

FRONT MATTER

Title

Mid-Holocene Antarctic sea-ice increase driven by marine ice sheet retreat

Authors

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30

31 **1. ABSTRACT**

32 Over recent decades Antarctic sea-ice extent has increased, alongside widespread ice shelf thinning and
33 freshening of waters along the Antarctic margin. In contrast, Earth system models generally simulate a
34 decrease in sea ice. Circulation of water masses beneath large cavity ice shelves is not included in current
35 Earth System models and may be a driver of this phenomena. We examine a Holocene sediment core off
36 East Antarctica that records the Neoglacial transition, the last major baseline shift of Antarctic sea-ice,
37 and part of a late-Holocene global cooling trend. We provide a multi-proxy record of Holocene glacial
38 meltwater input, sediment transport and sea-ice variability. Our record, supported by high-resolution
39 ocean modelling, shows that a rapid Antarctic sea-ice increase during the mid-Holocene (~4.5 ka) occurred
40 against a backdrop of increasing glacial meltwater input and gradual climate warming. We suggest that
41 mid-Holocene ice shelf cavity expansion led to cooling of surface waters and sea-ice growth which slowed
42 basal ice shelf melting. Incorporating this feedback mechanism into global climate models will be
43 important for future projections of Antarctic changes.

44

45 **2. INTRODUCTION**

46 Ice shelves and sea ice are intrinsically linked and represent fundamental components of the global
47 climate system, impacting ice-sheet dynamics, large-scale ocean circulation, and the Southern Ocean
48 biosphere. Antarctic ice-shelves with large sub-shelf cavities (e.g. Ross, Filchner-Ronne) play a key role
49 in regional sea-ice variations, by cooling and freshening surface ocean waters for hundreds of kilometres
50 beyond the ice shelf edge (Hellmer, 2004; Hughes *et al.*, 2014). Antarctic sea ice has expanded over the
51 past few decades, particularly in the western Ross Sea region (Turner *et al.*, 2016), alongside widespread
52 thinning of ice shelves (Paolo *et al.*, 2015) and freshening along the Antarctic margin (Jacobs *et al.*,
53 2002; Aoki *et al.*, 2013). The drivers and feedbacks involved in these decadal trends are still poorly
54 understood, hampered by the sparse and short-term nature of meteorological, oceanographic and
55 glaciological observations (Jones *et al.*, 2016), and thus establishing the long-term trajectory for
56 Antarctic sea ice on the background of accelerated ice sheet loss remains a challenge. Marine sediment
57 cores provide a longer-term perspective and highlight a major baseline shift in East Antarctic coastal sea
58 ice ~4.5 ka ago (Steig *et al.*, 1998; Crosta *et al.*, 2008; Denis *et al.*, 2010) which characterizes the mid-
59 Holocene ‘Neoglacial’ transition in the Antarctic. A mechanistic driver for this climate shift currently

60 remains unresolved, but we propose that two interrelated aspects of the last deglaciation are significantly
61 underrepresented in current models of this transition: (i) the retreat of grounded ice sheets from the
62 continental shelves of Antarctica, and (ii) the subsequent development of large ice shelf cavities during
63 the Holocene. Both factors would significantly alter water mass formation on Antarctica's continental
64 shelves, which today are major source regions of Antarctic Bottom Water (AABW) and Antarctic
65 Surface Water (AASW). These interrelated processes are underrepresented in coupled ocean-atmosphere
66 models which currently do not simulate the timing, magnitude and rapid onset of the Neoglacial
67 (Supplementary Materials).

68
69 Integrated Ocean Drilling Program (IODP) Expedition 318 cored a 171 m thick deposit of laminated
70 diatomaceous ooze at Site U1357 offshore Adélie Land (Fig. 1), deposited over the past 11,400 years.
71 Here, we present a new Holocene record of glacial meltwater, sedimentary input and local sea ice
72 concentrations from Site U1357 using compound-specific hydrogen isotopes of fatty acid biomarkers
73 ($\delta^2\text{H}_{\text{FA}}$), terrigenous grain size, natural gamma radiation, biogenic silica accumulation, highly-branched
74 isoprenoid alkenes (HBIs) and Ba/Ti ratios (Fig. 4 and 5).

75
76 We interpret $\delta^2\text{H}_{\text{FA}}$ (Fig. 4) fluctuations in Adélie Drift sediments as a record of meltwater input from
77 isotopically-depleted glacial ice. Antarctic glacial ice is highly depleted in ^2H compared to ocean water,
78 thus creating highly contrasting end-member values for the two major H source pools. Grain size,
79 natural gamma radiation (NGR) and terrigenous and biosiliceous mass accumulation rates (MARs)
80 reflect changing sediment delivery either driven via local glacial meltwater discharge or advection of
81 suspended sediment by oceanic currents. The diene/triene HBI ratio is used as a proxy for coastal sea ice
82 presence (Massé *et al.*, 2011). Ba/Ti enrichment is considered to reflect enhanced primary productivity.
83 These records allow a unique opportunity to reconstruct the magnitude of the coupled response of the
84 ocean and ice sheet during the Neoglacial transition. Details on all proxies and associated uncertainties
85 can be found in Section S2 of the Supplementary Information.

86 87 **3. MATERIALS AND METHODS**

88 89 **3.1 Organic geochemical analyses**

90 **3.1.1 Fatty acid extraction**

91 Lipid extraction of sediment samples was performed at the Royal Netherlands Institute for Sea Research
92 (NIOZ). Freeze-dried and homogenized samples were extracted by Dionex™ accelerated solvent
93 extraction (DIONEX ASE 200) using a mixture of dichloromethane (DCM)/methanol (MeOH) (9:1,
94 v/v) at a temperature of 100°C and a pressure of 7.6×10^6 Pa (Kim *et al.*, 2010).

95
96 Compound separation was undertaken at University of Glasgow, UK. The total lipid extract was
97 separated over an aminopropyl silica gel column and the total acid fraction was eluted into an 8ml vial
98 with 4% acetic acid in ethyl-ether solution (Huang *et al.*, 1999). Derivatisation to Fatty Acid Methyl
99 Esters was achieved by adding 200 µl of MeOH containing 14% v/v Boron trifluoride to the 8ml vial
100 containing the TAF. FAMES were recovered and cleaned up by eluting through a pre-cleaned 3cm silica
101 gel column (60 Å; 35-70) with 4ml of hexane and 4ml of DCM (containing the FAMES). $\delta^2\text{H}$ values
102 indicate depletion against the international standards: Vienna Pee Dee Belemnite (V-PDB) is the
103 standard for $\delta^{13}\text{C}$ and Vienna Standard Mean Ocean Water (V-SMOW) for $\delta^2\text{H}$.

104

105 3.1.2 Fatty acid hydrogen isotope analysis

106 Compound specific hydrogen isotope analyses of FAMES was performed at the Institute of Low
107 Temperature Science, Hokkaido University. $\delta^2\text{H}$ values were obtained using a CS-IRMS system with a
108 HP 6890 gas chromatograph and a ThermoQuest Finnigan MAT Delta Plus XL mass spectrometer.
109 Separation of the FAMES was achieved with a HP-5 MS fused silica capillary column (30 m x 0.32 mm
110 i.d., film thickness of 0.25 µm) with a cooled on-column injector. An *n*-alkane and a reference gas
111 whose isotopic values were known was co-injected with the samples as an internal isotopic standard for
112 $\delta^2\text{H}$. A laboratory standard (Mix F8 of FAMES from Indiana University) containing C₁₀–C₃₀ FAMES
113 was analyzed daily to check the accuracy and the drift of the instrument and to normalize the data to the
114 SMOW/SLAP isotopic scale. The H^{3+} factor was measured every three days.

115

116 3.1.3 HBIs

117 Highly branched isoprenoids (HBI) alkenes were extracted at Laboratoire d'Océanographie et du
118 Climat: Experimentations et Approches Numériques (LOCEAN), separately from the fatty acids, using a
119 mixture of 9mL CH₂Cl₂/MeOH (2:1, v:v) to which 7 hexyl nonadecane (m/z 266) was added as an
120 internal standard, following the Belt *et al.* (2007) and Massé *et al.* (2011) protocols. Several sonication
121 and centrifugation steps were applied in order to properly extract the selected compounds (Etourneau *et*

122 *al.*, 2013). After drying with N₂ at 35°C, the total lipid extract was fractionated over a silica column into
123 an apolar and a polar fraction using 3 mL hexane and 6 mL CH₂Cl₂/MeOH (1:1, v:v), respectively. HBIs
124 were obtained from the apolar fraction by the fractionation over a silica column using hexane as eluent
125 following the procedures reported by Belt *et al.* (2007; Massé *et al.*, 2011). After removing the solvent
126 with N₂ at 35°C, elemental sulfur was removed using the TBA (Tetrabutylammonium) sulfite method
127 (Jensen *et al.*, 1977; Riis and Babel, 1999). The obtained hydrocarbon fraction was analyzed within an
128 Agilent 7890A gas chromatograph (GC) fitted with 30 m fused silica Agilent J&C GC column (0.25 mm
129 i.d., 0.25 µm film thickness), coupled to an Agilent 5975C Series mass selective detector (MSD).
130 Spectra were collected using the Agilent MS-Chemstation software. Individual HBIs were identified on
131 the basis of comparison between their GC retention times and mass spectra with those of previously
132 authenticated HBIs (Johns *et al.*, 1999) using the Mass Hunter software. Values are expressed as
133 concentration relative to the internal standard.

134 135 **3.2 Inorganic geochemical analysis and electronic microscopy**

136 Major element concentrations were obtained using X-Ray Fluorescence Scanner on 412 analyses
137 measured directly over undisturbed sediment sections. The bulk major element composition included in
138 this study was measured between sections U1357B-1H-2 to U1357-19H-5 continuously each 50 cm. We
139 used an Avaatech X-ray fluorescence (XRF-Scanner) core scanner at the IODP-Core Repository/Texas
140 A&M University laboratories (USA) during December 2010. Non-destructive XRF core-scanning
141 measurements were performed over 1 cm² area with slit size of 10 mm, a current of 0.8 mA and
142 sampling time of 45 seconds at 10 kV in order to measure the relative content of titanium (Ti) and
143 barium (Ba).

