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## **Biotic transformation of methylmercury at the onset of the Arctic spring bloom**

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## 19 **Keywords**

20 Barents Sea, mercury, seasonality, Arctic Ocean, spring bloom

## 21 **Abstract**

22 Despite the lack of local anthropogenic mercury sources, methylated mercury (MeHg)  
23 concentrations in Arctic biota are higher than in biota from lower latitudes. The main entry route  
24 occurs during the bioconcentration of seawater monomethylmercury (MMHg) into phytoplankton.  
25 Despite the known seasonal changes in biological activity in the region, little is known about the  
26 seasonal cycling of total mercury (THg) and MeHg in the Arctic Ocean. Here, we report the  
27 concentrations of THg and MeHg in seawater sampled from the northwestern Barents Sea water  
28 column during late winter and spring. In the upper 500 m, the THg concentrations are significantly  
29 higher in spring ( $0.64 \pm 0.09$  pmol L<sup>-1</sup>) compared to late winter ( $0.53 \pm 0.07$  pmol L<sup>-1</sup>), driven by  
30 seasonal inputs to surface waters from atmospheric deposition and the dynamics of changing sea  
31 ice conditions. Contrastingly, the MeHg concentrations in spring were significantly lower ( $41 \pm$   
32  $39$  fmol L<sup>-1</sup>) compared to late winter ( $85 \pm 42$  fmol L<sup>-1</sup>). We suggest that most MeHg is biotically  
33 demethylated by both phytoplankton and bacteria, with additional losses from photodemethylation  
34 and evasion. Our observations highlight the importance of demethylation during potential uptake  
35 of methylmercury coinciding with the Arctic spring bloom. Lastly, we use our new data together  
36 with previously published seasonal data in the region to construct a simplified seasonal mercury  
37 cycle in an Arctic marginal ice zone.

## 38 **1 Introduction**

39  
40 The bioaccumulation of mercury (Hg) in the Arctic Ocean continues to remain an issue of concern  
41 for the marine ecosystem and indigenous peoples (Basu et al., 2022). Specifically, the  
42 bioconcentration of toxic monomethylmercury (MMHg) into phytoplankton and its subsequent  
43 bioaccumulation in the marine food web represents a risk to higher trophic level organisms (Dietz  
44 et al., 2022). The development and peak of primary productivity in the Arctic Ocean is restricted  
45 to the spring and summer (April to August), when light is available. In the spring, the  
46 phytoplankton bloom develops in the marginal ice zone (Strass and Nöthig, 1996; Reigstad et al.,  
47 2002). Several studies have highlighted phytoplankton blooms below fully consolidated sea ice as  
48 a result of the thinning and more dynamic icescape (Assmy et al., 2017, Ardyna et al., 2020).

49 Concurrently, the Arctic Ocean receives seasonal inputs of mostly inorganic Hg (iHg) via  
50 atmospheric deposition, riverine input, erosion, snow, glacial and sea ice melt (Dastoor et al.,  
51 2022). However, seasonal cycling of Hg in the water column in the Arctic region is still poorly  
52 understood (Kohler et al., 2022a). As the Arctic spring bloom provides a crucial transfer of energy  
53 and potentially Hg into higher trophic levels, more knowledge on the amount of Hg available for  
54 bioconcentration prior and during the Arctic spring bloom is needed. Seasonal inputs from thawing  
55 permafrost, glacial melt, and erosion are expected to deliver greater amounts of iHg to the  
56 biologically productive Arctic Ocean shelf seas in the future (Chételat et al., 2022) with most Hg  
57 expected to remain in the marine environment (Araujo et al., 2022). Hg can be scavenged to shelf  
58 sediments, advected with the halocline, cascaded into the deep basin (Dastoor et al., 2022) or  
59 potentially transported into the Central Arctic Ocean via the transpolar drift (Charette et al., 2020).

60

61 Several major Hg inputs to the Arctic region have strong seasonal patterns. Atmospheric mercury  
62 depletion events (AMDEs) (Schroeder et al., 1998) occur during polar sunrise in March and  
63 frequently occur throughout April, depositing iHg to snow, sea ice, and surface waters. Although  
64 a substantial fraction is photochemically reduced and reemitted back to the atmosphere as gaseous  
65 Hg<sup>0</sup> (Durnford and Dastoor 2011, Araujo et al., 2022), high concentrations of total Hg (THg) are  
66 seen in snow, sea ice and frost flowers (Dommergue et al., 2010, Chaulk et al., 2011). Sea ice,  
67 melting snow and glacial runoff can also provide additional Hg to the coastal and open ocean  
68 (Schartup et al., 2020, Kim et al., 2020a, Dastoor et al., 2022). Rivers also deliver large amounts  
69 of iHg from the snowpack, surface soils, thawing permafrost, and erosion to the Arctic shelf seas  
70 during a freshet in springtime (Sonke et al., 2018, Zolkos et al., 2020). While new evidence  
71 suggests seasonal cycling of iHg in the upper water column during autumn and early winter  
72 (Kohler et al., 2022a) with suspected surface losses to deeper waters and sediments with sinking  
73 particles, water column measurements of Hg species from late winter are scarce (Wang et al.,  
74 2012).

75

76 Arctic surface waters are elevated in THg during the summer (Heimbürger et al., 2015) from  
77 seasonal iHg inputs. Light reduces iHg to Hg<sup>0</sup> and photolytically degrades MeHg (Sellers et al.,  
78 1996). Gaseous Hg species can evade from the marginal ice zones (Yue et al., 2023) while ionic  
79 Hg species can be transported downward with sinking particles (Tesán Onrubia et al., 2020). Ionic

80 Hg is also often complexed to dissolved organic matter (DOM) and reduced sulfur compounds,  
81 which can affect Hg uptake and biogeochemical transformations between iHg and MeHg  
82 (Ravichandran 2004). During and after the phytoplankton bloom, bacterial remineralization of  
83 sinking particulate organic matter fuels in situ MeHg production, leading to elevated subsurface  
84 concentrations of MeHg in the marginal ice zones and the permanently ice-covered central Arctic  
85 Ocean (Heimbürger et al., 2015). However, sea ice hinders both photodemethylation (Point et al.,  
86 2009) and the evasion of gaseous species ( $\text{Hg}^0$  and dimethylmercury (DMHg)) (DiMento et al.,  
87 2019). Gaseous Hg species potentially build up under and within the ice throughout winter  
88 (Andersson et al., 2008, Schartup et al., 2020). Hg that is removed from surface waters via sinking  
89 particles and delivered to bottom sediments may ultimately be methylated to MeHg (Kim et al.,  
90 2020b). Resuspension of fine-grained sediment and diffusion of porewater can deliver both iHg  
91 and MeHg back to subsurface Arctic waters (Kim et al., 2020b). While in situ incubations indicate  
92 Arctic waters can be in net states of methylation or demethylation (Lehnher et al., 2011), the  
93 advection of outflowing Arctic waters indicate that the Arctic Ocean is a net producer of MeHg  
94 (Petrova et al., 2020, Jonsson et al., 2022a). Consequently, understanding the timing and  
95 magnitude of both Hg methylation and demethylation is important. Due to a limited number of  
96 observations, the fate of marine MeHg throughout the Arctic winter and during the transition to  
97 the spring bloom is unknown.

98  
99 We hypothesized the potential seasonality of pelagic Hg species in the northern Barents Sea in the  
100 late winter to spring transition due to seasonal Hg inputs and biological development. We  
101 measured unfiltered THg (the sum of all mercury species) and unfiltered MeHg (the sum of MMHg  
102 and DMHg) in seawater, under ice water, and sea ice on two seasonal cruises in late winter (March)  
103 and spring (end of April-May). We report Hg concentrations from seven stations along a south-  
104 north transect and explore how hydrography, biological parameters, and seasonal processes affect  
105 our observations using literature and statistical relationships. We combine our Hg and  
106 environmental data with that of two other seasonal cruises in the region (Kohler et al., 2022a) to  
107 propose a simple seasonal model of the pelagic Hg cycle in surface waters of an Arctic marginal  
108 ice zone.

109

## 110 **2 Materials & Methods**

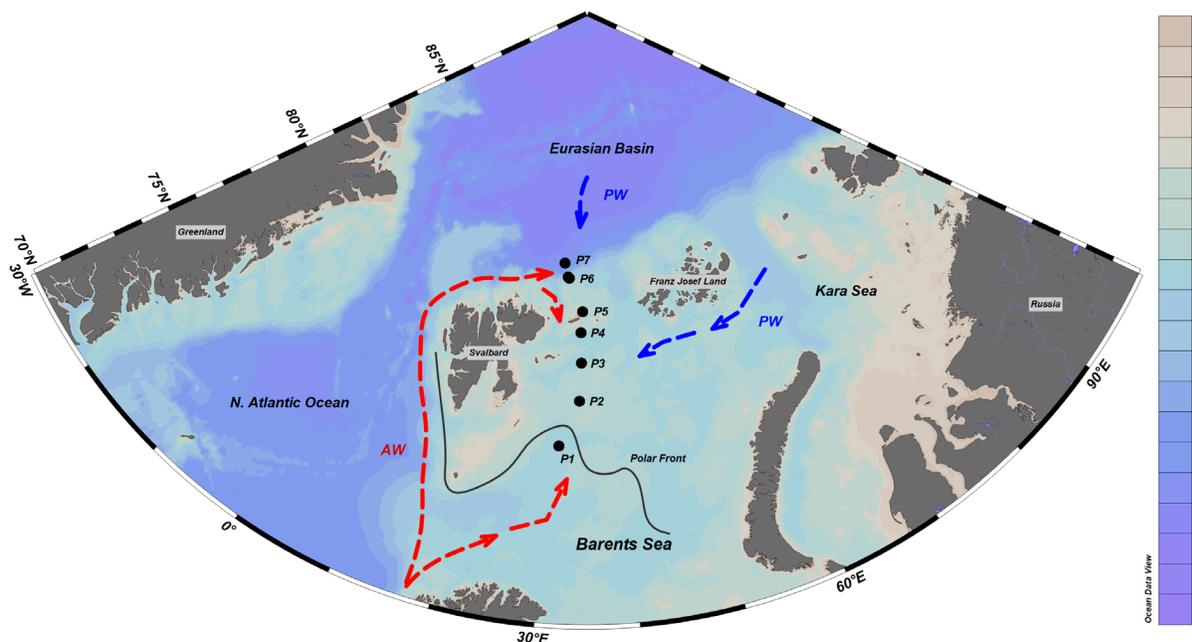
### 111 **2.1 Oceanographic Conditions**

112 The northern Barents Sea consists of water masses with both Atlantic Water (AW) and Polar Water  
113 (PW) origins (Sundfjord et al., 2020). AW, characteristic of warmer water with higher salinity,  
114 flows into the northern Barents Sea from the south through the Barents Sea Opening and through  
115 troughs in the north (Lundesgaard et al., 2022). This AW sinks below colder and fresher PW which  
116 arrives from the central Arctic Ocean and the Kara Sea in the east (Loeng 1991). The boundary  
117 between these two water masses is often referred to as the polar front, and generally delineates the  
118 extent of sea ice in the region. Surface PW with colder temperatures entering the northern Barents  
119 Sea leads to sea ice formation and can advect additional sea ice into the region. In the summer, sea  
120 ice melt and surface heating form a stratified meltwater layer in the upper water column. By  
121 autumn, the beginnings of sea ice formation, cooling, and turbulence lead to water column mixing.  
122 This overturning of the water column continues throughout the polar night and into spring. Dense  
123 water formation on the Barents Sea shelf enters the Arctic Basin through deep water plumes along  
124 the continental shelf (Rudels et al., 2000).

125 On the western coast of Svalbard, the West Spitsbergen Current brings AW into the Eurasian  
126 Basin. This current continues parallel along the Barents Sea shelf break and past Franz Josef Land  
127 in the east. This AW shows a distinct seasonality, with warmer temperatures in autumn and colder  
128 temperatures in early spring (Pérez-Hernández et al., 2019). As AW enters the Arctic Basin, it  
129 begins to cool and sinks below PW, forming an AW core at around 300 m depth in the water  
130 column north of Svalbard. As mixing occurs between AW and PW, continued temperature losses  
131 lead to the formation of modified Atlantic Water (mAW) below the pycnocline and AW core. As  
132 this water continues to cool below 0°C and sinks, typically below 1000 m, it forms Eurasian Basin  
133 Deep Water (EBDW). In addition, extensions of the West Spitsbergen Current flowing onto the  
134 northern Barents Sea shelf (Lundesgaard et al., 2022) can form warm Polar Water (wPW). This  
135 wPW is not as salty as AW yet warmer than PW, and can be found in deeper depths of the northern  
136 Barents Sea shelf or at the surface during summer stratification.

137

138 **2.2 Field Sampling**



139  
140 **Figure 1:** Map of sampling locations (stations P1-P7) in the northern Barents Sea region. A solid  
141 black line indicates the approximate location of the Polar Front, while the dashed colored arrows  
142 show a simplified water mass scheme and current direction, with Atlantic Water (AW) in red and  
143 Polar Water (PW) in blue. The color bar indicates water depth. Map created with Ocean Data View  
144 (Schlitzer 2021).

145 All samples were collected on a south-north transect in the northern Barents Sea and Nansen Basin  
146 (76 – 83°N) on two cruises, Q1 from March 2-24, 2021 (hereafter referred to as late winter), and  
147 Q2 from April 27 - May 20, 2021 (hereafter referred to as spring), on R/V *Kronprins Haakon* as  
148 part of The Nansen Legacy project (Fig. 1). Seawater samples for Hg were taken at seven stations  
149 at 8 depths down to 500 m depth using Teflon-coated GOFLO bottles attached sequentially to a  
150 600 m nonmetallic wire. At depths deeper than 500 m, Hg samples were collected with 10 L Niskin  
151 bottles mounted to a rosette. Upon retrieval, GOFLO bottles were transported immediately to a  
152 custom-made clean lab onboard. THg samples were collected unfiltered into pre-cleaned (Cutter  
153 et al., 2017) 40 mL borosilicate glass bottles with PTFE-lined caps with no headspace and kept  
154 unpreserved. MeHg samples were collected unfiltered into brand new 125 or 250 mL PET bottles  
155 and were acidified under a Class 100 clean air laminar flow hood to 0.4% v/v with double-distilled  
156 HCl. Upon acidification, dimethylmercury (DMHg) converts quantitatively to

157 monomethylmercury (MMHg) (Black et al., 2009); as a result, the sum of MMHg and DMHg is  
158 measured as total methylated mercury (MeHg). All Hg samples were double-bagged and stored in  
159 the dark at 4°C until analysis. Field blanks for THg were collected by bringing laboratory MilliQ  
160 onboard and subsampling at the time of field sampling into precleaned borosilicate bottles and  
161 treated identically to seawater samples. However, as we could not guarantee trace metal clean  
162 Niskin bottles for samples collected below 500 m, these data are reported (SI Fig. 1) but not  
163 included in subsequent statistical processing when noted.

164  
165 Under ice water (UIW) was collected using a handheld 10 L Niskin bottle lowered into a hole cut  
166 into the sea ice at stations P4, P6, and P7 on both cruises. UIW was collected directly from the  
167 Niskin similar to the other seawater samples. Sea ice was collected at two stations, P4 and P7,  
168 using a 9 cm diameter ice corer (Kovacs Enterprises, Inc., USA), bagged whole and taken directly  
169 onboard into a -20°C cold lab. The bottom 10 cm of the ice core was cut using a handheld carbon  
170 steel saw. Approximately 3 mm of ice was shaved off from all sides of the ice section with a  
171 ceramic knife to remove any potential contamination before being double bagged and melted at  
172 room temperature. One frost flower sample was collected in late winter using plastic equipment  
173 and melted at 4°C. After melting, all samples were decanted into precleaned 40 mL borosilicate  
174 glass bottles and 125 or 250 mL PET bottles and stored in the dark at 4°C until analysis.

### 175 **2.3 Mercury Analysis**

176 The concentration of THg in the samples was measured by cold vapor atomic fluorescence  
177 spectroscopy (CVAFS) according to USEPA Method 1631 at the Mediterranean Institute of  
178 Oceanography (MIO) using a custom-made single gold trap setup described elsewhere  
179 (Heimbürger et al., 2015, Kohler et al., 2022a), within one year after sampling. As samples were  
180 unfiltered to reduce contamination risk from filtration equipment, THg concentration includes both  
181 particulate and dissolved species of Hg. The THg concentration in all samples was calculated from  
182 a 6-point calibration curve traceable to NIST 3133 (National Institute of Standards and  
183 Technology) standard reference material in previously purged, Hg-free seawater. Standard  
184 bracketing of samples with a NIST 3133 matrix spike were used to correct for eventual instrument  
185 signal drift and ensure ongoing precision and recovery. Aliquots of certified reference materials

186 ORMS-5 (Research Council of Canada) and ERM-CA400 (European Reference Materials) were  
187 spiked into previously purged Hg-free seawater matrix with recoveries of  $93 \pm 2\%$  and  $125 \pm 4\%$ ,  
188 respectively. Selected seawater samples run in triplicate from both cruises had an average standard  
189 deviation of  $0.05 \text{ pmol L}^{-1}$ . Field blanks of MilliQ water were measured for THg for both late  
190 winter ( $0.31 \pm 0.02 \text{ pmol L}^{-1}$ ,  $n = 5$ ) and spring ( $0.23 \pm 0.02 \text{ pmol L}^{-1}$ ,  $n = 3$ ) cruises. As MilliQ  
191 water can potentially contain traces of Hg, we concluded that there was no significant  
192 contamination affecting our THg measurements.

193 The MeHg concentrations in seawater samples was measured using the isotope dilution (ID)  
194 technique via gas chromatography (GC) – sector field ICP-MS (GC-SF-ICP-MS) at MIO within  
195 one year after sampling (Heimbürger et al., 2015). Briefly, approximately 125 mL of seawater  
196 sample was spiked with enriched isotopes of  $i^{199}\text{Hg}$  and  $\text{MM}^{201}\text{Hg}$  (ISC Science) and equilibrated  
197 for 15 minutes. Samples were then adjusted to pH 3.9 using a sodium acetate buffer solution  
198 (ULTREX® II Ultrapure Reagent, J.T. Baker, USA) and  $\text{NH}_3$  (ULTREX® II Ultrapure Reagent,  
199 J.T. Baker, USA). A fresh aliquot of 1% sodium tetrapropylborate solution (Merseburger  
200 Spezialchemikalien) and 150  $\mu\text{L}$  of isooctane (Sigma Aldrich) were added to derivatize and  
201 preconcentrate Hg species. Samples were sealed and shaken for 15 minutes before the organic  
202 extract was transferred to GC vials for analysis on a coupled GC (THERMO GC 1300 with GC220  
203 transfer module) SF-ICP-MS (Thermo Element XR) system. Ultra-trace clean techniques and  
204 coupling to a high resolution ICP-MS allows a detection limit of  $2 \text{ fmol L}^{-1}$  and a limit of  
205 quantitation of  $7 \text{ fmol L}^{-1}$  ( $\text{LOQ} = 3.3 * \text{LOD}$ ). Selected seawater samples run in duplicate from  
206 both cruises had an average standard deviation of  $7 \text{ fmol L}^{-1}$ . The  $\text{MM}^{201}\text{Hg}$  spike concentration  
207 used to calculate the sample MeHg concentrations had an accuracy of  $94.4 \pm 8.4\%$ . The accuracy  
208 of our spike was determined using reverse isotope dilution using the Brooks Rand  $\text{MMHgCl}$   
209 standard, traceable to NIST1641E.

