
  

<b>Environmental Compartment</b>	<b>MeHg Pool</b>	<b>References</b>
Atmosphere	1.6 (0–3.2) Mg	Baya et al., 2015; Table 3.8.1 in AMAP (2021)
Surface soils	510 (270–749) Mg	St. Pierre et al., 2015; Tarbier et al., 2021
Snowpack	2.4 (0.43–4.3) Mg	St. Pierre et al., 2019, Varty et al., 2021; Jonsson et al.,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

Glaciers	133 ( $\pm 1.2$ ) 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 ( $\pm 2$ ) Mg	Agather et al., 2019;
20–200 m depth	52 ( $\pm 34$ ) Mg	Charette et al., 2020; Kim et al., 2020b; Petrova et al., 2020, Jonsson et al., 2022;
200–500 m depth	73 ( $\pm 44$ ) Mg	Kohler et al., 2022
>500 m depth–bottom)	461 ( $\pm 232$ ) 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/m<sup>2</sup> /day) similar to rates in already identified Hg  
345 methylation hotspots. High concentrations of dissolved MeHg (ranging up to 22 ng L<sup>-1</sup>), 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/km<sup>2</sup>/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

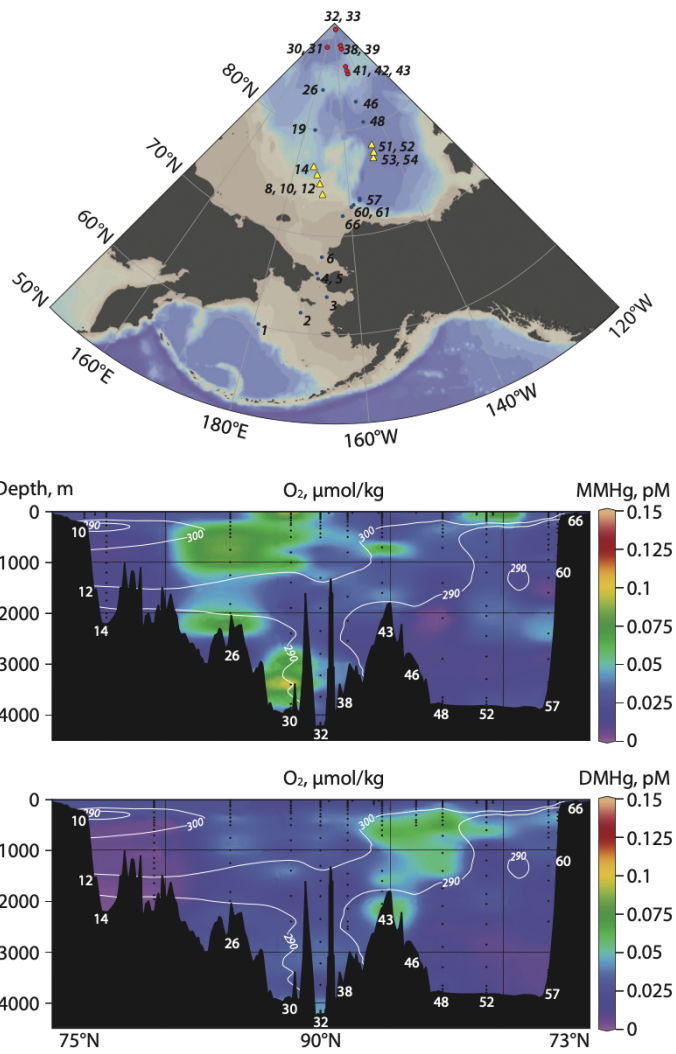
### 487 4.2.1 Seawater

488 MeHg concentrations in the western Arctic Ocean have been reported to be  $0.011\pm 0.010$  ng/L in  
489 filtered seawater (Agather et al., 2019, Figure 3) and  $0.046\pm 0.024$  ng/L in unfiltered seawater (Wang  
490 et al., 2018). Heimbürger et al. (2015) found similar levels of  $0.045\pm 0.017$  ng/L and  $0.034\pm 0.022$  ng/L  
491 in unfiltered seawater in the Eastern Arctic and Central Ocean. Methylmercury (MeHg) inflow to the  
492 Arctic Ocean totalled  $9\pm 2$  Mg/yr, most of which enters with deep and mid-depth waters of the Atlantic  
493 Ocean ( $7.5\pm 2$  Mg/yr; Petrova et al., 2020). Similar to THg, more MeHg flows out of the Arctic Ocean  
494 into the North Atlantic ( $16.5\pm 3$  Mg/yr,  $14\pm 2$  Mg/yr via the Fram Strait,  $2.5\pm 1$  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 (Lehnerr 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  $7.5\pm 2$  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 (O<sub>2</sub>) 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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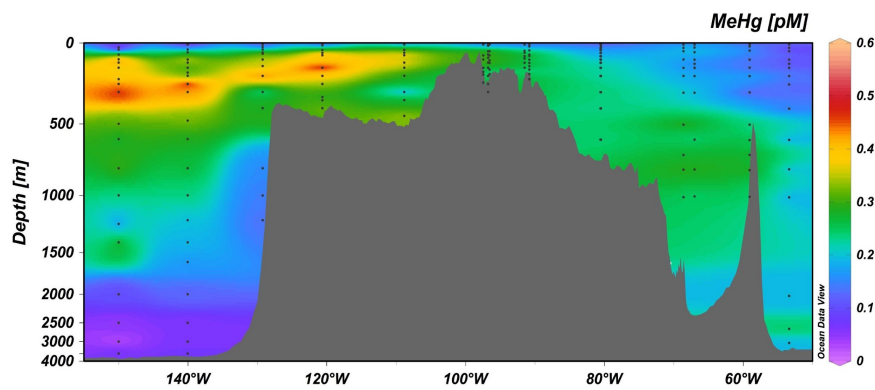
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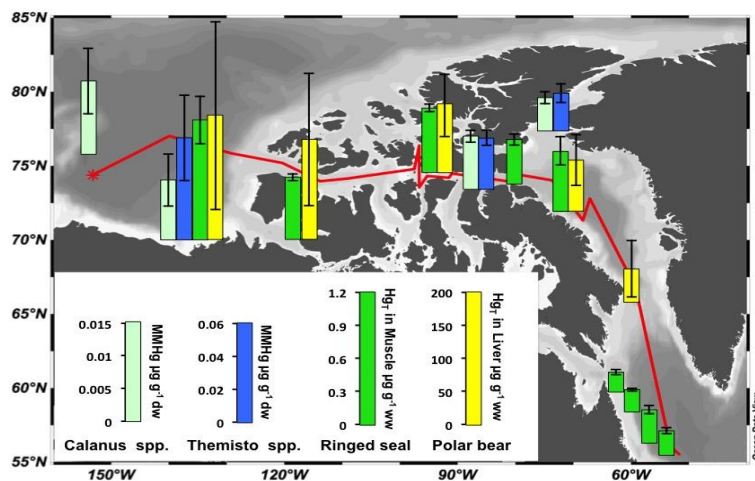
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**Figure 4.** Methylmercury (MeHg) concentrations in unfiltered seawater (top panel) and the marine food web (bottom panel) along a longitudinal (west-to-east) section (the red line in the bottom panel) across the Canadian Arctic and Labrador Sea. Bar charts in the bottom panel show mean concentrations of monomethylmercury (MMHg) in *Calanus* spp. and *Themisto* spp. collected from 1998 to 2012 ( $\pm 1$ 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  $4.6 \pm 0.2$  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**

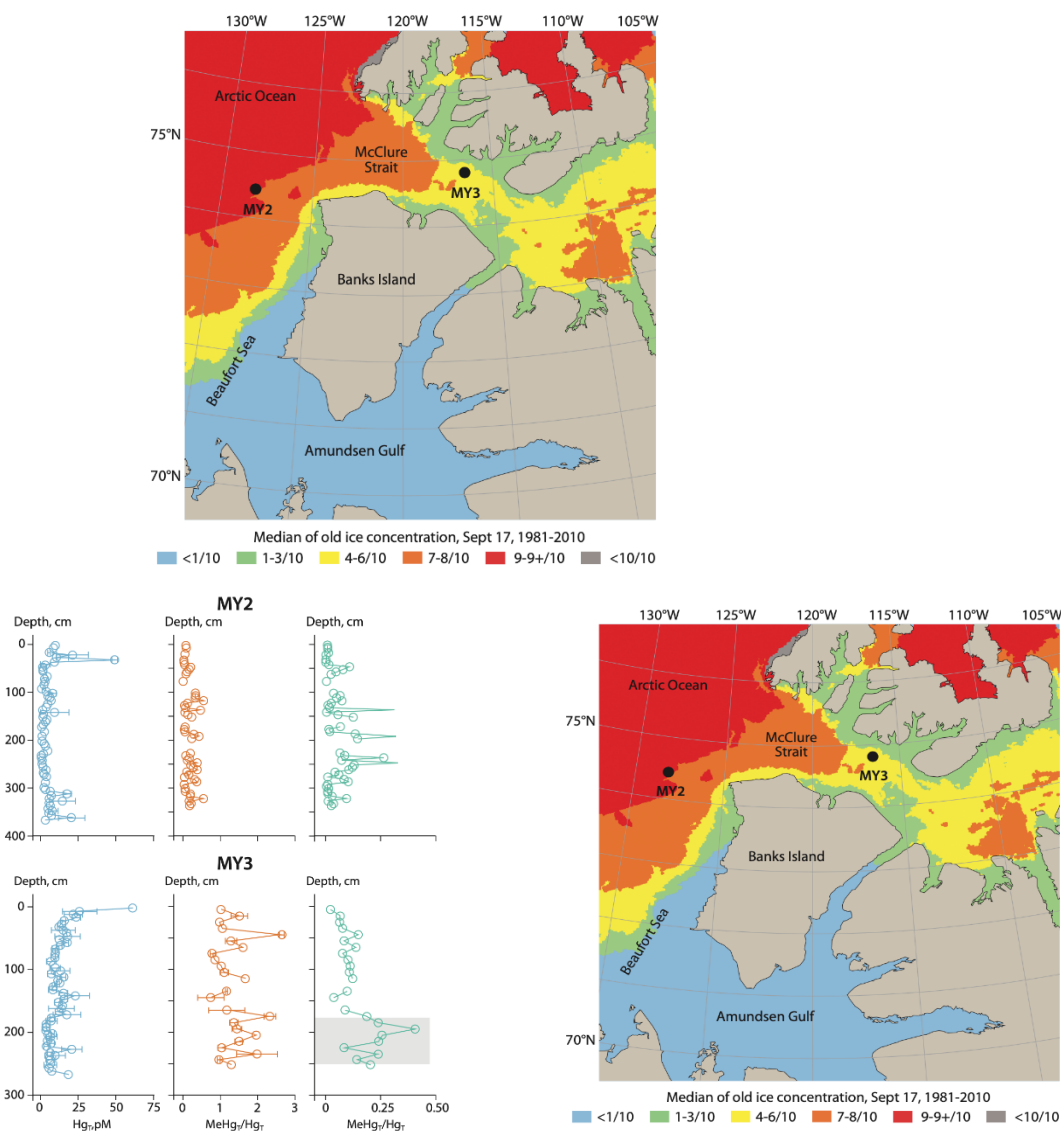
580 The sea ice environment, including first-year ice, multi-year ice and the overlying snow cover,  
581 provides a spatially and temporally dynamic interface between the ocean and the atmosphere in the  
582 Arctic Ocean where physical transport, chemical reactions, and biological uptake occur (Wang 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 SUVA<sub>254</sub>, 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

## 1013 Acknowledgements

1014 This review is a result of efforts led by the Arctic Monitoring and Assessment Programme (AMAP)  
1015 and the AMAP Mercury Expert Group to produce the 2021 AMAP Mercury Assessment. We thank  
1016 Rune Dietz (Aarhus University) and Simon Wilson (AMAP) for provided guidance and support.  
1017 Graphics production and technical editing were supported by AMAP. We also acknowledge Adam  
1018 Kirkwood (Laurentian University), Daniel Obrist (University of Massachusetts), Peter Outridge  
1019 (Geological Survey of Canada/ University of Manitoba), Kyra A. St Pierre (University of British  
1020 Columbia) and Christian Zdanowicz (Uppsala University) for their contributions to chapter 4 of the  
1021 2021 AMAP Mercury Assessment. This work was supported by the Swedish Research Council (2017-  
1022 05275 to SJ), the Swedish Research Council for Environment, Agricultural Sciences and Spatial  
1023 Planning (2020-01868 to S.J.), Environment and Climate Change Canada (to J.C.), Chantier Arctique  
1024 Francais (Pollution in the Arctic System to L.E.H.B) and the AXA Research Fund (to L.E.H.B).