144
145 Field emission scanning electron microscopy (FESEM) images and corresponding spectrum were
146 obtained with an AURIGA FIB-FESEM Carl Zeiss SMT at Centro de Instrumentación Científica,
147 Granada University, Spain

148 149 **3.3 Grain size analyses**

150 A total of 341 samples were prepared for grain size analysis. Samples were treated for removal of
151 biogenic opal with a 1M sodium hydroxide NaOH solution and incubated in a water bath at 80°C for 24
152 hours. This procedure was repeated twice due to an incomplete dissolution of diatoms observed in smear

153 slides. The samples were then treated with H₂O₂ to remove organic material at 80°C for 24 hours.
154 Samples were measured using a Beckman Coulter LS 13 320 Laser Diffraction Particle Size Analyser
155 (LPSA). Prior to grain size analysis, ~30 mL of 0.5 g/L Calgon (sodium hexametaphosphate) was added
156 to the samples, and sonicated and stirred in order to disperse the grains and prevent clumping.

158 **3.4 Biogenic silica**

159 Biogenic silica concentrations (wt% BSi) were measured on 349 discrete samples using a molybdate
160 blue spectrophotometric method modified from (Strickland and Parsons, 1970; DeMaster, 1981).
161 Analytical runs included replicates from the previous sample group and from within the run, and each
162 run was controlled by 10 standards and a blank with dissolved silica concentrations ranging from 0 µM
163 to 1200 µM. For each analysis, ~7 mg of dry, homogenized sediment was leached in 0.1M NaOH at
164 85°C, and sequential aliquots were collected after 2, 3, and 4 hours. Following addition of reagents,
165 absorbance of the 812 nm wavelength was measured using a Shimadzu UV-1800 spectrophotometer.
166 Dissolved silica concentration of each unknown was calculated using the standard curve, and data from
167 the three sampling hours were regressed following the method of DeMaster (1981) to calculate wt%
168 BSi. In our U1357B samples, wt% BSi ranges from maximum of ~60% in early and mid-Holocene light
169 laminae to a minimum of 31% in late Holocene dark laminae. The average standard deviation of
170 replicate measurements is 0.5%.

172 **3.5 Model simulations**

173 All numerical calculations were performed using the Massachusetts Institute of Technology general
174 circulation model (MITgcm) (Marshall *et al.*, 1997); a three-dimensional, ocean sea-ice, hydrostatic,
175 primitive equation model. The experiments presented here were integrated on a global domain projected
176 onto a cube-sphere grid to permit a relatively even grid spacing and to avoid polar singularities (Adcroft
177 *et al.*, 2004; Condron and Winsor, 2012). The ocean grid has a mean, eddy-permitting, horizontal grid
178 spacing of 1/6° (18-km) with 50 vertical levels ranging in thickness from 10m near the surface to
179 approximately 450m at the maximum model depth. The ocean model is coupled to a sea-ice model in
180 which ice motion is driven by forces generated by the wind, ocean, Coriolis force, and surface elevation
181 of the ocean, while internal ice stresses are calculated using a viscous-plastic (VP) rheology, as
182 described in Zhang and Hibler (1997). In all experiments, the numerical model is configured to simulate
183 present-day (modern) conditions: Atmospheric forcings (wind, radiation, rain, humidity etc.) are

184 prescribed using 6-hourly climatological (1979-2000) data from the ERA-40 reanalysis product
185 produced by the European Centre for Medium-range Weather Forecasts and background rates of runoff
186 from the ice sheet to the ocean are based on the numerical ice sheet model of Pollard and Deconto
187 (2016) integrated over the same period (1979-2000). To study the pathway of meltwater in the ocean,
188 additional fresh (i.e. 0 psu) water was released into the surface layer of the ocean model at the grid
189 points closest to the front of the Ross Ice Shelf. Five different discharge experiments were performed by
190 releasing meltwater into this region at rates of 0.01 Sv ($Sv = 10^6 \text{ m}^3/\text{s}$), 0.05 Sv, 0.1 Sv, 0.5 Sv, and 1 Sv
191 for the entire duration of each experiment (~3.5 years).

192

193 **4. ENVIRONMENTAL SETTING AND INTERPRETATION OF PROXY DATA**

194 We utilize a 180 m thick sediment core that was recovered from the Wilkes Land Margin continental
195 shelf in the Adélie Basin (IODP Site U1357). This core targeted an expanded sediment drift (Adélie
196 Drift) and provides a high-resolution Holocene record of climate variability. Below we provide pertinent
197 details on this unique site and on our application of compound specific $\delta^2\text{H}$ measurements on algal
198 biomarkers as a novel meltwater proxy. Further details on proxy interpretation (Ba/Ti, grain size, HBIs)
199 are given in the Supplementary Materials.

200

201 **4.1 The Adélie Drift**

202 Site U1357 is located in the Dumont d'Urville Trough of the Adélie Basin, ca. 35 km offshore from
203 Adélie Land ($66^\circ 24.7990'S$, $140^\circ 25.5705'E$; Fig 1). This is a >1000 m deep, glacially scoured
204 depression on the East Antarctic continental shelf, bounded to the east by the Adélie Bank. Further east
205 lays the Adélie Depression and the Mertz Bank, the latter located north of the Mertz Glacier floating ice
206 tongue. The Adélie Land region is dissected by several glaciers which could potentially contribute
207 terrigenous sediment into the coastal zone with the core site located 40 km to the north of the Astrolabe
208 Glacier, and ca. 75 and 300 km northwest of the Zélée and Mertz glaciers, respectively.

209

210 The site itself is located within the Dumont d'Urville polynya (DDUP), which has a summer (winter)
211 extent of $13,020 \text{ km}^2$ (920 km^2), but is also directly downwind and downcurrent of the much larger and
212 highly productive Mertz Glacier polynya (MGP) to the east, with a summer (winter) extent of $26,600$
213 km^2 (591 km^2) (Arrigo and van Dijken, 2003). The MGP forms as a result of reduced sea-ice westward
214 advection due to the presence of the Mertz Glacier Tongue (Massom *et al.*, 2001) and strong katabatic

215 winds which blow off the Antarctic ice sheet with temperatures below -30°C (Bindoff *et al.*, 2000).
216 Katabatic winds freeze the surface waters and blow newly formed ice away from the coast, making the
217 polynya an efficient sea-ice ‘factory’, with higher rates of sea-ice formation in comparison to non-
218 polynya ocean areas which undergo seasonal sea ice formation (Kusahara *et al.*, 2010). The MGP
219 produces 1.3% of the total Southern Ocean sea ice volume despite occupying less than 0.1% of total
220 Antarctic sea ice extent (Marsland *et al.*, 2004).

221
222 As a result of the upwelling polynya environments, the area along the Adélie Coast is characterized by
223 extremely high primary productivity, with the water column known to host significant amounts of
224 phytoplankton, dominated by diatoms (Beans *et al.*, 2008). The Mertz Glacier zone is generally
225 characterized by stratified waters in the summer, due to seasonal ice melt, with these conditions
226 corresponding to the highest phytoplankton biomass. The lack of ice cover means polynyas are the first
227 polar marine systems exposed to spring solar radiation, making them regions of enhanced biological
228 productivity compared to adjacent waters. A considerable amount of resultant sedimentation is focused
229 via the westward flowing currents from both of these polynyas within the deep, protected Adélie Basin,
230 resulting in a remarkably high sedimentation rate of ca. $1.5\text{-}2\text{ cm year}^{-1}$ at Site U1357 (Escutia *et al.*,
231 2011).

232
233 Although biogenic and terrigenous sediment is interpreted to be sourced locally in the Adélie Land
234 region, the mass accumulation rate of these sediments in this drift is associated with the intensity of
235 westward flowing currents (S2.2). Critically, these westward currents also act to transport water masses
236 from further afield, and Site U1357 is directly oceanographically downstream of the Ross Sea, meaning
237 the continental shelf in this region receives significant Antarctic Surface Water (ASSW) transported by
238 the Antarctic Slope Current (ASC) and Antarctic Coastal Current from the Ross Sea embayment . Thus,
239 changes in the surface waters of the Ross Sea influence Site U1357. Whitworth *et al.* (1998) confirm the
240 continuity of the westward flowing ASC between the Ross Sea and the Wilkes Land margin. This flow
241 is largely associated with the Antarctic Slope Front, which reflects the strong density contrast between
242 AASW and Circumpolar Deep Water (CDW). McCartney and Donohue (2007) estimate that the
243 transport in the westward ASC, which links the Ross Sea to the Wilkes Land margin, reaches 76 Sv (Sv
244 = $10^6\text{ m}^3\text{ s}^{-1}$). However, Peña-Molino *et al.* (2016) measured a highly variable ASC flow at 113°E
245 ranging from 0 to 100 Sv with a mean of -21.2 Sv . This contributes to a cyclonic gyre, which together

246 with the ASC dominate the circulation at Site U1357. The gyre transport is around 35 Sv, and comes
247 mainly from the Ross Sea region, with a lesser contribution from a westward flow associated with the
248 Antarctic Circumpolar Current (McCartney and Donohue, 2007).

249 250 **4.2 Site specific interpretation of $\delta^2\text{H}_{\text{FA}}$ as a glacial meltwater proxy**

251 252 **4.2.1. Source of fatty acids**

253 To best interpret the hydrogen isotope signal recorded by the C_{18} FA, it is important to determine the
254 most likely source these compounds are derived from, and thus the habitat in which they are produced.
255 The C_{18} FA, however, is known to be produced by a wide range of organisms and so we cannot preclude
256 the possibility of multiple sources, especially in a highly diverse and productive region such as the
257 surface waters of offshore Adélie Land. However, we can attempt to determine the most dominant
258 producer(s), which will help us understand the main signal being recorded by the isotopes.

259
260 An analysis of the FAs within eight classes of microalgae by Dalsgaard *et al.* (2003) (compiling results
261 from multiple studies) showed *Cryptophyceae*, *Chlorophyceae*, *Prasinophyceae* and *Prymnesiophyceae*
262 to be the most dominant producers of total C_{18} FAs. The *Bacillariophyceae* class, on the other hand,
263 which includes the diatoms, were found to produce only minor amounts of C_{18} FA, instead synthesizing
264 abundant $\text{C}_{16:1}$ FAs. Thus, despite the water column offshore Adélie Land being dominated by diatoms,
265 these are unlikely to be a major source of the C_{18} FA within U1357B (Beans *et al.*, 2008; Riaux-Gobin
266 *et al.*, 2011).