## 210 **2.4 Additional Data and Parameters**

211 Water temperature and salinity were recorded on a Seabird SBE 9 CTD unit attached to a rosette.  
212 Quality controlled temperature, salinity, sea ice concentration, nutrients, chlorophyll *a*, particulate  
213 organic carbon, nutrients, and phytoplankton abundance data for all seasonal cruises were retrieved  
214 from the Norwegian Marine Data Centre and the Norwegian Polar Institute (Gerland 2022,

215 Ludvigsen 2022, Reigstad 2022, Søreide 2022, Steer and Divine 2022, Marquardt 2022b, 2022c,  
216 2022d, 2022e, Müller 2023a, 2023b, 2023c, 2023d, Chierici et al. 2021a, 2021b, Jones et al.,  
217 2022a, 2022b, Vader 2022, Assmy 2022a, 2022b). Part of the late winter MeHg data (station  
218 average, upper 400 m only) presented in this analysis was previously published in the bottom  
219 image in Fig. 6 in Dietz et al., 2022. Flow cytometry, bacterial community analysis, and dissolved  
220 organic matter analysis details are presented in the Supplementary Information. Latitude and  
221 longitude coordinates for CTD stations differ slightly from GOFLO casts due to ship drift.  
222 Conservative temperature ( $\Theta$ ), absolute salinity ( $S_A$ ), and potential density anomaly ( $\sigma_\theta$ ) were  
223 calculated in Ocean Data View. Water mass definitions were assigned based on the most recent  
224 report from the Nansen Legacy project for the northern Barents Sea (Sundfjord et al., 2020).

225  
226 Statistical processing was performed for data above 500 m depth in Minitab software. Spearman  
227 rank correlation coefficients were calculated using a 95% CI with significant p-value  $< 0.05$ . Data  
228 selected for Student's t-test were log transformed for non-normal data distributions prior to  
229 evaluation using a 95% CI with significant p-value  $< 0.05$ . Log-log regression was performed on  
230 selected environmental variables with significance evaluated by F-test with a 95% CI and  
231 significant p-value  $< 0.05$ . Selected environmental parameters were chosen for Principal  
232 Component Analysis, where data is standardized prior to dimension reduction and plotted  
233 orthogonally as loadings plots for the first two principal components.

234

## 235 **3 Results & Discussion**

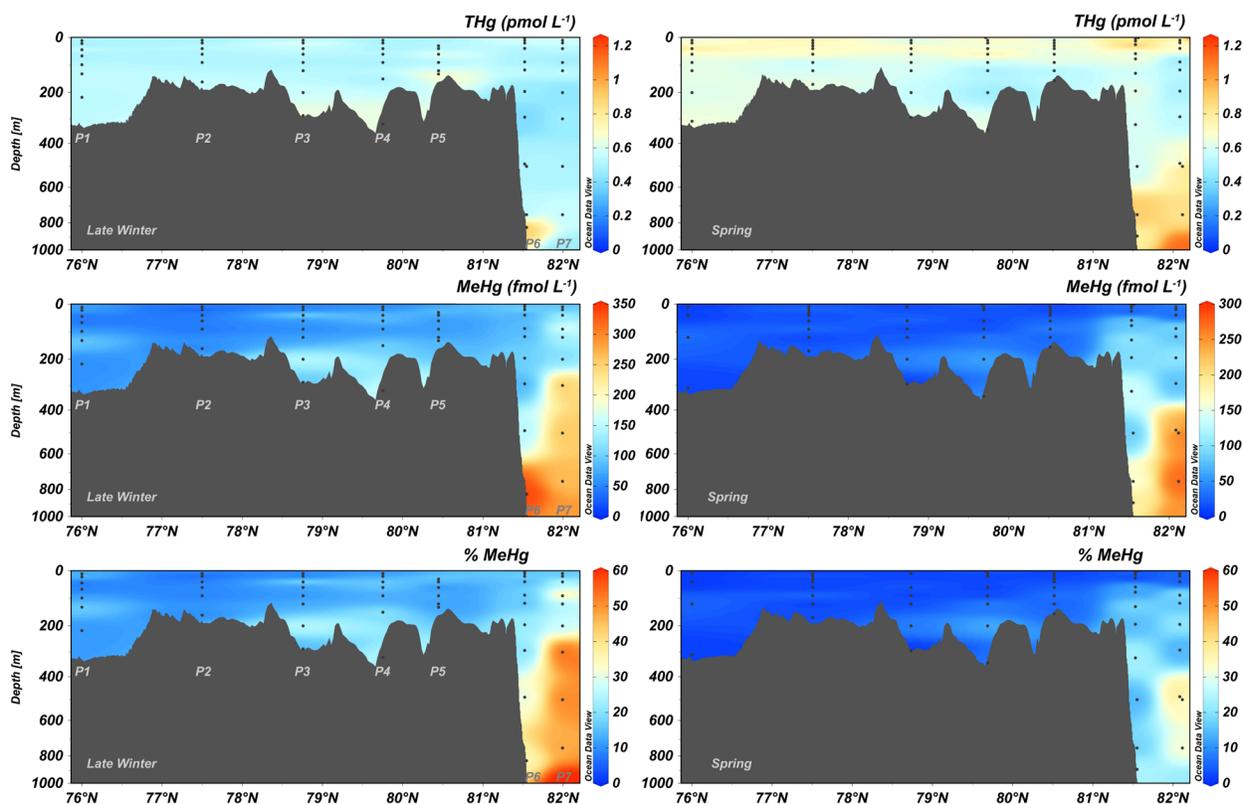
### 236 **3.1 Hydrography & sea ice**

237 In general, all stations in the northern Barents Sea showed both similarities and differences in  
238 water mass distributions (SI Figs. 2 & 3) in both late winter and spring, with changes influenced  
239 by sea ice coverage or temperature shifts (Jones et al., 2023, Van Engeland et al., 2023). In the late  
240 winter and spring sampling periods, station P1 south of the polar front had a mAW signature, with  
241  $S_A$  greater than  $35.06 \text{ g kg}^{-1}$  and  $\Theta > 0^\circ\text{C}$ . Stations P2, P3, and P4, all north of the polar front, had  
242 PW ( $\Theta < 0^\circ\text{C}$ ,  $\sigma_\theta < 27.97 \text{ kg m}^{-3}$ ) in surface waters and wPW ( $\Theta > 0^\circ\text{C}$ ,  $S_A < 35.06 \text{ g kg}^{-1}$ ) in deeper  
243 waters extending to the bottom. A deepening of PW in spring at stations P2-P5 could suggest  
244 rapidly changing sea ice conditions (SI Fig. 4), with small increases in salinity indicating  
245 refreezing and brine rejection. Station P5 had low temperatures, with PW throughout the entire  
246 water column in both seasons. Stations P6 and P7 intersect the West Spitsbergen Current, with PW  
247 and wPW in surface waters extending until approximately 200 m depth, where both mAW and  
248 AW ( $\Theta > 2^\circ\text{C}$ ,  $S_A > 35.06 \text{ g kg}^{-1}$ ) extends down to about 500 m. In waters below 500 m, mAW  
249 persists until about 800 m depth and EBDW ( $\Theta < 0^\circ\text{C}$ ,  $S_A > 35.06 \text{ g kg}^{-1}$ ) fills the Nansen Basin.  
250 At the time of sampling in late winter, first-year sea ice covered all stations except for P1  
251 (Marquardt et al., 2023a). Some sea ice changes were observed during sampling (SI Fig. 4), but in  
252 general most stations were covered by close drift ice. Minor changes in ice coverage occurred  
253 before sampling for shelf stations, while P6 and P7 showed significant variations in ice coverage.  
254 However, by the end of the late winter campaign, P6 and P7 were also covered by close drift ice.  
255 Shelf stations P2-P5 showed dynamic ice conditions in between the late winter and spring cruises,  
256 while P6 and P7 were covered by slightly older sea ice in spring (Marquardt et al., 2023a).

### 257 **3.2 THg and MeHg distributions in the water column during late winter and spring**

258 THg concentrations in late winter (Fig. 2, top left) in the northern Barents Sea and Nansen Basin  
259 were on average  $0.56 \pm 0.17 \text{ pmol L}^{-1}$  (Table 1). In spring (Fig. 2, top right), THg concentrations  
260 in the northern Barents Sea and Nansen Basin were on average  $0.69 \pm 0.24 \text{ pmol L}^{-1}$  ( $n = 67$ ,  $\pm$   
261 1SD, range:  $0.46 - 2.37 \text{ pmol L}^{-1}$ ). In the upper 500 m, our average THg concentrations on the  
262 spring cruise ( $0.64 \pm 0.09 \text{ pmol L}^{-1}$ ,  $n = 56$ ) were significantly higher (log transformed, Student's  
263 t-test,  $p < 0.05$ ) compared to late winter ( $0.53 \pm 0.07 \text{ pmol L}^{-1}$ ,  $n = 51$ ). On the shelf, our THg

264 concentrations in both late winter and spring (Table 1) are similar to THg concentrations in the  
 265 southern Barents Sea Opening ( $0.43 \pm 0.14 \text{ pmol L}^{-1}$ ) (Petrova et al., 2020) and comparable to  
 266 previous expeditions in this area ( $0.63 \pm 0.19 \text{ pmol L}^{-1}$  in summer, and  $0.46 \pm 0.07 \text{ pmol L}^{-1}$  in  
 267 early winter) (Kohler et al., 2022a). In late winter, shelf stations P1 - P5 exhibited relatively  
 268 constant THg concentration profiles with depth, similar to early winter 2019 (Kohler et al., 2022a)  
 269 while spring vertical THg profiles are similar to observed THg profiles in summer 2019 (Kohler  
 270 et al., 2022a), with enrichments in surface waters < 50 m.



271  
 272 **Figure 2:** THg concentrations (top,  $\text{pmol L}^{-1}$ ), MeHg concentrations (middle,  $\text{fmol L}^{-1}$ ), and %  
 273 MeHg as THg (bottom) along the shelf-basin transect in the northern Barents Sea during late winter  
 274 (left) and spring (right) in 2021. The maximum plotted THg concentration range is  $1.25 \text{ pmol L}^{-1}$ .  
 275 Depths where MeHg concentration < LOQ ( $7 \text{ fmol L}^{-1}$ ) have been removed from the visualization.  
 276 The x-axis displays latitude and the nonlinear y-axis is in units of depth (m). Transect created with  
 277 Ocean Data View (Schlitzer 2021).

278 MeHg concentrations (Table 1, Fig. 2 middle left) along the entire transect during late winter were  
 279  $112 \pm 76 \text{ fmol L}^{-1}$  ( $n = 58, \pm 1\text{SD}$ ). At station P6 and P7, MeHg concentrations were lowest near

280 the surface and directly below the ice ( $UIW = 59 \pm 11 \text{ fmol L}^{-1}$ ,  $n = 2$ ). At P7, MeHg concentrations  
281 greater than  $250 \text{ fmol L}^{-1}$  began at 300 m depth in the AW core of the West Spitsbergen Current.  
282 By the spring cruise, MeHg concentrations along the entire transect were  $69 \pm 75 \text{ fmol L}^{-1}$  ( $n = 60$ ,  
283  $\pm 1SD$ ) (Fig. 2, middle right) and covered a relatively large range (Table 1). In the upper 500 m,  
284 our average MeHg concentrations on the spring cruise ( $41 \pm 39 \text{ fmol L}^{-1}$ ,  $n = 49$ ) were significantly  
285 lower (log transformed, Student's t-test,  $p < 0.05$ ) compared to late winter ( $85 \pm 42 \text{ fmol L}^{-1}$ ,  $n =$   
286  $51$ ). Shelf stations in spring exhibited low MeHg concentrations in general (Table 1), with the  
287 lowest MeHg concentrations observed at P1 in mAW and some measurements below the LOQ of  
288  $7 \text{ fmol L}^{-1}$ . Previous seasonal expeditions on the northern Barents Sea shelf showed similarly  
289 constant MeHg distributions with depth, albeit with slightly higher average MeHg concentrations  
290 (Kohler et al., 2022a). Our spring MeHg concentrations on the shelf however, are also lower than  
291 both the Barents Sea Opening ( $\sim 60 \pm 30 \text{ fmol L}^{-1}$ ) (Petrova et al., 2020) and among the lowest  
292 reported for the Arctic region (Jonsson et al., 2022a). Percent MeHg (%MeHg) in both seasons  
293 follows a similar distribution pattern as MeHg concentrations (Figure 2, bottom), with higher  
294 %MeHg observed in subsurface waters. In late winter, %MeHg is  $20 \pm 13\%$  ( $n = 58$ ) while spring  
295 %MeHg is  $10 \pm 10\%$  ( $n = 60$ ). Similar to MeHg, there is a significant difference in %MeHg in the  
296 upper 500 m (log transformed, Student's t-test,  $p < 0.05$ ) between late winter ( $16 \pm 9\%$ ,  $n = 51$ )  
297 and spring cruises ( $7 \pm 8\%$ ,  $n = 49$ ).

298 The highest THg concentrations for both late winter and spring were observed in deep waters of  
299 P6 and P7 between 500 - 1500 m depth and both maxima are observed 10 m from the bottom of  
300 the Nansen Basin (SI Fig. 1). Our unfiltered THg observations in basin waters in both late winter  
301 and spring are within the ranges reported in the central Arctic Ocean (Heimbürger et al., 2015,  
302 Wang et al., 2018, Agather et al., 2019, Jonsson et al., 2022b) and the Fram Strait (Petrova et al.,  
303 2020). Similarly, relatively high MeHg concentrations were observed in basin waters  $> 500 \text{ m}$  (SI  
304 Fig. 1). These findings in late winter are similar to deep water MeHg concentrations observed in  
305 the Fram Strait (Petrova et al., 2020) and the central Arctic Ocean (Jonsson et al., 2022b).  
306 However, as we could not guarantee trace metal clean Niskin bottles for our deep water sampling  
307 ( $> 500 \text{ m}$ ), some caution is needed when interpreting these results.

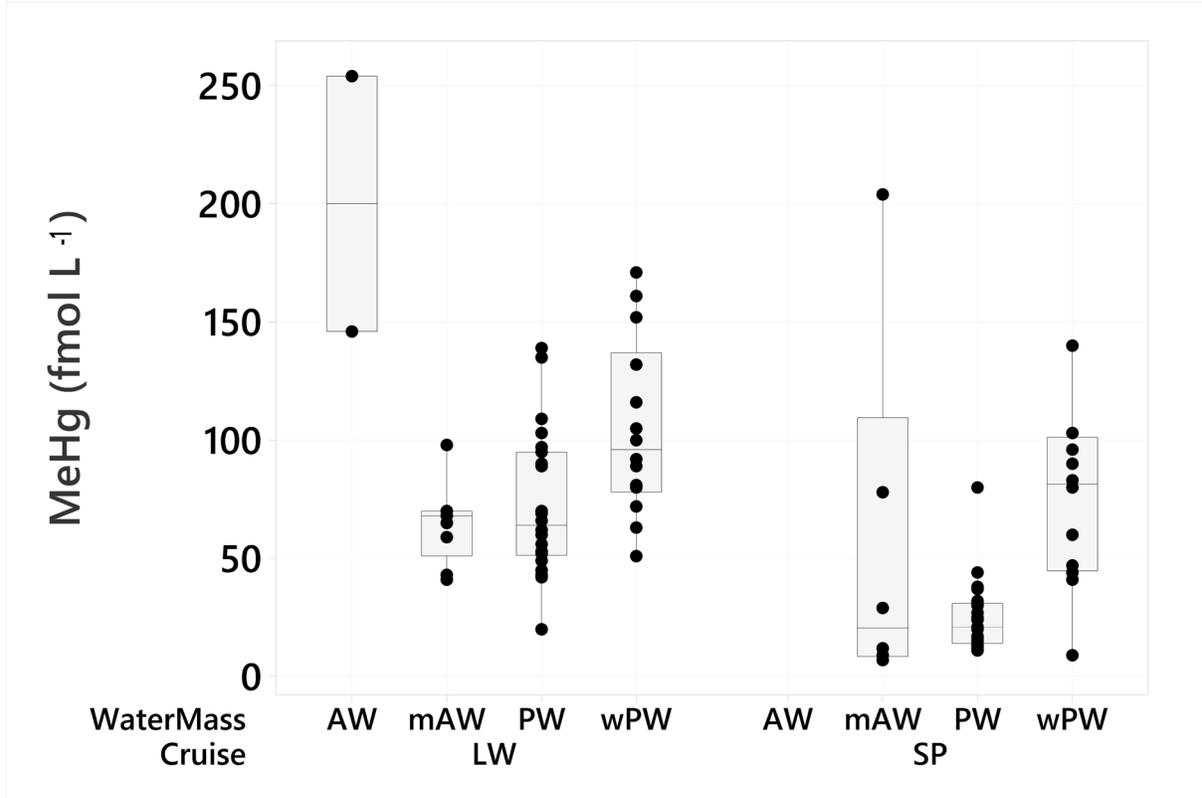
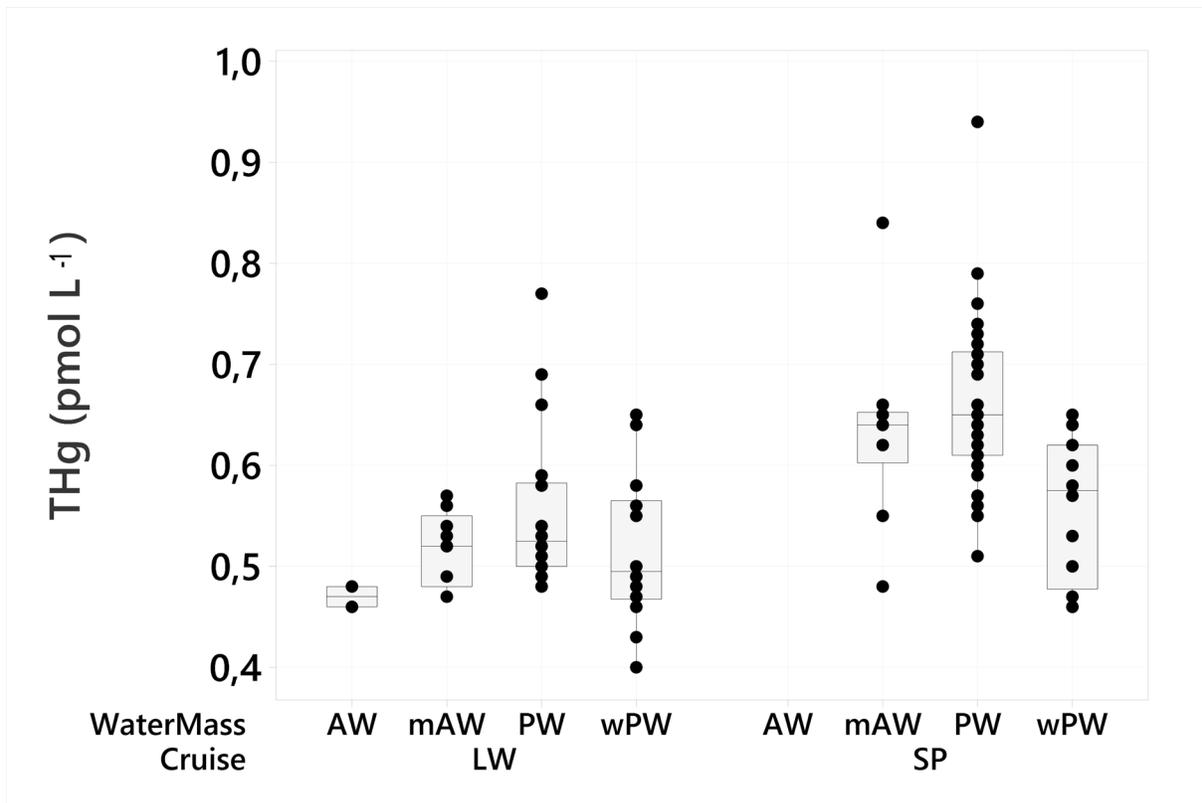
308

309 **Table 1:** Average ( $\pm$  one standard deviation) and range of THg, MeHg, concentrations and %  
 310 MeHg on both late winter and spring cruises for all water column samples, shelf stations (P1 - P5),  
 311 basin stations (P6, P7), under ice water (UIW), ice cores, and frost flowers (FF).