1025

1026

## 1027 References

1028 Adam D. Morris, Frank R. Rigét, Simon Wilson, Rob Fryer, Philippe J. Thomas, et al. 2022. Temporal  
1029 trends of mercury in Arctic biota: 10 more years of progress in Arctic monitoring. *Science of the Total*  
1030 *Enviroment*, in review.

1031 Agather, A.M., Bowman, K.L., Lamborg, C.H. and Hammerschmidt, C.R., 2019. Distribution of  
1032 mercury species in the Western Arctic Ocean (U.S. GEOTRACES GN01). *Marine Chemistry*,  
1033 216:103686.

1034 Albert, C., Helgason, H.H., Brault-Favrou, M., Robertson, G.J., Descamps, S., Amélineau, F., et al.  
1035 2021. Seasonal variation of mercury contamination in Arctic seabirds: A pan-Arctic assessment. *Science*  
1036 *of the Total Environment* 750, 142201.

1037 AMAP, 2011. AMAP Assessment 2011: Mercury in the Arctic. Arctic Monitoring and Assessment  
1038 Programme (AMAP), Oslo, Norway. xiv + 193 pp.

1039 AMAP/UNEP, 2013. Technical Background Report to the Global Atmospheric Mercury Assessment  
1040 2013. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway/United Nations  
1041 Environment Programme (UNEP), Chemicals Branch, Geneva, Switzerland. vi + 263pp.

1042 AMAP, 2021. Assessment 2021: Mercury in the Arctic. Arctic Monitoring and Assessment Programme  
1043 (AMAP), Oslo, Norway.

- 1044 Amélineau, F., Grémillet, D., Harding, A.M.A., Walkusz, W., Choquet, R. and Fort, J., 2019. Arctic  
1045 climate change and pollution impact little auk foraging and fitness across a decade. *Scientific Reports*,  
1046 9 (1):1014.
- 1047 Baldi, F., Pepi, M. and Filippelli, M., 1993. Methylmercury resistance in *Desulfovibrio desulfuricans*  
1048 strains in relation to methylmercury degradation. *Applied and Environmental Microbiology*, 59  
1049 (8):2479-2485.
- 1050 Barkay, T., Miller, S.M. and Summers, A.O., 2003. Bacterial mercury resistance from atoms to  
1051 ecosystems. *FEMS Microbiology Reviews*, 27 (2-3):355-384.
- 1052 Baya, P.A., Gosselin, M., Lehnher, I., St. Louis, V.L. and Hintelmann, H., 2015. Determination of  
1053 monomethylmercury and dimethylmercury in the Arctic marine boundary layer. *Environmental*  
1054 *Science & Technology*, 49 (1):223-232
- 1055 Beattie, S.A., Armstrong, D., Chaulk, A., Comte, J., Gosselin, M. and Wang, F., 2014. Total and  
1056 methylated mercury in Arctic multiyear sea ice. *Environmental Science & Technology*, 48 (10):5575-  
1057 5582.
- 1058 Bechshoft, T., Derocher, A.E., Richardson, E., Lunn, N.J. and St. Louis, V.L., 2016. Hair mercury  
1059 concentrations in Western Hudson Bay polar bear family groups. *Environmental Science & Technology*,  
1060 50 (10):5313-5319.
- 1061 Beldowska, M. and Kobos, J., 2016. Mercury concentration in phytoplankton in response to warming  
1062 of an autumn - winter season. *Environmental Pollution*, 215:38-47.
- 1063 Bidleman, T.F., Stern, G.A., Tomy, G.T., Hargrave, B.T., Jantunen, L.M. and MacDonald, R.W.,  
1064 2013. Scavenging amphipods: sentinels for penetration of mercury and persistent organic chemicals  
1065 into food webs of the deep Arctic Ocean. *Environmental Science & Technology*, 47 (11):5553-5561.
- 1066 Black, F.J., Conaway, C.H. and Flegal, A.R., 2009. Stability of dimethyl mercury in seawater and its  
1067 Conversion to monomethyl mercury. *Environmental Science & Technology*, 43 (11):4056-4062.
- 1068 Blais, J. M., Macdonald, R.W., Mackay, D., Webster, E., Harvey, C. and Smol, J. P., 2007. Biologically  
1069 mediated transport of contaminants to aquatic systems. *Environmental Science & Technology*, 41  
1070 (4):1075-1084.
- 1071 Blum, J.D., Popp, B.N., Drazen, J.C., Anela Choy, C. and Johnson, M.W., 2013. Methylmercury  
1072 production below the mixed layer in the North Pacific Ocean. *Nature Geoscience*, 6 (10):879-884.
- 1073 Bocharova, N., Treu, G., Czirják, G.Á., Krone, O., Stefanski, V., Wibbelt, G., Unnsteinsdóttir, E.R.,  
1074 Hersteinsson, P., Schares, G., Doronina, L., Goltsman, M. and Greenwood, A.D., 2013. Correlates  
1075 between feeding ecology and mercury levels in historical and modern Arctic foxes (*Vulpes lagopus*).  
1076 *PLOS ONE*, 8 (5):e60879.
- 1077 Bowman, K.L., Hammerschmidt, C.R., Lamborg, C.H. and Swarr, G., 2015. Mercury in the North  
1078 Atlantic Ocean: the U.S. GEOTRACES zonal and meridional sections. *Deep Sea Research Part II:*  
1079 *Topical Studies in Oceanography*, 116:251-26.
- 1080 Bowman, K.L., Hammerschmidt, C.R., Lamborg, C.H., Swarr, G.J. and Agather, A.M., 2016.  
1081 Distribution of mercury species across a zonal section of the eastern tropical South Pacific Ocean  
1082 (U.S. GEOTRACES GP16). *Marine Chemistry*, 186:156-166.
- 1083 Bowman, K.L., Collins, R.E., Agather, A.M., Lamborg, C.H., Hammerschmidt, C.R., Kaul, D.,  
1084 Dupont, C.L., Christensen, G.A. and Elias, D.A., 2020a. Distribution of mercury-cycling genes in the

- 1085 Arctic and equatorial Pacific Oceans and their relationship to mercury speciation. *Limnology and*  
1086 *Oceanography*, 65 (S1):S310-S320.
- 1087 Boetius, A., Albrecht, S., Bakker, K., Bienhold, C., Felden, J., Fernández-Méndez, M., Hendricks, S.,  
1088 Katlein, C., Lalande, C., Krumpfen, T., Nicolaus, M., Peeken, I., Rabe, B., Rogacheva, A., Rybakova,  
1089 E., Somavilla, R., Wenzhöfer, F., and RV Polarstern ARK27-3-Shipboard Science Party: Export of  
1090 algal biomass from the melting Arctic sea ice, *Science*, 339, 1430–1432,  
1091 doi:10.1126/science.1231346, 2013.
- 1092 Bowman, K.L., Lamborg, C.H. and Agather, A.M., 2020b. A global perspective on mercury cycling in  
1093 the ocean. *Science of the Total Environment*, 710:136166.
- 1094 Braaten, H.F.V., de Wit, H.A., Larssen, T. and Poste, A.E., 2018. Mercury in fish from Norwegian lakes:  
1095 the complex influence of aqueous organic carbon. *Science of the Total Environment*, 627:341-348.
- 1096 Bradley MA, Barst BD, Basu N. A Review of Mercury Bioavailability in Humans and Fish. *Int J Environ*  
1097 *Res Public Health*. 2017 Feb 10;14(2):169. doi: 10.3390/ijerph14020169. PMID: 28208586; PMCID:  
1098 PMC5334723.
- 1099 Bratkič, A., Vahčić, M., Kotnik, J., Obu Vazner, K., Begu, E., Woodward, E.M.S. and Horvat, M., 2016.  
1100 Mercury presence and speciation in the South Atlantic Ocean along the 40°S transect. *Global*  
1101 *Biogeochemical Cycles*, 30 (2):105-119.
- 1102 Broadley, H.J., Cottingham, K.L., Baer, N.A., Weathers, K.C., Ewing, H.A., Chaves-Ulloa, R.,  
1103 Chickering, J., Wilson, A.M., Shrestha, J. and Chen, C.Y., 2019. Factors affecting MeHg  
1104 bioaccumulation in stream biota: the role of dissolved organic carbon and diet. *Ecotoxicology*, 28  
1105 (8):949-963.
- 1106 Brown, T.M., Fisk, A.T., Wang, X., Ferguson, S.H., Young, B.G., Reimer, K.J. and Muir, D.C.G.,  
1107 2016. Mercury and cadmium in ringed seals in the Canadian Arctic: influence of location and diet.  
1108 *Science of the Total Environment*, 545-546:503-511.
- 1109 Brown, T.M., Macdonald, R.W., Muir, D.C.G. and Letcher, R.J., 2018. The distribution and trends of  
1110 persistent organic pollutants and mercury in marine mammals from Canada's Eastern Arctic. *Science*  
1111 *of the Total Environment*, 618:500-517.
- 1112 Burns, D.A., Aiken, G.R., Bradley, P.M., Journey, C.A. and Schelker, J., 2013. Specific ultra-violet  
1113 absorbance as an indicator measurement of mercury sources in an Adirondack River basin.  
1114 *Biogeochemistry*, 113 (1-3):451-466.
- 1115 Burt, A., Wang, F., Pučko, M., Mundy, C.-J., Gosselin, M., Philippe, B., Poulin, M., Tremblay, J.-É.  
1116 and Stern, G.A., 2013. Mercury uptake within an ice algal community during the spring bloom in first-  
1117 year Arctic sea ice. *Journal of Geophysical Research: Oceans*, 118 (9):4746-4754.
- 1118 Camacho, A., Rochera, C., Hennebelle, R., Ferrari, C. and Quesada, A., 2015. Total mercury and  
1119 methyl-mercury contents and accumulation in polar microbial mats. *Science of the Total Environment*,  
1120 509-510:145-153.
- 1121 Capo, E., Bravo, A.G., Soerensen, A.L., Bertilsson, S., Pinhassi, J., Feng, C., Andersson, A.F., Buck,  
1122 M. and Björn, E., 2020. Deltaproteobacteria and spirochaetes-like bacteria are abundant putative  
1123 mercury methylators in oxygen-deficient water and marine particles in the Baltic Sea. *Frontiers in*  
1124 *Microbiology*, 11:2277.
- 1125 Charette, M.A., Kipp, L.E., Jensen, L.T., Dabrowski, J.S., Whitmore, L.M., Fitzsimmons, J.N.,  
1126 Williford, T., Ulfsbo, A., Jones, E., Bundy, R.M., Vivancos, S.M., Pahnke, K., John, S.G., Xiang, Y.,  
1127 Hatta, M., Petrova, M.V., Heimbürger-Boavida, L.-E., Bauch, D., Newton, R., Pasqualini, A., Agather,