267
268 Of the four microalgae classes dominating C_{18} production (Dalsgaard *et al.*, 2003), species from the
269 Chlorophyceae and Prymnesiophyceae classes have been observed within surface waters offshore
270 Adélie Land after spring sea-ice break-up (Riaux-Gobin *et al.*, 2011). Here, *Phaeocystis antarctica* of
271 the Prymnesiophytes was the most abundant of the surface water phytoplankton community
272 (representing 16% of the phytoplankton assemblage), whereas Cryptophyceae spp. were found in only
273 minor abundances (Riaux-Gobin *et al.*, 2011). In the Antarctic, *Phaeocystis* is thought to be the most
274 dominant producer of C_{18} FAs (Dalsgaard *et al.*, 2003), and thus is likely to be a key producer of the C_{18}
275 FA in U1357B samples.

276

277 To investigate this further, we measured compound-specific carbon isotopes of the C₁₈ FAs in U1357B
278 samples, which gives an average $\delta^{13}\text{C}$ value of $-29.8 \pm 1.0 \text{‰}$ (n=85). Budge *et al.* (2008) measured a
279 similar $\delta^{13}\text{C}$ value of $-30.7 \pm 0.8\text{‰}$ from C₁₆ FAs derived from Arctic pelagic phytoplankton, while sea
280 ice algae and higher trophic level organisms all had much higher $\delta^{13}\text{C}$ values (sea ice algae having
281 values of $-24.0 \pm 2.4\text{‰}$). Assuming similar values apply for the C₁₈ FA and for organisms within the
282 water column at our site, this suggests that our C₁₈ FA is predominantly derived from pelagic
283 phytoplankton.

284

285 Furthermore, $\delta^{13}\text{C}$ measurements of suspended particulate organic matter (SPOM) near Prydz Bay, East
286 Antarctica by Kopczynska *et al.* (1995) showed that sites with high $\delta^{13}\text{C}$ SPOM values (-20.1 to -
287 22.4‰) were characterized by diatoms and large heterotrophic dinoflagellates, whereas the lowest $\delta^{13}\text{C}$
288 SPOM values (-29.7 to -31.85‰) were associated with *Phaeocystis*, naked flagellates and autotrophic
289 dinoflagellates. Wong and Sackett (1978) measured the carbon isotope fractionation of seventeen
290 species of marine phytoplankton and showed that Haptophyceae (of which *Phaeocystis* belongs) had the
291 largest fractionation of -35.5‰ .

292

293 Therefore, based on the known producers of C₁₈ FAs, observed phytoplankton assemblages within
294 modern surface waters offshore Adélie Land, and the $\delta^{13}\text{C}$ value of C₁₈ FAs in U1357B samples, as
295 discussed above, we argue that the C₁₈ FA here is predominantly produced by *Phaeocystis* (most likely
296 *P. antarctica*), but with potential minor inputs from other algal species such as Cryptophytes or diatoms.

297

298 *Phaeocystis antarctica* is a major phytoplankton species within the Antarctic, dominating spring
299 phytoplankton blooms, particularly in the Ross Sea (DiTullio *et al.*, 2000; Schoemann *et al.*, 2005). It is
300 known to exist both within sea ice and in open water (Riaux-Gobin *et al.*, 2013) and has been observed
301 in surface waters in great abundance following spring sea-ice break-up, at both coastal and offshore sites
302 in Adélie Land (Riaux-Gobin *et al.*, 2011).

303

304 Although a large proportion of organic matter produced in the surface water is recycled in the upper
305 water column, the small fraction which is deposited in the sediment reaches the sea floor through large
306 particles sinking from above as “marine snow”. This export production includes large algal cells, fecal
307 pellets, zooplankton carcasses and molts, and amorphous aggregates (Mayer, 1993). In the Ross Sea,

308 aggregates of *P. antarctica*, have been observed to sink at speeds of more than 200 m day⁻¹, meaning
309 they could reach deep water very quickly (Asper and Smith, 1999). In this way, a proportion of the lipid
310 content of *P. antarctica* and other algae is transported and sequestered in the sediments.

311
312 Initial diagenesis is characterized by the preferential degradation of more labile organic compounds e.g.
313 sugars, proteins, amino acids. Proportionally, lipids are relatively recalcitrant compared to other
314 compounds (e.g. amino acids, proteins) and thus are more likely to be preserved as molecular
315 biomarkers on geological timescales, even where the rest of the organism may be completely degraded
316 (Peters and Moldowan, 1993). The final proportion of lipids that are preserved within sediments are
317 affected by factors including the export production, O₂ content, residence time in the water column and
318 at the sediment/water interface before deposition, molecular reactivity, formation of macromolecular
319 complexes, adsorption to mineral surfaces and bioturbation (Meyers and Ishiwatari, 1993; Killops and
320 Killops, 2004). Within lacustrine sediments, a significant shift in FA distributions has been shown to
321 occur within 100 years due to early diagenesis, after which the FA distribution remains relatively
322 unaffected by diagenesis (Matsuda, 1978), thus major changes are assumed to reflect primary
323 environmental signals on longer timescales such as in our Holocene record. Due to the
324 hyperproductivity of the surface waters offshore Adélie land, we assume the dominant inputs of the C₁₈
325 FA are from algal sources in overlying waters and upcurrent regions. Allochthonous inputs e.g. long-
326 range aeolian transport of plant material are assumed to be minimal.

327

328 **4.2.2. Interpretation of hydrogen isotopes**

329 Compound-specific H isotopes of algal biomarkers are a well-used climate proxy in sediments
330 throughout the Cenozoic (e.g. Pagani *et al.*, 2006; Feakins *et al.*, 2012). Although diagenetic alteration,
331 including H-exchange, is possible within sedimentary archives, this has shown to be minimal in
332 sediments younger than 20 Ma (Sessions *et al.*, 2004). Furthermore, if H-exchange had occurred, we
333 would expect $\delta^2\text{H}$ values between different FA chain lengths and closely spaced samples to be driven
334 towards homogeneity, yet large variability remains, suggesting this is not the case. Thus, we are
335 confident that our measured H isotopes are indicating a primary signal throughout the Holocene.

336

337 The $\delta^2\text{H}$ value preserved in biomarkers is known to be correlated, but offset, with the $\delta^2\text{H}$ of the water
338 from which the hydrogen was derived. Measured $\delta^2\text{H}$ can therefore be described as a function of either
339 the $\delta^2\text{H}$ of the water source, or the fractionation occurring between source water and the lipid ($\varepsilon_{l/w}$) (i.e.
340 vital effects), in which various environmental factors play a part (Sachse *et al.*, 2012).

341
342 The main environmental factors controlling $\varepsilon_{l/w}$ are salinity and temperature, with which $\delta^2\text{H}$ increases
343 by 1-4‰ per increase in practical salinity unit (psu) (Schouten *et al.*, 2006; Sachse *et al.*, 2012) and
344 decreases by 2-4‰ per degree C increase (Zhang *et al.*, 2009), respectively. The $\delta^2\text{H}_{\text{FA}}$ record from Site
345 U1357 displays an absolute range of ca. 123‰, and millennial to centennial scale variability with an
346 amplitude of ca. 50‰, throughout the core. This would imply extremely large and pervasive variations
347 in temperature (up to ca. 60°C) and salinity (up to 123 psu) if fractionation driven by either of these
348 factors were the main control. One study has shown the salinity of present day Adélie shelf waters to
349 vary between 34 and 34.8 psu (Bindoff *et al.*, 2000), while tetraether-lipid based subsurface (50-200 m)
350 temperature estimates from nearby Site MD03-2601 (about 50 km west of Site U1357) range from -0.17
351 to 5.35°C over the Holocene (Kim *et al.*, 2010). Therefore, fractionation changes driven by temperature
352 or salinity cannot be invoked as a major control on $\delta^2\text{H}_{\text{FA}}$ in the Holocene.

353
354 Thus, the most parsimonious explanation relates to changes in $\delta^2\text{H}_{\text{FA}}$ of the water source (Sachse *et al.*,
355 2012). In the Adélie Basin, the most apparent controls on this are advection, upwelling or inputs of
356 isotopically depleted glacial meltwater. The $\delta^2\text{H}_{\text{FA}}$ value within Antarctic glaciers is highly depleted
357 relative to sea water due to the Rayleigh distillation process, leading to highly negative isotope values
358 for precipitation over the continent.

359
360 The glacial meltwater originating from the Ross Ice Shelf is likely to combine ice precipitated
361 throughout the Holocene and glacial period, and from both the East and West Antarctic Ice Sheets.
362 However, as noted by Shackleton and Kennett (1975) in their first oxygen isotope record of the
363 Cenozoic (see their Fig. 6), most of the ice that melts around the margin has been coastally precipitated
364 (due to higher accumulation rates). Since ice precipitated further inland has a greater residence time
365 (Shackleton and Kennett, 1975) and significantly lower accumulation rates it will contribute
366 significantly less to this signal. Thus, the ice that was melting along this margin is best represented by
367 average values of coastal ice dome records at a similar latitude to that which melted since the LGM

368 (such as TALDICE and Siple Dome) than more southerly locations. Glacial to Holocene $\delta^2\text{H}_{\text{FA}}$ values
369 from TALDICE, located on the western edge of the Ross Sea in the East Antarctic, for example, vary
370 between -276.2 and -330.3‰ (Steig *et al.*, 1998) (converted from $\delta^{18}\text{O}$ values following the global
371 meteoric water line (GMWL): $\delta^2\text{H}_{\text{FA}} = 8.13 (\delta^{18}\text{O}) + 10.8$), while values from Siple Dome on the
372 eastern edge of the Ross Sea in the West Antarctic, vary from ca. -200 to -293‰ (Brook *et al.*, 2005).
373 Taking the average of these values as an approximate estimate for the meltwater gives a $\delta^2\text{H}$ value of ca.
374 -275‰. We note our calculations are based on averages of set time periods, which we expect would
375 integrate ice of various ages - rather than extreme values which could relate to specific melt events of ice
376 or biases to certain ages/regions. This seems reasonable - the isotopic signal of coastal surface waters
377 masses advected from the RIS to the Adélie land (as illustrated in Fig. 2 and 3) must integrate a range of
378 source areas across the RIS and from the coast around to Adélie Land.

