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1 ***TITLE***

2 Arctic Ocean's wintertime Mercury concentrations limited by seasonal loss on the shelf

3

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21

22 ***ABSTRACT***

23 High biota mercury levels are persisting in the Arctic, threatening ecosystem and human health.
24 The Arctic Ocean receives large pulsed mercury inputs from rivers and the atmosphere. Yet, the
25 fate of those inputs and possible seasonal variability of mercury in the Arctic Ocean remain
26 uncertain. Until now, seawater observations were only possible during summer and fall. Here we
27 report polar night mercury seawater observations on a gradient from the shelf into the Arctic
28 Ocean. We observed lower and less variable total mercury concentrations during the polar night
29 (0.46 ± 0.07 pmol L⁻¹) compared to summer (0.63 ± 0.19 pmol L⁻¹) and no significant changes in
30 methylmercury concentrations (summer, 0.11 ± 0.03 pmol L⁻¹ and winter, 0.12 ± 0.04 pmol L⁻¹).
31 Seasonal changes were estimated by calculating the difference in the integrated mercury pools.
32 We estimate losses of inorganic mercury of 208 ± 41 pmol m⁻² d⁻¹ on the shelf driven by seasonal
33 particle scavenging. Persistent methylmercury concentrations (-1 ± 16 pmol m⁻² d⁻¹) are likely
34 driven by gaseous species and a lower affinity for particles. Our results update the current
35 understanding of Arctic mercury cycling and require budgets and models to be reevaluated with a
36 seasonal aspect.

37

38 *MAIN TEXT*

39 The Arctic Ocean and its biota exhibit elevated levels of toxic mercury (Hg)¹ despite an absence
40 of important local anthropogenic sources. Mercury is delivered to surface waters of the Arctic
41 Ocean through atmospheric deposition, inputs from other oceans, riverine discharge, snow and sea
42 ice melt². High inputs of mainly inorganic mercury (iHg) coincide with the Arctic's biologically
43 active summer season³. However, data on inter- and intra-seasonal Hg concentrations in the Arctic
44 Ocean, especially during the winter, is lacking so far. The discovery of springtime atmospheric Hg
45 depletion events⁴ over 20 years ago highlighted a distinct seasonal aspect to Hg biogeochemical

46 cycling in the Arctic. Recently, increased attention on pan-Arctic rivers have also confirmed
47 riverine Hg inputs to the Arctic shelf seas during spring flood events⁵. With increasing evidence
48 of a highly seasonal Arctic Hg cycle, a lack of seawater observations in winter indicates that the
49 current paradigm of the Arctic Ocean Hg budget and fluxes are only a “summer snapshot”.

50

51 The seasonal transition from summer to winter in the Arctic Ocean is marked by decreased
52 biological activity, weakened stratification, and sea ice growth. However, our understanding of Hg
53 cycling during the Arctic winter is still largely based on assumptions with considerable
54 uncertainty. Biogeochemical models suggest that water mixing during autumn resupplies Hg to
55 upper waters⁶. The presence of sea ice restricts Hg exchange with the atmosphere⁷ throughout
56 autumn and winter, allowing potential buildup of gaseous mercury and dimethylmercury (DMHg)
57 under sea ice⁸. As temperatures rise and light returns, sea ice melt and atmospheric inputs may
58 explain increased springtime Hg concentrations in polar waters compared to winter Hg
59 concentrations⁹. The spring freshet delivers additional Hg to surface waters from the major pan-
60 Arctic rivers¹⁰. As a result, summer Hg observations in Arctic surface waters are elevated
61 compared to deeper waters¹¹.

62

63 Arctic surface waters lose significant amounts of Hg via atmospheric evasion or scavenging with
64 sinking particles. During the summer, photochemical reduction of riverine Hg and subsequent
65 evasion emits a significant fraction of gaseous Hg back to the atmosphere¹². However, recent
66 observations indicate low Hg evasion to the atmosphere in late summer and autumn⁸, suggesting
67 particle scavenging dominates Hg losses from surface waters in autumn and winter. In addition,
68 Arctic summer and autumn exhibit the highest total particulate matter fluxes due to peak primary

69 productivity^{13,14}. Although particle fluxes decline in winter, dissolved Hg has a high affinity to
70 particles due to high partitioning coefficients (K_D)¹⁵. Previous observations, models, and
71 budgets^{2,12,16-19} have hinted at the importance of scavenging of Hg on the shelf yet lack
72 confirmation, especially during winter.

73 **Seasonal loss of Hg on the Arctic shelves**

74 We investigated a shelf to deep basin meridional transect in the northern Barents Sea in August –
75 summer, and December – winter (Fig. 1). We sampled seawater for unfiltered total Hg (THg), the
76 sum of all inorganic and organic Hg species, including both particulate and dissolved. We also
77 collected seawater for unfiltered methylated mercury (MeHg), the sum of organic Hg species,
78 monomethylmercury (MMHg) and DMHg.

79
80 Average THg concentrations (Fig. 2) in summer are 0.63 ± 0.19 pmol L⁻¹ (range: 0.38 – 1.36 pmol
81 L⁻¹, n = 60) and show a scavenged-type profile, similar to THg observations on previous
82 summertime Arctic expeditions^{16,17,20-23}. Our total Hg concentrations on the shelf (< 400 m depth,
83 P1 – P5) are slightly higher and more variable compared to the shelf break (P6) and Nansen Basin
84 (P7) stations (Fig. 2). We measured the highest THg concentrations in the polar mixed layer (< 20
85 m) under the sea ice. The higher Hg concentrations are most likely from Hg contained in pico- and
86 nanophytoplankton in addition to Hg input from melting sea ice. Elevated surface THg
87 concentrations in the southerly open water stations (P1 – P3) might also suggest atmospheric
88 deposition²⁴. Average THg concentrations on our transect trend lower than the Canadian
89 Archipelago and the Siberian shelf seas²⁵, although this difference may be explained by the
90 region's lack of major riverine sources and mixing of Hg-depleted Atlantic Water¹⁷ and Hg-
91 enriched Polar Water^{11,26,27}.

92
93 Our average THg concentrations in winter (Fig. 2) are 0.46 ± 0.07 pmol L⁻¹ (range: 0.34 – 0.66
94 pmol L⁻¹, n = 61) and display less variability than summer concentrations (range: 0.38 – 1.36 pmol
95 L⁻¹). The THg concentrations in the stratified Nansen Basin (P7) continue to exhibit a scavenged-
96 type profile, while THg concentrations on the shelf stations (P1 – P5) have changed to a more
97 conservative-type profile. Average winter THg concentrations at shelf stations (P1 – P5) including
98 all sampled depths were 28 – 41% less than summer. Our overall average winter THg
99 concentrations are similar to summer THg concentrations in Atlantic Water (0.43 ± 0.14 pmol L⁻¹)
100 ¹⁷ yet less variable. However, all shelf stations except P1 retain Polar Water signatures with
101 additional sea ice cover. Colder and saltier surface waters in winter display weaker stratification
102 and suggest mixing throughout the autumn (Extended Data Figs. 1 and 2). Therefore, our lower
103 winter THg concentrations in surface waters suggest THg is not resupplied from subsurface waters
104 during autumn as previously thought. Weak stratification and mixing promote downward transport
105 of particles^{28,29}, where surface THg may be scavenged to deeper waters and sediments.

106 **Seasonal scavenging of inorganic Hg**

107 We investigated the scavenging dynamics of THg by calculating iHg (pmol L⁻¹), the difference
108 between THg and MeHg concentrations at each discrete sampling depth. Temporal changes in iHg
109 between summer and winter are represented as Δ iHg using the integrated seasonal difference
110 between iHg concentrations at two depth intervals (Fig. 3). These calculated Δ iHg quantities
111 indicate a gain or loss in the total iHg pool. Six stations (P1 – P6) showed a negative Δ iHg, or loss
112 of iHg, by winter (Fig. 3). On average, Δ iHg was -208 ± 41 pmol m⁻² d⁻¹ in the upper 100 m on the
113 shelf (P1 – P5). By taking the absolute value of Δ iHg, we can compare to directional fluxes. Our
114 average $|\Delta$ iHg| on the shelf is comparable to recent estimates of particulate Hg fluxes (173 ± 78

115 pmol m⁻² d⁻¹) using a radiotracer approach¹⁹. While particulate Hg is approximately 17% of THg
116 concentrations in the Barents Sea shelf¹⁹, our average seasonal iHg decrease of 39 – 50% in the
117 upper 100 m (P1 – P5) suggests scavenging of both particulate and dissolved iHg species.