- 1128 A.M., Amon, R.M.W., Anderson, R.F., Andersson, P.S., Benner, R., Bowman, K.L., Edwards, R.L.,  
1129 Gdaniec, S., Gerringa, L.J.A., González, A.G., Granskog, M., Haley, B., Hammerschmidt, C.R.,  
1130 Hansell, D.A., Henderson, P.B., Kadko, D.C., Kaiser, K., Laan, P., Lam, P.J., Lamborg, C.H., Levier,  
1131 M., Li, X., Margolin, A.R., Measures, C., Middag, R., Millero, F J., Moore, W S., Paffrath, R.,  
1132 Planquette, H., Rabe, B., Reader, H., Rember, R., Rijkenberg, M.J.A., Roy-Barman, M., Rutgers van  
1133 der Loeff, M., Saito, M., Schauer, U., Schlosser, P., Sherrell, R M., Shiller, A.M., Slagter, H., Sonke,  
1134 J.E., Stedmon, C., Woosley, R.J., Valk, O., van Ooijen, J. and Zhang, R., 2020. The Transpolar Drift as  
1135 a source of riverine and shelf-derived trace elements to the central Arctic Ocean. *Journal of Geophysical*  
1136 *Research: Oceans*, 125 (5):e2019JC015920.
- 1137 Chaulk, A.; Stern, G. A.; Armstrong, D.; Barber, D. G.; Wang, F. Mercury distribution and transport  
1138 across the ocean-sea-ice-atmosphere interface in the Arctic Ocean. *Environ. Sci. Technol.* 2011,  
1139 45:1866–1872.
- 1140 Chételat, J., Ackerman, J.T., Eagles-Smith, C.A. and Hebert, C.E., 2020. Methylmercury exposure in  
1141 wildlife: a review of the ecological and physiological processes affecting contaminant concentrations  
1142 and their interpretation. *Science of the Total Environment*, 711:135117.
- 1143 Chételat, J. and Amyot, M., 2009. Elevated methylmercury in High Arctic *Daphnia* and the role of  
1144 productivity in controlling their distribution. *Global Change Biology*, 15 (3):706-718.
- 1145 Chételat, J., Amyot, M. and Cloutier, L., 2012. Shifts in elemental composition, methylmercury content  
1146 and  $\delta^{15}\text{N}$  ratio during growth of a High Arctic copepod. *Freshwater Biology*, 57 (6):1228-1240.
- 1147 Chételat, J., Cloutier, L. and Amyot, M., 2010. Carbon sources for lake food webs in the Canadian  
1148 High Arctic and other regions of Arctic North America. *Polar Biology*, 33 (8):1111–1123.
- 1149 Chételat, J., Richardson, M.C., MacMillan, G.A., Amyot, M. and Poulain, A.J., 2018. Ratio of  
1150 methylmercury to dissolved organic carbon in water explains methylmercury bioaccumulation across a  
1151 latitudinal gradient from north-temperate to Arctic lakes. *Environmental Science & Technology*, 52  
1152 (1):79-88.
- 1153 Chételat, J., Amyot, M., Arp, P., Blais, J.M., Depew, D., Emmerton, C.A., Evans, M., Gamberg, M.,  
1154 Gantner, N., Girard, C., Graydon, J., Kirk, J., Lean, D., Lehnerr, I., Muir, D., Nasr, M., Poulain, A.J.,  
1155 Power, M., Roach, P., Stern, G., Swanson, H. and van der Velden, S., 2015. Mercury in freshwater  
1156 ecosystems of the Canadian Arctic: recent advances on its cycling and fate. *Science of the Total*  
1157 *Environment*, 509-510:41-66.
- 1158 Chételat, J., McKinney, M.A., Amyot, M., Dastoor, A., Douglas, T.A., Heimbürger-Boavida, L.-E.,  
1159 Kirk, J., Kahilainen, K.K., Outridge, P.M., Pelletier, N., Skov, H., St Pierre, K., Vuorenmaa, J., Wang,  
1160 F., 2022. Climate change and mercury in the Arctic: Abiotic interactions. *Sci. Total Environ.* 824,  
1161 153715
- 1162 Chiasson-Gould, S.A., Blais, J.M. and Poulain, A.J., 2014. Dissolved Organic Matter Kinetically  
1163 Controls Mercury Bioavailability to Bacteria. *Environmental Science & Technology*, 48 (6):3153-3161.
- 1164 Choy ES, Gauthier M, Mallory ML, Smol JP, Douglas MSV, Lean D, et al. An isotopic investigation  
1165 of mercury accumulation in terrestrial food webs adjacent to an Arctic seabird colony. *Science of the*  
1166 *Total Environment* 2010; 408: 1858-1867.
- 1167 Clayden, M.G., Arsenault, L.M., Kidd, K.A., O’Driscoll, N.J. and Mallory, M.L., 2015. Mercury  
1168 bioaccumulation and biomagnification in a small Arctic polynya ecosystem. *Science of the Total*  
1169 *Environment*, 509-510:206-215.
- 1170 Cook, J., Edwards, A., Takeuchi, N. and Irvine-Fynn, T., 2016. Cryoconite: the dark biological secret  
1171 of the cryosphere. *Progress in Physical Geography: Earth and Environment*, 40 (1):66-111.

- 1172 Cossa, D., Heimbürger, L.-E., Lannuzel, D., Rintoul, S.R., Butler, E.C.V., Bowie, A.R., Averty, B.,  
1173 Watson, R.J. and Remenyi, T., 2011. Mercury in the Southern Ocean. *Geochimica et Cosmochimica*  
1174 *Acta*, 75 (14):4037-4052.
- 1175 Cossa, D., Averty, B. and Pirrone, N., 2009. The origin of methylmercury in open Mediterranean waters.  
1176 *Limnology and Oceanography*, 54 (3):837-844.
- 1177 Cossa, D., Madron, X.D. de, Schäfer, J., Lancelour, L., Guédron, S., Buscail, R., Thomas, B., Castelle,  
1178 S. and Naudin, J.-J., 2017. The open sea as the main source of methylmercury in the water column of  
1179 the Gulf of Lions (Northwestern Mediterranean margin). *Geochimica et Cosmochimica Acta*, 199:222-  
1180 237.
- 1181 Cossa, D., Harmelin-Vivien, M., Mellon-Duval, C., Loizeau, V., Averty, B., Crochet, S., Chou, L. and  
1182 Cadiou, J.-F., 2012. Influences of bioavailability, trophic Position, and growth on methylmercury in  
1183 hakes (*Merluccius merluccius*) from northwestern Mediterranean and northeastern Atlantic.  
1184 *Environmental Science & Technology*, 46 (9):4885-4893.
- 1185 Cossa, D., Garnier, C., Buscail, R., Elbaz-Poulichet, F., Mikac, N., Patel-Sorrentino, N., Tessier, E.,  
1186 Rigaud, S., Lenoble, V. and Gobeil, C., 2014. A Michaelis–Menten type equation for describing  
1187 methylmercury dependence on inorganic mercury in aquatic sediments. *Biogeochemistry*, 119 (1):35-  
1188 43
- 1189 Cyr, A., López, J.A., Rea, L., Wooller, M.J., Loomis, T., Mcdermott, S. and O'Hara, T.M., 2019.  
1190 Mercury concentrations in marine species from the Aleutian Islands: spatial and biological determinants.  
1191 *Science of the Total Environment*, 664:761-770.
- 1192 Dastoor A, Ryzhkov A, Durnford D, Lehnerr I, Steffen A, Morrison H. 2015, Atmospheric mercury  
1193 in the Canadian Arctic. Part II: Insight from modeling. *Science of The Total Environment*, 509-510:  
1194 16-27.
- 1195 Dastoor A., Angot H., Bieser J., Christensen J.H., Douglas T.A., Heimbürger-Boavida L-E., Jiskra M.,  
1196 Mason R.P., McLagan D.S., Obrist D., Outridge P.M, Petrova M.V, Ryjkov A., St. Pierre K.A.,  
1197 Schartup A.T., Soerensen A.L., Toyota K., Travnikov O., Wilson S.J., Zdanowicz C., 2022, Arctic  
1198 mercury cycling, *Nature Reviews Earth & Environment* volume 3, pages270–286 (2022)
- 1199 Desforges J-P, Outridge P, Hobson KA, Heide-Jørgensen MP, Dietz R. Anthropogenic and Climatic  
1200 Drivers of Long-Term Changes of Mercury and Feeding Ecology in Arctic Beluga (*Delphinapterus*  
1201 *leucas*) Populations. *Environmental Science & Technology* 2022; 56: 271-281.
- 1202 DiMento BP, Mason RP, Brooks S, Moore C. The impact of sea ice on the air-sea exchange of mercury  
1203 in the Arctic Ocean. *Deep Sea Research Part I: Oceanographic Research Papers* 2019; 144: 28-  
1204 38. Loseto, L.L., Lean, D.R. and Siciliano, S.D., 2004. Snowmelt sources of methylmercury to High  
1205 Arctic ecosystems. *Environmental Science & Technology* 38 (11):3004-3010.
- 1206 Douglas, T.A., Loseto, L.L., Macdonald, R.W., Outridge, P., Dommergue, A., Poulain, A., Amyot, M.,  
1207 Barkay, T., Berg, T., Chételat, J., Constant, P., Evans, M., Ferrari, C., Gantner, N., Johnson, M.S., Kirk,  
1208 J., Kroer, N., Larose, C., Lean, D., Nielsen, T.G., Poissant, L., Rognerud, S., Skov, H., Sørensen, S.,  
1209 Wang, F.Y., Wilson, S. and Zdanowicz, C.M., 2012. The fate of mercury in Arctic terrestrial and aquatic  
1210 ecosystems, a review. *Environmental Chemistry*, 9 (4):321-355.
- 1211 Douglas, T.A., Sturm, M., Blum, J.D., Polashenski, C., Stuefer, S., Hiemstra, C., Steffen, A., Filhol, S.  
1212 and Prevost, R., 2017. A pulse of mercury and major ions in snowmelt runoff from a small Arctic  
1213 Alaska watershed. *Environmental. Science & Technology*, 51 (19):11145-11155.
- 1214 Douglas, T.A. and Blum, J.D., 2019. Mercury isotopes reveal atmospheric gaseous mercury deposition  
1215 directly to the Arctic coastal snowpack. *Environmental Science & Technology Letters*, 6 (4):235-242.

- 1216 Douglas TA, Hiemstra CA, Anderson JE, Barbato RA, Bjella KL, Deeb EJ, Gelvin AB, Nelsen PE,  
1217 Newman SD, Saari SP, Wagner AM. 2021, Recent degradation of Interior Alaska permafrost mapped  
1218 with ground surveys, geophysics, deep drilling, and repeat airborne LiDAR. *The Cryosphere*  
1219 *Discussions*. Feb 22:1-39.
- 1220 Driscoll, C.T., Yan, C., Schofield, C.L., Munson, R. and Holsapple, J., 1994. The mercury cycle and  
1221 fish in the Adirondack lakes. *Environmental Science & Technology*, 28 (3):136A-143A.
- 1222 Driscoll CT, Mason RP, Chan HM, Jacob DJ, Pirrone N. 2013. Mercury as a Global Pollutant: Sources,  
1223 Pathways, and Effects. *Environmental Science & Technology*, 47: 4967-4983
- 1224 Drott, A., Lambertsson, L., Björn, E. and Skjällberg, U., 2007. Importance of dissolved neutral mercury  
1225 sulfides for methyl mercury production in contaminated sediments. *Environmental Science &*  
1226 *Technology*, 41 (7):2270-2276.
- 1227 Durnford, D., Dastoor, A., Figueras-Nieto, D., and Ryjkov, A. 2010. Long range transport of mercury  
1228 to the Arctic and across Canada, *Atmos. Chem. Phys.*, 10, 6063–6086, [https://doi.org/10.5194/acp-10-](https://doi.org/10.5194/acp-10-6063-2010)  
1229 [6063-2010](https://doi.org/10.5194/acp-10-6063-2010).
- 1230 Dietz R, Letcher R.J., Aars J., Andersen M., Andriashek et al. 2022 What are the toxicological effects  
1231 of mercury in Arctic marine and terrestrial mammals? *Science of the Total Environment*, in review.
- 1232 Ebinghaus, R., Kock, H.H., Temme, C., Einax, J.W., Löwe, A.G., Richter, A., Burrows, J.P. and  
1233 Schroeder, W.H., 2002. Antarctic springtime depletion of atmospheric mercury. *Environmental*  
1234 *Science & Technology*, 36 (6):1238-1244.
- 1235 Eckbo, N., Le Bohec, C., Planas-Bielsa, V., Warner, N.A., Schull, Q., Herzke, D., Zahn, S., Haarr, A.,  
1236 Gabrielsen, G.W. and Borgå, K., 2019. Individual variability in contaminants and physiological status  
1237 in a resident Arctic seabird species. *Environmental Pollution*, 249:191-199.
- 1238 Emmerton, C.A., Graydon, J.A., Gareis, J.A.L., St. Louis, V.L., Lesack, L.F.W., Banack, J.K.A.,  
1239 Hicks, F. and Nafziger, J., 2013. Mercury export to the Arctic Ocean from the Mackenzie River,  
1240 Canada. *Environmental Science & Technology*, 47 (14):7644-7654.
- 1241 Evans, M.S., Muir, D.C.G., Keating, J. and Wang, X., 2015. Anadromous char as an alternate food  
1242 choice to marine animals: A synthesis of Hg concentrations, population features and other influencing  
1243 factors. *Science of the Total Environment*, 509-510:175-194.
- 1244 Evers, D.C., Schmutz, J.A., Basu, N., DeSorbo, C.R., Fair, J., Gray, C.E., Paruk, J.D., Perkins, M.,  
1245 Regan, K., Uher-Koch, B.D. and Wright, K.G., 2014. Historic and contemporary mercury exposure  
1246 and potential risk to yellow-billed loons (*Gavia adamsii*) breeding in Alaska and Canada. *Waterbirds*,  
1247 37 (sp1):147-159.
- 1248 Ewald JD, Kirk JL, Li M, Sunderland EM. Organ-specific differences in mercury speciation and  
1249 accumulation across ringed seal (*Phoca hispida*) life stages. *Science of the Total Environment* 2019;  
1250 650: 2013-2020.
- 1251 Fahnestock, M.F., Bryce, J.G., McCalley, C.K., Montesdeoca, M., Bai, S., Li, Y., Driscoll, C.T., Crill,  
1252 P.M., Rich, V.I. and Varner, R.K., 2019. Mercury reallocation in thawing Subarctic peatlands.  
1253 *Geochemical Perspectives Letters*, 11:33-38.
- 1254 Fort, J., Robertson, G.J., Grémillet, D., Traisnel, G. and Bustamante, P., 2014. Spatial ecotoxicology:  
1255 migratory Arctic seabirds are exposed to mercury contamination while overwintering in the Northwest  
1256 Atlantic. *Environmental Science & Technology*, 48 (19):11560-11567.