	Total Hg (pmol L <sup>-1</sup> )					Methylated Hg (fmol L <sup>-1</sup> )					
	Late winter		Spring			Late winter			Spring		
	Average $\pm$ 1SD	Range	Average $\pm$ 1SD	Range		Average $\pm$ 1SD	Range	% MeHg as THg	Average $\pm$ 1SD	Range	% MeHg as THg
<b>All water column data</b>	0.56 $\pm$ 0.17 (n = 60)	0.40 - 1.61	0.69 $\pm$ 0.24	0.46 - 2.37 (n = 67)	<b>All water column data</b>	112 $\pm$ 76 (n = 58)	20 - 340	20 $\pm$ 13	69 $\pm$ 75 (n = 60)	< 7 - 288	10 $\pm$ 10
<b>Upper 500 m only</b>	0.53 $\pm$ 0.07 (n = 51)	0.40 - 0.77	0.64 $\pm$ 0.09	0.46 - 0.94 (n = 56)	<b>Upper 500 m only</b>	85 $\pm$ 42 (n = 51)	20 - 254	16 $\pm$ 9	41 $\pm$ 39 (n = 49)	< 7 - 204	7 $\pm$ 8
<b>Shelf (P1-P5) all depths</b>	0.54 $\pm$ 0.07 (n = 34)	0.47 - 0.77	0.64 $\pm$ 0.08	0.46 - 0.84 (n = 40)	<b>Shelf (P1-P5) all depths</b>	71 $\pm$ 32 (n = 34)	20 - 161	13 $\pm$ 5	23 $\pm$ 13 (n = 33)	< 7 - 60	4 $\pm$ 3
<b>Basin (P6, P7) all depths</b>	0.59 $\pm$ 0.24 (n = 26)	0.40 - 1.61	0.76 $\pm$ 0.36	0.47 - 2.37 (n = 27)	<b>Basin (P6, P7) all depths</b>	171 $\pm$ 82 (n = 24)	72 - 340	30 $\pm$ 15	127 $\pm$ 79 (n = 27)	21 - 288	18 $\pm$ 10
<b>UIW</b>	0.58 $\pm$ 0.02 (n = 3)	0.56 - 0.60	1.00 $\pm$ 0.30	0.73 - 1.32 (n = 3)	<b>UIW</b>	59 $\pm$ 11 (n = 2)	50 - 67	10 $\pm$ 2	36 $\pm$ 17 (n = 3)	26 - 55	4 $\pm$ 1
<b>Ice core</b>	0.27 $\pm$ 0.10 (n = 2)	0.20 - 0.34	1.83 $\pm$ 0.40	1.55 - 2.12 (n = 2)	<b>Ice core</b>	n/a			n/a		
<b>FF</b>	24.03 (n = 1)		n/a		<b>FF</b>	n/a			n/a		

312 To understand how changes in physical oceanography may affect our distributions of both THg  
 313 and MeHg species, we averaged Hg concentrations by water mass in the upper 500 m, visualized  
 314 in Fig. 3 and summarized in Table S1. Significant differences (log transformed, Student's t-test, p  
 315 < 0.05) for both THg and MeHg concentrations in late winter and spring were observed for PW.  
 316 A significant difference in THg concentrations was also observed for mAW, which was mainly  
 317 observed in waters at station P1. No difference was observed for MeHg in mAW, although we  
 318 assumed this is mainly due to local MeHg concentration differences in surface mAW at station P1

319 and subsurface mAW at stations P6 and P7. No statistically significant differences were observed  
320 for THg nor MeHg concentrations in wPW, which comprised only subsurface and bottom waters  
321 at our stations (Van Engeland et al., 2023). As PW comprises most of our shelf stations and surface  
322 waters at P6 and P7, our observed changes in THg concentrations are best explained by external  
323 inputs to surface waters such as atmospheric deposition or ice melt instead of changes in water  
324 mass. In addition, our general transect (Fig. 2) and insignificant differences in wPW (Table 2) do  
325 not suggest major inputs of benthic THg nor MeHg during this time period. We also cannot rule  
326 out potential concentration differences in individual water parcels between sampling periods, but  
327 we infer that our observed THg profiles and differences in THg concentrations in spring reflect  
328 seasonal THg inputs to surface waters. Similarly, we hypothesize our lower MeHg concentrations  
329 in spring are driven by internal biogeochemical processes rather than external inputs and/or water  
330 masses.



332 **Figure 3:** THg (top) and MeHg (bottom) concentrations averaged for water masses in both late  
333 winter (LW) and spring (SP) in the northern Barents Sea in 2021. Water masses are Atlantic Water  
334 (AW), modified Atlantic Water (mAW), Polar Water (PW), and warm Polar Water (wPW). Box  
335 plots display median ( $\pm$  IQR) and individual data points for each water mass. For averages and  
336 sample sizes, see Table S1.

### 337 **3.3 Seasonal inputs of THg to surface waters**

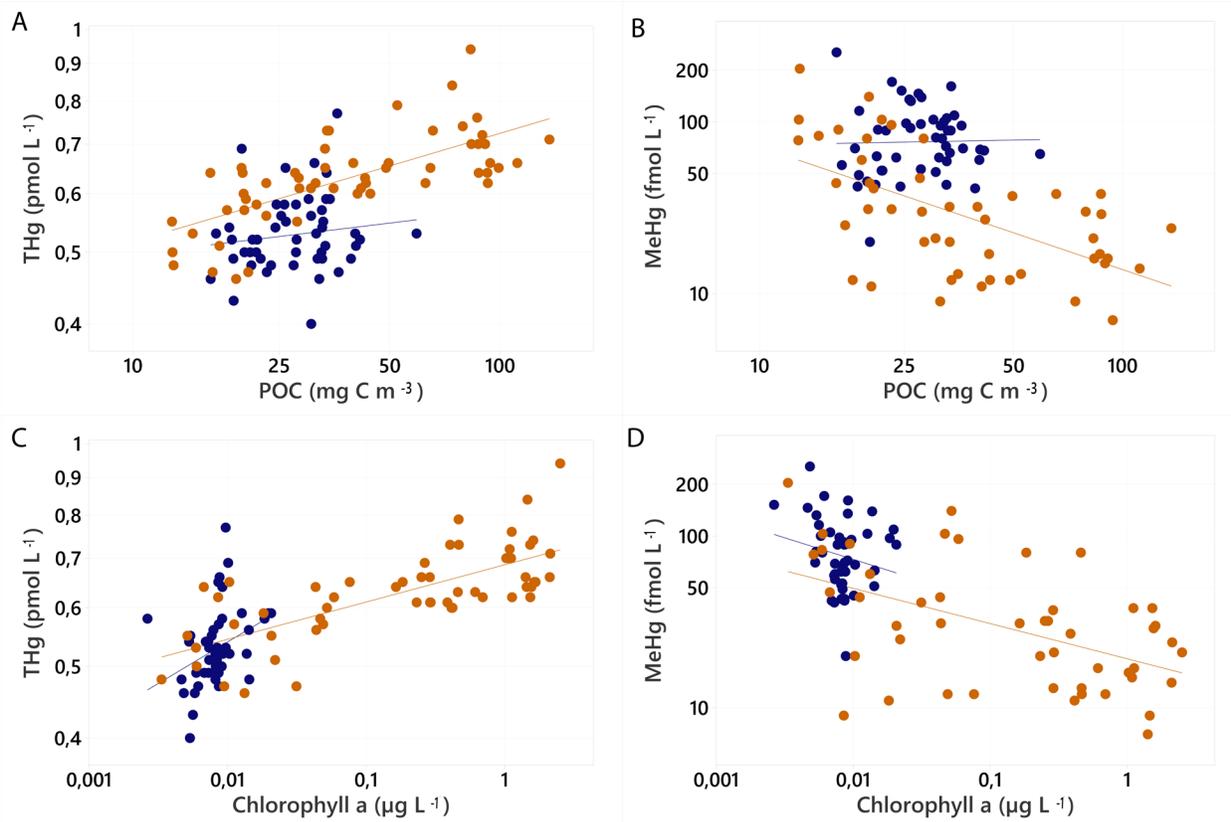
338 Elevated THg in surface waters in spring could suggest inputs from AMDEs or sea ice processes,  
339 where changing sea ice conditions facilitate atmospheric deposition to surface waters. Although  
340 much of the AMDE-derived Hg is photochemically re-emitted back to the atmosphere from the  
341 cryosphere during the summer (Dastoor et al., 2022), a temporary enrichment could exist during  
342 spring. Under ice water (UIW) taken at stations P4, P6 and P7, in late winter was  $0.58 \pm 0.02$  pmol  
343  $L^{-1}$  ( $n = 3$ ), similar to the remaining THg levels in the water column. However, the bottom 10 cm  
344 of ice cores taken at P4 and P7 had low THg concentrations, with values of only  $0.34$  pmol  $L^{-1}$  and  
345  $0.20$  pmol  $L^{-1}$ , respectively. As sea ice appeared to both melt and freeze in between our two  
346 sampling periods (SI Fig. 4), it is plausible that dynamic melt and freeze processes facilitated  
347 additional THg to surface waters. However, changes in integrated THg in seawater (upper 100 m)  
348 do not show a correlation with the average fraction of open water (SI Fig. 5). We suspect that the  
349 heterogeneity in sea ice and snow may deliver variable amounts of Hg to surface waters, as the sea  
350 ice covering our stations during the two seasonal cruises have different origins and drift patterns  
351 (Marquardt et al., 2023a). Previous studies in the Arctic found higher concentrations and a wide  
352 range of THg (range:  $0.089 - 60.8$  pmol  $L^{-1}$ ) in both first year ice and multiyear ice compared to  
353 seawater (Chaulk et al., 2011, Beattie et al., 2014, Schartup et al., 2020, Jonsson et al., 2022b).  
354 Although we measured low THg concentrations in the bottom sea ice in late winter, THg can vary  
355 by up to an order of magnitude throughout the core, with highest concentrations near the sea ice-  
356 atmosphere interface (Beattie et al., 2014). As sea ice drifts and snow and ice melts between cruise  
357 samplings (SI Fig. 4), these reservoirs could supply additional Hg to surface seawater (Huang et  
358 al., 2023). Furthermore, any leads in the ice pack during these changing ice conditions could also  
359 facilitate direct atmospheric deposition of AMDEs to surface seawater or deposit additional Hg to  
360 the cryosphere (Moore et al., 2014).

361 Refreezing also results in the rejection of brine between sampling periods (SI Fig. 4), with brine  
362 THg concentrations potentially up to 355 pmol L<sup>-1</sup> (Chaulk et al., 2011). Although we did not  
363 measure brine directly, a frost flower formed from brine expulsion in late winter had a  
364 concentration of 24 pmol L<sup>-1</sup> (n = 1). By spring, UIW concentrations were 1.00 ± 0.30 pmol L<sup>-1</sup> (n  
365 = 3), and higher than the average concentration for the remaining water column. Likewise, in sea  
366 ice, the THg concentration in the bottom 10 cm of ice cores taken at P4 and P7 were higher in  
367 spring, with values of 1.55 pmol L<sup>-1</sup> and 2.12 pmol L<sup>-1</sup>, respectively. Spring THg concentrations  
368 in the bottom of ice cores are higher compared to our late winter observations, which contrasts  
369 with a seasonal study in Antarctica (Nerentorp Mastromonaco et al., 2016). Local melt and freeze  
370 processes, downward brine movement, and varying sea ice advection are most likely responsible  
371 for the differences observed in the two studies. However, seasonal increases in THg in the  
372 Antarctic water column from winter to spring were suggested to be a result of both atmospheric  
373 and sea ice processes (Nerentorp Mastromonaco et al., 2017). Consequently, we conclude that our  
374 observed seasonal THg water column concentrations most likely reflect similar seasonal inputs  
375 and processes.

### 376 **3.4 Potential uptake of THg by the spring bloom**

377 Increased THg concentrations in surface waters and sea ice in spring could also be driven by the  
378 onset of primary productivity (SI Fig. 6). Development of ice algae during the spring bloom can  
379 take up THg from brine and seawater (Burt et al., 2013), explaining our increased THg  
380 concentrations in the bottom of sea ice. In the water column, Spearman's rank correlation analysis  
381 in spring indicated significant positive relationships between THg, particulate organic carbon  
382 (POC) and chlorophyll *a* (Table S3). Additionally, elevated chlorophyll *a*, pico-, nano-, and  
383 microphytoplankton in surface waters were observed in spring, indicating recent development of  
384 the phytoplankton spring bloom (SI Figs. 6 & 9). Log-log regression models between THg, POC,  
385 and chlorophyll *a* concentrations in the upper 500 m for all stations in spring (Fig. 4A, 4C) showed  
386 a positive and statistically significant relationship (F-test,  $p < 0.05$ ). Compared to late winter, this  
387 analysis suggests that THg is more associated with the particulate phase. If we assume POC as a  
388 proxy for total particulate matter and a constant partition coefficient  $K_d$  to calculate dissolved Hg  
389 (dHg) and particulate Hg (pHg) (SI Equations 3 and 4), this also indicates a greater percentage of  
390 pHg in spring when compared to late winter. A strong positive Spearman rank correlation also

391 exists between POC and chlorophyll *a* (Table S3,  $p < 0.05$ ), suggesting that the positive correlation  
392 of THg with both of these biological indicators may suggest adsorption and/or uptake of Hg species  
393 to primary producers during the spring bloom (Burt et al., 2014, Luengen and Flegal 2009).



394  
395 **Figure 4:** Log scale scatterplots with log-log regressions of THg (left) and MeHg (right)  
396 concentrations against POC (top) and chlorophyll *a* (bottom) concentrations in the upper 500 m  
397 for both late winter (blue) and spring (orange) in the northern Barents Sea in 2021.

398 However, it is important to note that higher THg concentrations in spring suggest that seasonal  
399 inputs increase both pHg and dHg concentrations, with a significant fraction of THg in the  
400 dissolved phase. While our THg relationships to biological parameters during the spring bloom  
401 suggests that THg is either taken up into primary producers by diffusion or adsorbed to cell  
402 surfaces, the remaining dHg in the water column can also be complexed by extracellular algal  
403 exudates (Cossart et al., 2022). In the dissolved phase, Hg shows strong affinity to DOM and  
404 especially to reduced sulfur ligands (Ravichandran 2004). Indeed, we saw increased relative  
405 intensities of dissolved organic sulfur compounds (CHOS) in surface waters in both sampling  
406 periods compared to deeper waters (SI Fig. 7), most likely released by phytoplankton. These

407 biologically driven processes, such as uptake by phytoplankton and organic matter complexation,  
408 could keep THg sourced from the atmosphere or sea ice in surface waters as well as facilitate  
409 transformations of Hg species.

### 410 **3.5 MeHg dynamics in late winter and spring**

411 Significant differences in MeHg concentrations from late winter to spring can be driven by several  
412 biogeochemical processes. As our spring MeHg concentrations showed a significant decline  
413 compared to late winter, the collective sinks of MeHg in the water column outweigh MeHg  
414 sources. Potential MeHg sinks to consider are photodemethylation, particle scavenging of MMHg,  
415 atmospheric evasion of DMHg, and dark demethylation processes. To systematically evaluate the  
416 contribution of each of these potential sinks, we used both literature and field data to calculate the  
417 percentage contribution to the observed difference in the upper 100 m. It is important to note that  
418 our observed differences in MeHg are net values, which can lead to high uncertainty when  
419 estimating the contribution from various sources and sinks.

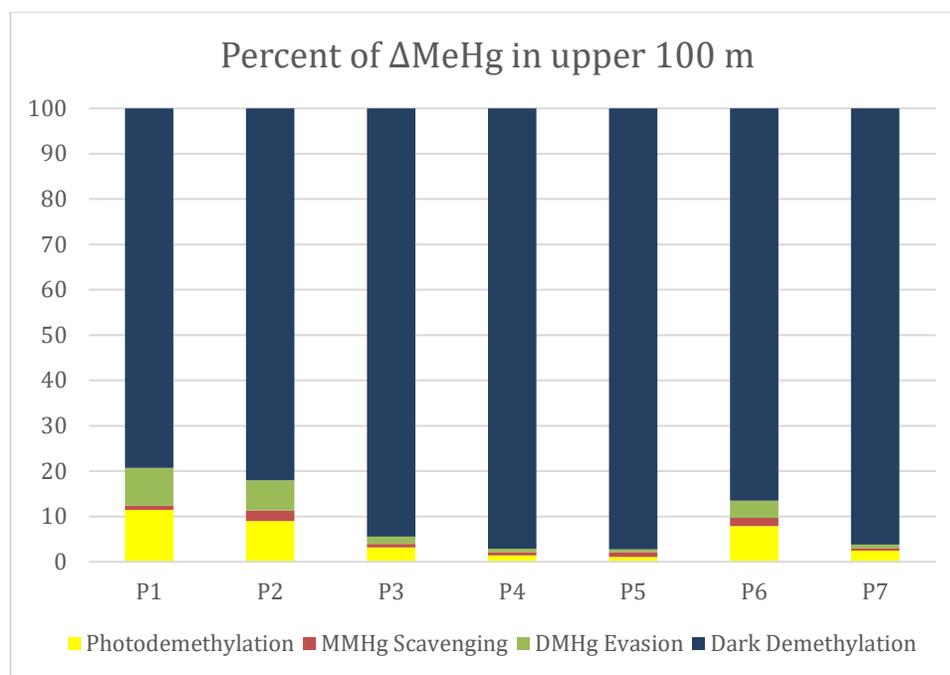
420 We integrated the water column for MeHg for each station to calculate the rate of change per day,  
421 called  $\Delta\text{MeHg}$  (Kohler et al., 2022a) for each station using the following equation:

$$422 \quad \Delta\text{MeHg} = \left( \int_0^z [\text{MeHg}]_{\text{SP}} dz - \int_0^z [\text{MeHg}]_{\text{LW}} dz \right) / dt$$

423 Here, we used the trapezoidal integration method to calculate from 0 to 100 m depth, where  $z =$   
424 depth, SP = spring, and LW = late winter, and  $t =$  time in days between sampling of each station.  
425 The total integrated pools are summarized in Table S4. All stations show a net decline, or loss, in  
426 the integrated pool from late winter to spring with an average of  $-81 \pm 36 \text{ pmol m}^{-2} \text{ d}^{-1}$  ( $n = 7$ ),  
427 demonstrating that sinks outweigh sources for observed change in MeHg concentrations.