118
119 Previous work has suggested Hg shelf scavenging along the continental margins³⁰ driven by strong
120 Hg-particle interactions. Updated estimates of Hg-particle partition coefficients (K_D) in the Arctic
121 Ocean indicate high affinity to manganese and iron oxides³¹. We found high dissolved manganese
122 (DMn) concentrations on the shelf in both summer and winter (Extended Data Fig. 3). Both DMn
123 and total manganese (TMn, Extended Data Fig. 4) concentrations are highest near sediments³²
124 (Extended Data Figs. 3 – 5). Suboxic reducing conditions in shelf sediments supply Mn²⁺ to the
125 overlying water column through mixing, sediment resuspension and diffusion³³. Here, we
126 speculate that seasonal decreases of DMn on the shelf in the upper 100 m (P1 – P5, Extended Data
127 Figs. 3 and 6) are due to oxidation to insoluble MnO₂. Colloidal MnO₂ can form precipitates that
128 can scavenge and shuttle iHg to deeper depths, similar to observations in the Baltic and the Black
129 Seas³⁴. In the absence of late summer POC fluxes, the DMn shuttle on the Arctic shelf may explain
130 our seasonal losses of DMn (Extended Data Fig. 6), iHg (Fig. 3) and total lead (TPb, Extended
131 Data Fig. 7). As TPb and THg have comparable K_D values for manganese particle phases in the
132 Arctic Ocean³⁵, our negative Δ TPb values on shelf stations (P1 – P5, Extended Data Fig. 8) reflect
133 similar trends to Δ iHg (Fig. 3). A seasonal loss of TPb on the Barents Sea shelf (P1 – P5) agrees
134 well with previous hypotheses of strong Pb scavenging on the Arctic shelves^{18,36-38}.

135
136 Losses of gaseous Hg from Arctic surface waters may also influence our seasonal loss of iHg. We
137 evaluated Hg⁰ losses from summer to winter by limiting Δ iHg to the wind mixed layer (< 20 m).

138 We assumed our iHg concentrations at the shallowest sampling depth (approx. 10 m) were
139 representative of the atmosphere-sea surface interface (0 m). Most stations (P1 – P6) exhibit a
140 negative Δ iHg, with an average Δ iHg of -80 ± 14 pmol m⁻² d⁻¹ on the shelf (P1 – P5). By
141 comparison, we calculated a net open water Hg⁰ evasion rate of only 11 pmol m⁻² d⁻¹ from a late
142 summer atmospheric evasion rate⁸ combined with their suggested equal Hg inputs from deposition
143 and meltwater. Accordingly, our low calculated Hg⁰ evasion rate compared to high wind-mixed
144 layer $|\Delta$ iHg| values suggest minimal losses of gaseous Hg species.

145 **Persistent seawater MeHg**

146 While THg concentrations showed significant seasonal differences, average MeHg concentrations
147 remained constant (Fig. 4). Average summer MeHg concentrations were 0.11 ± 0.03 pmol L⁻¹
148 (range; 0.08 – 0.23 pmol L⁻¹, n = 58) and winter concentrations were 0.12 ± 0.04 pmol L⁻¹ (range;
149 0.07 – 0.23 pmol L⁻¹, n = 55). We found that MeHg concentrations exhibit near uniform
150 distributions on the shelf (P1 – P5). In addition, a localized shallow MeHg maximum is seen at ~
151 80 – 120 m depth in the Nansen Basin (P6 – P7), indicative of in situ MeHg production^{16,17,21,23}.
152 Below 200 m, MeHg at P6 and P7 displayed the typical profile of MeHg observed in the Fram
153 Strait¹⁷ with deeper maxima between 400 – 1000 m depth. Overall, both of our seasonal MeHg
154 profiles are typical of the summer Arctic Ocean. Surprisingly, our unchanged seasonal MeHg
155 profiles are in stark contrast to our seasonal THg profiles.

156

157 While it is difficult to estimate, one could expect seasonal changes of MeHg due to the absence of
158 photochemistry³⁹, sea ice cover limiting evasion to the atmosphere⁸, net demethylation⁴⁰ and low
159 primary productivity during winter^{3,41}. Similar to Δ iHg, we calculated Δ MeHg at each station by
160 integrating the winter MeHg reservoir and comparing it to the integrated summer MeHg reservoir

161 at two depth intervals (Fig. 3). Our calculated ΔMeHg in the upper 100 m are small and show no
162 clear trend (range: -25 to 15 $\text{pmol m}^{-2} \text{d}^{-1}$, SI Table 1). We expect waters to be in a net state of
163 demethylation⁴⁰ as low particulate organic carbon (POC) fluxes on the shelf suggest minimal in
164 situ production of MeHg⁴². As a result, small variations in our ΔMeHg therefore suggest seasonal
165 MeHg distributions to be in a locally dynamic equilibrium.

166
167 On the other hand, our winter MeHg concentrations may remain the same as summer due to the
168 absence of photodegradation^{39,43}. Additionally, reduced losses of DMHg due to the presence of
169 sea ice may also explain static winter MeHg concentrations, although MMHg is typically the
170 dominant species on the shelf^{17,23}. Particle scavenging can also transport MMHg to deeper waters
171 and sediments, decreasing MeHg and THg concentrations in the water column. Higher amounts of
172 gaseous DMHg, with negligible affinity to sinking particles, could also explain the persistence of
173 MeHg concentrations. In addition, the K_D of MMHg may be an order of magnitude lower than
174 THg⁴⁴, reducing its affinity to sinking particles and explaining the difference in our THg and MeHg
175 trends. Generally, particulate MMHg concentrations in the Arctic Ocean are low, approximately
176 4% of MeHg²³. Consequently, our unchanged MeHg concentrations therefore suggest high
177 proportions of dissolved MeHg in both summer and winter.

178 **Implications for the Arctic Hg cycle**

179 The first observations during the polar night demonstrate a seasonal loss of THg. We suggest that
180 the seasonal loss is primarily driven by particle scavenging of iHg via an Arctic DMn shuttle.
181 Seasonal scavenging mechanisms on the entire Arctic shelf during the polar night may dominate
182 water column Hg losses to shelf sediments. We also suggest MeHg concentrations are maintained
183 in the water column in winter by reduced evasion, low particle affinities, slow demethylation rates

184 and a lack of photodegradation. Although in situ production is the dominant source of the
185 subsurface MeHg maximum during summer in the Arctic basin, small shelf to basin transport of
186 MeHg produced in the shelf sediments may contribute partially to the persistent shallow MeHg
187 maximum and positive ΔMeHg seen in the Arctic basin (P7) in winter (Fig. 3 & 4). Assuming a
188 shelf seasonal loss of 33 – 46% iHg including all sampled depths (P1 – P5), we estimate a residence
189 time for iHg of ~ 3 years. However, the persistence of MeHg extends the lifetime of THg in the
190 Arctic Ocean to ~ 9 years¹⁷. Our work highlights a potential decoupling of iHg and MeHg species
191 during the polar night and invites a new seasonally based paradigm of the Arctic Hg budget.

192

193 In the future, climate change is expected to significantly affect the biogeochemical cycle of Hg^{45,46}
194 and drive MeHg concentrations in the marine food chain⁴⁷. Sea ice loss will increase primary
195 productivity on the Arctic shelves⁴⁸ and lead to greater iHg export to deeper waters and sediments.
196 In addition, thawing permafrost and increased riverine flux will deliver high iHg inputs to the
197 Arctic shelves. Recent work has suggested increased Hg uptake in benthic feeding-organisms on
198 the Arctic shelf⁴⁹. We anticipate that seasonal scavenging during autumn and winter months will
199 deliver larger amounts of iHg to shelf sediments, driving increased benthic MeHg production and
200 MeHg accumulation in the future Arctic Ocean.

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213 ***AUTHOR CONTRIBUTIONS STATEMENT***

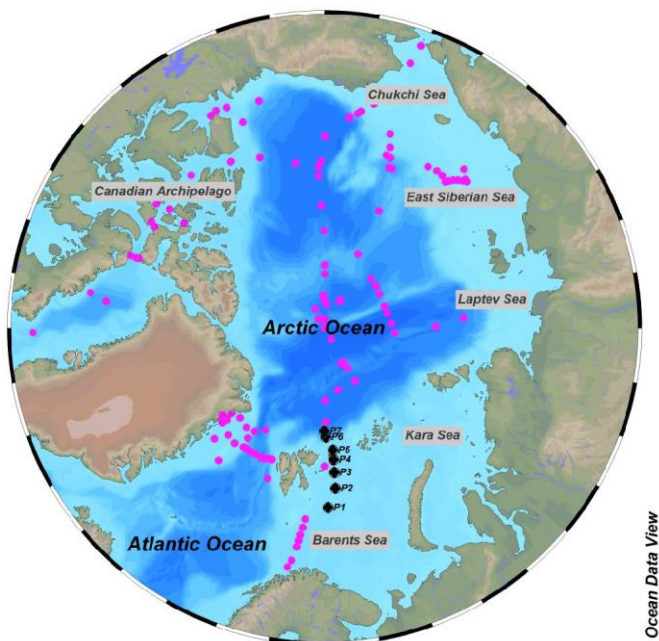
214 **SGK:** Conceptualization, formal analysis, investigation, validation, visualization, writing –
215 original draft. **LEHB:** Conceptualization, formal analysis, investigation, resources, writing –
216 review and editing, supervision. **MVP:** Formal analysis, investigation, validation, resources,
217 writing – review and editing. **MGD:** Investigation, writing – review and editing. **NS:** Formal
218 analysis, investigation, writing – review and editing. **AD:** Formal analysis, validation, writing –
219 review and editing. **AS:** Formal analysis, validation, writing – review and editing. **KN:**
220 Conceptualization, writing – review and editing, supervision. **MVA:** Conceptualization,
221 investigation, resources, writing – review and editing, supervision, funding acquisition.