379

380 In comparison to the highly negative glacial ice isotope composition, sea surface water $\delta^{18}\text{O}$
381 measurements taken near the Mertz Glacier offshore Adélie Land (140-150°E) in summer 2000-2001
382 ranged between -0.47 and 0.05‰ (Jacobs *et al.*, 2004), equivalent to $\delta^2\text{H}$ values of 6.9 to 11.2‰
383 (average = 9‰) following the GMWL. Thus, the two major hydrogen source pools (RIS glacial ice and
384 ocean water) have highly contrasting isotope values, meaning inputs of upstream glacial ice could have a
385 large effect on surface water $\delta^2\text{H}$ values in the Adélie Land region.

386

387 Taking the average glacial meltwater $\delta^2\text{H}$ value as -275‰ and the average modern Adélie surface water
388 $\delta^2\text{H}$ value of 9‰ as end-members, and assuming a biosynthetic offset between the FA and sea water of
389 173‰ (see below), we can use a simple mixing model to estimate the percentage of glacial meltwater
390 required in the surface waters to change the $\delta^2\text{H}_{\text{FA}}$ value to those recorded in U1357B samples. The
391 most negative values occur during the early Holocene, 11.4 – 8.2 ka, averaging -214.2‰ (n=18) which,
392 converted to a surface water value of -41‰, requires 17.6% of the surface water to be comprised of
393 glacial meltwater. During this time, we argue that large volumes of meltwater were reaching the core
394 site as local glaciers retreated, leading to intense surface-water stratification. Thus, a relatively high
395 percentage of meltwater in the Adélie Land surface waters seems reasonable. During the mid-Holocene
396 (5-4 ka), the average $\delta^2\text{H}_{\text{FA}}$ is very similar (-213.9‰, n=7), requiring 17.2% of the surface water to be
397 derived from glacial meltwater. During this time, we argue for the dominant meltwater source as coming

398 from the Ross Sea, and interpret this as a major period of glacial retreat (see section 5.2), during which
399 large volumes of meltwater are injected into the surface water and transported to the Adélie coast. In
400 contrast, the most recent samples (last 0.5 ka, n=7), which includes the most positive value of the record,
401 has an average $\delta^2\text{H}_{\text{FA}}$ value of -174.5‰. This brings the surface water value up to -1.5‰, which
402 approaches modern measured values, and requires just 3.7% (e.g. well within uncertainties) of the
403 surface waters in the Adélie Land to be glacial meltwater. However, it is also possible that the meltwater
404 was dominated by more LGM-aged ice. In either case, perturbation of the exact isotopic values still
405 indicate only significant changes in the flux of glacial meltwater can account for this signal. For
406 example, the use of -330‰ (LGM values) for the ice input gives an estimate of 3% of the surface water
407 being comprised of glacial meltwater for latest Holocene values, and 14.7% for pre 8 ka values. Taking -
408 240‰ (Holocene values) for the ice input gives an estimate of 4% for latest Holocene values, and 20%
409 for pre 8 ka values). Thus even with changing isotopic values though the deglacial, this signal of
410 changing meltwater flux would still dominate. We note these are semi-quantitative estimates, as the
411 salinity and temperature fractionation could reduce these estimates further (but salinity and temperature
412 can only ever account for a small part of the signal).

413

414 Surface water $\delta^{18}\text{O}$ values around Antarctica (below 60°S), measured between 1964 and 2006, ranged
415 from -8.52‰ to 0.42‰ (Schmidt *et al.*, 1999), the most negative value having been measured proximal
416 to the George VI Ice Shelf edge, where high melt rates have been observed (Potter and Paren, 1985). If
417 converted to $\delta^2\text{H}$ using the global meteoric water line, these values give a $\delta^2\text{H}$ range of 83.4‰. Thus,
418 our absolute $\delta^2\text{H}_{\text{FA}}$ range of 123‰ over the Holocene suggests a range of isotopically depleted
419 meltwater inputs to our core site over this time that are 1.5 times greater than that occurring in different
420 locations around the Antarctic in recent decades. This seems plausible based on geological evidence that
421 indicates large glacial retreat and ice mass loss occurred from the Ross Sea sector during the Holocene
422 (Anderson *et al.*, 2014; McKay *et al.*, 2016; Spector *et al.*, 2017), meaning resultant changes in surface
423 water are likely to be greater in magnitude than observed around the Antarctic in recent decades. This
424 assumes a relatively constant value for the isotopic composition of glacial meltwater, however, there is
425 likely to be some variability due to the possibility of melting ice of different $\delta^2\text{H}$ values. But, as
426 discussed above, the meltwater is best represented by the average values of the ice sheet, rather than
427 extreme values, since it must (over the broad expanse of the RIS) include an integrated signal, and thus
428 the actual variation in meltwater $\delta^2\text{H}$ will be significantly within the range of the end-members.

429

430 Although the biosynthetic fractionation of the C₁₈ FAs in U1357B is unknown, we assume that the offset
431 with surface water remains relatively constant throughout the record. Sessions *et al.* (1999) showed the
432 biosynthetic fractionation of hydrogen isotopes in the C₁₈ FA from four different marine algae to range
433 from -189 to -157‰. If we take the average of these values of 173‰ and apply this as a biosynthetic
434 offset to the youngest samples in U1357B (last 0.5 ka, n=7), which includes the most positive value of
435 the record, gives an average $\delta^2\text{H}_{\text{FA}}$ value of -174.5‰. This brings the surface water value up to -1.5‰,
436 which approaches modern measured values (Jacobs *et al.*, 2004).

437

438 Furthermore, it is interesting to note that the biosynthetic offsets measured by Sessions *et al.* (1999) for
439 the C₁₈ FA from different algal species have a total $\delta^2\text{H}$ range of 32‰. Although we cannot dismiss
440 changes in the relative contribution of C₁₈ from different species in U1357B samples (and thus different
441 biosynthetic fractionations), we argue this would only be a minor control on $\delta^2\text{H}$ compared to other
442 influences. As a thought experiment, taking the above end-members for biosynthetic fractionation from
443 Sessions *et al.* (1999), even with a 100% change in C₁₈ producer to a different algal source, this could
444 only explain a quarter of the observed $\delta^2\text{H}$ change (i.e. 32‰ of 123‰).

445

446 Therefore, we interpret the first order control on $\delta^2\text{H}_{\text{FA}}$ at Site U1357 as inputs of isotopically depleted
447 glacial meltwater. Such inputs are, in turn, influenced by the mass balance of the proximal or up-current
448 glaciers and ice-shelves.

449

450 **4.3 Other proxies**

451 Grain size, natural gamma radiation (NGR) and terrigenous and biosiliceous mass accumulation rates
452 (MARs) reflect changing sediment delivery either driven via local glacial meltwater discharge or
453 advection of suspended sediment by oceanic currents. The diene/triene HBI ratio is used as a proxy for
454 coastal sea ice presence (Massé *et al.*, 2011), in which high values indicate greater sea ice extent over
455 the core site. The HBI diene, also known as Ice Proxy for the Southern Ocean with 25 carbon atoms
456 (IPSO25), has been shown to derive from a sea-ice associated diatom (Belt *et al.*, 2016), whereas the
457 HBI triene is produced in the marginal ice zone (Smik *et al.*, 2016). Ba/Ti enrichment is considered to
458 reflect enhanced primary productivity. Interpretation of these proxies is discussed in more detail in
459 Supplementary Information S2.

460 **5 RESULTS**

461 **5.1. Model simulations**

462 We employed a series of sensitivity tests from a high-resolution numerical ocean model by releasing a
463 range of meltwater volumes (0.01 to 1 Sv) from along the front of the Ross Ice Shelf (RIS) to determine
464 its pathway. This demonstrates that, even under the lowest flux scenarios, freshwater is transported
465 anticlockwise, entrained within the coastal current (Fig. 2 and 3), and reaches Site U1357 within a year.
466 Moreover, although the higher input scenarios are not realistic values for the release of meltwater since
467 the LGM, the full range of simulations show a strong linear relationship between meltwater flux and
468 salinity change at the core site (Fig 3), suggesting the magnitude of the signal recorded at Site U1357 is
469 directly related to the magnitude of meltwater released. Thus, we argue that any changes in Ross Sea
470 water mass properties (salinity and temperature) would have a direct influence on surface water mass
471 properties at Site U1357 during the Holocene.

472 **5.2 Geochemical data**

473

474 The main datasets from Core U1357 are displayed in Fig 4 and 5. FA $\delta^{2}\text{H}$ (Fig 4a) shows an overall
475 trend towards more positive values over the course of the Holocene, indicating a decline in glacial
476 meltwater input. There is a notable deviation from this trend in the mid-Holocene involving a sustained
477 period of more negative $\delta^{2}\text{H}$ values, suggesting a peak in meltwater input, centred on ca. 4.4 ka. This
478 mid-Holocene deviation in FA $\delta^{2}\text{H}$ coincides with an increase in the HBI diene/triene ratio (Fig. 4c),
479 indicating a baseline shift in sea ice conditions whereby greater sea ice concentrations are sustained for
480 the rest of the Holocene. This is a similar pattern to the relative abundance of the *Fragilariopsis curta*
481 group (Fig 4b), a sea ice diatom group in core MD03-2601 also indicating a shift in sea ice
482 concentrations. Along the entire record, Ba/Ti ratios show persistent periodic fluctuations in marine
483 productivity, with values between 0.1 and 2.7 (Fig. 4g). A marked enrichment can be observed at ca. 4.4
484 ka reaching Ba/Ti ratio values over 36.1, suggesting a peak in primary productivity, before declining to
485 background levels again (Fig. 4).

486

487 **5.3 Sedimentological data**

488 The stratigraphy of U1357B is divided into three units: the lowermost 10 cm recovered Last Glacial
489 Maximum (LGM) till (Unit III), overlain by 15 m of laminated mud-rich diatom oozes with ice rafted
490 debris (IRD) (Unit II), and the upper most 171 m (Unit I) consists of laminated diatom ooze with a
491 general lack of IRD and a significant reduction in terrigenous sediment (Escutia *et al.*, 2011).