428 Our measured declines of MeHg pools by spring could be due to increased photodemethylation  
429 during the transition to polar day. However, both lower water temperatures and the presence and  
430 thickness of snow and sea ice hinder MeHg photodemethylation (Point et al., 2009). Previous  
431 estimates in the Arctic Ocean suggest photodemethylation rates of  $0.04 \text{ d}^{-1}$  (Lehnher et al., 2011)  
432 and  $0.0082 \text{ d}^{-1}$  (Zhang et al., 2020) at the sea surface. Assuming first-order kinetics and using  
433  $\Delta\text{MeHg}$ , our calculated rate constant for our seven stations in the upper 100 m between late winter

434 to spring is an average MeHg loss of  $0.021 \pm 0.013 \text{ d}^{-1}$  ( $n = 7$ ). This loss, although a net loss and  
 435 not directly comparable, agrees well with the estimated photodemethylation rate constants;  
 436 however, the photodemethylation rate would be attenuated with depth and dependent on available  
 437 light. To estimate the percentage of the MeHg pool potentially degraded due to  
 438 photodemethylation at each station, we used photosynthetically active radiation (PAR)  
 439 approximated to  $15 \text{ E m}^{-2} \text{ d}^{-1}$  between the months of March and May 2021 (SI Fig. 8) and rate  
 440 constant from Arctic waters of  $0.001 \text{ m}^2 \text{ E}^{-1}$  when expressed to solar irradiance (Lehnherr et al.,  
 441 2011). After accounting for the fraction of open water (with a simple assumption of zero  
 442 transmittance through snow-covered sea ice) and attenuation of PAR with depth (Table S6), our  
 443 calculations suggest that photodemethylation can only account for  $5 \pm 4\%$  of our observed  $\Delta\text{MeHg}$   
 444 in the upper 100 m (Fig. 5). The highest percentage of photodemethylation (11%) is observed at  
 445 station P1 due to no sea ice coverage. This calculation carries a high uncertainty due to additional  
 446 variables we could not account for, such as the partial transmittance of light through sea ice,  
 447 enhanced or inhibited photodemethylation in the presence of DOM, and scattering and absorption  
 448 of light by both particles and DOM. Nevertheless, potential photodemethylation plays a relatively  
 449 small role compared to our observed changes in MeHg pools.



451 **Figure 5:** Percent contribution of selected processes influencing the change in MeHg pools,  
452  $\Delta$ MeHg, between late winter and spring. Photodemethylation is in yellow, MMHg particle  
453 scavenging in red, DMHg evasion in green, and dark demethylation (all other unaccounted for  
454 processes) in dark blue.

455 Particulate scavenging could also potentially serve to transport particle-bound MMHg (pMMHg)  
456 to deeper waters. As the total amount of sinking particles, including POC, are low in late winter  
457 waters due to the lack of biological productivity (Dybwad et al., 2022), we could assume that  
458 scavenging of pMMHg is negligible. Furthermore, a previous study has estimated that only 4% of  
459 pHg is pMMHg, which might be overestimated due to several samples below the detection limit  
460 (Agather et al., 2019). Additionally, observed POC fluxes across our transect in late winter were  
461 low at  $27 \pm 9 \text{ mg C m}^{-2} \text{ d}^{-1}$  (Bodur et al., 2023) with no flux attenuation, suggesting minimal  
462 vertical pHg transport. For comparison, POC fluxes in spring were  $178 \pm 199 \text{ mg C m}^{-2} \text{ d}^{-1}$  (Bodur  
463 et al., 2023). While we only measured MeHg and do not know exactly when primary production  
464 began to develop, we made several assumptions to attempt to determine the contribution of  
465 pMMHg scavenging to  $\Delta$ MeHg (Table S6), with an estimate of  $1 \pm 1\%$  of  $\Delta$ MeHg (Fig. 5).

466 In surface waters, shifts in ice coverage could lead to evasion of DMHg trapped below the ice and  
467 help explain lower MeHg in surface waters during spring. In UIW, MeHg concentrations were  $59$   
468  $\pm 11 \text{ fmol L}^{-1}$  ( $n = 3$ ) in late winter and  $36 \pm 16 \text{ fmol L}^{-1}$  ( $n = 3$ ) in spring. As our average UIW  
469 concentrations of MeHg in spring were lower than in late winter, it is possible DMHg has evaded  
470 to the atmosphere during melting and freezing, or DMHg has built up in the overlying sea ice  
471 (Schartup et al., 2020). Previous studies have emphasized the importance of the evasion of DMHg  
472 from Arctic waters (Baya et al., 2015, Soerensen et al., 2016) but recent studies of DMHg in sea  
473 ice and surface seawater (Agather et al., 2019, Jonsson et al., 2022) suggest a relatively modest  
474 evasion rate. Although we did not measure DMHg, we expect a large proportion of our surface  
475 MeHg concentrations to consist of MMHg, similar to other Arctic Ocean studies (Baya et al., 2015,  
476 Agather et al., 2019, Jonsson et al., 2022b). We attempted to quantify the amount of DMHg evasion  
477 during our transition to spring with additional limitations and assumptions (Table S6), with  
478 atmospheric evasion making up only  $3 \pm 3\%$  of  $\Delta$ MeHg (Fig. 5). As this estimate is a gross value  
479 and does not account for atmospheric re-deposition of MMHg, it may be overestimated.

480 Water mass distribution along our transect did not change drastically from late winter to spring  
481 (Van Engeland et al., 2023), suggesting relatively minimal importance of advection of lower  
482 concentration MeHg waters in the upper 100 m. However, station P6 and P7 do intercept the West  
483 Spitsbergen Current, with slight shifts from wPW to PW in spring (Van Engeland et al., 2023).  
484 MeHg concentrations in PW are lower than wPW in late winter (One-way ANOVA with Tukey's  
485 test,  $p < 0.05$ ), and could contribute to the observed  $\Delta\text{MeHg}$ . Nevertheless, PW is significantly  
486 different between the two cruises, so for the sake of simplicity we have assumed advection plays  
487 a negligible role in explaining our observed  $\Delta\text{MeHg}$ .

488 The importance of dark demethylation processes in unfiltered waters has been highlighted in  
489 previous studies using spiked stable Hg isotopes to measure demethylation rates (Lehnherr et al.,  
490 2011, Monperrus et al., 2007). In addition, the recent MeHg annual budget for the East Siberian  
491 Sea attributed approximately 50% of potential MeHg sinks to gross dark demethylation (Kim et  
492 al., 2020b). Consequently, we attribute the remaining  $\Delta\text{MeHg}$  observed ( $90 \pm 8\%$ , Fig. 5) as dark  
493 demethylation processes and hypothesize that they are mainly driven by biological activity.

### 494 **3.6 Biotic demethylation of MeHg**

495 As POC fluxes were low in late winter (Bodur et al., 2023), it is likely that there is no substantial  
496 in situ biotic Hg methylation occurring throughout the water column. Additionally, despite a higher  
497 concentration of THg by spring, both MeHg concentrations and % MeHg have decreased (Fig. 2),  
498 suggesting methylation processes were dominated by demethylation processes. However, the  
499 increase in phytoplankton abundance by orders of magnitude in surface waters along the transect  
500 in spring could support biotic transformations of Hg species (SI Fig. 6). Upon development of the  
501 phytoplankton bloom, we observed that diatoms, particularly species of the genus *Thalassiosira*,  
502 had the highest relative abundance in the upper 90 m at five out of seven stations along our transect  
503 (SI Fig. 9). Our regression analysis (Fig. 4) supports the hypothesis that THg is indeed potentially  
504 taken up by primary producers. Unlike THg, however, MeHg has significant negative relationships  
505 with both POC and chlorophyll *a* in spring (Fig. 4B, 4D, F-test,  $p < 0.05$ ), supporting declining  
506 MeHg concentrations in association with biological particles. As diatoms and other marine  
507 phytoplankton passively take up MMHg (Mason et al., 1996, Lee and Fisher 2016), we  
508 hypothesize that our observed MeHg losses in surface waters could be due to phytoplankton uptake

509 and subsequent demethylation. This hypothesis is further supported by relatively low contributions  
510 of the other MeHg sinks in surface waters (Fig. 5) and possible biological demethylation  
511 mechanisms identified in literature. Previous research has observed reduction of inorganic  $\text{Hg}^{\text{II}}$  to  
512  $\text{Hg}^0$  in the diatom *Thalassiosira weissflogii* (Morelli et al., 2009) while the green alga  
513 *Chlamydomonas reinhardtii* was demonstrated to demethylate MMHg (Bravo et al., 2014).  
514 Another study in a freshwater lake has shown that demethylation of intracellular MMHg correlated  
515 positively with diatom abundance (Cossart et al., 2021). Recently, a study suggested that  
516 phytoplankton can intracellularly demethylate MeHg through the production of reactive oxygen  
517 species (Liang et al., 2023).

518 This MMHg uptake during the onset of the Arctic spring bloom could potentially facilitate the  
519 seasonal bioaccumulation of MMHg by higher trophic level organisms throughout the summer. A  
520 previous study in an Arctic lake observed decreasing MeHg levels in water from winter to spring,  
521 yet saw a seasonal increase in MeHg concentrations in freshwater zooplankton (Varty et al., 2021).  
522 Furthermore, another winter to spring increase of MeHg concentrations in zooplankton was  
523 observed in waters of the Canadian Arctic Archipelago (Pućko et al., 2014). While other  
524 mechanisms like zooplankton grazing activity or direct methylation in the gut could explain their  
525 observed seasonal Hg biota concentrations (Pućko et al., 2014), we speculate that our seasonal  
526 changes of water column MeHg partly reflect a MMHg biomagnification pathway for higher  
527 trophic level organisms in the Arctic's productive spring and summer seasons.

528 Further studies on Hg methylation activity in specific marine phytoplankton species demonstrated  
529 that MeHg demethylation positively correlated with thiol concentrations in algal exudates (Li et  
530 al., 2022). We observed more elevated relative intensities of CHOS compounds in surface waters  
531 compared to deeper waters (SI Fig. 7). Dissolved CHOS compounds could facilitate the  
532 transformation of MMHg to DMHg via localized reduced thiol groups (Jonsson et al., 2016), with  
533 a proposed disproportionation reaction mechanism of two MMHg molecules to form DMHg and  
534  $\text{iHg}$ . Inorganic Hg could be reduced to  $\text{Hg}^0$  or be scavenged, while DMHg could evade to the  
535 atmosphere as previously highlighted. Complexation of MMHg to dissolved thiols in DOM could  
536 also reduce uptake of MMHg by phytoplankton (Lee and Fisher 2017) reducing MMHg  
537 bioavailability. MMHg-DOM complexes can also undergo enhanced photodemethylation (Qian et  
538 al., 2014) in surface waters, although the specific mechanisms remain debated (Barkay and Gu

539 2021). Additional Arctic seasonal studies are needed to characterize the types of DOM and its role  
540 in driving MeHg biogeochemical processes in the water column. Collectively though, our data and  
541 literature support biotically mediated MeHg demethylation during the development of the spring  
542 bloom.

543 Interestingly, deeper waters (100 to 500 m) also show a net loss of MeHg concentrations, with an  
544 average loss rate of  $0.015 \pm 0.011 \text{ d}^{-1}$  ( $n = 7$ ) in the 100 - 200 m depth interval. While potential  
545 phytoplankton demethylation could explain MeHg losses in surface waters, we expect different  
546 biotic processes to affect MeHg losses in subsurface and bottom waters. Recent research has  
547 demonstrated that the presence of the *merB* gene, found in certain bacteria and responsible for the  
548 demethylation of MeHg to  $\text{Hg}^{\text{II}}$ , is present in deep waters of the Arctic Ocean (Bowman et al.,  
549 2020). In bacteria, the *mer* operon, encoding for *merA* and *merB* genes, exists primarily as a  
550 detoxifying mechanism for high concentrations of Hg, in a process called reductive demethylation.  
551 Lineages of a particular bacterial strain genome containing *merA* and *merB*, with high resistance  
552 and strong degradation capacity of MeHg, are present in polar regions (Sanz-Sáez et al., 2022).  
553 On our transect, potential reductive demethylators (*Alteromonas*, *Marinobacter*, and *Oleibacter*)  
554 are present, but in low relative abundance during both sampling periods compared to the entire  
555 bacterial and archaeal community (SI Text). However, bottom waters at shelf stations had higher  
556 total abundances of bacteria and higher relative abundances of potential demethylators than surface  
557 waters in late winter and could support MeHg degradation during the winter-spring transition (SI  
558 Text). This includes methanotrophic bacteria and sulfate reducing bacteria, which can also  
559 participate in oxidative demethylation of MeHg under anoxic conditions (Barkay and Gu, 2021).

560 While dark abiotic demethylation could influence our losses of MeHg in deep waters, more  
561 research is needed to uncover these mechanisms. Dissolved CHOS compounds, seen in both  
562 surface and deeper waters in both late winter and spring (SI Fig. 6), could facilitate transformation  
563 and demethylation of MMHg as previously discussed. Additionally, resuspension of sediments  
564 may supply manganese oxide to bottom waters, resulting in both the demethylation and scavenging  
565 of MMHg and iHg (Vlassopolous et al., 2018, Murphy et al., 2021, Kohler et al., 2022a). However,  
566 due to the uncertainty of these potential abiotic processes, we hypothesize much of our MeHg in  
567 deeper waters is also biotically demethylated from late winter into spring.

### 568 **3.7 Hg in basin deep waters (> 500 m)**

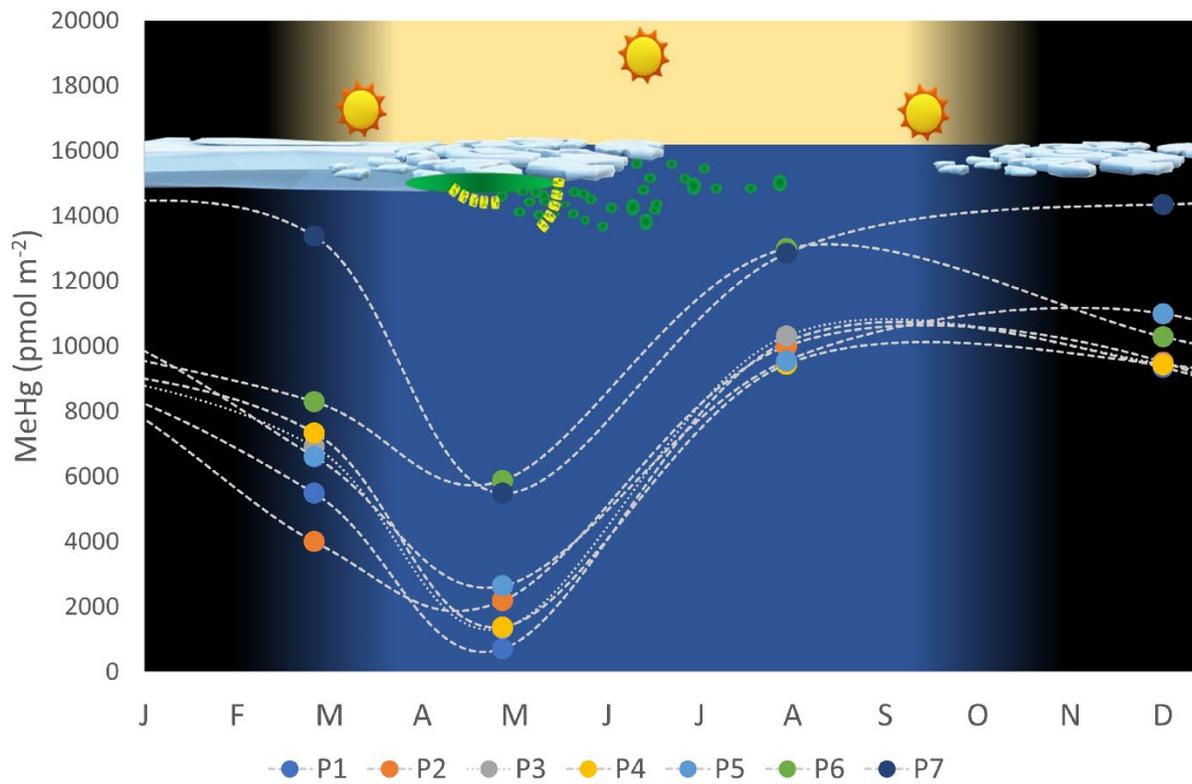
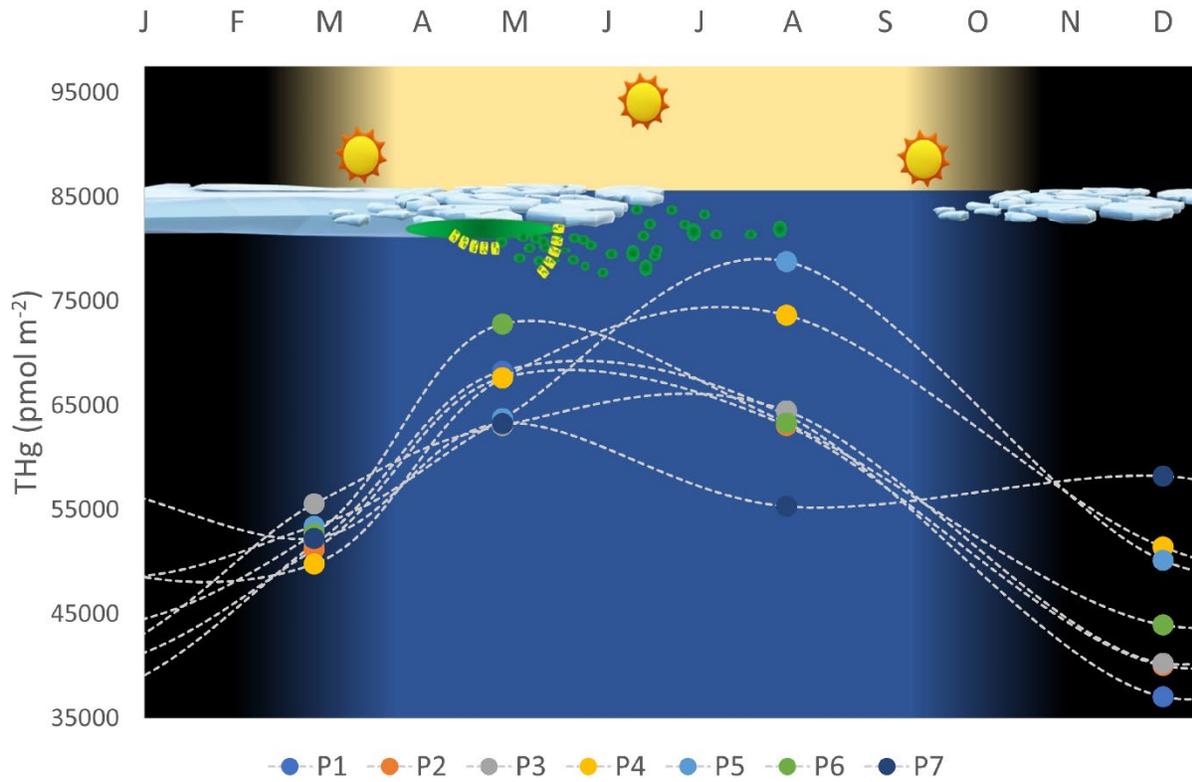
569 Bottom waters below 500 m at stations P6 and P7 are elevated in THg compared to the shelf  
570 stations (SI Fig. 1). These elevated concentrations in EBDW could contain additional Hg from  
571 resuspended sediment or sinking particles, as suggested in the Canadian Basin (Agather et al.,  
572 2019). As these basin stations are close to the shelf break, turbidity along the continental slope  
573 caused by the West Spitsbergen Current may resuspend and deliver Hg-rich particles into the basin.  
574 Throughout the Arctic winter (December to March), overturning on the Barents Sea shelf generates  
575 a nepheloid layer of suspended sediment which can sink as dense water plumes into the basin  
576 (Rudels et al., 2004, Gardner et al., 2022, Rogge et al., 2022). High concentrations of Hg and other  
577 trace elements in surface sediments in the Eurasian Basin have suggested hydrodynamic transport  
578 and scavenging of particles sourced from the shelf (Kohler et al., 2022b). Beam transmittance data  
579 (SI Fig. 10), however does not support significant resuspension of sediments in basin waters. For  
580 MeHg, stations P6 and P7 show similar patterns in late winter and spring, with the highest MeHg  
581 concentrations observed at 500 m and below. In EBDW, MeHg concentrations remain elevated  
582 above  $170 \text{ fmol L}^{-1}$  at P7 until 2500 m depth. Compared to the shelf, EBDW and deeper waters  
583 remain enriched in MeHg. These observations are in line with previous measurements in the Fram  
584 Strait with maximum MeHg between 400 – 1000 m (Petrova et al., 2020). Diffusion of porewater-  
585 derived MeHg could help explain elevated MeHg concentrations along the continental slope by  
586 station P6 (Kim et al., 2020b). Although both MeHg and THg concentrations in the deep basin  
587 agree with previous literature (Wang et al., 2012, Heimbürger et al., 2015), our sampling with  
588 standard Niskin bottles lends a degree of caution.