222 **COMPETING INTERESTS STATEMENT**

223 The authors declare no competing interests.

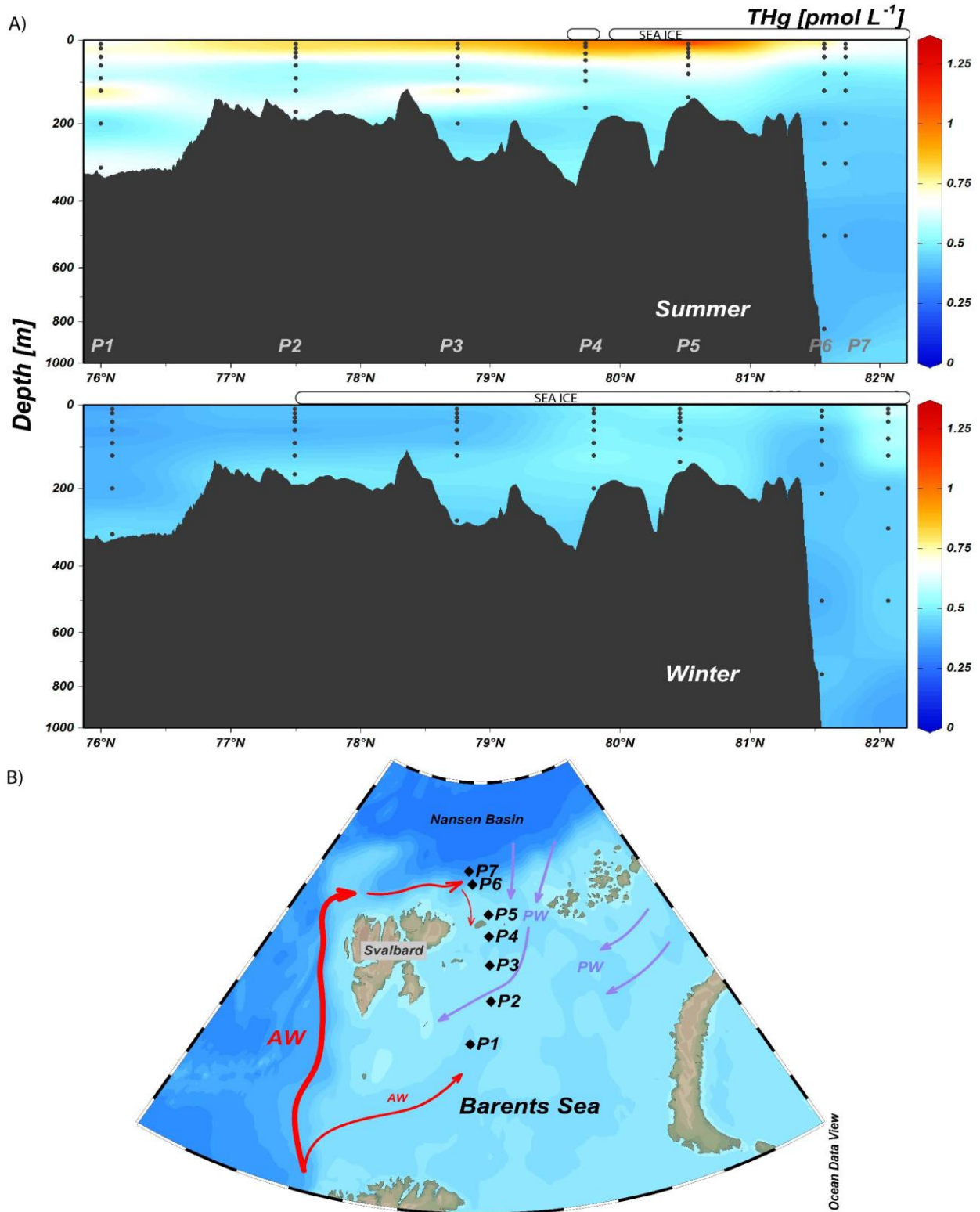
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225 **FIGURE LEGENDS/CAPTIONS**



226

227 **Figure 1: Map of water column Hg profiles in the Arctic Ocean.** Literature data marked with
228 pink circles^{1,16,17,20,21,23,25,50}. Stations (P1 – P7), sampled during the 2019 summer and winter
229 cruises, marked with black diamonds.



230

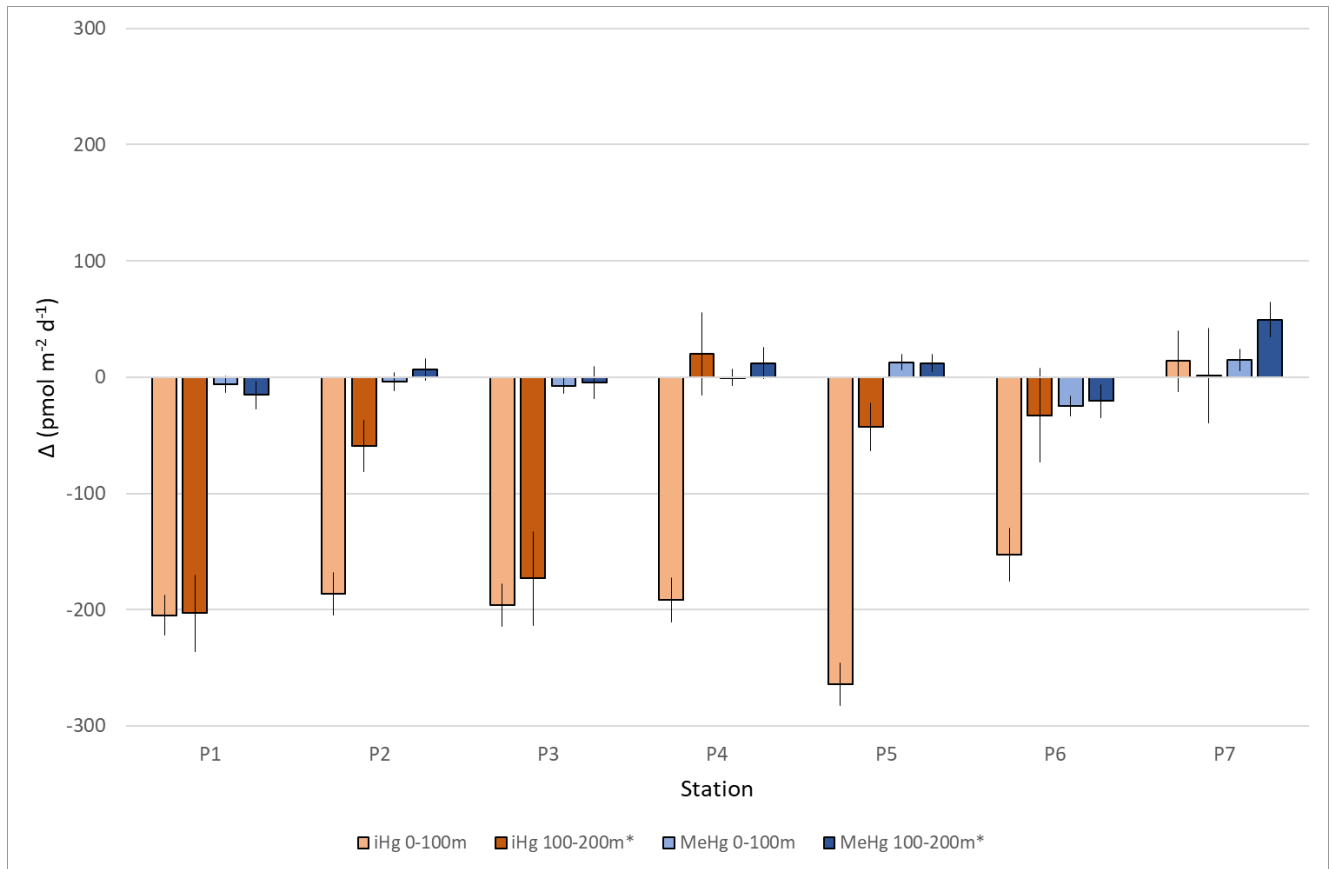
231 **Figure 2: Total mercury (THg) concentrations (pmol L⁻¹) along the shelf-deep basin gradient.**

232 A) Stations P1 – P7, (Latitude) are on the x-axis for summer 2019 and winter 2019 cruises. B)

233 Map of sampling stations with warmer Atlantic Water (AW) advected into the Barents Sea from
234 the south and west, and Polar Water (PW) advected from the north and east²⁷.