492 Between ca. 11.4 and 8 ka, U1357B has a relatively high terrigenous component (i.e. high Natural Gamma
493 Radiation (NGR) content and low BSi%; Fig 5). The grain size distribution contains coarse tails of fine (125-
494 250 μm) to medium sands (250-500 μm), but only one sample contains coarse sands (>500 μm) that may
495 represent ice-berg rafted debris (IBRD). However, terrigenous content and IBRD is more common in the
496 underlying Unit II. The fine-grained sands and muds have a distribution with similar modes to overlying
497 intervals, albeit with an increase in the size of the coarse silt and very fine sand modes. There is a subtle
498 increase in sorting up core between ca. 11.4 and ca.8 ka (from very poorly to poorly sorted, Fig. 5).

499 Between 9 and 4.5 ka, mass accumulation rates (MARs) (both biogenic and terrigenous; Fig. 5) are
500 relatively high, albeit with millennial scale variability. However, the mean grain size and sorting of the
501 terrigenous material is relatively stable throughout the entire interval, and as with the rest of Unit I there
502 is an almost complete lack of IBRD. There is a rapid increase in mud content at 4.5 ka coincident with a
503 reduction in both the biogenic and terrigenous MARs, although the terrigenous MAR curve shows higher
504 accumulation rates than the biogenic MAR curve (Fig. 5).

505

506 **6 DISCUSSION**

507

508 The sedimentology and geometry of the drift prior to ~11.4 ka (Unit II) is consistent with the calving
509 bay re-entrant model (Domack *et al.*, 2006; Leventer *et al.*, 2006) (Fig. 1 and Supplementary Fig. S4;
510 Supplementary Materials), whereby LGM ice retreated in the deeper troughs while remaining grounded
511 on shallower banks and ridges. Sediment laden meltwater and IRD content in Unit II (>11.4 ka) is thus
512 likely derived from local outlet glaciers. However, anomalously old radiocarbon ages due to glacial
513 reworking precludes development of a reliable age model prior to the Holocene (Supplementary
514 Materials).

515 The results of model simulations (Section 5.1) indicate that, although several small glaciers within
516 Adélie Land may contribute meltwater to the site, the region is also likely to be influenced significantly
517 by changes in Ross Sea waters. Freshwater release simulations from the Ross Ice Shelf (RIS) confirm

518 this oceanographic continuity between the Ross Sea and the Wilkes region (Fig 2). All five simulations
519 indicate that meltwater released from the edge of the RIS is almost completely entrained within the
520 westward coastal surface current and reaches Site U1357 within 4 months to 1 year (Fig 3). These fluxes
521 cover a wide range of meltwater inputs and show a strong linear relationship with salinity at the core site
522 (Fig. 4). This suggests that the magnitude of the signal recorded at Site U1357 is directly related to the
523 magnitude of the meltwater input.

524 Local processes do also play a critical role in this region. For example, episodic calving events of the
525 Mertz Glacier tongue release fast ice over the drill site and create strong surface water stratification,
526 cutting off local AABW production (Campagne *et al.*, 2015). Although appearing to be only a local
527 process, there is still a regional (Ross Sea) influence, as this fast ice that builds up behind the Mertz
528 Glacier is formed by the freezing of fresher AASW transported from the Ross Sea (Fig 2). Thus,
529 conditions in the Ross Sea, such as the melting of isotopically depleted glacial ice, would influence both
530 the isotopic composition and amount of this sea ice.

531

532 **6.1 Early Holocene**

533 The base of the drift deposit shows down-lapping of material suggesting a supply from the south,
534 indicating local focusing of meltwater and terrigenous material was the dominating influence until 11.4
535 ka (Supplementary Materials). This is overlain by onlapping strata (Unit I) with the drift forming an
536 east-west elongation on the northern flank of the Dumont d'Urville Trough, which is more consistent
537 with advection of material from the east than with delivery from local outlet glaciers to the south. Thus,
538 an increased meltwater influence from the Ross Sea is likely since this time.

539

540 Due to the potential for competing sources of glacial meltwater in the earliest Holocene, we focus our
541 study on Unit I, where there is less influence of calving bay processes (Escutia *et al.*, 2011). However,
542 the earliest part of Unit I (11.4 to 8 ka BP), which includes the most negative $\delta^2\text{H}_{\text{FA}}$ values, is
543 characterized by a very gradual up-core increase of sorting in the terrigenous sediment supply,
544 decreasing natural gamma ray (NGR) values (Fig. 5) and a general lack of IRD. We conservatively
545 interpret this as potentially maintaining some local glacial meltwater input from local outlet glaciers in
546 the lowermost interval of Unit I. Nevertheless, this process was probably greatly reduced relative to Unit

547 II deposition and it is likely much of this signal between 11.4 and 8 ka could still be derived from water
548 masses advecting to the site from the east (e.g. the Ross Sea).

549

550 This is supported by geological and cosmogenic evidence which demonstrates that the majority of the
551 margin of the East Antarctic, and also the Amundsen Sea margins, had retreated to their modern-day
552 positions by ~10 ka (Bentley et al., 2014; Mackintosh et al., 2014; Hillenbrand et al., 2017). Thus, these
553 margins are unlikely to contribute large scale shifts in meltwater fluxes to the Adélie Coast during most
554 of the Holocene. The history of grounding line retreat in the Ross sea is relatively well-constrained,
555 particularly in the Western Ross Sea, and the loss of residual ice caps appears to be largely complete by
556 ca. 7 ka to the immediate north of Ross Island, near present day calving line front of the Ross Ice Shelf
557 (Anderson et al., 2014; McKay et al. 2016). Indeed, the phase of isotopically depleted glacial meltwater
558 is apparent at Site U1357 between 8 and 7 ka could be sourced from the Ross Sea, reconciling our data
559 with these chronologies. Prior to 8 ka, any meltwater signal in U1357B is potentially influenced by local
560 glacier retreat, based on the caveats noted earlier in the grainsize and geophysical datasets (S2.2),
561 although we note a dominant Ross Sea contribution to this signal is possible.

562

563 Glacial retreat, however, persisted in the Ross Sea until at least 3 ka (Anderson et al., 2014; Spector et
564 al., 2017) providing a large upstream source of meltwater feeding into the Adélie Coast. We therefore
565 interpret our meltwater signal as being dominated by Ross Sea inputs since at least 8 ka, but potentially
566 as early as 11.4 ka. Furthermore, the retreat of grounded ice from the outer Ross Sea continental shelf
567 was accompanied by the growth of a significant floating ice shelf (which was not the case in the
568 Amundsen Sea or proximal East Antarctic coast) (Bentley et al., 2014).

569

570 An overall trend to more positive $\delta^2\text{H}_{\text{FA}}$ values, from the most negative value of the record at ~9.6 ka, to
571 ~8 ka indicates decreasing meltwater (Fig. 4a), thus suggesting a gradually diminished input from either
572 local outlet glaciers or the Ross Sea. This is associated with an increase in MARs, between 10 and 8 ka,
573 and is tentatively interpreted to represent the final retreat of residual ice from local bathymetric highs
574 allowing more material to advect into the drift (Fig. 5). Although there is millennial scale variability,
575 MARs remain relatively high until 4.5 ka. However, $\delta^2\text{H}_{\text{FA}}$ and MARs show greater coherence at the
576 millennial-scale after 7 ka BP, suggesting that increased fluxes of glacial meltwater broadly

577 corresponded to stronger easterly currents, which advected biogenic and terrigenous material into the
578 drift.

579

580 **6.2 Middle Holocene**

581 A negative excursion in $\delta^2\text{H}_{\text{FA}}$ starting from 6 ka and culminating at 4.5 ka is interpreted to record a
582 period of enhanced glacial meltwater flux to the site relating to a final retreat phase of the major ice
583 sheet grounding line in the Ross Sea embayment (Fig. 6). A marked enrichment of Ba/Ti ratios also
584 occurs at 4.5 ka, reaching values of 36.1, on a background of baseline fluctuations between 0.1 and 2.7
585 (Fig. 4g), which suggests enhanced primary productivity, potentially driven by meltwater-induced
586 stratification. Ongoing Holocene retreat in the Ross Sea is interpreted to be primarily the consequence of
587 marine ice sheet instability processes resulting from the overdeepened continental shelf in that sector
588 (McKay et al., 2016). We use the model presented by Lowry et al. (2019) to help constrain the pattern
589 and rate of retreat of the grounding line to the south of Ross Island. This model compares geological
590 data with ice sheet model experiments that were forced by a range of environmental conditions. These
591 experiments indicate that the Ross Ice shelf cavity only started to expand once the grounding line
592 retreated to the south of Ross Island. Furthermore, to reconcile these model experiments with geological
593 datasets, the cavity expansion was not completed until the mid-Holocene (ca. 5 ka). This reconciles well
594 with ^{10}Be exposure ages of erratics in coastal nunataks at the confluence of the Mercer Ice Stream and
595 Reedy Glacier indicate 105 m of ice sheet deflation since 6.8 ka, with 40 m of this after 4.9 ka (Todd et
596 al., 2010), indicating the most rapid phase of retreat occurred between 6.8 ka and 4.9 ka. More recent
597 deflation profiles for the Beardmore Glacier (84°S) and Scott Glacier (86°S) regions show sustained
598 thinning between ca. 9 and 8 ka, but the Scott Glacier experience a second phase of rapid thinning of ca.
599 200 m between 6.8 and 5.3 ka (Fig. 4), followed by a slower rate of thinning of between 5.3 and 3.5 ka
600 of ca. 100 m. Ages younger than this, near the modern surface are thought to be related to surface
601 ablation rather than dynamic thinning. This suggests that the grounding line was at its modern location
602 by ca. 3.5 ka (Spector et al., 2017) although it may have potentially retreated further south, followed by
603 a short duration readvance of the grounding line (Kingslake, et al., 2018). Glaciological evidence from
604 radar profiles suggests the development of divide flow on Roosevelt Island occurred sometime between
605 3 and 4 ka BP, suggesting that the ice sheet thickness was at least 500 m thicker until this time (Conway
606 et al., 1999). Combined, these lines of evidence suggest the majority of grounding line retreat south of
607 Ross Island occurred after 8 ka, with a sustained retreat occurring after 6.8 ka, consistent with the timing

608 of the largest inputs of glacial meltwater feeding the U1357 site. However, a younger age (e.g. 3 – 3.5
609 ka) for final establishment of the modern grounding line position is consistent with our interpretation, as
610 although the meltwater signal in $\delta^2\text{H}_{\text{FA}}$ peaks at 4.5 ka, it does not stabilise at lower levels until 3 ka.