### 589 **3.8 Seasonal cycle of Hg in the northern Barents Sea**

590 Our observations for both THg and MeHg suggest a distinct seasonality of Hg in the northern  
591 Barents Sea marginal ice zone from late winter to spring. Based on our analysis, we conclude that  
592 the main drivers of both THg and MeHg changes in surface waters are ultimately driven by  
593 seasonal Hg inputs and the biotically driven processes of the phytoplankton spring bloom. In  
594 deeper waters, biotic demethylation is likely to play a more important role than abiotic  
595 demethylation mechanisms. A previous study suggested iHg seasonality in this region during the  
596 transition from summer to winter, with no significant change in MeHg concentrations (Kohler et  
597 al., 2022a). By assuming Hg follows a distinct seasonal cycle in the Arctic Ocean every year, we

598 combined our observations from late winter and spring 2021 with that of summer and early winter  
599 2019 (Kohler et al., 2022c,d) to illustrate a simple seasonal cycle of Hg over the course of a year  
600 in the northern Barents Sea surface waters (Fig. 6). To best illustrate the seasonal dynamics of THg  
601 and MeHg, we integrated our results from the surface to 100 m depth for each station (Tables S4  
602 & S5), where most biological productivity and Hg input occurs.

# Seasonal Cycle of Hg in the Northern Barents Sea

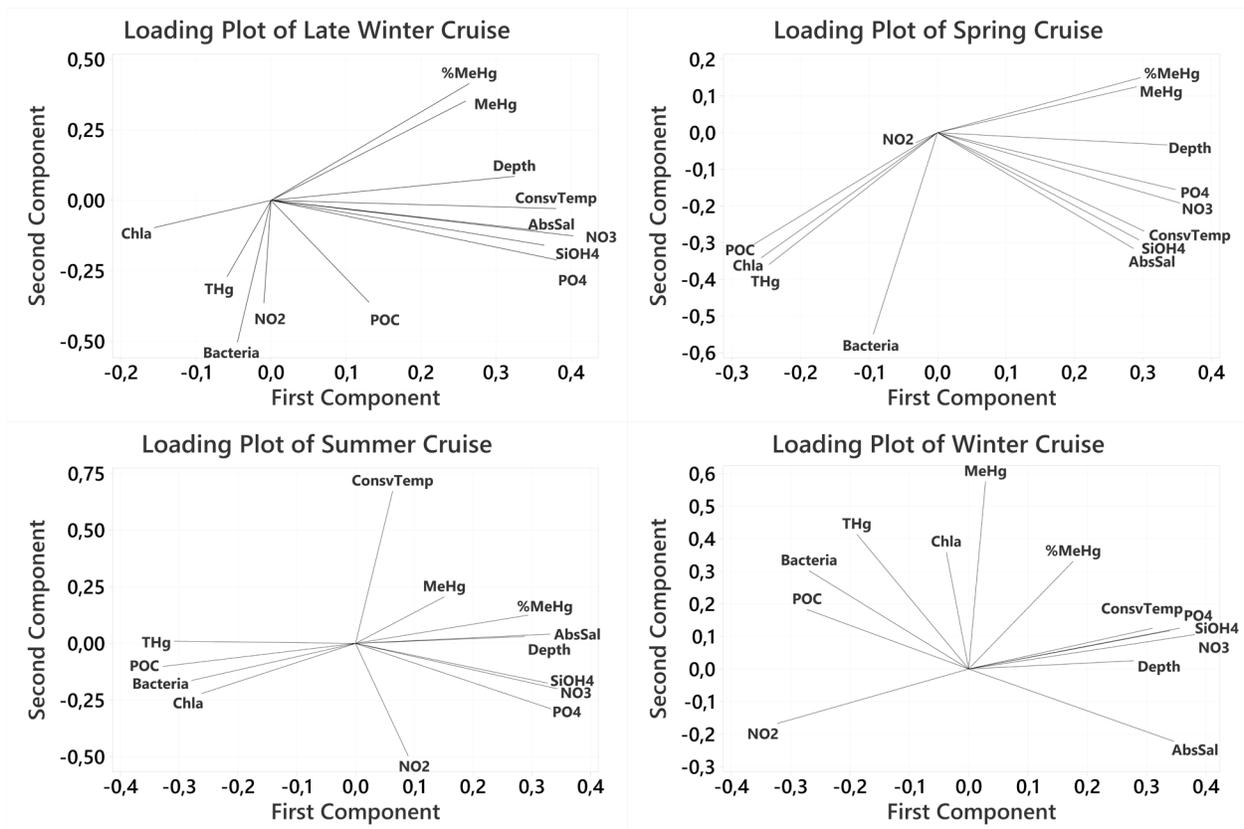


604 **Figure 6:** Integrated water column (0 to 100 m) observations of THg (top) and MeHg (bottom)  
605 from seasonal cruises (this study) and a previous study (Kohler et al., 2022a) in the northern  
606 Barents Sea. Graphs are superimposed on a simplified Arctic seasonal cycle in the marginal ice  
607 zone with polar day occurring between April and August and polar night beginning November  
608 until February. Green particles represent the development of the ice algal and phytoplankton spring  
609 blooms. Integrated pools for THg and MeHg are found in Tables S4 and S5. The x-axis is in  
610 months. Note the difference in scales on the y-axis.

611 Seasonal patterns of both THg and MeHg emerge and appear to be driven by the development of  
612 biological activity. The THg pools are higher during the polar day, while MeHg pools are at the  
613 minimum at the onset of the phytoplankton bloom and maximum in the second half of the year.  
614 As hypothesized, seasonal inputs and biological activity increase THg concentrations in surface  
615 waters. To better assess how relevant environmental drivers impact seasonal THg and MeHg  
616 concentrations, a PCA was conducted for each seasonal cruise (Fig. 7), with the first two principal  
617 components explaining > 60% of the variance for each cruise (Table S7). Clustering of THg with  
618 biological parameters POC and chlorophyll *a* in spring and summer suggest seasonal changes in  
619 THg concentrations are driven by biological activity and its partitioning to the particulate phase.  
620 Notably, this includes potential uptake of Hg species, transformation to MeHg during  
621 remineralization of POC, and downward transport with sinking particles. Sinking POC supports  
622 the production of subsurface in situ MeHg throughout polar day (Heimbürger et al., 2015),  
623 reflected in increasing MeHg pools progressing into summer (Fig. 6) and the general trend of  
624 positive MeHg loadings on the first component with depth, nutrients, and salinity for the spring  
625 and summer cruises. Late summer POC fluxes (Bodur et al., 2023), and increased proportions of  
626 manganese oxides leading into the polar night (Xiang and Lam 2020) lower THg pools by early  
627 winter (Kohler et al., 2022a).

628 Less clear is the behavior of both THg and MeHg throughout the polar night into late winter. It  
629 was proposed that potential scavenging by manganese oxides could be a driver of THg during  
630 Arctic winter (Kohler et al., 2022a), although a negative trend in THg pools from early winter to  
631 late winter is not observed (Fig. 6). We speculate that changes in Hg speciation, such as increased  
632 proportions of gaseous  $\text{Hg}^0$  and DMHg could protect against scavenging losses. A recent  
633 experimental study argued that gross dark reduction rates of  $\text{Hg}^{\text{II}}$  to  $\text{Hg}^0$  are significant in both

634 photic and aphotic zones, with dark reduction most likely dominant in the Arctic except during  
 635 polar day (Lamborg et al., 2021). Reduction could be driven by bacteria with the *merA* gene, and  
 636 potentially include bacteria with both *merA* and *merB*, facilitating declining MeHg pools  
 637 throughout the polar night (Fig. 6). The bacterial community in this region shifts from carbon  
 638 cycling in summer to nitrogen cycling in winter (Thiele et al., 2023), suggesting a possible link  
 639 between MeHg and the nitrogen cycle (Despins et al., 2023) during the polar night. In our PCA  
 640 analysis, we observed that MeHg and nitrite (NO<sub>2</sub>) exhibited opposite loadings on PC2 for all  
 641 cruises (Fig. 7). This observation could link nitrite-oxidizing *Nitrospina*, known to harbor and  
 642 express *hgcAB*-like proteins for Hg methylation (Gionfriddo et al., 2016, Villar et al., 2020), as a  
 643 driver for in situ seasonal MeHg concentrations in the Arctic. Our in situ concentration data  
 644 however, show a net loss of the MeHg pool from winter to late winter (Fig. 6). As our studies  
 645 comprise two different sampling years (2019 vs. 2021), more seasonal and interannual studies are  
 646 needed to fully understand the drivers of MeHg concentrations in the Arctic water column.



647

648 **Figure 7:** Principal component analysis (PCA) loadings plot for each seasonal cruise in the  
 649 northern Barents Sea from this study and previous cruises where Hg observations are available (<

650 500 m only) (Kohler et al., 2022a). Rotation is orthogonal with the first two principal components  
651 plotted. Selected environmental parameters included depth, conservative temperature  
652 (ConsvTemp), absolute salinity (AbsSal), bacterial abundance (bacteria), chlorophyll *a* (Chla),  
653 particulate organic carbon (POC), nitrate (NO<sub>3</sub>), nitrite (NO<sub>2</sub>), phosphate (PO<sub>4</sub>), and silicate  
654 (SiOH<sub>4</sub>), THg, MeHg, and %MeHg.

655 To the best of our knowledge, only one model has attempted to represent seasonal cycling of Hg  
656 in the Arctic Ocean (Fisher et al., 2012). In this model, sources and sinks of Hg are categorized by  
657 month, with subsurface waters resupplying Hg to surface waters through vertical mixing.  
658 However, the latest Arctic budget highlights that export of Hg to deeper waters is most likely  
659 underestimated (Dastoor et al., 2022), further supporting our hypothesis that seasonal changes in  
660 Hg may be driven by spring Hg inputs and changes in the particulate phase. Our new seasonal data  
661 help fill in this knowledge gap but continue to remain a “seasonal snapshot” from both a seasonal  
662 and interannual perspective. Recent studies on seasonality in this region highlight large seasonal  
663 and interannual variability in the northern Barents Sea regarding water masses and sea ice  
664 dynamics (Jones et al., 2023, Kohlbach et al., 2023, Van Engeland et al., 2023). Specifically,  
665 interannual variability may exert a greater impact on perceived seasonality, as our four cruises  
666 were sampled in two different years (2019 vs. 2021). As a result, our interpretations between the  
667 spring to summer transition and the early winter to late winter transition are subject to both  
668 seasonal and interannual variability, and thus more speculation. Nevertheless, we propose that our  
669 seasonal cycle of Hg in the northern Barents Sea is supported by our observational data and is  
670 reflected in current literature on Arctic Hg cycling.

## 671 **4 Conclusion**

672 We show that both THg and MeHg concentrations in the water column of the northern Barents  
673 Sea exhibit dynamic changes throughout the year. Variations in THg and MeHg are driven by  
674 seasonal processes during the early spring, such as Hg input to surface waters and biological  
675 activity. Notably, we observed a significant decline in MeHg concentrations during the  
676 development of the phytoplankton spring bloom. We suggest biotic demethylation mediated by  
677 phytoplankton and bacteria as the main mechanism underlying our observations. This  
678 demethylation by phytoplankton and bacteria during the spring bloom may mitigate the

679 bioaccumulation of Hg up the food web. Although it is difficult to completely disentangle these  
680 losses and transformations of MeHg, our data suggests that potential seasonal uptake of MeHg  
681 during the spring bloom represents an important entry point of Hg into the Arctic marine  
682 ecosystem.

683 Using our current data, we constructed a simplified seasonal Hg cycle in the northern Barents Sea  
684 marginal ice zone and conducted additional analysis to try and identify major environmental  
685 parameters driving both THg and MeHg concentrations. THg was clustered with chlorophyll *a* and  
686 POC in spring and summer, suggesting seasonal changes of THg are modulated by changes in  
687 primary productivity, particles, and food web processes. MeHg concentrations also exhibited a  
688 seasonal pattern, with potential uptake and demethylation during spring, followed by hypothesized  
689 in situ production throughout the summer. Our simple seasonal cycle also encompasses potential  
690 interannual variability, which may underestimate additional drivers of Hg concentrations such as  
691 sea ice coverage and changing water masses. In addition, the northern Barents Sea has limited  
692 riverine inputs, highly variable sea ice coverage and a unique water mass distribution compared to  
693 the other Arctic shelf seas. Thus, the applicability of our proposed seasonal cycle may have limited  
694 use in other Arctic regions, like the river-influenced Siberian shelf seas or less biologically  
695 productive Central Arctic Ocean. Nevertheless, our study represents an important first step in  
696 identifying Hg seasonal processes and drivers in Arctic waters, with additional seasonal  
697 environmental data from the Nansen Legacy project available for use in budgets and models. We  
698 suggest future Arctic Hg budgets and coupled ocean-atmosphere/earth system models include an  
699 element of seasonality.

700 The projected climate driven inputs of increasing amounts of Hg from melting glaciers and  
701 thawing permafrost to Arctic shelf seas during the onset of the productive season may facilitate  
702 greater uptake of MMHg in the marine food web (Chételat et al., 2022). As ongoing climate change  
703 continues to transform the Arctic region, more research is needed to elucidate how region-specific  
704 seasonal processes affect Hg distributions in the Arctic Ocean and the surrounding shelf seas.  
705 Specifically, we suggest more studies examining the connection between demethylation and  
706 biological uptake in the marginal ice zones as well as seasonal measurements of gaseous Hg  
707 species. We suspect that these transformation processes and rates that control MeHg distributions  
708 in the water column, and subsequently uptake in biota, follow a seasonal pattern.

709 **CRedit authorship contribution statement**

710 **Stephen G. Kohler:** Conceptualization, methodology, validation, formal analysis, investigation,  
711 writing - original draft, visualization, funding acquisition. **Lars-Eric Heimbürger-Boavida:**  
712 conceptualization, methodology, validation, investigation, writing - review and editing,  
713 visualization, supervision. **Philipp Assmy:** investigation, formal analysis, writing - review and  
714 editing, visualization. **Oliver Müller:** investigation, formal analysis, writing - review and editing,  
715 visualization. **Stefan Thiele:** investigation, formal analysis, writing - review and editing. **Maria**  
716 **G. Digernes:** Investigation, formal analysis, visualization. **Kuria Ndungu:** writing - review and  
717 editing, supervision. **Murat V. Ardelan:** Conceptualization, methodology, writing - review and  
718 editing, funding acquisition, supervision.

719

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730

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## Supplementary Material

### 1141 **Biotic transformation of methylmercury at the onset of the Arctic spring bloom**

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### 1154 **Integration of THg and MeHg**

1155 We used trapezoidal integration for the upper 100 m in the water column for both THg and MeHg  
1156 concentrations (Kohler et al., 2022). We assumed that the first available sampling depth (10 m or  
1157 UIW depending on station) represented the sea ice-ocean interface (0 m) or the atmosphere-ocean  
1158 interface (0 m). For 100 - 200 m integration, stations P2 and P5 were integrated to bottom sampling  
1159 depths less than 200 m. Stations were linearly interpolated to 100 m using the closest available  
1160 sampling depths. The exact sampling dates and days between for each station were used to  
1161 calculate the rate constants and  $\Delta\text{MeHg}$  values. Where MeHg samples were less than the LOD or  
1162 LOQ, the LOD ( $2 \text{ fmol L}^{-1}$ ) and LOQ ( $7 \text{ fmol L}^{-1}$ ) were used as the sample depth's concentration  
1163 for integration, respectively. As most samples are run in singles, uncertainty in  $\Delta$  ( $\pm 1\text{SD}$ ) for late  
1164 winter and spring  $\Delta\text{MeHg}$  and  $\Delta\text{THg}$  was calculated using the absolute THg reproducibility  
1165 reported ( $\pm 0.05 \text{ pmol L}^{-1}$ ) and absolute MeHg reproducibility reported ( $\pm 7 \text{ fmol L}^{-1}$ ) for all depths  
1166 and propagated using the sum of squares method for integration, and reported as  $\pm 1 \text{ SD}$ .

### 1167 **Depth matching for environmental parameters**

1168 THg and MeHg sampling geographic coordinates and depths differ slightly from environmental  
1169 parameter sampling geographic coordinates due to ship drift during multiple samplings. Station

1170 samplings were assumed to be identical despite multiple sampling CTD casts and deviance from  
1171 exact coordinates. For Spearman rank analysis, regression analysis, and PCA, environmental data  
1172 were linearly interpolated to sampling depths of THg and MeHg for each cruise using the closest  
1173 sampling depths when necessary. Otherwise, matching depths were assumed to be from the same  
1174 water parcel for data analysis.