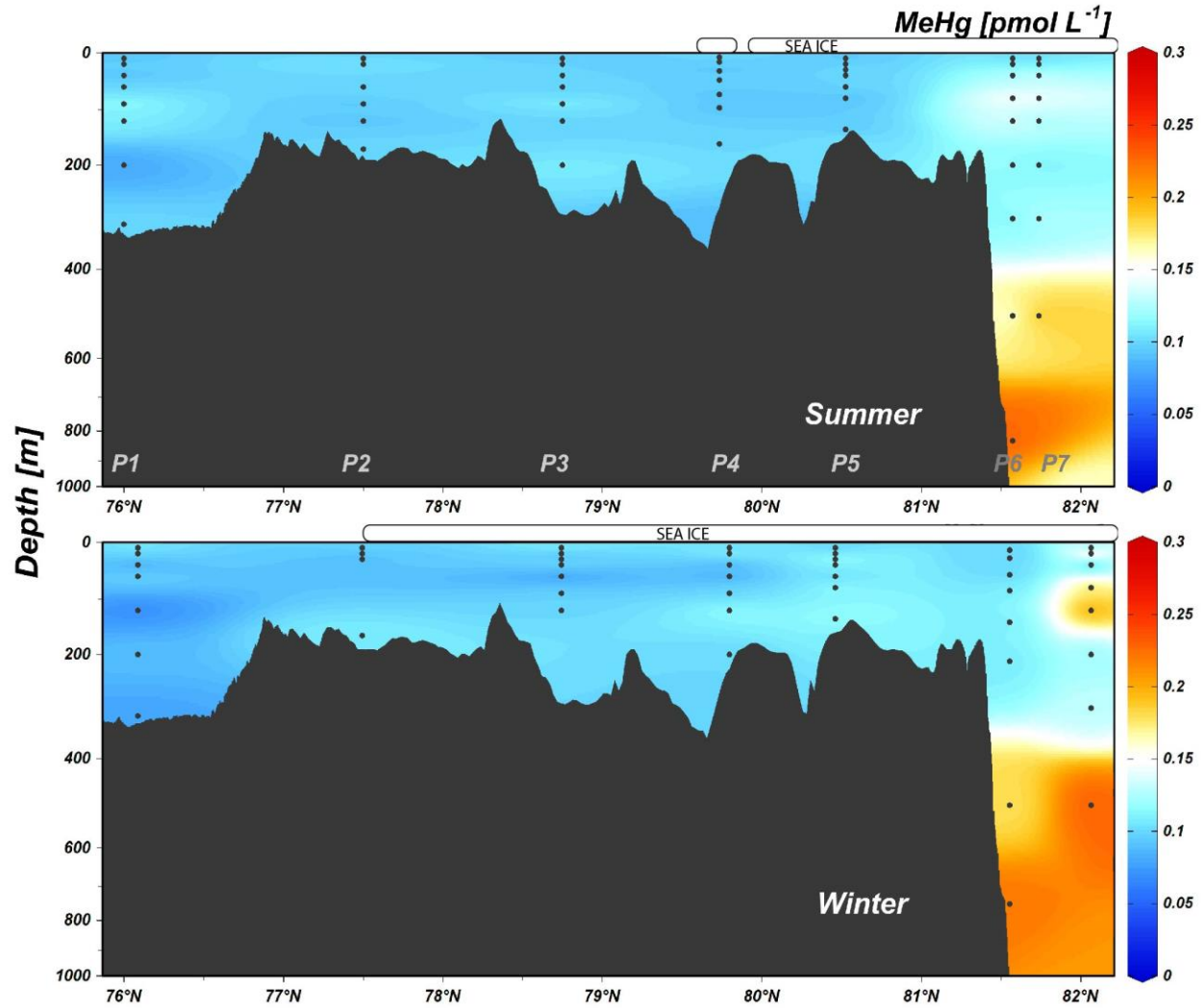
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237

238 **Figure 3: Δ iHg ($\text{pmol m}^{-2} \text{d}^{-1}$) and Δ MeHg ($\text{pmol m}^{-2} \text{d}^{-1}$) at specified depth intervals for each**
239 **station.** iHg 0-100 m is in light orange, iHg 100-200 m in dark orange, MeHg 0-100 m in light
240 blue, and MeHg 100-200 m in dark blue. A positive Δ value indicates a temporal gain in the iHg
241 or MeHg pool while a negative Δ value indicates a temporal loss in the iHg or MeHg pool. Error
242 bars represent combined standard uncertainty ($\pm 1\sigma$). Values associated with Figure 3 are compiled
243 in SI Table 1. *Stations P2 and P5 were integrated from 100 m to sample depth less than 200 m.



244

245 **Figure 4: Total methylated mercury (MeHg) concentrations (pmol L^{-1}) along the shelf-deep**
 246 **basin gradient.** Stations P1 – P7, (Latitude) are on the x-axis for summer 2019 (top) and winter
 247 2019 cruises (bottom).

248

249

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371 **METHODS**

372 **Sample collection**

373 The transect in the northern Barents Sea and Nansen Basin (76°N – 83°N) was sampled on two
374 cruises, Q3 and Q4, in August 2019 and December 2019 on *R/V Kronprins Haakon* as part of The
375 Nansen Legacy project. Water samples were collected manually using Teflon-coated GOFLO
376 bottles attached sequentially to a 600 m Aramid rope. Seawater was sampled at seven stations at 8
377 depths up to 500 m. Depths deeper than 500 m were sampled with 10 L Niskin bottles mounted to
378 a rosette with a SeaBird SBE 9 CTD unit. Upon GOFLO retrieval, bottles were transported
379 immediately to an onboard custom-made clean lab. High-efficiency particulate air filter cartridges
380 were attached to the pressure release valve. THg samples were collected unfiltered with no
381 headspace into pre-cleaned⁵¹ 40 mL borosilicate glass bottles with PTFE-lined caps and kept
382 unpreserved. MeHg samples were collected unfiltered into brand new 125 mL PET bottles and
383 were acidified to 0.4 % v/v double-distilled HCl under a Class 100 clean air laminar flow hood.
384 Upon acidification, dimethylmercury (DMHg) converts quantitatively to monomethylmercury
385 (MMHg)⁵²; thus, total methylated mercury (MeHg) is measured as the sum of MMHg and DMHg.
386 All samples were double-bagged and stored in the dark at 4°C until analysis.

387

388 **Hg measurements**

389 The THg concentration in the samples was measured according to USEPA Method 1631 at the
390 Mediterranean Institute of Oceanography (MIO) using a custom-made single gold trap setup
391 described elsewhere²¹ within six months after sampling. To adapt to the low concentrations
392 expected in seawater, potassium bromide (Sigma Aldrich, USA) and potassium bromate (Sigma
393 Aldrich, USA) are heated for 4h at 250°C to remove Hg traces before making up BrCl solution

394 with freshly double-distilled HCl. The BrCl solution was blank checked prior to use using a
395 standard addition protocol. Field blanks were measured for both summer (0.01 pmol L^{-1} , $n = 4$)
396 and winter (0.09 pmol L^{-1} , $n = 3$) cruises. Average THg concentrations are reported in the main
397 text plus the standard deviation of the mean ($\pm 1\text{SD}$).

398
399 THg measurements were calibrated against NIST 3133 (National Institute of Standards and
400 Technology), with a 6-point calibration curve in Hg-free seawater (R^2 values of $0.9998 - 1$),
401 accounting for matrix effects. A NIST quality control sample was spiked into previously purged
402 Hg-free sample seawater matrix and analyzed after every 6 samples (standard-bracketing) to
403 monitor instrumental drift. Initial recovery and precision of this quality control sample for our
404 batches is $94 \pm 7\%$. Ongoing recovery of this quality control sample was monitored and checked
405 against the recovery guidelines outlined in EPA Method 1631, Revision E, Table 2 ($77 - 123\%$).
406 The detection limit of $0.003 \text{ pmol L}^{-1}$ was evaluated as $3x$ the standard deviation of the
407 instrumental CVAFS “bubbler” blank. To test our analytical method, certified reference materials
408 ORMS-5 and BCR-579 were spiked into previously purged Hg-free seawater matrix to test for
409 recovery. Both standards were within the certified ranges, with additional analytical details
410 provided in Supplemental Information.

411
412 The MeHg concentration in seawater samples was measured using the isotope dilution (ID)
413 technique via gas chromatography – sector field ICP-MS (GC-SF-ICP-MS)²¹ at MIO within six
414 months after sampling. Briefly, enriched isotopic spikes of $i^{199}\text{Hg}$ and MM^{201}Hg (ISC Science)
415 were added to approximately 110 mL of seawater sample in pre-combusted borosilicate glass
416 bottles and equilibrated for 15 minutes. Samples were adjusted to pH 3.9 using a sodium acetate

417 buffer solution (ULTREX® II Ultrapure Reagent, J.T. Baker, USA) and NH₃ (ULTREX® II
 418 Ultrapure Reagent, J.T. Baker, USA). A freshly made 1% solution of sodium tetrapropylborate
 419 (Merseburger Spezialchemikalien) and 200 µL of hexane (Sigma Aldrich) were added to derivatize
 420 and extract Hg species. Bottles were sealed and shaken for 15 minutes before the organic layer
 421 was transferred to GC vials for injection on a coupled GC (THERMO GC 1300 with GC220
 422 transfer module) SF-ICP-MS (Thermo Element XR) system. The coupling to a high resolution
 423 ICP-MS and application of ultra-trace clean techniques allows reaching a detection limit of 0.001
 424 pmol L⁻¹. We determined that the accuracy of our MM²⁰¹Hg spike concentration compared to its
 425 theoretical concentration was 96.6 ± 7.9%, calculated with reverse isotope dilution using the
 426 Brooks Rand MeHgCl standard, traceable to NIST1641E. Additional analytical details are
 427 provided in Supplemental Information. Average MeHg concentrations are reported in the main
 428 text plus the standard deviation of the mean (±1SD).