611
612 The $\delta^2\text{H}_{\text{FA}}$ peak at 4.5 ka in U1357 coincides directly with a rapid shift in HBI biomarker ratios at the
613 site (Fig. 4a and c), as well as sea ice proxies recorded in nearby site MD03-2601 (Fig. 4b), in the Ross
614 embayment (Taylor Dome ice core on a revised age model) (Steig *et al.*, 1998; Baggenstos *et al.*, 2018)
615 (Fig. 4d) and other sectors of the East Antarctic margin in Prydz Bay (JPC24) (Denis *et al.*, 2010) (Fig.
616 4e), reflecting a widespread increase in coastal sea-ice concentration and duration. We interpret
617 decreasing MAR and finer-grained terrigenous content (e.g. increased mud percent) at Site U1357 after
618 4.5 ka (Fig. 5) to also be a consequence of increased coastal sea ice, reducing wind stress on the ocean
619 surface and limiting the easterly advection of detritus to the drift deposit.

620
621 Coastal sea-ice concentration and duration remain high throughout the rest of the Holocene (this study
622 and Steig *et al.*, 1998; Crosta *et al.*, 2008; Denis *et al.*, 2010), compared to the period before 4.5 ka,
623 despite a decrease in glacial meltwater flux to the U1357 site. In addition, meltwater input prior to 4.5 ka
624 does not have a major influence on sea ice extent. Thus, an increase in meltwater flux cannot explain the
625 Neoglacial intensification of sea ice at ~4.5 ka. Here, we propose that greater coastal sea ice cover since
626 4.5 ka is related to the development of a large ice-shelf cavity in the Ross Sea as the ice sheet retreats
627 (Fig. 6), which pervasively modified ice shelf-ocean interactions and increased sea ice production.
628 Models suggest a large cavity on the continental shelf increases contact between basal-ice and
629 circulating ocean water, driving the formation of a cool, fresh water mass feeding into the AASW,
630 stabilizing the water column and enhancing the production of sea ice (Hellmer, 2004) (Fig. 6). However,
631 under small cavities such as in the modern Amundsen Sea influenced by warm-water incursions, ice
632 shelf melting results in an “ice pump” enhancement of sub-ice shelf circulation. This increases flow of
633 warm Circumpolar Deep Water (CDW) under the ice shelf that is 100-500 times the rate of melt, and
634 this volume of water does not allow for supercooling. Small cavity ice shelf outflows are therefore warm
635 and act to restrict sea ice at the ice shelf front (Jourdain *et al.*, 2017). Thus, during the Holocene, the size
636 of the cavity must have reached a threshold after which this positive warming feedback switched to a
637 negative feedback. We argue that such a tipping point takes place at 4.5 ka BP, when our proxy data
638 suggest meltwater peaks, and would explain why the increase in sea-ice concentration appears rapid and

639 only occurs at the peak of the meltwater input, and not during its prior increase, or previous meltwater
640 inputs (Fig. 4a-g).

641

642 Although the glacial meltwater volume is greatly reduced after 4.5 ka BP, the volume of Ice Shelf Water
643 (ISW) produced beneath the modern RIS is estimated at 0.86 Sv-1.6 Sv (Holland *et al.*, 2003; Smethie
644 and Jacobs, 2005). We note that ISW is not glacial meltwater, but it is defined as a supercooled water
645 mass formed through interaction with the base of the RIS, but once formed acts to modify other water
646 masses in the Ross Sea. A significant proportion of ISW is high salinity and is thus advected northwards
647 at intermediate waters depth to ultimately form AABW. However, a significant volume of ISW is lower
648 salinity and buoyant, due to development of frazil ice, and acts to mixes with surface waters (Robinson
649 *et al.*, 2014). Currently, a 0.4 Sv plume of ISW in the western margin of the Ross Ice Shelf (Robinson *et al.*,
650 *et al.*, 2014) is directly delivered to the surface resulting in enhanced sea ice production, while seasonal
651 melt of this enhanced sea ice further acts to cool and freshen surface waters. Although unrealistic in the
652 context of a post-LGM meltwater flux from the Ross Sea alone, the larger meltwater release scenarios in
653 our simulations (0.5 to 1 Sv) show the potential pathways that a cool, fresher surface water mass
654 collecting and forming on the broad Ross Sea continental shelf would follow (Fig. 2). These waters are
655 transported in easterly coastal currents to the Weddell Sea and the Antarctic Peninsula. This eventually
656 retroflects to join the Antarctic Circumpolar Current (Fig. 2b), and thus has potential for cooling and
657 freshening in the South Atlantic far offshore, as the ice shelf cavity increased in the Ross Sea. Indeed,
658 offshore site TTN057-13 records increased lithics in the South Atlantic after 4.5 ka (Fig. 4f), relative to
659 the period before, suggested to have been predominantly transported by sea ice indicating a cooling in
660 sea surface temperatures and increase in sea-ice extent in the South Atlantic at this time (Hodell *et al.*,
661 2001; Nielsen *et al.*, 2007). However, it also is feasible that this circum-Antarctic cooling signal
662 indicates similar melt processes may have been occurring in the Weddell Sea at ~4.5 ka, as suggested by
663 cosmogenic nuclide data (Hein *et al.*, 2016).

664

665 **6.3 What Drove the Neoglacial Transition?**

666 Our observed coastal sea-ice increase is part of a widespread transition to Neoglacial conditions both
667 globally and at high southern latitudes (Kim *et al.*, 2002; Masson-Delmotte *et al.*, 2011; Marcott *et al.*,
668 2013; Solomina *et al.*, 2015). However, most current climate models do not simulate this cooling trend,
669 resulting in a significant data-model mismatch (Liu *et al.*, 2014) (Supplementary Fig. S6). Marine ice

670 sheet retreat along the Pacific margin of West Antarctic has previously been proposed to be triggered by
671 enhanced wind-driven incursions of warm CDW onto the continental shelves in the early Holocene
672 (Hillenbrand *et al.*, 2017), with continued retreat in the Ross Sea being the consequence of the
673 overdeepened continental shelf and marine ice sheet instability processes (McKay *et al.*, 2016). We
674 propose that a series of negative feedbacks was also associated with the latter phases of this retreat due
675 to the RIS cavity expansion that occurred in the mid-Holocene, with similar processes possibly
676 occurring in the Weddell Sea, leading to the onset and continuation of Neoglacial conditions.
677 Widespread albedo changes associated with increased coastal sea ice would have amplified regional
678 cooling trends (Masson-Delmotte *et al.*, 2011), whilst increased stratification resulting from seasonal
679 sea-ice melt and increased ISW production drove the deepening of the fresher water surface isopycnal at
680 the continental shelf break. Grounding line retreat creates new space for continental shelf water masses
681 to form, while ice shelf cavity expansion promotes supercooling of waters circulating beneath the ice
682 shelf, and freshening of AASW. Thus, as seasonal sea ice melt and ice shelf supercooling processes
683 played a greater role in enhancing AASW cooling and production on the continental shelf, they would
684 have acted to restrict warmer subsurface water transport onto the continental shelf (Smith Jr. *et al.*,
685 2012) (Fig. 6). Furthermore, the Neoglacial sea-ice increase itself may have been associated with a
686 stabilising feedback mechanism (which also is not resolved in ice-ocean models) through its role in
687 dampening ocean-induced wave flexural stresses at ice shelf margins, reducing their vulnerability to
688 catastrophic collapse (Massom *et al.*, 2018). We suggest that some combination of the above processes
689 could have acted to slow the rate of Ross Sea grounding line retreat and reduced basal ice shelf melt as
690 indicated by a trend towards more positive $\delta^2\text{H}_{\text{FA}}$ values in U1357 between 4.5 and 3 ka (Fig. 4a).
691 Furthermore, large Antarctic ice shelves currently have large zones of marine accreted ice resulting from
692 supercooling (Rignot *et al.*, 2013), thus the signature of $\delta^2\text{H}_{\text{FA}}$ is anticipated to become more positive as
693 the ice shelf approaches a steady state of mass balance, relative to the thinning phases when basal melt
694 rates exceed that of accretion. The stabilization of $\delta^2\text{H}_{\text{FA}}$ values observed at 3 ka in U1357 suggests the
695 Ross Ice Shelf has maintained a relatively steady state of mass balance since this time.

696
697 A recent study implies that the late Holocene shift in patterns of coastal versus open water sea ice
698 patterns in the Ross Sea was driven an increase in katabatic winds since at least 3.6 ka in the Ross Sea
699 (Mezgec *et al.*, 2017), leading to enhanced polynya activity. During colder Antarctic climates, increased
700 latitudinal temperature gradients enhanced katabatic winds in the Ross Sea (Rhodes *et al.*, 2012). This is

701 consistent with our hypothesis, as we interpret this katabatic wind and polynya activity signal to be a
702 response to the preceding Neoglacial cooling at 4.5 ka and evolution of the modern ocean-ice shelf
703 connectivity, which our data suggest was primarily driven by ice shelf cavity expansion. Furthermore,
704 this transition takes place on the background of declining winter insolation (Berger and Loutre, 1991)
705 which would have acted to further enhance and maintain these changes. This insolation decline has
706 previously been hypothesised as a driver of the Neoglacial increase in coastal sea ice in both Prydz Bay
707 and the Adélie Land regions (Denis *et al.*, 2010), however this monotonic decrease contrasts with the
708 markedly rapid increase in sea ice observed in many records (Fig 4). Our mechanism of ice shelf cavity
709 expansion, reaching a threshold that promoted significant supercooling of continental shelf water
710 masses, reconciles both the rapidity and timing of Neoglacial onset in the middle Holocene.