- 1257 Foster, K.L., Stern, G.A., Pazerniuk, M.A., Hickie, B., Walkusz, W., Wang, F. and Macdonald, R.W.,  
1258 2012. Mercury biomagnification in marine zooplankton food webs in Hudson Bay. *Environmental*  
1259 *Science & Technology*, 46 (23):12952-12959.
- 1260 Fox, A.L., Hughes, E.A., Trocine, R.P., Trefry, J.H., Schonberg, S.V., McTigue, N.D., Lasorsa, B.K.,  
1261 Konar, B. and Cooper, L.W., 2014. Mercury in the northeastern Chukchi Sea: distribution patterns in  
1262 seawater and sediments and biomagnification in the benthic food web. *Deep Sea Research Part II:*  
1263 *Topical Studies in Oceanography*, 102:56-67.
- 1264 Fox, A.L., Trefry, J.H., Trocine, R.P., Dunton, K.H., Lasorsa, B.K., Konar, B., Ashjian, C.J., Cooper,  
1265 L.W., 2017. Mercury biomagnification in food webs of the northeastern Chukchi Sea, Alaskan Arctic.  
1266 *Deep Sea Research Part II: Topical Studies in Oceanography*, 144:63-77.
- 1267 French, T.D., Houben, A.J., Desforges, J.P.W., Kimpe, L.E., Kokelj, S.V., Poulain, A.J., Smol, J.P.,  
1268 Wang, X. and Blais, J.M., Dissolved organic carbon thresholds affect mercury bioaccumulation in Arctic  
1269 lakes. *Environmental Science & Technology*, 48 (6):3162-3168.
- 1270 Frey, K.E. and McClelland, J.W., 2009. Impacts of permafrost degradation on Arctic river  
1271 biogeochemistry. *Hydrological Processes*, 23 (1):169-182.
- 1272 Gamberg, M., Cuyler, C. and Wang, X., 2016. Contaminants in two West Greenland caribou  
1273 populations. *Science of the Total Environment*, 554-555:329-336.
- 1274 Gantner, N., Muir, D.C., Power, M., Iqaluk, D., Reist, J.D., Babaluk, J.A., Meili, M., Borg, H.,  
1275 Hammar, J., Michaud, W., Dempson, B. and Solomon, K.R., 2010. Mercury concentrations in  
1276 landlocked Arctic char (*Salvelinus alpinus*) from the Canadian Arctic. Part II: Influence of lake biotic  
1277 and abiotic characteristics on geographic trends in 27 populations. *Environmental Toxicology and*  
1278 *Chemistry*, 29 (3):633-643.
- 1279 Gascón Díez, E., Loizeau, J.-L., Cosio, C., Bouchet, S., Adatte, T., Amouroux, D. and Bravo, A.G.,  
1280 2016. Role of settling particles on mercury methylation in the oxic water column of freshwater  
1281 systems. *Environmental Science & Technology*, 50 (21):11672-11679.
- 1282 Gilmour, C.C., Podar, M., Bullock, A.L., Graham, A.M., Brown, S.D., Somenahally, A.C., Johs, A.,  
1283 Hurt, R.A., Bailey, K.L. and Elias, D.A., 2013. Mercury methylation by novel microorganisms from  
1284 new environments. *Environmental Science & Technology*, 47 (20):11810-11820.
- 1285 Gilmour, C.C., Bullock, A.L., McBurney, A., Podar, M., Elias, D.A., 2018, Robust Mercury  
1286 Methylation across Diverse Methanogenic Archaea, *mBio*, 9(2), 10.1128/mBio.02403-17
- 1287 Gionfriddo, C.M., Tate, M.T., Wick, R.R., Schultz, M.B., Zemla, A., Thelen, M.P., Schofield, R.,  
1288 Krabbenhoft, D.P., Holt, K.E. and Moreau, J.W., 2016. Microbial mercury methylation in Antarctic  
1289 sea ice. *Nature Microbiology*, 1 (10):16127.
- 1290 Gionfriddo C.M., Stott, M.B., Power, J.F., Ogorek, J.M., Krabbenhoft, D.P., Wick, R., Holt, K., Chen,  
1291 L.-X., Thomas, B.C., Banfield, J.F., Moreau, J.W. and Cann, I., 2020. Genome-resolved  
1292 metagenomics and detailed geochemical speciation analyses yield new insights into microbial mercury  
1293 cycling in geothermal springs. *Applied and Environmental Microbiology*, 86 (15):e00176-20.
- 1294 Girard, C., Leclerc, M. and Amyot M., 2016. Photodemethylation of methylmercury in Eastern  
1295 Canadian Arctic thaw pond and lake ecosystems. *Environmental Science & Technology*, 50 (7):3511-  
1296 3520.
- 1297 Gordon, J., Quinton, W., Branfireun, B.A. and Olefeldt, D., 2016. Mercury and methylmercury  
1298 biogeochemistry in a thawing permafrost wetland complex, Northwest Territories, Canada.  
1299 *Hydrological Processes*, 30 (20):3627-3638.



- 1300 Góngora, E., Braune, B.M. and Elliott, K.H., 2018. Nitrogen and sulfur isotopes predict variation in  
1301 mercury levels in Arctic seabird prey. *Marine Pollution Bulletin*, 135:907-914.
- 1302 Graham, A.M., Aiken, G.R. and Gilmour, C.C., 2012. Dissolved organic matter enhances microbial  
1303 mercury methylation under sulfidic conditions. *Environmental Science & Technology*, 46 (5):2715-  
1304 2723.
- 1305 Hallanger, I.G., Fuglei, E., Yoccoz, N.G., Pedersen, Å.Ø., König, M. and Routti, H., 2019. Temporal  
1306 trend of mercury in relation to feeding habits and food availability in Arctic foxes (*Vulpes lagopus*) from  
1307 Svalbard, Norway. *Science of the Total Environment*, 670:1125-1132.
- 1308 Halm, D.R. and Dornblaser, M.M., 2007. Water and sediment quality in the Yukon River and its  
1309 tributaries between Atlin, British Columbia, Canada, and Eagle, Alaska, USA, 2004 (Open-File Report  
1310 2007-1197). U.S. Geological Survey. Available at: <https://doi.org/10.3133/ofr20071197>.
- 1311 Hammerschmidt, C.R. and Fitzgerald, W.F., 2006. Photodecomposition of methylmercury in an Arctic  
1312 Alaskan Lake. *Environmental Science & Technology*, 40 (4):1212-1216.
- 1313 Hammerschmidt, C.R. and Bowman, K.L., 2012. Vertical methylmercury distribution in the subtropical  
1314 North Pacific Ocean. *Marine Chemistry*, 132-133:77-82.
- 1315 Hammerschmidt, C.R. and Fitzgerald, W.F., 2010. Iron-mediated photochemical decomposition of  
1316 methylmercury in an Arctic Alaskan Lake. *Environmental Science & Technology*, 44 (16):6138-6143.
- 1317 Hawkings, J. R. Linhoff B.S., Wadham J.L., Stibal M., Lamborg C.H., Carling G. T., Lamarche-  
1318 Gagnon G., Kohler T.J., Ward R., Hendry K.R., Falteisek L., Kellerman A. M., Cameron K. A.,  
1319 Hatton J. E., Tingey S., Holt A.D., Vinšová P., Hofer S., Bulínová M., Větrovský T., Meire L. &  
1320 Spencer R.G. M., 2021, Large subglacial source of mercury from the southwestern margin of the  
1321 Greenland Ice Sheet. *Nat. Geosci.* 14, 496–502
- 1322 Heimbürger, L.-E., Sonke, J.E, Cossa, D., Point, D., Lagane, C., Laffont, L, Galfond, B., Nicolaus, M.,  
1323 Rabe, B. and Rutgers van der Loeff, M., 2015. Shallow methylmercury production in the marginal sea  
1324 ice zone of the central Arctic Ocean. *Scientific Report*, 5 (1):10318.
- 1325 Heimbürger, L.-E., Cossa, D., Marty, J.-C., Migon, C., Averty, B., Dufour, A. and Ras, J., 2010.  
1326 Methyl mercury distributions in relation to the presence of nano- and picophytoplankton in an oceanic  
1327 water column (Ligurian Sea, North-western Mediterranean). *Geochimica et Cosmochimica Acta*, 74  
1328 (19):5549-5559.
- 1329 Hollweg, T.A., Gilmour, C.C. and Mason, R.P., 2010. Mercury and methylmercury cycling in  
1330 sediments of the mid-Atlantic continental shelf and slope. *Limnology and Oceanography*, 55 (6):2703-  
1331 2722.
- 1332 Hood, E., Fellman, J., Spencer, R.G.M., Hernes, P.J., Edwards, R., D'Amore, D. and Scott, D., 2009.  
1333 Glaciers as a source of ancient and labile organic matter to the marine environment. *Nature*, 462  
1334 (7276):1044-1047.
- 1335 Hood, E., Battin, T.J., Fellman, J., O'Neel, S. and Spencer, R.G.M., 2015. Storage and release of organic  
1336 carbon from glaciers and ice sheets. *Nature Geoscience*, 8 (2):91-96.
- 1337 Hopwood, M.J., Carroll, D., Dunse, T., Hodson, A., Holding, J.M., Iriarte, J.L., Ribeiro, S., Achterberg,  
1338 E.P., Cantoni, C., Carlson, D.F., Chierici, M., Clarke, J.S., Cozzi, S., Fransson, A., Juul-Pedersen, T.,  
1339 Winding, M.H.S. and Meire, L., 2020. How does glacier discharge affect marine biogeochemistry and  
1340 primary production in the Arctic? *The Cryosphere*, 14 (4):1347-1383.