429

430 **Δ calculations**

431 We calculate iHg for each depth as the difference between THg and MeHg. iHg and MeHg in the
 432 upper 200 m water column at each station were depth integrated using the trapezoidal method.
 433 ΔMeHg (Equation 1) and ΔiHg (Equation 2) were calculated from summer (s) to winter (w) using
 434 the specific sampling dates. We assumed the same concentrations of THg and MeHg at 0 m until
 435 observations begin at 10 m. When sampling depths differ or are missing, concentrations were
 436 extrapolated by linear regression from the closest available sampling depths to calculate Δ values.

437

$$438 \quad \Delta MeHg = \left(\int_0^z [MeHg]_s dz - \int_0^z [MeHg]_w dz \right) / dt \quad (1)$$

$$439 \quad \Delta iHg = \left(\int_0^z ([THg]_s - [MeHg]_s) dz - \int_0^z ([THg]_w - [MeHg]_w) dz \right) / dt \quad (2)$$

440

441 A positive Δ value indicates a temporal gain in iHg and MeHg while a negative Δ value indicates
442 a temporal loss in iHg and MeHg. To compare directly to published fluxes, we present absolute
443 values of Δ iHg and Δ MeHg as $|\Delta$ iHg| and $|\Delta$ MeHg|. Uncertainty in Δ values is reported as
444 combined standard uncertainty ($\pm 1\sigma$) calculated from the uncertainty in measured THg
445 concentrations and uncertainty in measured MeHg concentrations.

446 **Data availability statement**

447 Figures 1, 2, and 4 and Extended Data Figures 1-5 and 7 were created by the publicly available
448 Ocean Data View⁵⁵ and edited in Adobe Illustrator. Temperature and salinity data from seasonal
449 cruises are publicly available from the Norwegian Marine Data Centre (<https://nmdc.no>)^{53,54}.
450 Mercury and trace element concentration data for all depths and stations are publicly available
451 from the Norwegian Marine Data Centre (<https://nmdc.no>)^{56,57}. Figure 3 and Extended Data
452 Figures 6 and 8 were created in Microsoft Excel with data displayed in Supplementary Tables 1
453 and 3.

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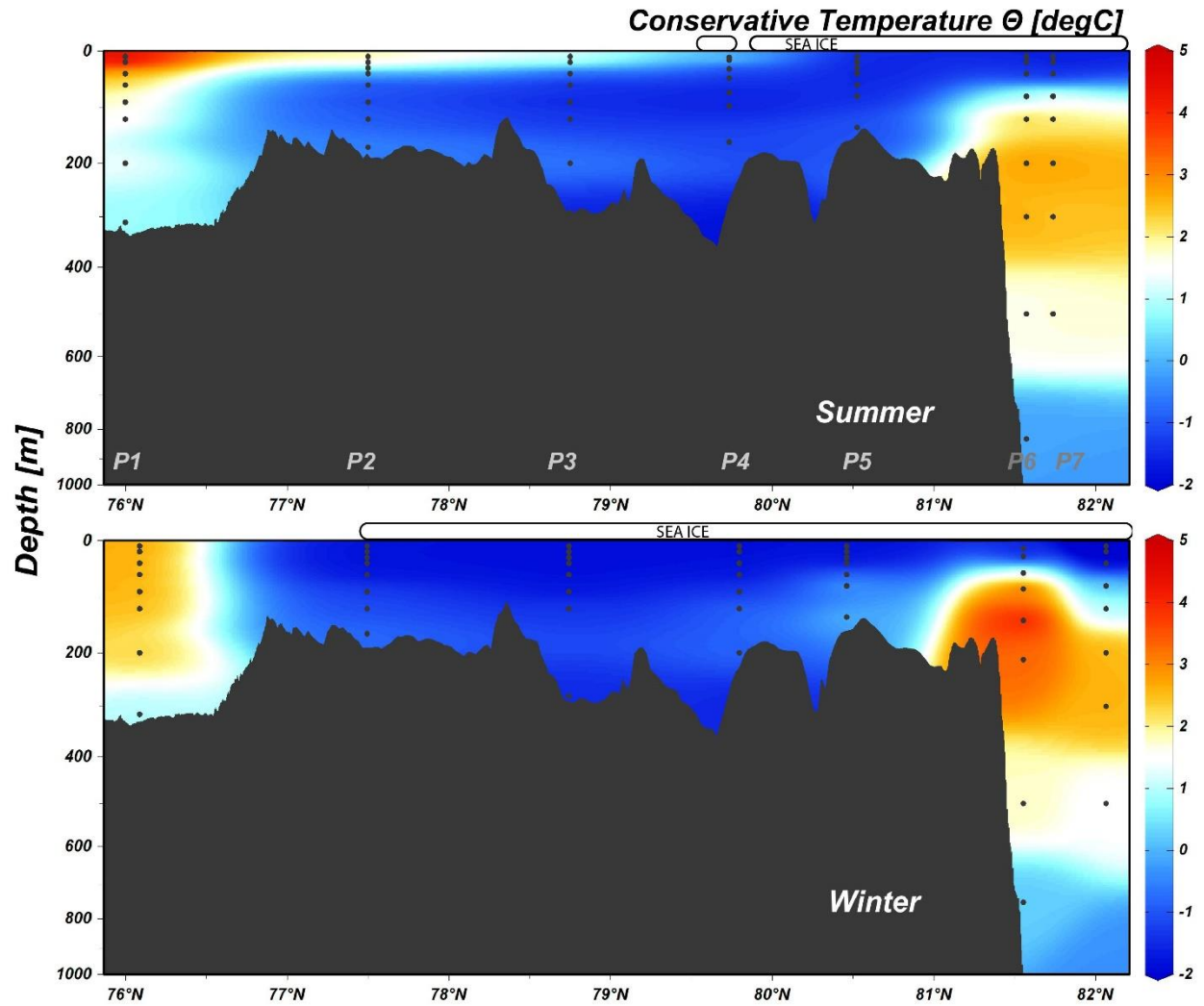
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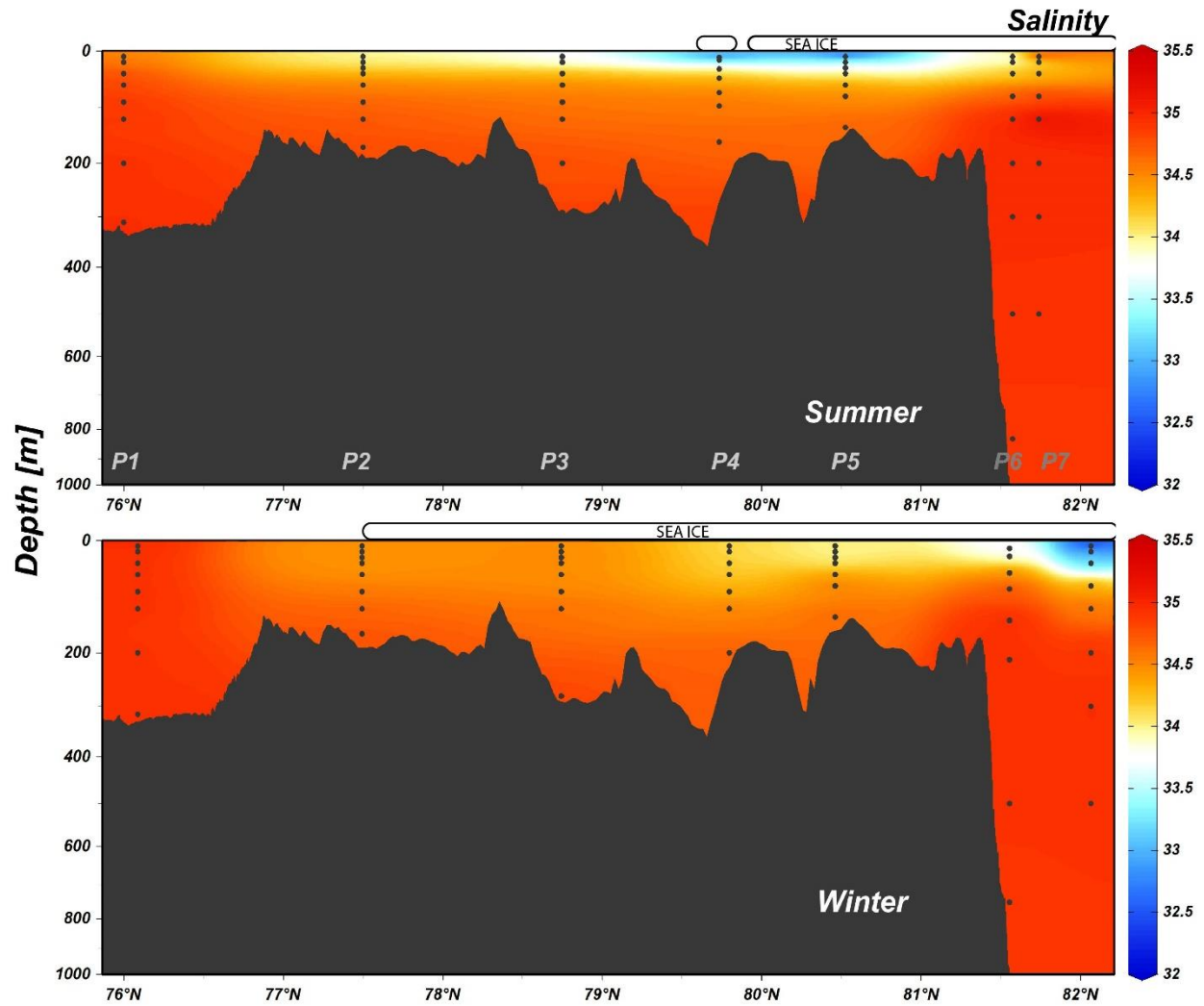
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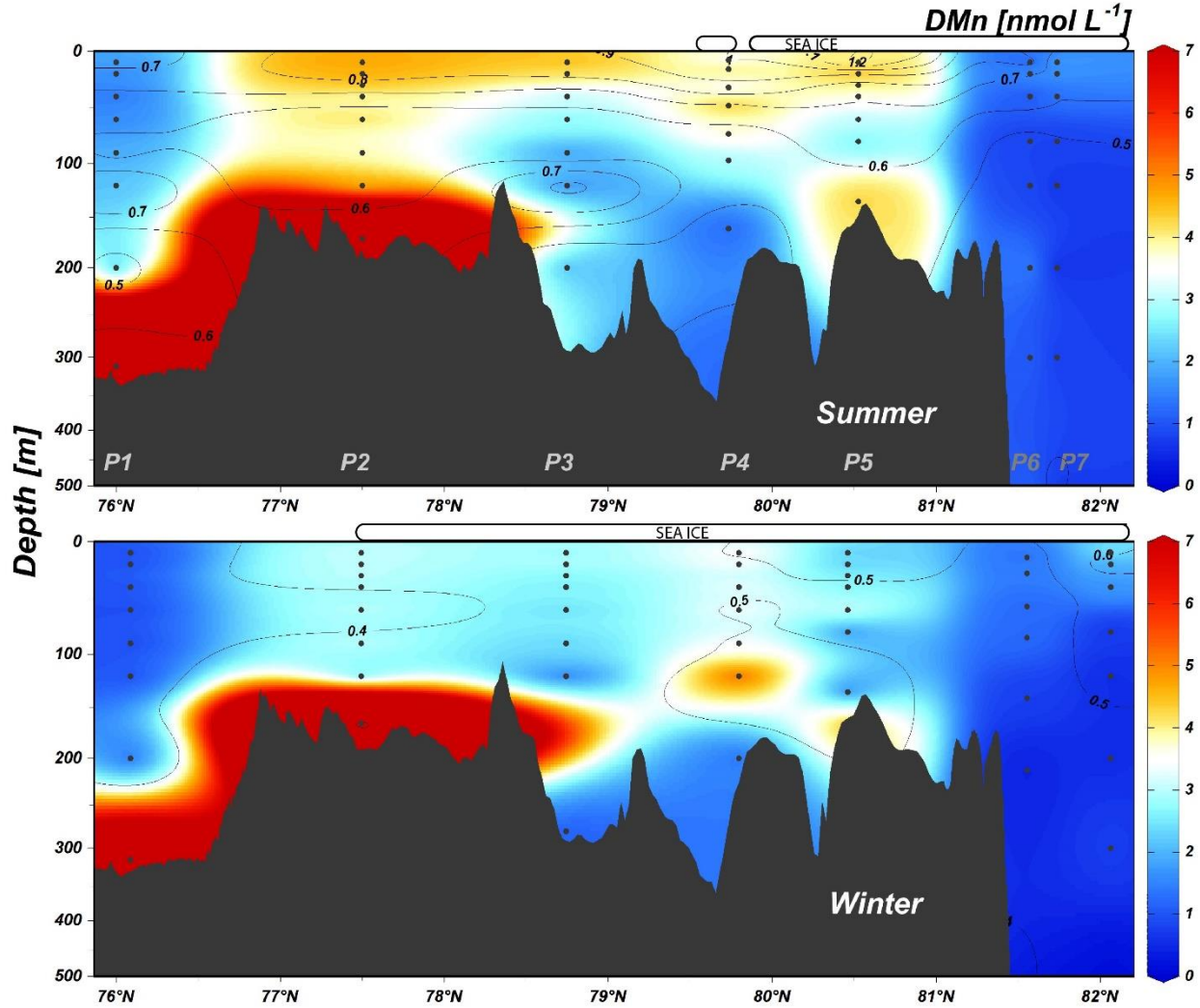
478