## 1175 **Percentage of $\Delta$ MeHg calculations**

### 1176 *Photodemethylation*

1177 Delta photodemethylation (Table S6,  $\Delta$ MeHg<sub>pd</sub>, pmol m<sup>-2</sup> d<sup>-1</sup>) was estimated using a similar  
1178 equation (1) as Kim et al., 2020. MeHg concentrations ( $C_{MeHg}$ , pmol L<sup>-1</sup>) at each station were  
1179 linearly interpolated for each depth in the surface 100 m in late winter using our observed  
1180 concentrations at discrete depths. The photodemethylation rate was estimated based on satellite  
1181 PAR ( $I_0$ , 15 E m<sup>-2</sup> d<sup>-1</sup>) in the region in April 2021 multiplied by the photodemethylation rate  
1182 constant ( $k_{pd}$ , 0.001 m<sup>2</sup> E<sup>-1</sup>) (Lehnherr et al., 2011). The  $k_{pd}$  was assumed constant over 100 m,  
1183 while PAR ( $I$ ) was attenuated by depth ( $z$ ) using equation (2) found in Kim et al., 2020, with  
1184 original reference Paulson and Simpson 1977, type I (clear water). We accounted for the sea ice  
1185 coverage by multiplying by the average fraction of open water ( $f_{ow}$ ) at each station in the previous  
1186 60 days from sampling in May, assuming zero transmittance through snow-covered sea ice.

$$1187 \quad \Delta MeHg_{pd} = \int_0^{100} C_{MeHg} * (f_{ow} * PAR * k_{pd}) \quad (1)$$

$$1188 \quad \frac{I}{I_0} = R e^{\frac{z}{\xi_1}} + (1 - R) e^{\frac{z}{\xi_2}} \quad (2)$$

### 1189 *Scavenging of MMHg*

1190 Delta MMHg scavenging (Table S5,  $\Delta$ MeHg<sub>scav</sub>, pmol m<sup>-2</sup> d<sup>-1</sup>) was estimated using several  
1191 assumptions. First we calculated pHg in the upper 100 m using our THg concentrations at each  
1192 discrete depth in late winter. To do this, we used in situ POC concentrations (Marquardt 2022a,  
1193 2022b) at each sampling depth as a proxy for total particulate matter and  $K_d$  for Hg in the Arctic  
1194 shelf ( $1.38 \cdot 10^6$  L kg<sup>-1</sup>, Tesán Onrubia et al., 2020) to calculate dHg (3) and pHg (4). We also  
1195 assumed that  $K_d$  does not change seasonally nor with depth. Then, we assumed that pMMHg makes

1196 up approximately 4% of pHg (Agather et al., 2019). We used our estimated pMMHg and POC  
 1197 concentrations to estimate a pMMHg/TPM ratio in the upper 100 m, by assuming that POC in late  
 1198 winter makes up approximately 25% of TPM (Dybwad et al., 2022). We used the average late  
 1199 winter POC fluxes reported in Bodur et al., 2023 using a short term sediment trap method (27 mg  
 1200 C m<sup>-2</sup> d<sup>-1</sup>) and assumed it represented 25% TPM flux of 108 mg m<sup>-2</sup> d<sup>-1</sup>, which is in agreement  
 1201 with previous TPM fluxes in the area determined by long term moorings (Dybwad et al., 2022).  
 1202 As POC fluxes were not attenuated with depth in late winter (Bodur et al., 2023), we multiplied  
 1203 our average pMMHg/TPM ratio by our estimated TPM flux to calculate a pMMHg flux at 100 m  
 1204 depth (5), for an average of 0.76 ± 0.06 pmol m<sup>-2</sup> d<sup>-1</sup> for ΔMeHg<sub>scav</sub>.

$$1205 \quad dHg = \frac{THg}{((K_d * POC) + 1)} \quad (3)$$

$$1206 \quad pHg = THg - dHg \quad (4)$$

$$1207 \quad \Delta MeHg_{scav} = pMMHg_{TPM} * TPM \text{ flux} \quad (5)$$

#### 1208 *Evasion of DMHg*

1209 We followed a similar approach to Kim et al., 2020 (and references therein) for calculating delta  
 1210 DMHg evasion (Table S6, ΔMeHg<sub>ev</sub>) (6). This included using estimated windspeeds from  
 1211 Longyearbyen airport in March and April 2021 (~ 8 m s<sup>-1</sup>), surface water temperatures (~ 10 m  
 1212 depth), at each station in late winter, temperature dependent Schmidt numbers for CO<sub>2</sub> and DMHg  
 1213 to calculate the DMHg transfer coefficient (K<sub>w</sub>), and temperature dependent Henry's law constant  
 1214 for DMHg (H). We estimated the DMHg (C<sub>DMHg water</sub>) concentration in surface waters using the %  
 1215 DMHg as MeHg as reported by Jonsson et al., 2022 (~ 6% in primary mixed layer) at shallowest  
 1216 sample depth for MeHg in late winter, and a literature value of 0.023 pmol m<sup>-3</sup> for DMHg in air  
 1217 (C<sub>DMHg air</sub>, Baya et al., 2015). We multiplied our estimated DMHg evasion flux by the average  
 1218 fraction of open water (f<sub>ow</sub>) in the previous 60 days at each station from sampling in May,  
 1219 assuming sea ice completely restricts atmospheric evasion (DiMento et al., 2019).

$$1220 \quad \Delta MeHg_{ev} = K_w * [C_{DMHg water} - \left(\frac{C_{DMHg air}}{H}\right)] * f_{ow} \quad (6)$$

1221

## 1222 *Dark demethylation*

1223 We calculated percentage dark methylation ( $\Delta\text{MeHg}_{\text{dark}}$ ) as the proportion of  $\Delta\text{MeHg}$  unaccounted  
1224 for from the sum of  $\Delta\text{MeHg}_{\text{pd}}$ ,  $\Delta\text{MeHg}_{\text{scav}}$  and  $\Delta\text{MeHg}_{\text{ev}}$  for each station.

## 1225 **Flow cytometry (FCM) Analysis**

1226 To measure the abundance of microorganisms (prokaryotes [bacteria and archaea], pico- and  
1227 nanophytoplankton), four replicate water samples (1.8 mL) were taken, fixed with glutaraldehyde  
1228 (0.5 % final concentration) at 4 °C for a minimum of 2 h, flash-frozen in liquid nitrogen and stored  
1229 at -80 °C until analysis. Samples for the analysis of prokaryote abundance were first diluted 10  
1230 times with 0.2- $\mu\text{m}$ -filtered TE buffer (Tris 10 mM and EDTA 1 mM, pH 8), then stained with a  
1231 green fluorescent nucleic acid dye (SYBR Green I; Molecular Probes, Eugene, OR, USA) and  
1232 incubated for 10 min at 80 °C and measured on a FACS Calibur (Becton Dickinson, Oxford, UK)  
1233 flow cytometer (Marie et al., 1999). Samples were counted at a low flow rate of around 60  $\mu\text{L}$   
1234  $\text{min}^{-1}$  and groups discriminated on a biparametric plot of green fluorescence (BL1) vs. side scatter  
1235 (SSC). Pico- and nanosized phytoplankton abundance was measured using an Attune® Acoustic  
1236 Focusing Flow cytometer (Applied Biosystems by Thermo Fisher Scientific, Waltham, MA, USA)  
1237 and the different phytoplankton groups were discriminated based on their red fluorescence (BL3)  
1238 vs. orange fluorescence (BL2) (Paulsen et al., 2016). The data for all cruises can be found at the  
1239 Norwegian Marine Data Centre (Müller et al., 2023a, 2023b, 2023c, 2023d).

## 1240 **Bacterial Community Analysis**

1241 We investigated the bacterial and archaeal community with respect to potential demethylators  
1242 (Christakis et al., 2018), methanotrophs, and sulfate reducing bacteria (SRB) using 16S rRNA gene  
1243 amplicon sequences collected on our transect to help explain the MeHg concentrations at different  
1244 depths. Using PICRUST2 with standard parameters in bioconda, the metabolic potential of the  
1245 different amplicon sequence variants was inferred based on their location in a phylogenetic tree of  
1246 fully sequenced organisms and the genomic assets of the closest relative in this tree (Grüning et  
1247 al., 2018, Douglas et al., 2020). Using KEGG-numbers, the marker genes *merA* and *merB*  
1248 (reductive demethylation), *dsrA* (sulfate reduction), and *mmoX* (methanotrophy) were extracted  
1249 and used to identify SRB, methanogens, and methanotrophs. For *merA* and *merB*, only *Oleibacter*

1250 was identified from the samples and was complemented with manual searches for *Alteromonas*  
1251 and *Marinobacter* (Sanz-Sáez et al., 2022). Similarly, no *mmoX* genes were predicted and known  
1252 methanotrophs were extracted based on recent literature (Guerrero-Cruz et al., 2021). SRB were  
1253 well covered by the PICRUSt2 results. In the water column, the relative abundance of potential  
1254 reductive demethylators and SRB was highest in the deepest sample of station P7 with 1.9% and  
1255 0.7% (potential reductive demethylators) and 4.8% and 3.9% (SRB) in late winter and spring.  
1256 Potential reductive demethylators were otherwise low in relative abundance, rarely exceeding  
1257 0.1% of the total bacterial and archaeal community. Higher abundance was found in most samples  
1258 at 10 m and below 200 m depth as compared to the samples in between, regardless of bottom depth  
1259 for both seasons, although on average the abundance was higher in late winter (~0.1%) than in  
1260 spring (~0.06%). SRB were similarly abundant in both seasons with 0.39% in late winter and  
1261 0.37% in spring but were lower in surface samples (10 and 20 m) as compared to deeper samples.  
1262 Methanotrophs were also low in abundance with ~0.15% both in late winter and spring. The  
1263 highest abundance was found in both seasons at P1 (near bottom depth) with 1.3% in spring as  
1264 compared to 0.7% in late winter. Generally, the abundance was higher in samples > 20 m.

#### 1265 **Dissolved Organic Matter (DOM) Analysis**

1266 DOM seawater and under ice water samples were collected at stations P1, P4, P6, and P7 on both  
1267 late winter and spring sampling expeditions. Water samples were immediately extracted after  
1268 collection using solid phase extraction (Dittmar et al., 2008) and stored at -20°C until analysis.  
1269 Extracts were analyzed by LTQ-Velos-Pro Orbitrap mass spectrometer (Thermo Scientific,  
1270 Germany) with an electrospray ionization source (ESI) in negative mode. Each sample spectrum  
1271 was calibrated internally in mass range 200-800 m/z with data resolution of 100 000. Additional  
1272 detailed instrument parameters are noted in Hawkes et al., 2016. Dissolved organic sulfur  
1273 compounds (CHOS) formulas are determined based on the following elemental criteria: C<sub>4-50</sub> H<sub>4-</sub>  
1274 <sub>100</sub>O<sub>2-40</sub>N<sub>0-2</sub>S<sub>0-1</sub>, hydrogen to carbon ratio (H/C) = 0.3 - 2.2, oxygen to carbon ratio (O/C) = 0-1,  
1275 double bond equivalent-oxygen (DBE-O) = -10 - 10, valence electron as even number, mass to  
1276 charge ratio (m/z) = 150-800 and mass defect (mass after nominal mass) = -0.1 - 0.3.

1277

1278 **References**

1279

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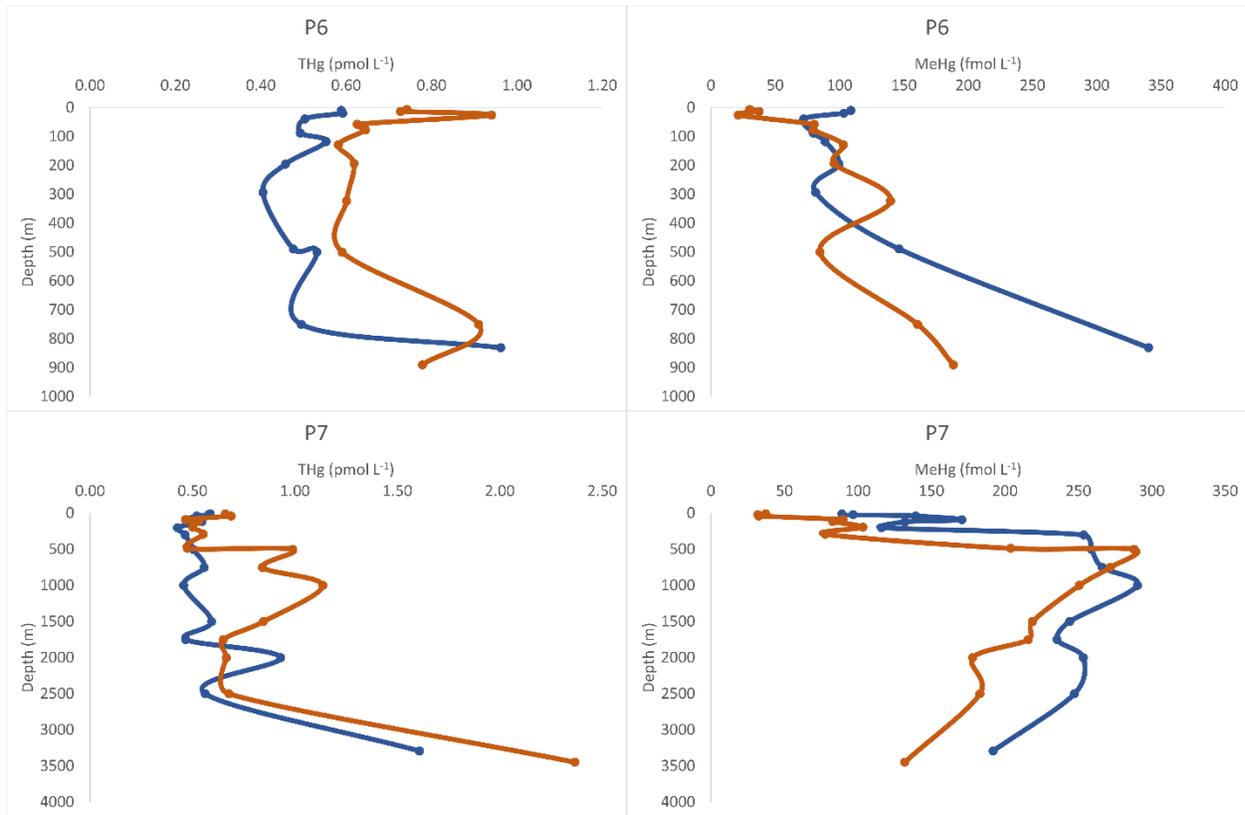
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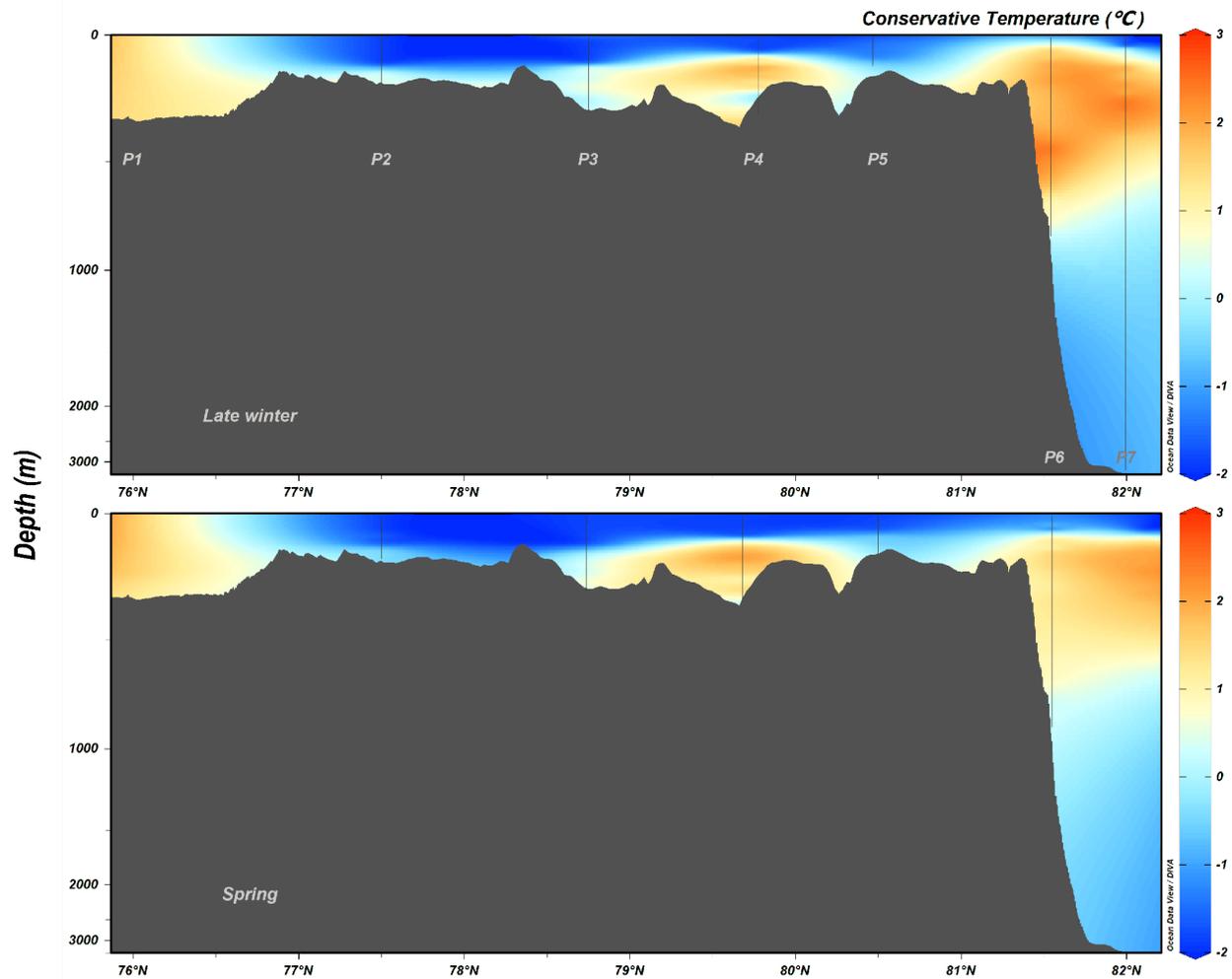
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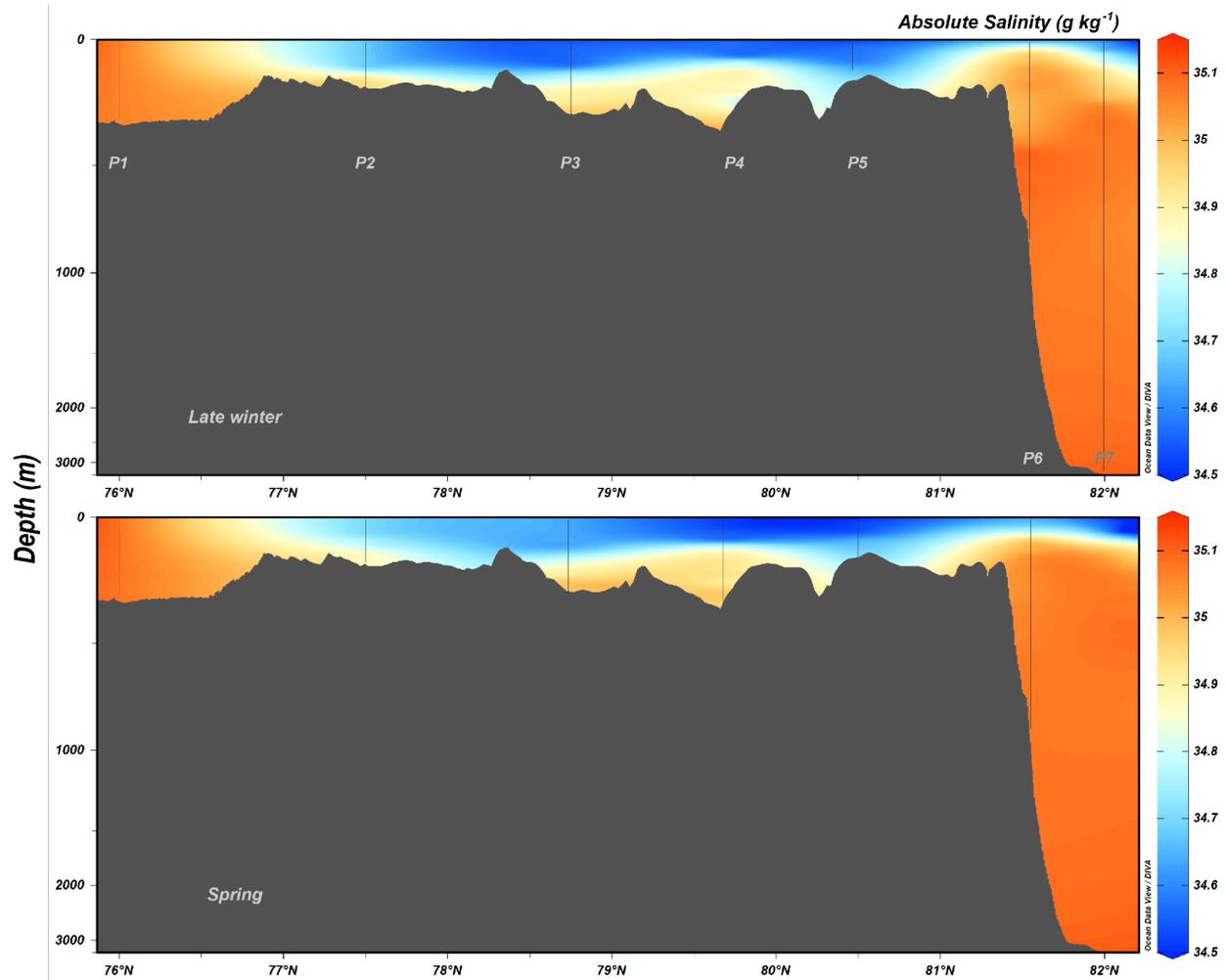
1383 **SI Figure 1:** Complete depth profiles of THg (left, pmol L<sup>-1</sup>) and MeHg (right, fmol L<sup>-1</sup>) for  
1384 stations P6 (top) and P7 (bottom) in late winter (blue) and spring (orange) in the northern Barents  
1385 Sea in 2021.