- 1341 Houben, A.J., French, T.D., Kokelj, S. V., Wang, X., Smol, J.P. and Blais, J.M., 2016. The impacts of  
1342 permafrost thaw slump events on limnological variables in upland tundra lakes, Mackenzie Delta region.  
1343 *Fundamental and Applied Limnology*, 189:11-35.
- 1344 Hsu-Kim, H., Eckley, C.S., Achá, D., Feng, X., Gilmour, C.C., Jonsson, S. and Mitchell, C.P.J., 2018.  
1345 Challenges and opportunities for managing aquatic mercury pollution in altered landscapes. *Ambio*, 47  
1346 (2):141-169.
- 1347 Hudelson, K.E., Drevnick, P.E., Wang, F., Armstrong, D. and Fisk, A.T., 2020. Mercury methylation  
1348 and demethylation potentials in Arctic lake sediments. *Chemosphere*, 248:126001.
- 1349 Hudelson, K.E., Muir, D.C.G., Drevnick, P.E., Köck, G., Iqaluk, D., Wang, X., Kirk, J.L., Barst, B.D.,  
1350 Grgicak-Mannion, A., Shearon, R. and Fisk, A.T., 2019. Temporal trends, lake-to-lake variation, and  
1351 climate effects on Arctic char (*Salvelinus alpinus*) mercury concentrations from six High Arctic lakes  
1352 in Nunavut, Canada. *Science of the Total Environment*, 678:801-812.
- 1353 Imura, N., Sukegawa, E., Pan, S.-K., Nagao, K., Kim, J.-Y., Kwan, T. and Ukita, T., 1971. Chemical  
1354 methylation of inorganic mercury with methylcobalamin, a vitamin B<sub>12</sub> analog. *Science*, 172  
1355 (3989):1248-1249.
- 1356 Isles, P.D.F., Jonsson, A., Creed, I.F. and Bergström, A.-K., 2020. Does browning affect the identity of  
1357 limiting nutrients in lakes? *Aquatic Sciences*, 82 (2):45.
- 1358 Jiang, T., Wang, D., Meng, B., Chi, B., Laudon, H. and Liu, J., 2020. The concentrations and  
1359 characteristics of dissolved organic matter in high-latitude lakes determine its ambient reducing  
1360 capacity. *Water Research*, 169:115217.
- 1361 Jiskra, M., Sonke, J.E., Obrist, D., Bieser, J., Ebinghaus, R., Myhre, C.L., Pfaffhuber, K.A.,  
1362 Wängberg, I., Kyllönen, K., Worthy, D., Martin, L.G., Labuschagne, C., Mkololo, T., Ramonet, M.,  
1363 Magand, O., Dommergue, A., 2018. A vegetation control on seasonal variations in global atmospheric  
1364 mercury concentrations. *Nat. Geosci.* 11, 244–250
- 1365 Jiskra, M., Heimbürger-Boavida, L.-E., Desgranges, M.-M., Petrova, M.V., Dufour, A., Ferreira-  
1366 Araujo, B., Masbou, J., Chmeleff, J., Thyssen, M., Point, D., Sonke, J.E., 2021. Mercury stable  
1367 isotopes constrain atmospheric sources to the ocean. *Nature* 597, 678–682.
- 1368 Jones, D.S., Walker, G.M., Johnson, N.W., Mitchell, C.P.J., Coleman Wasik, J.K. and Bailey, J.V.,  
1369 2019. Molecular evidence for novel mercury methylating microorganisms in sulfate-impacted lakes.  
1370 *The ISME Journal*, 13 (7):1659-1675.
- 1371 Jonsson, S., Skjellberg, U., Nilsson, M.B., Westlund, P.-O., Shchukarev, A., Lundberg, E. and Björn,  
1372 E., 2012. Mercury methylation rates for geochemically relevant HgII species in sediments.  
1373 *Environmental Science & Technology*, 46 (21):11653-11659.
- 1374 Jonsson, S., Skjellberg, U., Nilsson, M.B., Lundberg, E., Andersson, A. and Björn, E., 2014.  
1375 Differentiated availability of geochemical mercury pools controls methylmercury levels in estuarine  
1376 sediment and biota. *Nature Communications*, 5 (1):4624.
- 1377 Jonsson, S., Mazrui, N.M. and Mason, R.P., 2016. Dimethylmercury formation mediated by inorganic  
1378 and organic reduced sulfur surfaces. *Scientific Reports*, 6 (1):27958.
- 1379 Jonsson, S., Nerentorp Mastromonaco, M.G, Gårdfeldt, K., Mason, R.P., Distribution of Total  
1380 Mercury and Methylated Mercury Species in Central Arctic Ocean Water and Ice, 2022, *Marine*  
1381 *Chemistry*, 242, 104105.

- 1382 Jorgenson, M.T., Racine, C.H., Walters, J.C. and Osterkamp, T.E., 2001. Permafrost degradation and  
1383 ecological changes associated with a warming climate in central Alaska. *Climatic Change*, 48 (4):551-  
1384 579.
- 1385 Jorgenson, M.T., Shur, Y.L. and Pullman, E.R., 2006. Abrupt increase in permafrost degradation in  
1386 Arctic Alaska. *Geophysical Research Letters*, 33 (2): L02503.
- 1387 Jörundsdóttir, H.Ó., Jensen, S., Hylland, K., Holth, T.F., Gunnlaugsdóttir, H., Svavarsson, J.,  
1388 Ólafsdóttir, Á., El-Taliawy, H., Rigét, F., Strand, J., Nyberg, E., Bignert, A., Hoydal, K.S. and  
1389 Halldórsson, H.P., 2014. Pristine Arctic: background mapping of PAHs, PAH metabolites and  
1390 inorganic trace elements in the North-Atlantic Arctic and Sub-Arctic coastal environment. *Science of  
1391 the Total Environment*, 493:719-728.
- 1392 Kahilainen, K.K., Thomas, S.M., Keva, O., Hayden, B., Knudsen, R., Eloranta, A.P., Tuohiluoto, K.,  
1393 Amundsen, P.,-A., Malinen, T. and Järvinen, A., 2016. Seasonal dietary shift to zooplankton  
1394 influences stable isotope ratios and total mercury concentrations in Arctic charr (*Salvelinus alpinus*  
1395 (L.)). *Hydrobiologia*, 783 (1):47-63.
- 1396 Kahilainen, K.K., Thomas, S.M., Nystedt, E.K.M., Keva, O., Malinen, T. and Hayden, B., 2017.  
1397 Ecomorphological divergence drives differential mercury bioaccumulation of polymorphic European  
1398 whitefish (*Coregonus lavaretus*) populations of Subarctic lakes. *Science of the Total Environment*, 599-  
1399 600:1768-1778.
- 1400 Keva, O., Hayden, B., Harrod, C. and Kahilainen, K.K., 2017. Total mercury concentrations in liver and  
1401 muscle of European whitefish (*Coregonus lavaretus* (L.)) in a Subarctic lake - assessing the factors  
1402 driving year-round variation. *Environmental Pollution*, 231 (Part 2):1518-1528.
- 1403 Khan, M.A. and Wang, F., 2010. Chemical demethylation of methylmercury by selenoamino acids.  
1404 *Chemical Research in Toxicology*, 23 (7):1202-1206.
- 1405 Kim, H., Soerensen, A.L., Hur, J., Heimbürger, L.-E., Hahm, D., Rhee, T.S., Noh, S. and Han, S.,  
1406 2017. Methylmercury mass budgets and distribution characteristics in the western Pacific Ocean.  
1407 *Environmental Science & Technology*, 51 (3):1186-1194.
- 1408 Kim, H., Kwon, S.Y., Lee, K., Lim, D., Han, S., Kim, T.-W., Joo, Y.J., Lim, J., Kang, M.-H. and Nam,  
1409 S.-I., 2020a. Input of terrestrial organic matter linked to deglaciation increased mercury transport to the  
1410 Svalbard fjords. *Scientific Reports*, 10 (1):3446.
- 1411 Kim, J., Soerensen, A.L., Kim, M.S., Eom, S., Rhee, T.S., Jin, Y K. and Han, S., 2020b. Mass budget  
1412 of methylmercury in the east Siberian Sea: the importance of sediment sources. *Environmental Science  
1413 and Technology*, 54 (16):9949–9957.
- 1414 Kirk, J.L., St. Louis, V.L., Hintelmann, H., Lehnerr, I., Else, B. and Poissant, L., 2008. Methylated  
1415 mercury species in marine waters of the Canadian High and Sub Arctic. *Environmental Science &  
1416 Technology*, 42 (22):8367-8373.
- 1417 Klapstein, S.J. and O’Driscoll, N.J., 2018. Methylmercury biogeochemistry in freshwater ecosystems:  
1418 a review focusing on DOM and photodemethylation. *Bulletin of Environmental Contamination and  
1419 Toxicology*, 100 (1):14-25.
- 1420 Klapstein, S.J., Ziegler, S.E. and O’Driscoll, N.J., 2017. Methylmercury photodemethylation is  
1421 inhibited in lakes with high dissolved organic matter. *Environmental Pollution*, 232:392-401.
- 1422 Kohlbach, D., Graeve, M., Lange, B.A., David, C., Peeken, I. and Flores, H., 2016. The importance of  
1423 ice algae-produced carbon in the central Arctic Ocean ecosystem: food web relationships revealed by  
1424 lipid and stable isotope analyses. *Limnology and Oceanography*, 61 (6):2027-2044.

- 1425 Kohler, Stephen G., Heimbürger-Boavida, L.E., Petrova, M.V., Digernes, M.G., Sanchez, N., Dufour,  
1426 A., Simić, A., Ndungu, K., Ardelan, M.V., 2022. Arctic Ocean's wintertime mercury concentrations  
1427 limited by seasonal loss on the shelf. in press
- 1428 Kristiansen SM, Leinaas HP, Herzke D, Hylland K, Gabrielsen GW, Harju M, et al. Seabird-  
1429 Transported Contaminants Are Reflected in the Arctic Tundra, but Not in Its Soil-Dwelling Springtails  
1430 (Collembola). *Environmental Science and Technology* 2019; 53: 12835-12845.
- 1431 Kåresdotter, E., Destouni, G., Ghajarnia, N., Hugelius, G., & Kalantari, Z., 2021. Mapping the  
1432 vulnerability of Arctic wetlands to global warming. *Earth's Future*, 9, e2020EF001858.
- 1433 Lantz, T.C. and Kokelj, S.V., 2008. Increasing rates of retrogressive thaw slump activity in the  
1434 Mackenzie Delta region, N.W.T., Canada. *Geophysical Research Letters*, 35 (6):L06502.
- 1435 Lavoie, R.A., Jardine, T.D., Chumchal, M.M., Kidd, K.A. and Campbell, L.M., 2013.  
1436 Biomagnification of mercury in aquatic food webs: a worldwide meta-analysis. *Environmental*  
1437 *Science & Technology*, 47 (23):13385-13394.
- 1438 Lavoie, R.A., Amyot, M., Lapierre, J.-F., 2019. Global meta-analysis on the relationship between  
1439 mercury and dissolved organic carbon in freshwater environments. *Journal of Geophysical Research:*  
1440 *Biogeosciences*, 124 (6):1508-1523.
- 1441 Lee, C.-S. and Fisher, N.S., 2016. Methylmercury uptake by diverse marine phytoplankton.  
1442 *Limnology and Oceanography*, 61 (5):1626-1639.
- 1443 Lehnherr, I., St. Louis, V.L., Hintelmann, H. and Kirk, J.L., 2011. Methylation of inorganic mercury in  
1444 polar marine waters. *Nature Geoscience*, 4 (5):298-302.
- 1445 Lehnherr, I., St. Louis, V.L. and Kirk, J.L., 2012a. Methylmercury cycling in High Arctic wetland  
1446 ponds: sources and sinks. *Environmental Science & Technology*, 46 (19):10514-10522.
- 1447 Lehnherr, I., St. Louis, V.L. and Kirk, J.L., 2012b Methylmercury cycling in High Arctic wetland ponds:  
1448 controls on sedimentary production. *Environmental Science & Technology*, 46 (19):10523-10531.
- 1449 Lehnherr, I., 2014. Methylmercury biogeochemistry: a review with special reference to Arctic aquatic  
1450 ecosystems. *Environmental Reviews*, 22 (3):229-243.
- 1451 Lescord, G.L., Emilson, E.J.S., Johnston, T.A., Branfireun, B.A. and Gunn, J.M., 2018. Optical  
1452 properties of dissolved organic matter and their relation to mercury concentration in water and biota  
1453 across a remote freshwater drainage basin. *Environmental Science & Technology*, 52 (6): 3344-3353.
- 1454 Lescord, G.L., Kidd, K.A., Kirk, J.L., O'Driscoll, N.J., Wang, X. and Muir, D.C.G., 2015. Factors  
1455 affecting biotic mercury concentrations and biomagnification through lake food webs in the Canadian  
1456 High Arctic. *Science of the Total Environment*, 509-510:195-205.
- 1457 Li, C., Sonke, J.E., Le Roux, G., Piotrowska, N., Van der Putten, N., Roberts, S.J., Daley, T.,  
1458 Rice, E., Gehrels, R., Enrico, M., Mauquoy, D., Roland, T.P., De Vleeschouwer, F., 2020.  
1459 Unequal Anthropogenic Enrichment of Mercury in Earth's Northern and Southern  
1460 Hemispheres. *ACS Earth Space Chem.* 4, 2073–2081
- 1461 Liem-Nguyen, V., Wild, B., Gustafsson, Ö., Semiletov, I., Dudarev, O. and Jonsson, S., 2022. Spatial  
1462 patterns and distributional controls of total and methylated mercury off the Lena River in the Laptev Sea  
1463 sediments. *Marine Chemistry*, 238, 104052