479 **Extended Data Fig. 1. Conservative temperature (Θ) in degrees Celsius ($^{\circ}\text{C}$) along the shelf-**
 480 **deep basin gradient.** Stations P1 – P7, (Latitude) are on the x-axis for summer 2019 (top) and
 481 winter 2019 cruises (bottom). Data retrieved from the Norwegian Marine Data Centre^{53,54}.



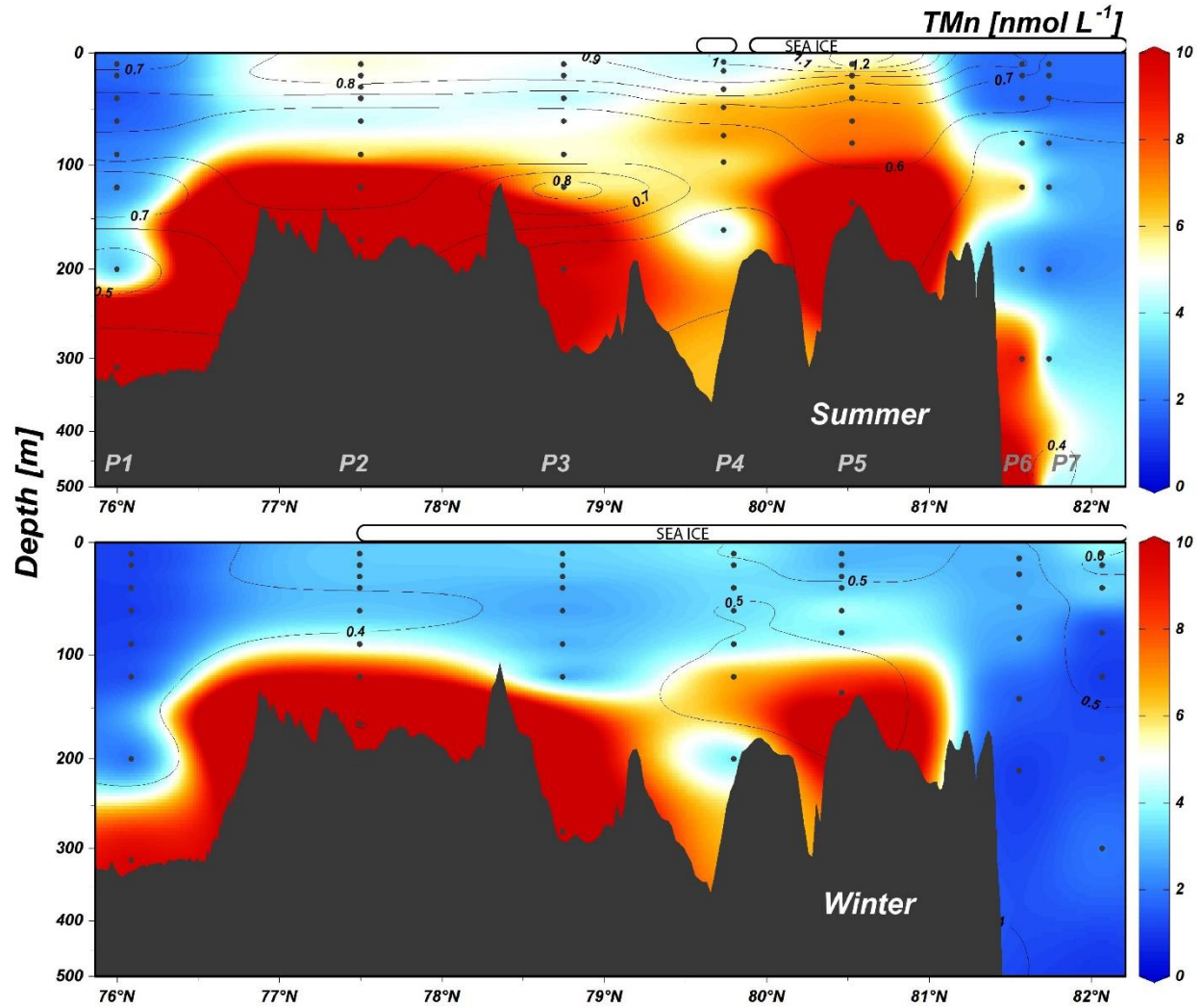
482

483 **Extended Data Fig. 2. Salinity along the shelf-deep basin gradient.** Stations P1 – P7, (Latitude)
 484 are on the x-axis for summer 2019 (top) and winter 2019 cruises (bottom). Data retrieved from the
 485 Norwegian Marine Data Centre^{53,54}.