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712 **7. Conclusions and Implications for Antarctic Climate, Sea-Ice and Ice Shelf Behaviour**

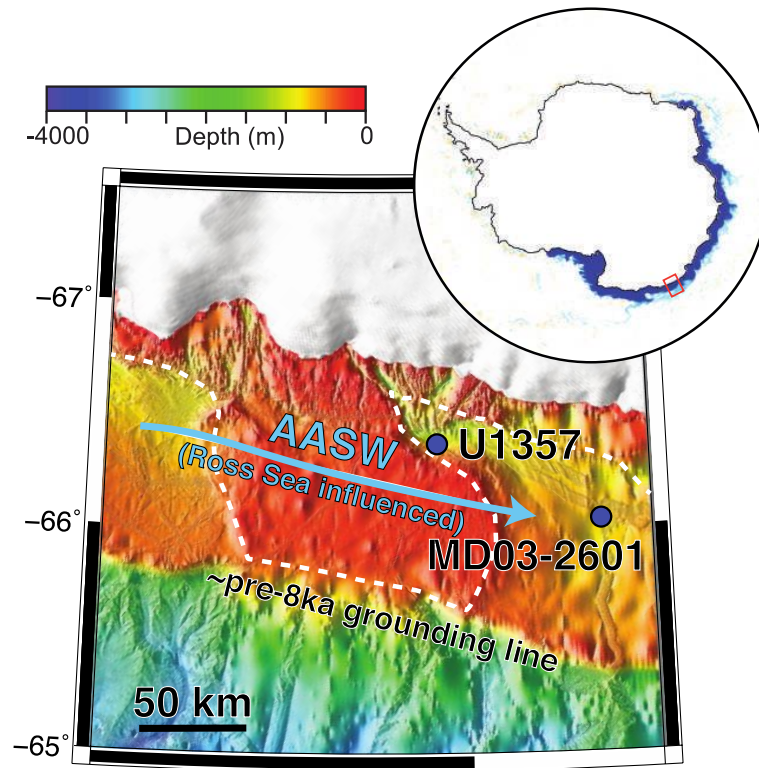
713 Our multiproxy record of changing oceanographic conditions in the Adélie Land region indicates a
714 significant meltwater event during the middle Holocene. Comparison of this record with pre-existing
715 studies from around the Antarctic margin indicates this was likely associated with final phases of
716 deglaciation of the Ross Sea embayment. Expansion of the Ross Ice Shelf cavity at this time is proposed
717 to have led to modification of surface water mass formation processes on the continental shelves of the
718 Ross Sea and Adélie Land, and contributed to widespread Antarctic surface water cooling and increased
719 coastal sea ice during the late Holocene Neoglacial. The lack of these coupled ice-ocean processes is
720 apparent in recent Earth system model experiments, in particular the incorporation of evolving ice shelf
721 cavities, with Trace-21k for example, instead simulating a decrease in Antarctic sea-ice extent and
722 thickness after 5 ka (Supplementary Fig. S6). These model outputs are in direct contrast to multiple lines
723 of proxy data in this study and previous work (Steig *et al.*, 1998; Crosta *et al.*, 2008; Denis *et al.*, 2010).
724 Consequently, our results provide insights into the magnitude of this data-model mismatch, as well as a
725 mechanism for rapid sea-ice change and grounding line stabilisation on the background of a warming
726 climate (Liu *et al.*, 2014), both on modern and Holocene time scales. Better representation of the role of
727 evolving ice shelf cavities on oceanic water mass evolution and sea-ice dynamics, which our data
728 indicate acted as a strong negative feedback, will be fundamental to understanding the oceanographic
729 and glaciological implications of future ice shelf loss in the Antarctic.

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732 **Figures**

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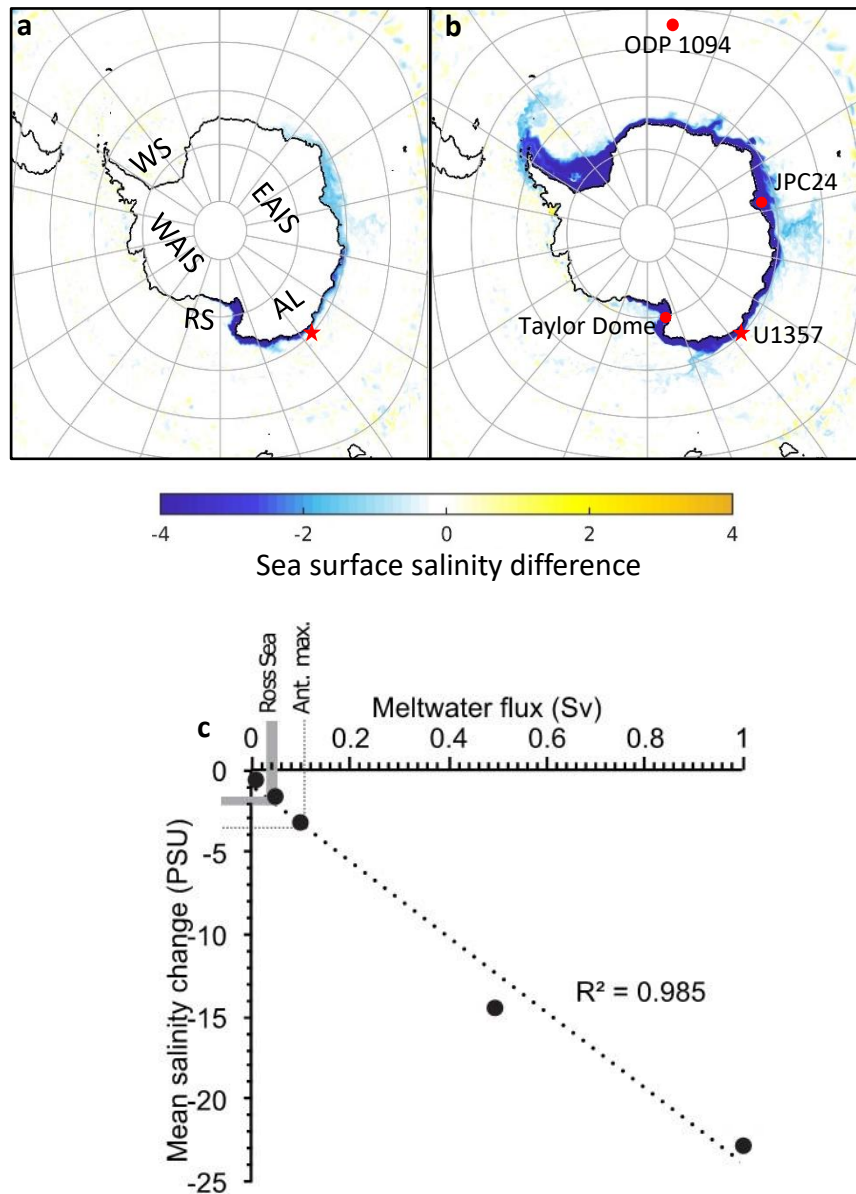
735 **Figure 1: Location of Sites U1357 and MD03-2601 (blue dots).** The ice sheet grounding line formed a
736 calving-bay environment (dashed white line) prior to 11.4 ka, but since at least 8.2 ka Antarctic Surface
737 Water flow is largely advected from the Ross Sea (blue line). Inset map: pathway of freshwater (dark
738 blue) after 1 year of 1 Sv meltwater released from along the edge of the Ross Ice Shelf in a model
739 simulation.