- 1464 Lim, A.G., Sonke, J.E.; Krickov, I.V.; Manasypov, R.M., Loiko, S.V. and Pokrovsky, O.S., 2019.  
1465 Enhanced particulate Hg export at the permafrost boundary, western Siberia. *Environmental Pollution*,  
1466 254, (Part B):113083.
- 1467 Lim, A. G., Jiskra, M., Sonke, J. E., Loiko, S. V., Kosykh, N., and Pokrovsky, O. S., 2020, A revised  
1468 pan-Arctic permafrost soil Hg pool based on Western Siberian peat Hg and carbon observations,  
1469 *Biogeosciences*, 17, 3083–3097
- 1470 Lin H., Ascher D.B., Myung Y., Lamborg C.H., Hallam S.J., Gionfriddo C.M., Holt K.E., Moreau  
1471 J.W., 2021. Mercury methylation by metabolically versatile and cosmopolitan marine bacteria. *The*  
1472 *ISME Journal* 15:1810–1825
- 1473 Liu, Y., Chai, X., Hao, Y., Gao, X., Lu, Z., Zhao, Y., Zhang, J. and Cai, M., 2015. Total mercury and  
1474 methylmercury distributions in surface sediments from Kongsfjorden, Svalbard, Norwegian Arctic.  
1475 *Environmental Science and Pollution Research* , 22 (11):8603-8610.
- 1476 Lockhart WL, Stern GA, Low G, Hendzel M, Boila G, Roach P, et al. A history of total mercury in  
1477 edible muscle of fish from lakes in northern Canada. *Science of The Total Environment* 2005; 351-352:  
1478 427-463.
- 1479 Loose, B., SchlosseR, P., Perovich, D., Ringelberg, D., HO, D., Takahashi, T., Richter-Menge, J.,  
1480 Reynolds, C., McGillis, W. and Tison, J.-L. (2011), Gas diffusion through columnar laboratory sea  
1481 ice: implications for mixed-layer ventilation of CO<sub>2</sub> in the seasonal ice zone. *Tellus B*, 63: 23-39.
- 1482 Loseto LL, Stern GA, Deibel D, Connelly TL, Prokopowicz A, Lean DRS, et al. Linking mercury  
1483 exposure to habitat and feeding behaviour in Beaufort Sea beluga whales. *Journal of Marine Systems*  
1484 2008; 74: 1012-1024.
- 1485 Loseto LL, Stern GA, Macdonald RW. Distant drivers or local signals: Where do mercury trends in  
1486 western Arctic belugas originate? *Science of The Total Environment* 2015; 509-510: 226-236.
- 1487 Ma, M., Du, H. and Wang, D., 2019. Mercury methylation by anaerobic microorganisms: a review.  
1488 *Critical Reviews in Environmental Science and Technology*, 49 (20):1893-1936.
- 1489 Macdonald, R.W., Harner, T. and Fyfe, J., 2005. Recent climate change in the Arctic and its impact on  
1490 contaminant pathways and interpretation of temporal trend data. *Science of the Total Environment*, 342  
1491 (1-3):5-86.
- 1492 Mackelprang, R., Waldrop, M.P., DeAngelis, K.M., David, M.M., Chavarria, K.L., Blazewicz, S.J.,  
1493 Rubin, E.M. and Jansson, J.K., 2011. Metagenomic analysis of a permafrost microbial community  
1494 reveals a rapid response to thaw. *Nature*, 480 (7377):368-371.
- 1495 MacMillan, G.A., 2018. Hunting for trace metals in the rapidly changing North: using limnological,  
1496 ecological and collaborative approaches. Doctoral thesis, Department of Biological Sciences,  
1497 University of Montreal, Quebec, Canada. Available at:  
1498 <https://papyrus.bib.umontreal.ca/xmlui/handle/1866/21777>.
- 1499 MacMillan, G.A., Girard, C., Chételat, J., Laurion, I., and Amyot, M., 2015. High methylmercury in  
1500 Arctic and Subarctic ponds is related to nutrient levels in the warming eastern Canadian Arctic.  
1501 *Environmental Science & Technology*, 49 (13):7743-7753.
- 1502 Mangal, V., Stenzler, B.R., Poulain, A.J. and Guéguen, C., 2019. Aerobic and anaerobic bacterial  
1503 mercury uptake is driven by algal organic matter composition and molecular weight. *Environmental*  
1504 *Science & Technology*, 53 (1):157-165.

- 1505 Masbou, J., Sonke, J.E., Amouroux, D., Guillou, G., Becker, P.R. and Point, D., 2018. Hg-stable isotope  
1506 variations in marine top predators of the western Arctic Ocean. *ACS Earth and Space Chemistry* 2  
1507 (5):479-490.
- 1508 Mazrui, N.M., Jonsson, S., Thota, S., Zhao, J. and Mason, R.P., 2016. Enhanced availability of  
1509 mercury bound to dissolved organic matter for methylation in marine sediments. *Geochimica et*  
1510 *Cosmochimica Acta*, 194:153-162.
- 1511 Martyniuk, M.A.C., Couture, P., Tran, L., Beaupré, L. and Power, M., 2020. Seasonal variation of total  
1512 mercury and condition indices of Arctic charr (*Salvelinus alpinus*) in Northern Quebec, Canada. *Science*  
1513 *of the Total Environment*, 738:139450.
- 1514 Mason, R.P., Choi, A.L., Fitzgerald, W.F., Hammerschmidt, C.R., Lamborg, C.H., Soerensen, A.L. and  
1515 Sunderland, E.M., 2012. Mercury biogeochemical cycling in the ocean and policy implications.  
1516 *Environmental Research*, 119:101-117.
- 1517 Mason, R.P. and Sullivan, K.A., 1999. The distribution and speciation of mercury in the South and  
1518 equatorial Atlantic. *Deep Sea Research Part II: Topical Studies in Oceanography* 46 (5):937-956.
- 1519 McDaniel, E.A., Peterson, B.D., Stevens, S.L.R., Tran, P.Q., Anantharaman, K., McMahon, K.D. and  
1520 Kent, A.D., 2020. Expanded phylogenetic diversity and metabolic flexibility of mercury-methylating  
1521 microorganisms. *mSystems*, 5 (4):e00299-20.
- 1522 McGrew, A.K., Ballweber, L.R., Moses, S.K., Stricker, C.A., Beckmen, K.B., Salman, M.D. and  
1523 O'Hara, T.M., 2014. Mercury in gray wolves (*Canis lupus*) in Alaska: increased exposure through  
1524 consumption of marine prey. *Science of the Total Environment*, 468-469:609-613.
- 1525 McKinney, M.A., Atwood, T.C., Pedro, S. and Peacock, E., 2017. Ecological change drives a decline in  
1526 mercury concentrations in southern Beaufort Sea polar bears. *Environmental Science & Technology*, 51  
1527 (14):7814-7822.
- 1528 McMeans, B.C., Arts, M.T. and Fisk, A.T., 2015. Impacts of food web structure and feeding behavior  
1529 on mercury exposure in Greenland Sharks (*Somniosus microcephalus*). *Science of the Total*  
1530 *Environment*, 509-510:216–225.
- 1531 Michelutti N, Blais JM, Mallory ML, Brash J, Thienpont J, Kimpe LE, et al. Trophic position  
1532 influences the efficacy of seabirds as metal biovectors. *Proceedings of the National Academy of*  
1533 *Sciences of the United States of America* 2010; 107: 10543-10548.
- 1534 Moslemi-Aqdam M, Baker LF, Baltzer JL, Branfireun BA, Evans MS, Laird BD, et al. Understanding  
1535 among-lake variability of mercury concentrations in Northern Pike (*Esox lucius*): A whole-ecosystem  
1536 study in subarctic lakes. *Science of The Total Environment* 2022; 822: 153430.
- 1537 Munson, K.M., Lamborg, C.H., Swarr, G.J. and Saito, M.A., 2015. Mercury species concentrations  
1538 and fluxes in the central tropical Pacific Ocean. *Global Biogeochemical Cycles*, 29 (5):656-676.
- 1539 Munthe, J., Bodaly, R.A., Branfireun, B.A., Driscoll, C.T., Gilmour, C.C., Harris, R., Horvat, M.,  
1540 Lucotte, M. and Malm, O., 2007. Recovery of mercury-contaminated fisheries. *Ambio*, 36 (1):33-44.
- 1541 Nagorski, S.A., Engstrom, D.R., Hudson, J.P., Krabbenhoft, D.P., Hood, E., DeWild, J.F. and Aiken,  
1542 G.R., 2014. Spatial distribution of mercury in southeastern Alaskan streams influenced by glaciers,  
1543 wetlands, and salmon. *Environmental Pollution*, 184:62-72.
- 1544 Obrist, D., Agnan, Y., Jiskra, M., Olson, C.L., Colegrove, D.P., Hueber, J., Moore, C.W., Sonke, J.E.  
1545 and Helmig, D., 2017. Tundra uptake of atmospheric elemental mercury drives Arctic mercury pollution.  
1546 *Nature*, 547 (7662):201-204.

- 1547 Olefeldt, D., Goswami, S., Grosse, G., Hayes, D., Hugelius, G., Kuhry, P., McGuire, A.D.,  
1548 Romanovsky, V.E., Sannel, A.B.K., Schuur, E.A.G. and Turetsky, M.R., 2016. Circumpolar distribution  
1549 and carbon storage of thermokarst landscapes. *Nature Communications*, 7 (1):13043.
- 1550 Olefeldt D, Heffernan L, Jones MC, Sannel AB, Treat CC, Turetsky MR. 2021. Permafrost thaw in  
1551 northern peatlands: rapid changes in ecosystem and landscape functions. *Ecosystem Collapse and*  
1552 *Climate Change*:27-67.
- 1553 Olson, C., Jiskra, M., Biester, H., Chow, J. and Obrist, D., 2018. Mercury in active-layer tundra soils of  
1554 Alaska: concentrations, pools, origins, and spatial distribution. *Global Biogeochemical Cycles*, 32  
1555 (7):1058-1073.
- 1556 Parks, J.M., Johs, A., Podar, M., Bridou, R., Hurt, R.A., Jr., Smith, S.D., Tomanicek, S.J., Qian, Y.,  
1557 Brown, S.D., Brandt, C.C., Palumbo, A.V., Smith, J.C., Wall, J.D., Elias, D.A. and Liang, L., 2013. The  
1558 genetic basis for bacterial mercury methylation. *Science*, 339 (6125):1332-1335.
- 1559 Pedro, S., Fisk, A.T., Ferguson, S.H., Hussey, N.E., Kessel, S.T. and McKinney, M.A., 2019. Limited  
1560 effects of changing prey fish communities on food quality for aquatic predators in the eastern Canadian  
1561 Arctic in terms of essential fatty acids, methylmercury and selenium. *Chemosphere*, 214:855-865.
- 1562 Peterson, S.H., Ackerman, J.T., Crocker, D.E. and Costa, D.P., 2018. Foraging and fasting can  
1563 influence contaminant concentrations in animals: an example with mercury contamination in a free-  
1564 ranging marine mammal. *Proceedings of the Royal Society B: Biological Sciences*, 285  
1565 (1872):20172782.
- 1566 Petrova, M.V., Krisch, S., Lodeiro, P., Valk, O., Dufour, A., Rijkenberg, M.J.A., Achterberg, E.P., Rabe,  
1567 B., Rutgers van der Loeff, M., Hamelin, B., Sonke, J.E., Garnier, C., Heimbürger-Boavida, L.-E., 2020.  
1568 Mercury species export from the Arctic to the Atlantic Ocean. *Marine Chemistry*, 225:103855.
- 1569 Piquet, A.M.-T., van de Poll, W.H., Visser, R.J.W., Wiencke, C., Bolhuis, H., Buma, A.G.J., 2014.  
1570 Springtime phytoplankton dynamics in Arctic Krossfjorden and Kongsfjorden (Spitsbergen) as a  
1571 function of glacier proximity. *Biogeosciences*, 11 (8):2263–2279.
- 1572 Point, D., Sonke, J. E., Day, R. D., Roseneau, D. G., Hobson, K. A., Vander Pol, S. S., Moors, A. J.,  
1573 Pugh, R. S., Donard, O. F. X., Becker P. R., 2011, Methylmercury photodegradation influenced by  
1574 sea-ice cover in Arctic marine ecosystems, *Nature Geoscience*, 4, pages188–194
- 1575 Podar, M., Gilmour, C.C., Brandt, C.C., Soren, A., Brown, S.D., Crable, B.R., Palumbo, A.V.,  
1576 Somenahally, A.C. and Elias, D.A., 2015. Global prevalence and distribution of genes and  
1577 microorganisms involved in mercury methylation. *Science Advances*, 1 (9):e1500675.
- 1578 Pomerleau, C., Matthews, C.J.D., Gobeil, C., Stern, G.A., Ferguson, S.H. and Macdonald, R.W., 2018.  
1579 Mercury and stable isotope cycles in baleen plates are consistent with year-round feeding in two  
1580 bowhead whale (*Balaena mysticetus*) populations. *Polar Biology*, 41 (9):1881-1893.
- 1581 Pomerleau, C., Stern, G.A., Pućko, M., Foster, K.L., Macdonald, R.W. and Fortier, L., 2016. Pan-  
1582 Arctic concentrations of mercury and stable isotope ratios of carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) in  
1583 marine zooplankton. *Science of the Total Environment*, 551-552:92-100.
- 1584 Pongratz, R. and Heumann, K.G., 1998. Production of methylated mercury and lead by polar  
1585 macroalgae — a significant natural source for atmospheric heavy metals in clean room compartments.  
1586 *Chemosphere*, 36 (9):1935-1946.
- 1587 Pokrovsky, O.S., Bueno, M., Manasypov, R.M., Shirokova, L.S., Karlsson, J. and Amouroux, D.,  
1588 2018. Dissolved organic matter controls seasonal and spatial selenium concentration variability in  
1589 thaw lakes across a permafrost gradient. *Environmental Science & Technology*, 52 (18):10254-10262.