486

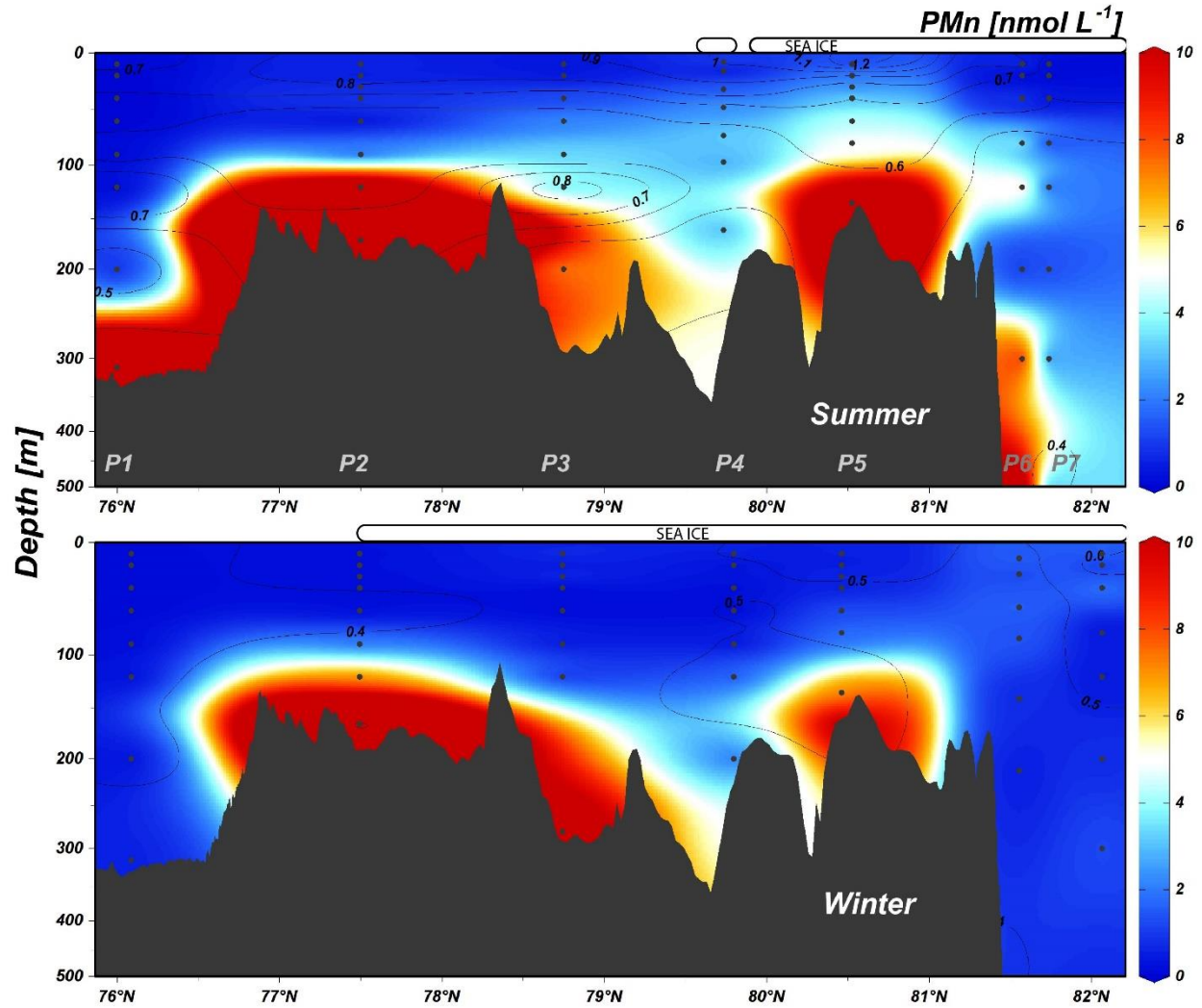
487 **Extended Data Fig. 3. Dissolved manganese (DMn) concentrations (nmol L^{-1}) along the shelf-**
 488 **deep basin gradient.** Stations P1 – P7, (Latitude) are on the x-axis for summer 2019 (top) and
 489 winter 2019 cruises (bottom). Black contour lines for THg (pmol L^{-1}) from each corresponding
 490 season are overlain. Concentrations greater than the presented range (7 nmol L^{-1}) are plotted as the
 491 maximum.



492

493 **Extended Data Fig. 4. Total acid-leachable manganese (TMn) concentrations (nmol L⁻¹)**
 494 **along the shelf-deep basin gradient.** Stations P1 – P7, (Latitude) are on the x-axis for summer
 495 2019 (top) and winter 2019 cruises (bottom). Black contour lines for THg (pmol L⁻¹) from each
 496 corresponding season are overlain. Concentrations greater than the presented range (10 nmol L⁻¹)
 497 are plotted as the maximum.