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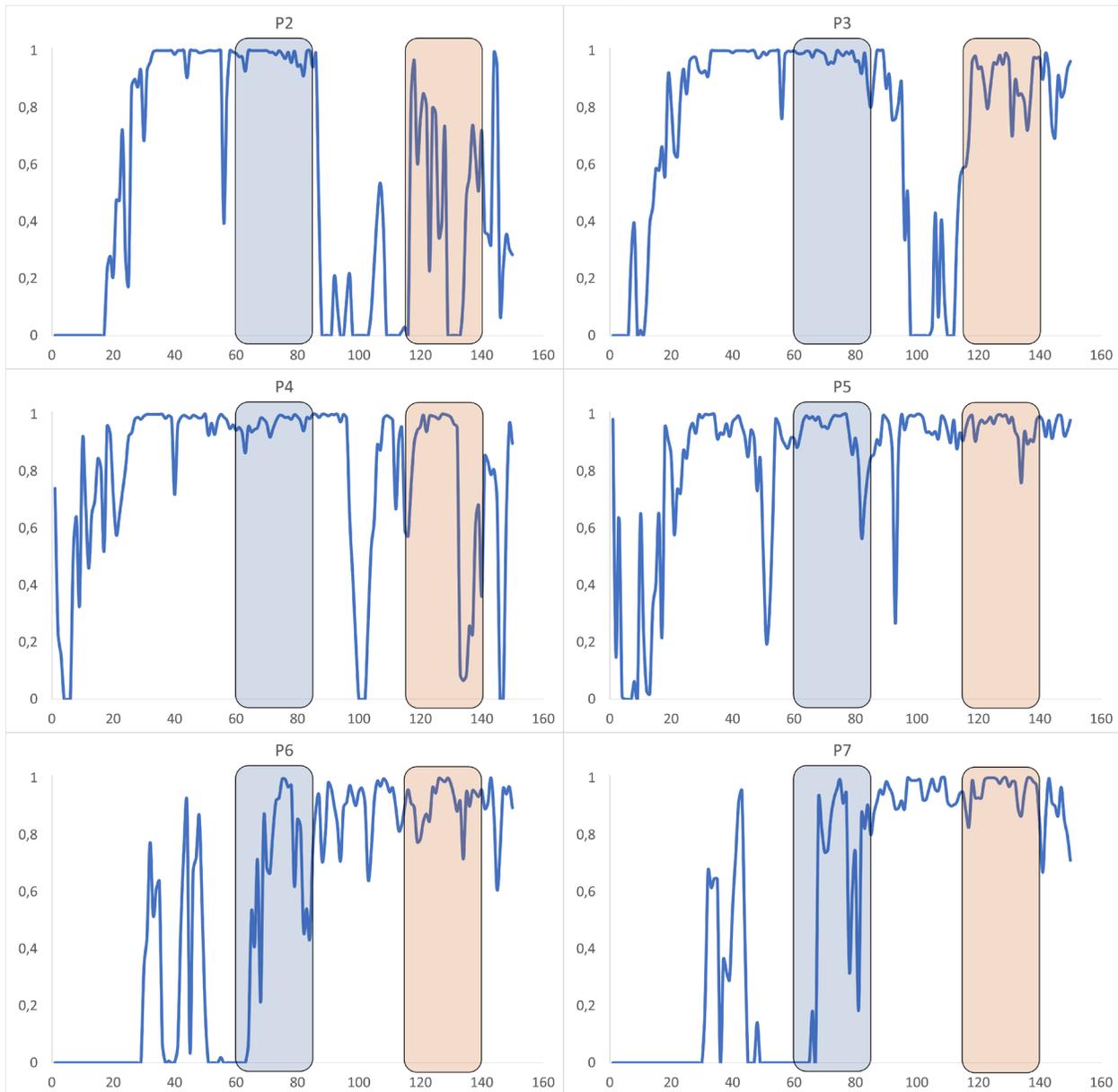
1388 **SI Figure 2:** Conservative temperature ( $\Theta$ , °C) along the shelf-basin transect in the northern  
 1389 Barents Sea during late winter (top) and spring (bottom) in 2021. The x-axis displays latitude and  
 1390 the nonlinear y-axis is in units of depth (m). Transect created with Ocean Data View (Schlitzer  
 1391 2021).



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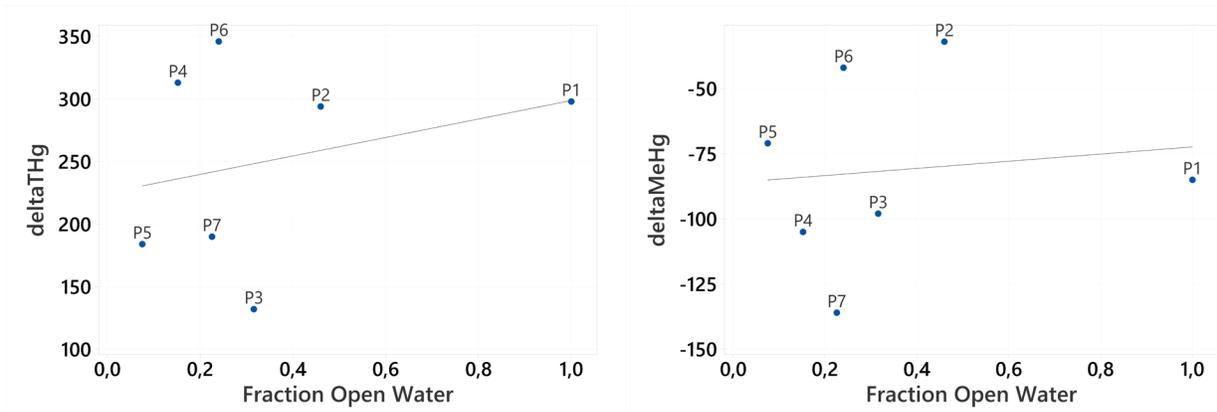
1393 **SI Figure 3:** Absolute salinity ( $S_A$ ,  $\text{g kg}^{-1}$ ) along the shelf-basin transect in the northern Barents  
 1394 Sea during late winter (top) and spring (bottom) in 2021. The x-axis displays latitude and the  
 1395 nonlinear y-axis is in units of depth (m). Transect created with Ocean Data View (Schlitzer 2021).

## Sea Ice Coverage Fraction



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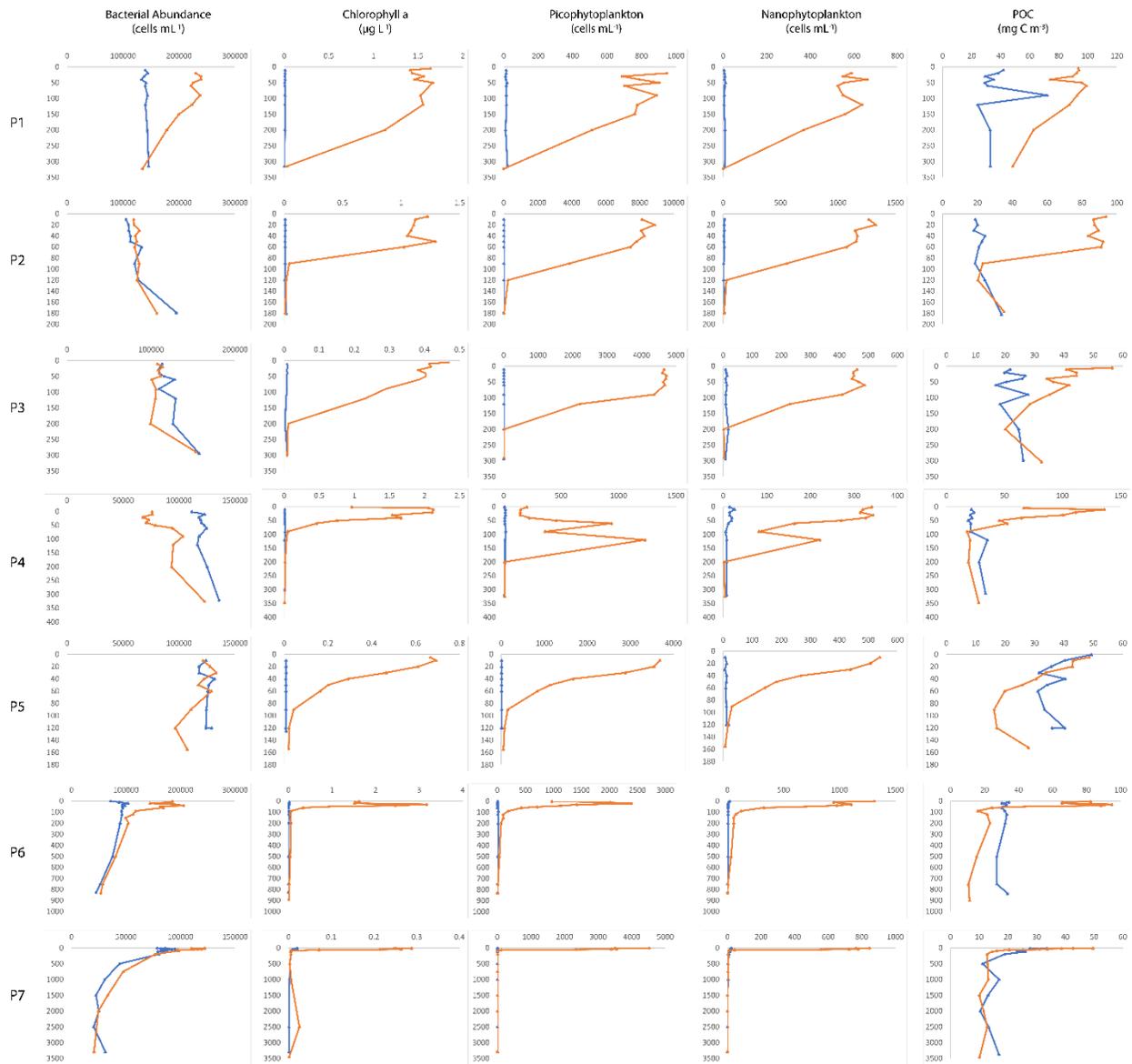
1397 **SI Figure 4:** Fraction of sea ice coverage in the northern Barents Sea for stations P2 - P7 during  
1398 January 1 - May 30, 2021. P1 had no ice coverage. An ice fraction of 1 indicates complete sea ice  
1399 coverage and 0 indicates open water. Sampling periods are outlined in rectangles, with late winter  
1400 (blue) and spring (orange). The y-axis is fraction of sea ice coverage (0 - 1) and the x-axis is in  
1401 days starting from January 1, 2021.



1402

1403 **SI Figure 5:** Linear regression of  $\Delta\text{THg}$  ( $\text{pmol m}^{-2} \text{d}^{-1}$ , left) and  $\Delta\text{MeHg}$  ( $\text{pmol m}^{-2} \text{d}^{-1}$ , right) in  
 1404 the upper 100 m against the average fraction of open water in between sampling periods (60 day-  
 1405 average) in the northern Barents Sea in 2021. No significance (F-test,  $p > 0.05$ ) was observed for  
 1406 either regression.

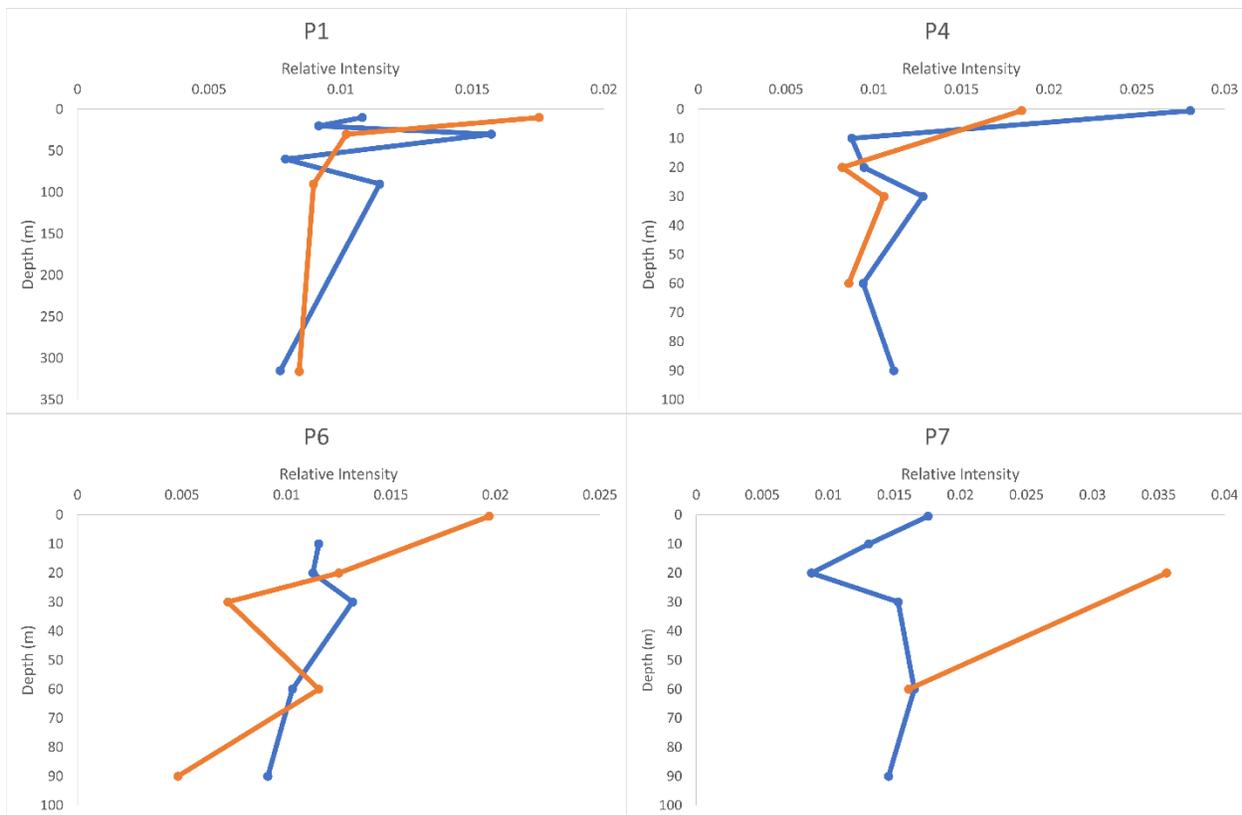
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1409 **SI Figure 6:** Bacterial abundance (cells mL<sup>-1</sup>), chlorophyll *a* (µg L<sup>-1</sup>), picophytoplankton  
 1410 abundance (cells mL<sup>-1</sup>), nanophytoplankton abundance (cells mL<sup>-1</sup>), and POC (mg C m<sup>-3</sup>) for all  
 1411 seven stations (horizontal rows). Sampling periods are blue (late winter) and orange (spring). Each  
 1412 y-axis is in units of depth (m). Note the different scales in both x and y axes. Data from Marquardt  
 1413 2022a, 2022b, Müller 2023a, 2023b, and Vader 2022.

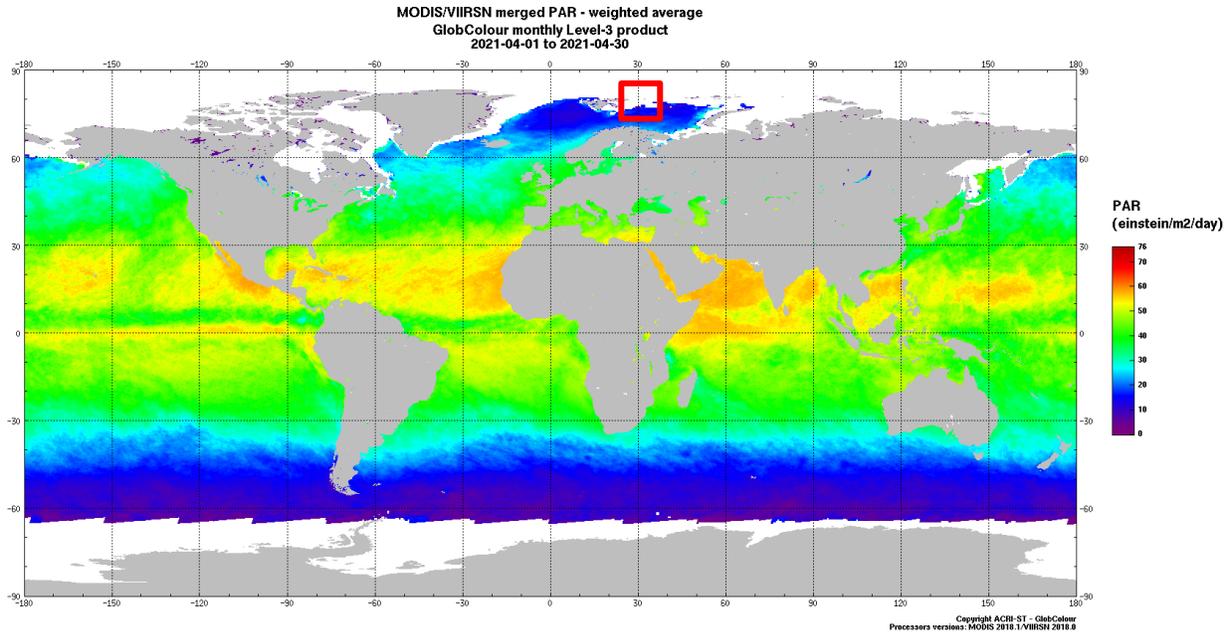
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1416 **SI Figure 7:** Relative intensities of dissolved organic sulfur compounds (CHOS) at selected  
 1417 stations on our transect in the northern Barents Sea for late winter (blue) and spring (orange). The  
 1418 y-axis is in units of depth and the x-axis is relative intensity. Note the different scales in both x  
 1419 and y axes.