- 1590 Poste, A.E., Braaten, H.F.V., de Wit, H.A., Sørensen, K. and Larssen, T., 2015. Effects of  
1591 photodemethylation on the methylmercury budget of boreal Norwegian lakes. *Environmental*  
1592 *Toxicology and Chemistry*, 34 (6):1213-1223.
- 1593 Poste, A.E., Hoel, C.S., Andersen, T., Arts, M.T., Frærøvig, P-J. and Borgå, K., 2019. Terrestrial organic  
1594 matter increases zooplankton methylmercury accumulation in a brown-water boreal lake. *Science of the*  
1595 *Total Environment*, 674:9-18.
- 1596 Provencher, J.F., Forbes, M.R., Hennin, H.L., Love, O.P., Braune, B.M., Mallory, M.L., and Gilchrist,  
1597 2016. Implications of mercury and lead concentrations on breeding physiology and phenology in an  
1598 Arctic bird. *Environmental Pollution*, 218:1014-1022.
- 1599 Pućko, M., Burt, A., Walkusz, W., Wang, F., Macdonald, R.W., Rysgaard, S., Barber, D.G., Tremblay,  
1600 J.-É. and Stern, G.A., 2014. Transformation of mercury at the bottom of the Arctic food web: an  
1601 overlooked puzzle in the mercury exposure narrative. *Environmental Science & Technology*, 48  
1602 (13):7280-7288.
- 1603 Rautio, M., Dufresne, F., Laurion, I., Bonilla, S., Vincent, W.F. and Christoffersen, K.S., 2011. Shallow  
1604 freshwater ecosystems of the circumpolar Arctic. *Écoscience*, 18 (3):204-222.
- 1605 Richardson M, Chételat J, MacMillan GA, Amyot M. Mercury concentrations and associations with  
1606 dissolved organic matter are modified by water residence time in eastern Canadian lakes along a 30°  
1607 latitudinal gradient. *Limnology and Oceanography* 2021; 66: S64-S80.
- 1608 Rohonczy, J., Cott, P.A., Benwell, A., Forbes, M.R., Robinson, S.A., Rosabal, M., Amyot, M. and  
1609 Chételat, 2020. Trophic structure and mercury transfer in the Subarctic fish community of Great Slave  
1610 Lake, Northwest Territories, Canada. *Journal of Great Lakes Research* 46 (2):402-413.
- 1611 Rosati, G., Heimbürger, L.E., Melaku Canu, D., Lagane, C., Laffont, L., Rijkenberg, M.J.A., Gerringa,  
1612 L.J.A., Solidoro, C., Gencarelli, C.N., Hedgecock, I.M., De Baar, H.J.W. and Sonke, J.E., 2018.  
1613 Mercury in the Black Sea: new insights from measurements and numerical modeling. *Global*  
1614 *Biogeochemical Cycles*, 32 (4):529-550.
- 1615 Ruus, A., Øverjordet, I.B., Braaten, H.F.V., Evenset, A., Christensen, G., Heimstad, E.S., Gabrielsen,  
1616 G.W. and Borgå, K., 2015. Methylmercury biomagnification in an Arctic pelagic food web.  
1617 *Environmental Toxicology & Chemistry*, 34 (11):2636-2643.
- 1618 Ruuskanen M.O., Colby G., St. Pierre K.A., St.Louis V.AL., Aris-Brosou S., Poulain A.J. 2020.,  
1619 Microbial genomes retrieved from High Arctic lake sediments encode for adaptation to cold and  
1620 oligotrophic environments, *Limnol. Oceanogr.*65, S233–S247
- 1621 Rysgaard, S. and Glud, R.N., 2004. Anaerobic N<sub>2</sub> production in Arctic sea ice. *Limnology and*  
1622 *Oceanography*, 49 (1):86-94.
- 1623 Sandheinrich, M.B. and Drevnick, P.E., 2016. Relationship among mercury concentration, growth rate,  
1624 and condition of northern pike: A tautology resolved? *Environmental Toxicology and Chemistry*, 35  
1625 (12):2910-2915.
- 1626 Saros, J.E., Osburn, C.L., Northington, R.M., Birkel, S.D., Auger, J.D., Stedmon, C.A. and Anderson,  
1627 N.J., 2015. Recent decrease in DOC concentrations in Arctic lakes of southwest Greenland. *Geophysical*  
1628 *Research Letters*, 42 (16):6703-6709.
- 1629 Schaefer, J.K. and Morel, F.M.M., 2009. High methylation rates of mercury bound to cysteine by  
1630 *Geobacter sulfurreducens*. *Nature Geoscience*, 2 (2):123-126.



- 1631 Schaefer, K., Elshorbany, Y., Jafarov, E., Schuster, P.F., Striegl, R.G., Wickland, K.P. and Sunderland,  
1632 E.M., 2020. Potential impacts of mercury released from thawing permafrost. *Nature Communications*,  
1633 11 (1):4650.
- 1634 Schartup, A.T., Balcom, P.H., Soerensen, A.L., Gosnell, K.J., Calder, R.S.D., Mason, R.P. and  
1635 Sunderland, E.M., 2015a. Freshwater discharges drive high levels of methylmercury in Arctic marine  
1636 biota. *Proceedings of the National Academy of Sciences*, 112 (38):11789-11794.
- 1637 Schartup, A.T., Ndu, U., Balcom, P.H., Mason, R.P. and Sunderland, E.M., 2015b. Contrasting effects  
1638 of marine and terrestrially derived dissolved organic matter on mercury speciation and bioavailability  
1639 in seawater. *Environmental Science & Technology*, 49 (10):5965-5972.
- 1640 Schartup, A.T., Qureshi, A., Dassuncao, C., Thackray, C.P., Harding, G. and Sunderland, E.M., 2018.  
1641 A model for methylmercury uptake and trophic transfer by marine plankton. *Environmental Science &  
1642 Technology*, 52 (2):654-662.
- 1643 Schartup, A.T., Soerensen, A.L. and Heimbürger-Boavida, L.-E., 2020. Influence of the Arctic sea-ice  
1644 regime shift on sea-ice methylated mercury trends. *Environmental Science & Technology Letters*, 7  
1645 (10):708-713.
- 1646 Seewagen, C.L., Cristol, D.A. and Gerson, A.R., 2016. Mobilization of mercury from lean tissues  
1647 during simulated migratory fasting in a model songbird. *Scientific Reports*, 6 (1):25762.
- 1648 Seller, P., Kelly, C.A., Rudd, J.W.M. and MacHutchon, A.R., 1996. Photodegradation of  
1649 methylmercury in lakes. *Nature*, 380 (6576):694-697.
- 1650 Skrobonja, A., Gojkovic, Z., Soerensen, A.L., Westlund, P.-O., Funk, C. and Björn, E., 2019. Uptake  
1651 kinetics of methylmercury in a freshwater alga exposed to methylmercury complexes with  
1652 environmentally relevant thiols. *Environmental Science & Technology*, 53 (23):13757-13766.
- 1653 Skyllberg, U., Westin, M.B., Meili, M. and Björn, E., 2009. Elevated concentrations of methyl  
1654 mercury in streams after forest clear-cut: a consequence of mobilization from soil or new methylation?  
1655 *Environmental Science & Technology*, 43 (22):8535-8541.
- 1656 Skyllberg U., 2012, Chemical Speciation of Mercury in Soil and Sediment, in: Liu, G., Cai, Y.,  
1657 O'Driscoll, N.J. (Eds.), *Environmental Chemistry and Toxicology of Mercury*, pp. 219-258.
- 1658 Smith, S.D., Bridou, R., Johs, A., Parks, J.M., Elias, D.A., Hurt, R.A., Brown, S.D., Podar, M., Wall,  
1659 J.D. and Kivisaar, M., 2015. Site-directed mutagenesis of *hgcA* and *hgcB* reveals amino acid residues  
1660 important for mercury methylation. *Applied and Environmental Microbiology*, 81 (9):3205-3217.
- 1661 Sommar, J., Feng, X. and Lindqvist, O., 1999. Speciation of volatile mercury species present in digester  
1662 and deposit gases. *Applied Organometallic Chemistry*, 13 (6):441-445.
- 1663 Soerensen, A.L., Schartup, A.T., Gustafsson, E., Gustafsson, B.G., Undeman, E. and Björn, E., 2016a.  
1664 Eutrophication increases phytoplankton methylmercury concentrations in a coastal sea—a Baltic Sea  
1665 case study. *Environmental Science & Technology*, 50 (21):11787-11796.
- 1666 Soerensen, A.L., Jacob, D.J., Schartup, A.T., Fisher, J.A., Lehnher, I., St. Louis, V.L., Heimbürger, L.-  
1667 E., Sonke, J.E., Krabbenhoft, D.P. and Sunderland, E.M., 2016b. A mass budget for mercury and  
1668 methylmercury in the Arctic Ocean. *Global Biogeochemical Cycles*, 30 (4):560-575.
- 1669 Soerensen, A.L., Schartup, A.T., Skrobonja, A. and Björn, E., 2017. Organic matter drives high  
1670 interannual variability in methylmercury concentrations in a Subarctic coastal sea. *Environmental  
1671 Pollution*, 229:531-538.