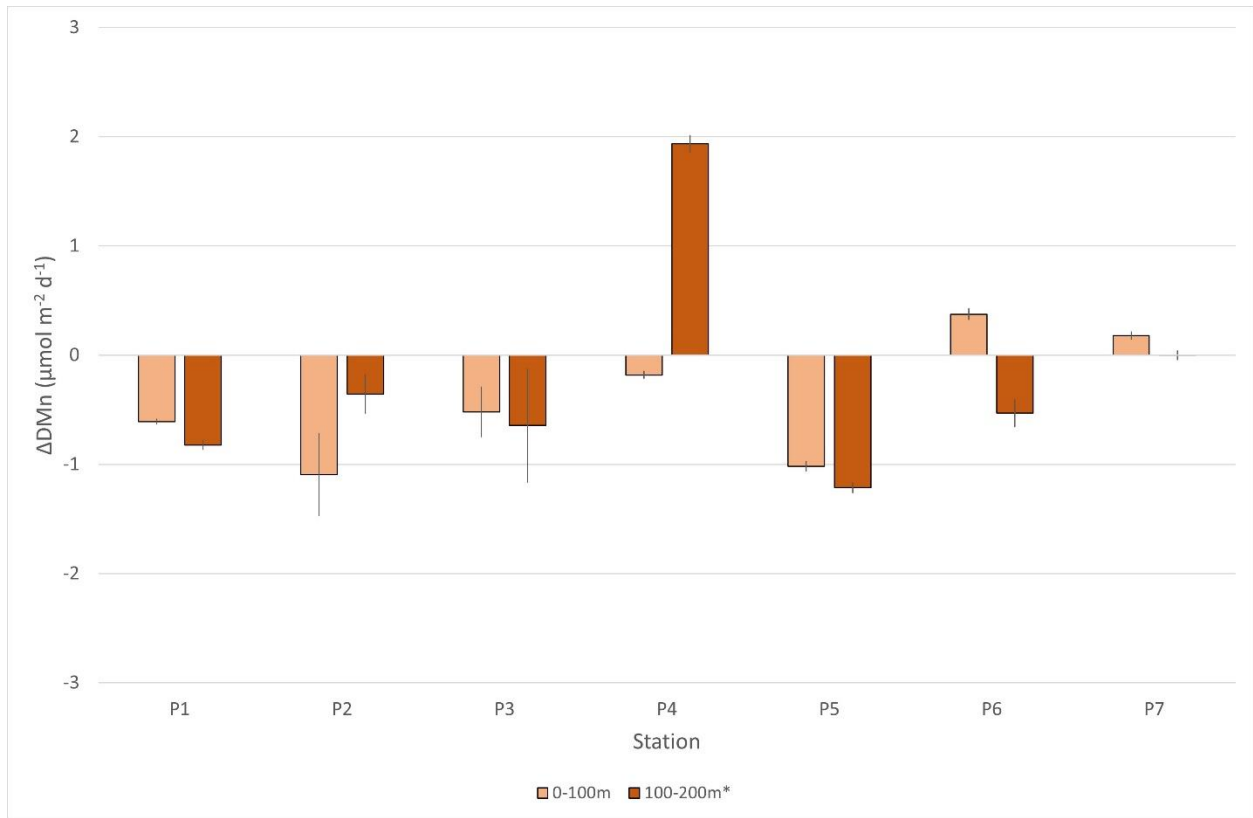
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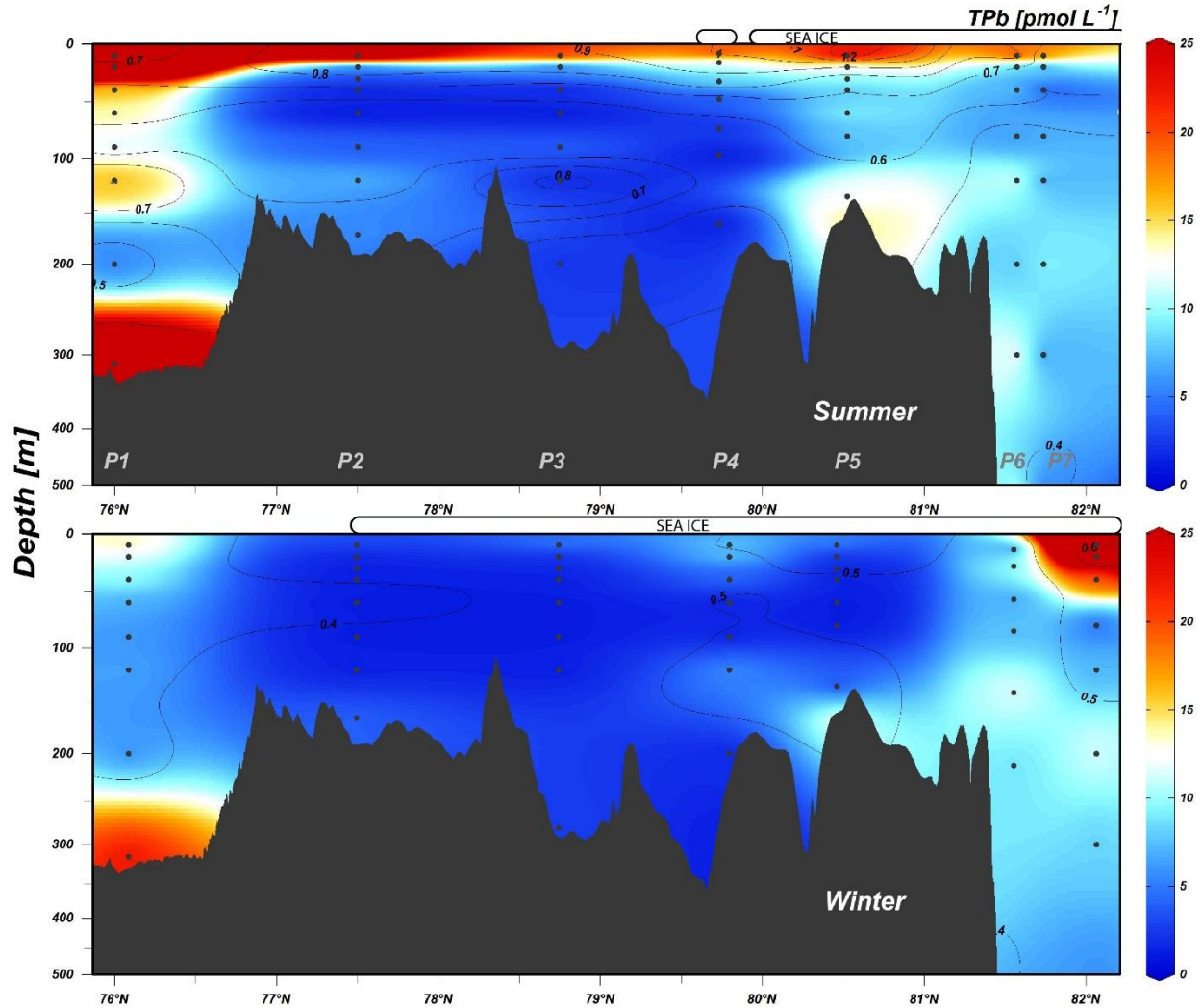
500 **Extended Data Fig. 5. Particulate manganese (PMn) concentrations (nmol L⁻¹) along the**
 501 **shelf-deep basin gradient.** Stations P1 – P7, (Latitude) are on the x-axis for summer 2019 (top)
 502 and winter 2019 cruises (bottom). Black contour lines for THg (pmol L⁻¹) from each corresponding
 503 season are overlain. Concentrations greater than the presented range (10 nmol L⁻¹) are plotted as
 504 the maximum.

505



506

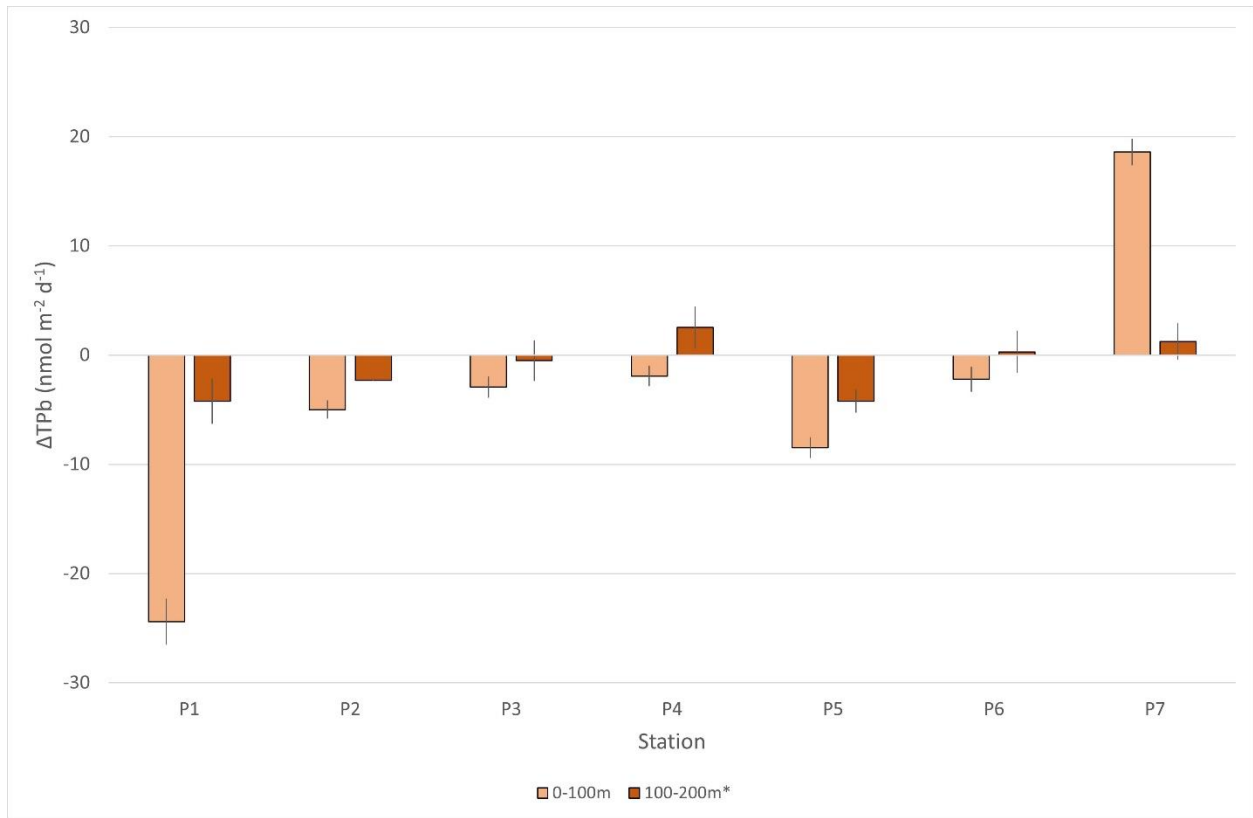
507 **Extended Data Fig. 6. Δ DMn ($\mu\text{mol m}^{-2} \text{d}^{-1}$) at specified depth intervals for each station. A**
 508 **positive Δ value indicates a temporal gain in the DMn pool while a negative Δ value indicates a**
 509 **temporal loss in the DMn pool. Δ values are reported with error bars as combined standard**
 510 **uncertainty ($\pm 1\sigma$). *Stations P2 and P5 were integrated from 100 m to sample depth less than 200**
 511 **m.**



512

513 **Extended Data Fig. 7. Total acid-leachable lead (TPb) concentrations (pmol L^{-1}) along the**
 514 **shelf-deep basin gradient.** Stations P1 – P7, (Latitude) are on the x-axis for summer 2019 (top)
 515 and winter 2019 cruises (bottom). Data points at or below the detection limit were assigned the
 516 value of the detection limit (1.08 pmol L^{-1}) determined by the seaFAST-pico ICP-MS for plotting
 517 purposes. Black contour lines for THg (pmol L^{-1}) from each corresponding season are overlain.
 518 Concentrations greater than the presented range (25 pmol L^{-1}) are plotted as the maximum.

519



520

521 **Extended Data Fig. 8. Extended Data Figure 8: Δ TPb (nmol m⁻² d⁻¹) at specified depth**
 522 **interval for each station.** A positive Δ value indicates a temporal gain in the TPb pool while a
 523 negative Δ value indicates a temporal loss in the TPb pool. Δ values are reported with error bars
 524 as combined standard uncertainty ($\pm 1\sigma$). *Stations P2 and P5 were integrated from 100 m to
 525 sample depth less than 200 m.

526

527