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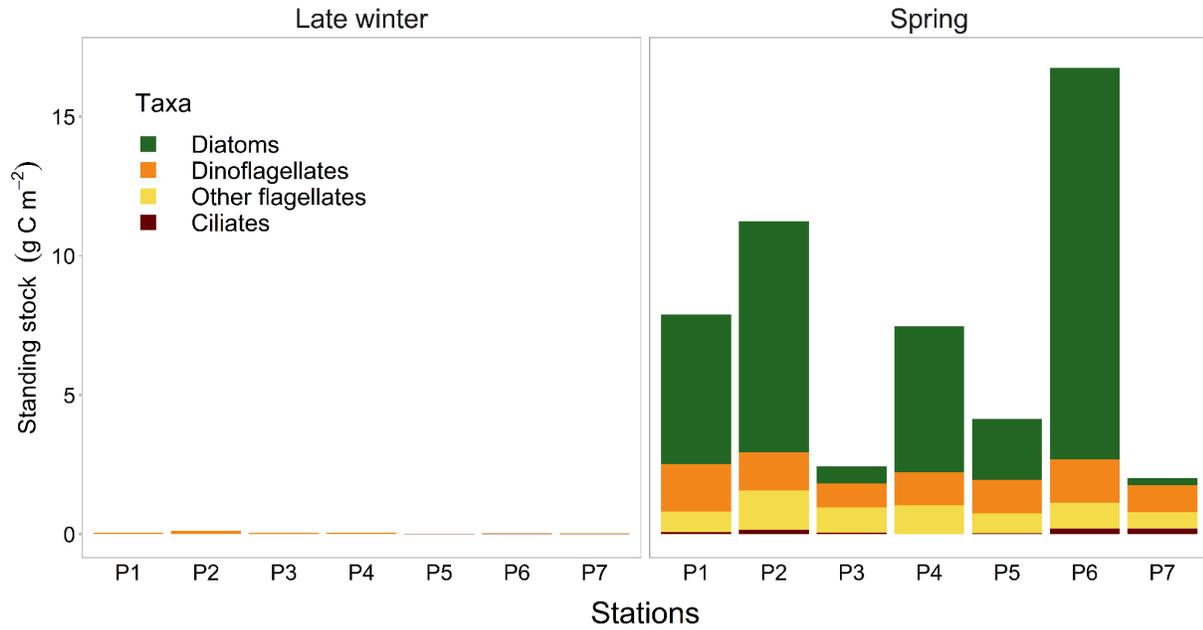


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1422 **SI Figure 8:** Satellite PAR ( $E m^{-2} d^{-1}$ ) during the late winter and spring sampling periods (April  
 1423 2021). PAR in the transect east of Svalbard (in red box), estimated to be approximately  $15 E m^{-2}$   
 1424  $d^{-1}$  for all stations, despite significant ice coverage. Retrieved from GlobColour, Hermes, ocean  
 1425 surface optical PAR, © ACRI-ST – GlobColour (<https://hermes.acri.fr/index.php?class=archive>).

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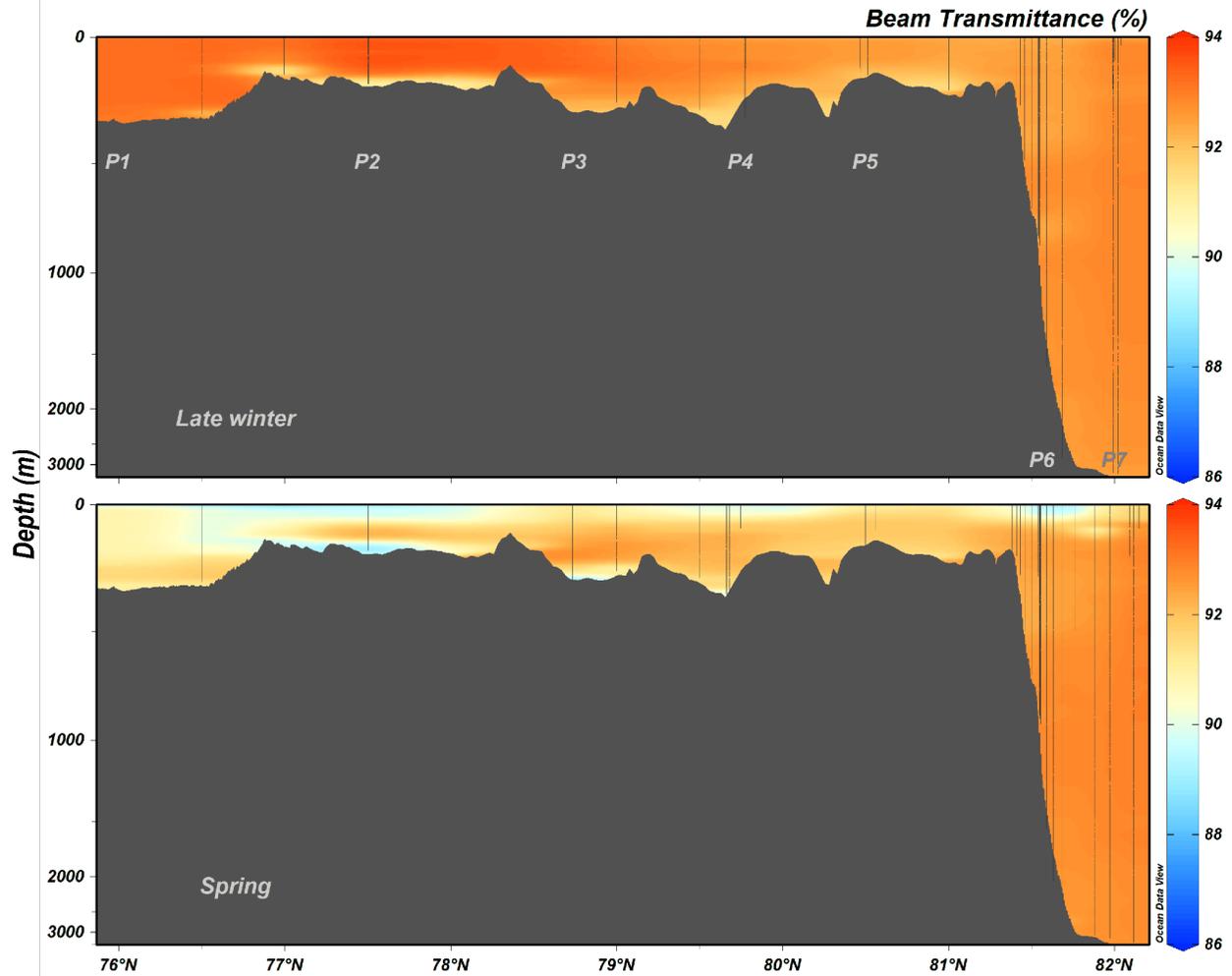
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1429 **SI Figure 9:** Integrated standing stocks (upper 90 m) of major protist groups (g C m<sup>-2</sup>) during the  
 1430 late winter and spring sampling periods in the northern Barents Sea. The colors illustrate diatoms  
 1431 (dark green), dinoflagellates (orange), other flagellates (yellow) and ciliates (dark red). The x-axis  
 1432 shows each individual station. Data from Assmy 2022a, 2022b.

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 1435 **SI Figure 10:** Raw beam transmittance (%), the fraction of light that reaches the detector, along  
 1436 the shelf-basin transect in the northern Barents Sea during late winter (top) and spring (bottom). A  
 1437 lower beam transmittance can indicate greater scattering or absorption by particles. The x-axis  
 1438 displays latitude and the nonlinear y-axis is in units of depth (m). Transect created with Ocean  
 1439 Data View (Schlitzer 2021).

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1441 **Table S1:** Average ( $\pm$  one standard deviation) of THg and MeHg concentrations on both late  
 1442 winter and spring cruises for water column samples above 500 m, categorized by water masses;  
 1443 Atlantic Water (AW), modified Atlantic Water (mAW), Polar Water (PW), warm Polar Water  
 1444 (wPW). Significant differences (log-transformed, Student's t-test,  $p < 0.05$ ) between THg and  
 1445 MeHg in each respective season when categorized by water mass have each seasonal average  
 1446 marked with an asterisk (\*).

Water mass	Total Hg (pmol L <sup>-1</sup> )		Methylated Hg (fmol L <sup>-1</sup> )	
	Late winter	Spring	Late winter	Spring
<b>AW</b>	0.47 $\pm$ 0.01 (n = 2)	N/A	200 $\pm$ 76 (n = 2)	N/A
<b>mAW</b>	0.53 $\pm$ 0.04* (n = 7)	0.64 $\pm$ 0.09* (n = 10)	63 $\pm$ 19 (n = 7)	57 $\pm$ 77 (n = 6)
<b>PW</b>	0.55 $\pm$ 0.07* (n = 26)	0.66 $\pm$ 0.08* (n = 34)	72 $\pm$ 30* (n = 26)	25 $\pm$ 14* (n = 31)
<b>wPW</b>	0.52 $\pm$ 0.07 (n = 14)	0.56 $\pm$ 0.07 (n = 12)	105 $\pm$ 37 (n = 14)	75 $\pm$ 36 (n = 12)

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1448 **Table S2:** Spearman rank correlation analysis (95% CI) for select environmental parameters for all stations and depths less than 500 m  
 1449 on the late winter cruise, with depth, total mercury (THg) methylated mercury (MeHg) particulate organic carbon (POC), chlorophyll *a*  
 1450 (Chla), nitrite (NO<sub>2</sub>), nitrate (NO<sub>3</sub>), phosphate (PO<sub>4</sub>), silicate (SiOH<sub>4</sub>), conservative temperature (Θ), absolute salinity (S<sub>A</sub>), and bacteria.  
 1451 A significant p-value (p < 0.05) marked in bold.

	<b>Depth</b>	<b>THg</b>	<b>MeHg</b>	<b>POC</b>	<b>Chla</b>	<b>NO<sub>2</sub></b>	<b>NO<sub>3</sub></b>	<b>PO<sub>4</sub></b>	<b>SiOH<sub>4</sub></b>	<b>Θ</b>	<b>S<sub>A</sub></b>
THg	-0.097										
MeHg	<b>0.364</b>	0.03									
POC	0.037	0.201	0,095								
Chla	<b>-0.545</b>	<b>0.459</b>	-0.117	0,2							
NO <sub>2</sub>	-0.077	<b>0.317</b>	<b>-0.315</b>	0.213	0.217						
NO <sub>3</sub>	<b>0.623</b>	-0.053	<b>0.497</b>	<b>0.42</b>	<b>-0.323</b>	0.153					
PO <sub>4</sub>	<b>0.545</b>	-0.025	<b>0.417</b>	<b>0.523</b>	-0.277	0.237	<b>0,957</b>				
SiOH <sub>4</sub>	<b>0.588</b>	0.061	<b>0.565</b>	<b>0.455</b>	-0.204	0.135	<b>0,912</b>	<b>0.867</b>			
Θ	<b>0.63</b>	-0.243	<b>0.475</b>	<b>0.38</b>	<b>-0.429</b>	0	<b>0,889</b>	<b>0.833</b>	<b>0.817</b>		
S <sub>A</sub>	<b>0.559</b>	-0.241	0.205	<b>0.294</b>	<b>-0.329</b>	0.082	<b>0,785</b>	<b>0.757</b>	<b>0.654</b>	<b>0.8</b>	
Bacteria	0.105	<b>0.298</b>	<b>-0.418</b>	0.267	0.133	<b>0.618</b>	0,113	0.249	0.037	-0.082	0.077

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1462 **Table S3:** Spearman rank correlation analysis (95% CI) for select environmental parameters for all stations and depths less than 500 m  
 1463 on the spring cruise, with depth, total mercury (THg) methylated mercury (MeHg) particulate organic carbon (POC), chlorophyll *a*  
 1464 (Chla), nitrite (NO<sub>2</sub>), nitrate (NO<sub>3</sub>), phosphate (PO<sub>4</sub>), silicate (SiOH<sub>4</sub>), conservative temperature (Θ), absolute salinity (S<sub>A</sub>), and bacteria.  
 1465 A significant p-value (p < 0.05) marked in bold.  
 1466

	<b>Depth</b>	<b>THg</b>	<b>MeHg</b>	<b>POC</b>	<b>Chla</b>	<b>NO<sub>2</sub></b>	<b>NO<sub>3</sub></b>	<b>PO<sub>4</sub></b>	<b>SiOH<sub>4</sub></b>	<b>Θ</b>	<b>S<sub>A</sub></b>
THg	<b>-0.609</b>										
MeHg	<b>0.394</b>	<b>-0.42</b>									
POC	<b>-0.622</b>	<b>0.714</b>	<b>-0.582</b>								
Chla	<b>-0.707</b>	<b>0.708</b>	<b>-0.467</b>	<b>0.895</b>							
NO <sub>2</sub>	-0.05	0.045	<b>-0.308</b>	0.212	0.251						
NO <sub>3</sub>	<b>0.66</b>	<b>-0.417</b>	<b>0.51</b>	<b>-0.489</b>	<b>-0.534</b>	-0.22					
PO <sub>4</sub>	<b>0.724</b>	<b>-0.484</b>	<b>0.542</b>	<b>-0.59</b>	<b>-0.64</b>	-0.235	<b>0.937</b>				
SiOH <sub>4</sub>	<b>0.668</b>	<b>-0.324</b>	<b>0.438</b>	<b>-0.389</b>	<b>-0.472</b>	-0.162	<b>0.938</b>	<b>0.897</b>			
Θ	<b>0.603</b>	<b>-0.414</b>	<b>0.378</b>	<b>-0.311</b>	<b>-0.315</b>	-0.081	<b>0.802</b>	<b>0.674</b>	<b>0.823</b>		
S <sub>A</sub>	<b>0.649</b>	<b>-0.329</b>	<b>0.319</b>	-0.155	-0.255	-0.057	<b>0.723</b>	<b>0.621</b>	<b>0.724</b>	<b>0.856</b>	
Bacteria	-0.249	<b>0.469</b>	<b>-0.389</b>	<b>0.468</b>	<b>0.438</b>	-0.061	0.107	0.021	0.188	0.166	0.249

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1469 **Table S4:** Integrated THg pools ( $\text{pmol m}^{-2}$ ) from 0 -100 m from all seasonal cruises completed in  
 1470 the area. We used data from Kohler et al. 2022a for summer and early winter.

	<b>Integrated Total Hg pool (<math>\text{pmol m}^{-2}</math>)</b>			
<b>0 - 100 m</b>	<b>Late winter (March 2021)</b>	<b>Spring (May 2021)</b>	<b>Summer (August 2019)</b>	<b>Early winter (December 2019)</b>
<b>P1</b>	51647 $\pm$ 1696	68280 $\pm$ 1620	64033 $\pm$ 869	37050 $\pm$ 1736
<b>P2</b>	51197 $\pm$ 1620	67652 $\pm$ 1541	63050 $\pm$ 865	40050 $\pm$ 1800
<b>P3</b>	55570 $\pm$ 1620	63074 $\pm$ 1620	64483 $\pm$ 922	40267 $\pm$ 1829
<b>P4</b>	49786 $\pm$ 1601	67616 $\pm$ 1601	73635 $\pm$ 838	51400 $\pm$ 1866
<b>P5</b>	53409 $\pm$ 1677	63737 $\pm$ 1541	78773 $\pm$ 886	50136 $\pm$ 1753
<b>P6</b>	52700 $\pm$ 1994	72807 $\pm$ 1743	63300 $\pm$ 1115	43955 $\pm$ 2049
<b>P7</b>	52244 $\pm$ 2016	63251 $\pm$ 1989	55300 $\pm$ 1089	58200 $\pm$ 2200

1471 **Table S5:** Integrated MeHg pools ( $\text{pmol m}^{-2}$ ) from 0 -100 m from all seasonal cruises completed  
 1472 in the area. We used data from Kohler et al. 2022 for summer and early winter.

	<b>Integrated Methylated Hg pool (<math>\text{pmol m}^{-2}</math>)</b>			
<b>0 - 100 m</b>	<b>Late winter (March 2021)</b>	<b>Spring (May 2021)</b>	<b>Summer (August 2019)</b>	<b>Early winter (December 2019)</b>
<b>P1</b>	5486 $\pm$ 237	710 $\pm$ 293	10133 $\pm$ 583	9350 $\pm$ 648
<b>P2</b>	3999 $\pm$ 227	2187 $\pm$ 216	10000 $\pm$ 604	9513 $\pm$ 731
<b>P3</b>	6912 $\pm$ 227	1341 $\pm$ 260	10317 $\pm$ 583	9450 $\pm$ 555
<b>P4</b>	7339 $\pm$ 224	1378 $\pm$ 245	9444 $\pm$ 548	9433 $\pm$ 583
<b>P5</b>	6611 $\pm$ 235	2640 $\pm$ 216	9550 $\pm$ 547	11000 $\pm$ 547
<b>P6</b>	8297 $\pm$ 279	5878 $\pm$ 244	13000 $\pm$ 679	10290 $\pm$ 624
<b>P7</b>	13381 $\pm$ 282	5485 $\pm$ 278	12850 $\pm$ 679	14350 $\pm$ 679

1473 **Table S6:**  $\Delta\text{MeHg}$  ( $\text{pmol m}^{-2} \text{d}^{-1}$ ) for 0-100 m, calculated from 0 – 100 m integrated pools for late  
 1474 winter and spring. A negative value assigned in front of each  $\Delta\text{MeHg}$  indicates in a negative  
 1475 change in the MeHg pool from late winter to spring, or a loss of MeHg.

<b>Station</b>	<b>Days between sampling</b>	<b><math>\Delta\text{MeHg}</math> <math>\text{pmol m}^{-2} \text{d}^{-1}</math></b>	<b><math>\Delta\text{MeHg}_{\text{pd}}</math> <math>\text{pmol m}^{-2} \text{d}^{-1}</math></b>	<b><math>\Delta\text{MeHg}_{\text{scav}}</math> <math>\text{pmol m}^{-2} \text{d}^{-1}</math></b>	<b><math>\Delta\text{MeHg}_{\text{ev}}</math> <math>\text{pmol m}^{-2} \text{d}^{-1}</math></b>
P1	56	$-85 \pm 7$	-10	-1	-7
P2	56	$-32 \pm 6$	-3	-1	-2
P3	57	$-98 \pm 6$	-3	-1	-2
P4	57	$-105 \pm 6$	-1	-1	-1
P5	56	$-71 \pm 6$	-1	-1	0
P6	58	$-42 \pm 6$	-3	-1	-2
P7	58	$-136 \pm 7$	-3	-1	-2

1476 **Table S7:** Percent of variance explained for the first two plotted principal components for each  
 1477 cruise.

<b>Cruise</b>	<b>% of Variance</b>	
	<b>PC 1</b>	<b>PC 2</b>
<b>Late Winter 2021 (LW)</b>	43.3	20.2
<b>Spring 2021 (SP)</b>	52.8	17.0
<b>Summer 2019 (SU)</b>	56.5	12.1
<b>Winter 2019 (W)</b>	46.8	16.2

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