- 1672 Sonke, J.E., Teisserenc, R., Heimbürger-Boavida, L.-E., Petrova, M.V., Maruszczak, N., Le Dantec, T.,  
1673 Chupakov, A.V., Li, C., Thackray, C.P., Sunderland, E.M., Tananaev, N. and Pokrovsky, O.S., 2018.  
1674 Eurasian river spring flood observations support net Arctic Ocean mercury export to the atmosphere and  
1675 Atlantic Ocean. *Proceedings of the National Academy of Sciences*, 115 (50):E11586-E11594.
- 1676 Steffen, A., Bottenheim, J., Cole, A., Ebinghaus, R., Lawson, G. and Leitch, W.R., 2014. Atmospheric  
1677 mercury speciation and mercury in snow over time at Alert, Canada. *Atmospheric Chemistry and*  
1678 *Physics*, 14 (5):2219-2231.
- 1679 Streets, D.G., Horowitz, H.M., Lu, Z., Levin, L., Thackray, C.P., Sunderland, E.M., 2019. Five hundred  
1680 years of anthropogenic mercury: spatial and temporal release profiles, *Environ. Res. Lett.* 14 084004
- 1681 St. Louis, V.L., Derocher, A.E., Stirling, I., Graydon, J.A., Lee, C., Jocksch, E., Richardson, E.,  
1682 Ghorpade, S., Kwan, A.K., Kirk, J.L., Lehnerr, I. and Swanson, H.K., 2011. Differences in mercury  
1683 bioaccumulation between polar bears (*Ursus maritimus*) from the Canadian High- and Sub-Arctic.  
1684 *Environmental Science & Technology*, 45 (14):5922-5928.
- 1685 St. Louis, V.L., Sharp, M.J., Steffen, A., May, A., Barker, J., Kirk, J.L., Kelly, D.J.A., Arnott, S.E.,  
1686 Keatley, B. and Smol, J.P., 2005. Some sources and sinks of monomethyl and inorganic mercury on  
1687 Ellesmere Island in the Canadian High Arctic. *Environmental Science & Technology*, 39 (8):2686-2701.
- 1688 St. Louis, V.L., Hintelmann, H., Graydon, J.A., Kirk, J.L., Barker, J., Dimock, B., Sharp, M.J. and  
1689 Lehnerr, I., 2007. Methylated mercury species in Canadian High Arctic marine surface waters and  
1690 snowpacks. *Environmental Science & Technology*, 41 (18):6433-6441.
- 1691 St. Pierre, K.A., St. Louis, V.L., Kirk, J.L., Lehnerr, I., Wang, S. and La Farge, C., 2015. Importance  
1692 of open marine waters to the enrichment of total mercury and monomethylmercury in lichens in the  
1693 Canadian High Arctic. *Environmental Science & Technology*, 49 (10):5930-5938.
- 1694 St. Pierre, K.A., Zolkos, S., Shakil, S., Tank, S.E., St. Louis, V.L. and Kokelj, S.V., 2018.  
1695 Unprecedented increases in total and methyl mercury concentrations downstream of retrogressive thaw  
1696 slumps in the western Canadian Arctic. *Environmental Science & Technology*, 52 (24):14099-14109.
- 1697 St. Pierre, K.A., St. Louis, V.L., Lehnerr, I., Gardner, A.S., Serbu, J.A., Mortimer, C.A., Muir, D.C.G.,  
1698 Wiklund, J.A., Lemire, D., Szostek, L. and Talbot, C., 2019. Drivers of mercury cycling in the rapidly  
1699 changing glacierized watershed of the High Arctic's largest lake by volume (Lake Hazen, Nunavut,  
1700 Canada). *Environmental Science & Technology*, 53 (3):1175-1185.
- 1701 St. Pierre, K.A., Chételat, J., Yumvihoze, E. and Poulain, A. J., 2014. Temperature and the sulfur cycle  
1702 control monomethylmercury cycling in High Arctic coastal marine sediments from Allen Bay, Nunavut,  
1703 Canada. *Environmental Science & Technology*, 48 (5):2680-2687.
- 1704 Sunderland, E.M., Gobas, F.A.P.C., Branfireun, B.A. and Heyes, A., 2006. Environmental controls on  
1705 the speciation and distribution of mercury in coastal sediments. *Marine Chemistry*, 102 (1-2):111-123.
- 1706 Sunderland, E.M., Krabbenhoft, D.P., Moreau, J.W., Strode, S.A. and Landing, W.M., 2009. Mercury  
1707 sources, distribution, and bioavailability in the North Pacific Ocean: insights from data and models.  
1708 *Global Biogeochemical Cycles*, 23 (2):GB2010.
- 1709 Swanson, H., Gantner, N., Kidd, K.A., Muir, D.C.G. and Reist, J.D., 2011. Comparison of mercury  
1710 concentrations in landlocked, resident, and sea-run fish (*Salvelinus* spp.) from Nunavut, Canada.  
1711 *Environmental Toxicology and Chemistry*, 30 (6):1459-1467.
- 1712 Tank, S.E., Vonk, J.E., Walvoord, M.A., McClelland, J.W., Laurion, I. and Abbott, B.W., 2020.  
1713 Landscape matters: predicting the biogeochemical effects of permafrost thaw on aquatic networks with  
1714 a state factor approach. *Permafrost and Periglacial Processes*, 31 (3):358-370.

- 1715 Tarbier, B., Hugelius, G., Sannel, A.B.K., Baptista-Salazar, C. and Jonsson, S., 2021. Permafrost thaw  
1716 increases methylmercury formation in Subarctic Fennoscandia. *Environmental Science & Technology*,  
1717 55 (10):6710–6717.
- 1718 Trudel, M. and Rasmussen, J.B., 2006. Bioenergetics and mercury dynamics in fish: a modelling  
1719 perspective. *Canadian Journal of Fisheries and Aquatic Sciences*, 63 (8):1890-1902.
- 1720 Tsui, M.T.K. and Finlay, J.C., 2011. Influence of dissolved organic carbon on methylmercury  
1721 bioavailability across Minnesota stream ecosystems. *Environmental Science & Technology*, 45  
1722 (14):5981-5987.
- 1723 Tsz-Ki Tsui, M., Liu, S., Brasso, R.L., Blum, J.D., Kwon, S.Y., Ulus, Y., Nollet, Y.H., Balogh, S.J.,  
1724 Eggert, S.L. and Finlay, J.C., 2019. Controls of methylmercury bioaccumulation in forest floor food  
1725 webs. *Environmental Science & Technology*, 53 (5):2434-2440.
- 1726 Vadeboncoeur, Y., Jeppesen, E., Zanden, M.J.V., Schierup, H.-H., Christoffersen, K. and Lodge,  
1727 D.M., 2003. From Greenland to green lakes: cultural eutrophication and the loss of benthic pathways  
1728 in lakes. *Limnology and Oceanography*, 48 (4):1408-1418.
- 1729 van der Velden, S., Evans, M.S., Dempson, J.B., Muir, D.C.G. and Power, M., 2013a. Comparative  
1730 analysis of total mercury concentrations in anadromous and non-anadromous Arctic charr (*Salvelinus*  
1731 *alpinus*) from eastern Canada. *Science of the Total Environment*, 447:438-449.
- 1732 van der Velden, S., Dempson, J.B., Evans, M.S., Muir, D.C.G. and Power, M. 2013b. Basal mercury  
1733 concentrations and biomagnification rates in freshwater and marine food webs: effects on Arctic charr  
1734 (*Salvelinus alpinus*) from eastern Canada. *Science of the Total Environment*, 444: 531-542.
- 1735 Varty S, Lehnherr I, St. Pierre K, Kirk J, Wisniewski V. Methylmercury Transport and Fate Shows  
1736 Strong Seasonal and Spatial Variability along a High Arctic Freshwater Hydrologic Continuum.  
1737 *Environmental Science and Technology* 2021; 55: 331-340.
- 1738 Vermilyea, A.W., Nagorski, S.A., Lamborg, C.H., Hood, E.W., Scott, D. and Swarr, G.J., 2017.  
1739 Continuous proxy measurements reveal large mercury fluxes from glacial and forested watersheds in  
1740 Alaska. *Science of the Total Environment*, 599-600:145-155.
- 1741 Villar, E., Cabrol, L. and Heimbürger-Boavida, L.-E., 2020. Widespread microbial mercury  
1742 methylation genes in the global ocean. *Environmental Microbiology Reports*, 12 (3):277-287.
- 1743 Vonk, J.E., Tank, S.E., Bowden, W.B., Laurion, I., Vincent, W.F., Alekseychik, P., Amyot, M., Billet,  
1744 M.F., Canário, J., Cory, R.M., Deshpande, B.N., Helbig, M., Jammet, M., Karlsson, J., Larouche, J.,  
1745 MacMillan, G., Rautio, M., Walter Anthony, K.M. and Wickland, K.P., 2015. Reviews and syntheses:  
1746 effects of permafrost thaw on Arctic aquatic ecosystems. *Biogeosciences*, 12 (23):7129-7167.
- 1747 Walvoord, M.A., Voss, C.I., Ebel, B.A. and Minsley, B.J., 2019. Development of perennial thaw  
1748 zones in boreal hillslopes enhances potential mobilization of permafrost carbon. *Environmental*  
1749 *Research Letters*, 14:015003.
- 1750 Wang, F., Macdonald, R.W., Armstrong, D.A. and Stern, G.A., 2012b. Total and methylated mercury  
1751 in the Beaufort Sea: the role of local and recent organic remineralization. *Environmental Science &*  
1752 *Technology*, 46 (21):11821-11828.
- 1753 Wang F., Lemes M. and Khan M.A.K., 2012a. Metallomics of mercury: role of thiol- and selenol-  
1754 containing biomolecules. In: Cai, Y. and O'Driscoll, N. (Eds.). *Environmental Chemistry and*  
1755 *Toxicology of Mercury*, pp. 517-544. John Wiley & Sons, Ltd.

- 1756 Wang, F., Pućko, M. and Stern, G., 2017. Chapter 19: Transport and transformation of contaminants in  
1757 sea ice. In: Thomas, D.N. (Ed.). *Sea Ice*, Third Edition, pp. 472-491 John Wiley & Sons, Ltd.
- 1758 Wang, K., Munson, K.M., Beaupré-Laperrière, A., Mucci, A., Macdonald, R.W. and Wang, F., 2018.  
1759 Subsurface seawater methylmercury maximum explains biotic mercury concentrations in the Canadian  
1760 Arctic. *Scientific Reports*, 8 (1):14465.
- 1761 West, J., Graham, A.M., Liem-Nguyen, V. and Jonsson, S., 2020. Dimethylmercury degradation by  
1762 dissolved sulfide and mackinawite. *Environmental Science & Technology*, 54 (21):13731-13738.
- 1763 West J., Gindorf S., Jonsson S., 2022, Photochemical Degradation of Dimethylmercury in Natural  
1764 Waters, *Environ. Sci. Technol.* 56, 9, 5920–5928
- 1765 Wietrzyk-Pełka, P., Rola, K., Szymański, W. and Węgrzyn, M.H., 2020. Organic carbon accumulation  
1766 in the glacier forelands with regard to variability of environmental conditions in different ecogenesis  
1767 stages of High Arctic ecosystems. *Science of the Total Environment*, 717:135151.
- 1768 Willis C.E., Kirk J., St. Louis V.L., Lehnerr I., Ariya P.A., Rangel-Alvarado R.B., 2018. Sources of  
1769 Methylmercury to Snowpacks of the Alberta Oil Sands Region: A Study of In Situ Methylation and  
1770 Particulates. *Environmental Science & Technology* 52,2,531-540
- 1771 Wu, P., Kainz, M.J., Bravo, A.G., Åkerblom, S., Sonesten, L. and Bishop, K., 2019. The importance  
1772 of bioconcentration into the pelagic food web base for methylmercury biomagnification: a meta-  
1773 analysis. *Science of the Total Environment*, 646:357-367.
- 1774 Yang, Z., Fang, W., Lu, X., Sheng, G.-P., Graham, D.E., Liang, L., Wullschleger, S.D. and Gu, B.,  
1775 2016. Warming increases methylmercury production in an Arctic soil. *Environmental Pollution*,  
1776 214:504-509.
- 1777 Zdanowicz, C. M., E. M. Krümmel, A. J. Poulain, E. Yumvihoze, J. Chen, M. Štok, M. Scheer, and H.  
1778 Hintelmann., 2016, Historical variations of mercury stable isotope ratios in Arctic glacier firn and ice  
1779 cores, *Global Biogeochem. Cycles*, 30, 1324–1347
- 1780 Zhang, B., Chen, T., Guo, J., Wu, M., Yang, R., Chen, X., Wu, X., Zhang, W., Kang, S., Liu, G. and  
1781 Dyson, P., 2020. Microbial mercury methylation profile in terminus of a high-elevation glacier on the  
1782 northern boundary of the Tibetan Plateau. *Science of the Total Environment*, 708: 135226.
- 1783 Zhou J, Obrist D. 2021. Global Mercury Assimilation by Vegetation. *Environmental Science &*  
1784 *Technology*; 55: 14245-14257.