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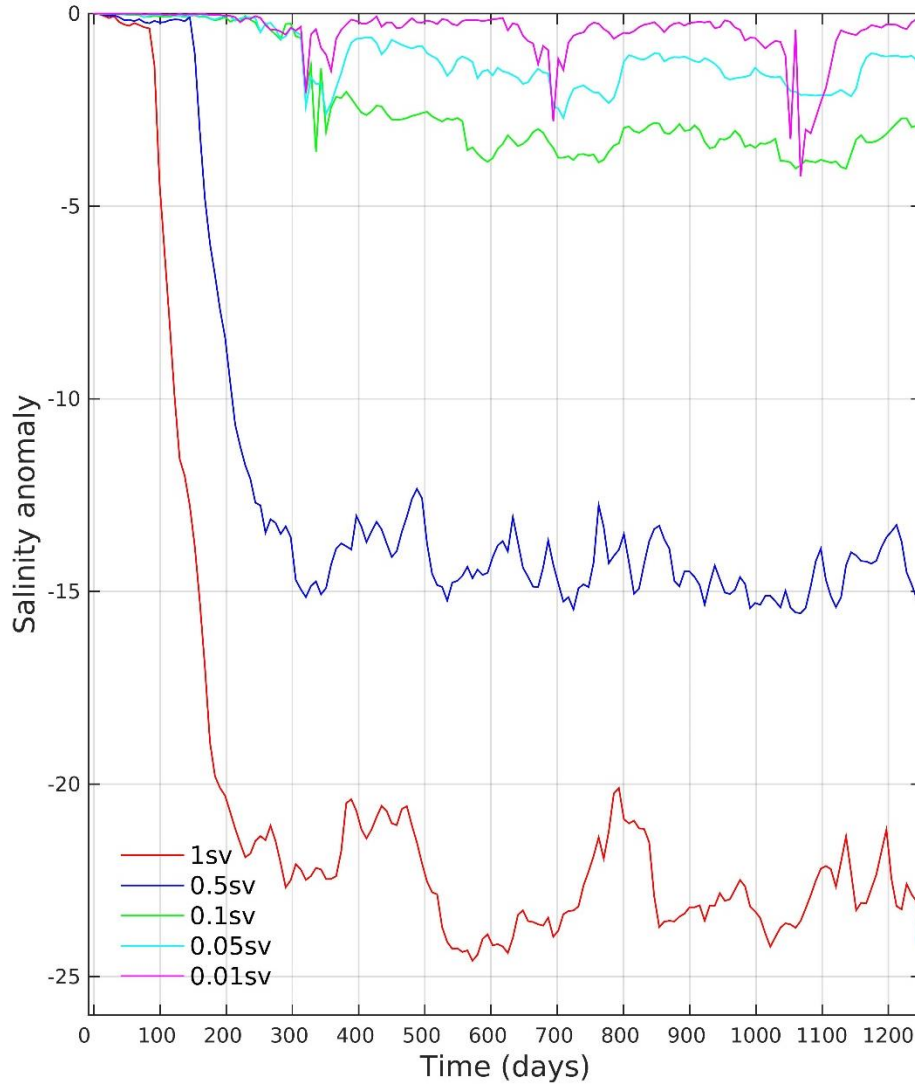
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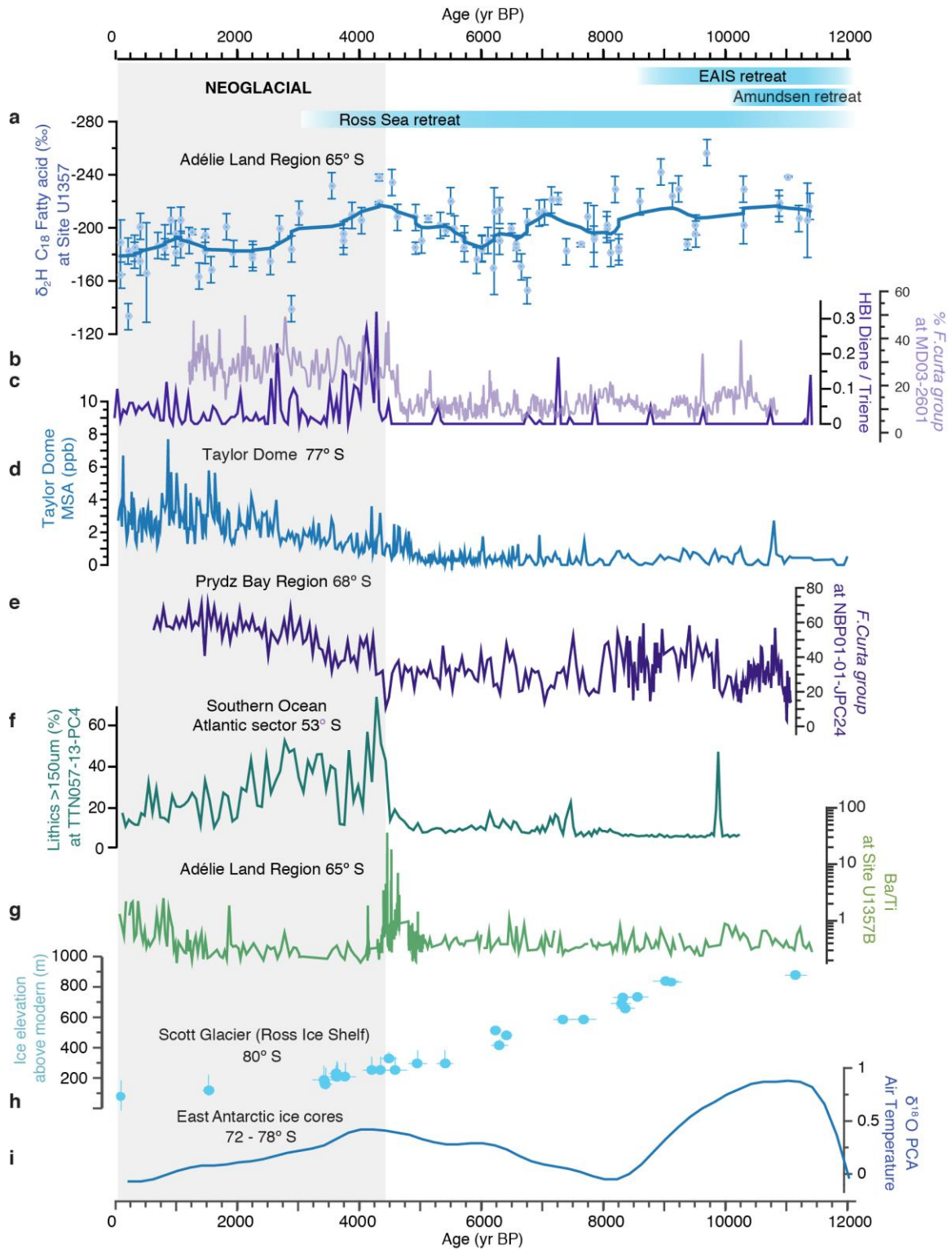


744 **Figure 2: MITgcm simulations of meltwater release from along the edge of the Ross Ice Shelf.** First
 745 two images show sea-surface salinity difference (in practical salinity units) after 3.5 model years
 746 resulting from meltwater release volumes of a) 0.1 Sv (2×10^{13} m³ total ice volume equivalent) and b) 0.5
 747 Sv (1×10^{14} m³ total ice volume equivalent). Red star indicates position of Site U1357 (this study) and
 748 red dots show positions of other core sites mentioned in this study where a Mid-Holocene increase in sea
 749 ice and/or cooling is recorded: Taylor Dome (Steig *et al.*, 1998; Baggenstos *et al.*, 2018), JPC24 (Denis
 750 *et al.*, 2010) and ODP 1094 (Nielsen *et al.*, 2007). AL = Adélie Land, RS = Ross Sea, WS = Weddell
 751 Sea, EAIS = East Antarctic Ice Sheet, WAIS = West Antarctic Ice Sheet. c) Scatter plot of simulated
 752 meltwater flux (Sv) against mean salinity difference at U1357 core site. Grey band indicates range of

753 plausible Holocene to deglacial Ross Sea meltwater inputs. Dotted line indicates maximum Antarctic
754 meltwater during the Holocene.



776 **Figure 3** Simulated salinity anomalies over time at Site U1357 for the five meltwater release
777 experiments.



785

786 **Figure 4: Holocene Adélie Land proxy records from IODP Site U1357 and other circum-Antarctic**

787 **sites.** Glacial retreat chronologies are shown as bars at the top as discussed in the text. a) $\delta^2\text{H}$ C₁₈ fatty

788 acid at Site U1357 (errors bars based on replicates), with robust locally weighted smoothing (rflowss). b)
789 *Fragilariopsis curta* group (*F. curta* and *F. cylindrus*) relative abundance at MD03-2601, as a proxy of
790 sea-ice conditions (Crosta *et al.*, 2008) c) di-unsaturated HBI (C_{25:2}; Diene)/tri-unsaturated HBI isomer
791 (C_{25:3}; Triene) ratio at Site U1357 d) Methanesulfonate (MSA) concentrations (ppb) from Taylor Dome
792 ice core e) *F. curta* group relative abundances in core NBP-01-JPC24 f) Coarse lithic (ice-rafted)
793 content at TTN057-13-PC4 (Hodell *et al.*, 2001) g) Ba/Ti (logarithmic scale) at Site U1357 h) ¹⁰Be
794 cosmogenic nuclide ages from Scott Glacier in the SW Ross Ice Shelf region (Spector *et al.*, 2017) i)
795 Temperature signal from principal component analyses of five δ¹⁸O records in five East Antarctic ice
796 cores (Vostok, EPICA Dome C, EPICA Dronning Maud Land, Dome Fuji and Talos Dome) (Masson-
797 Delmotte *et al.*, 2011).

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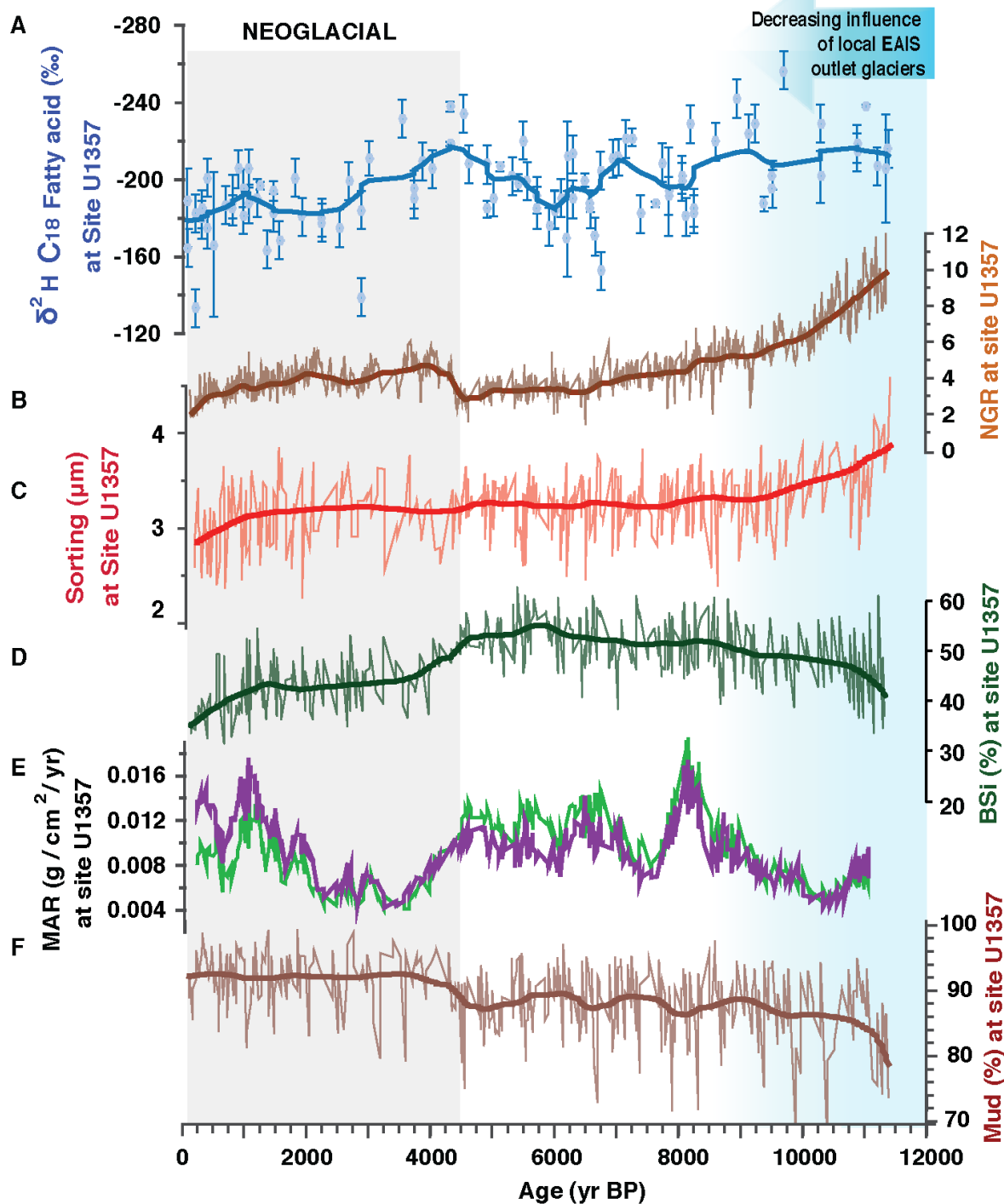
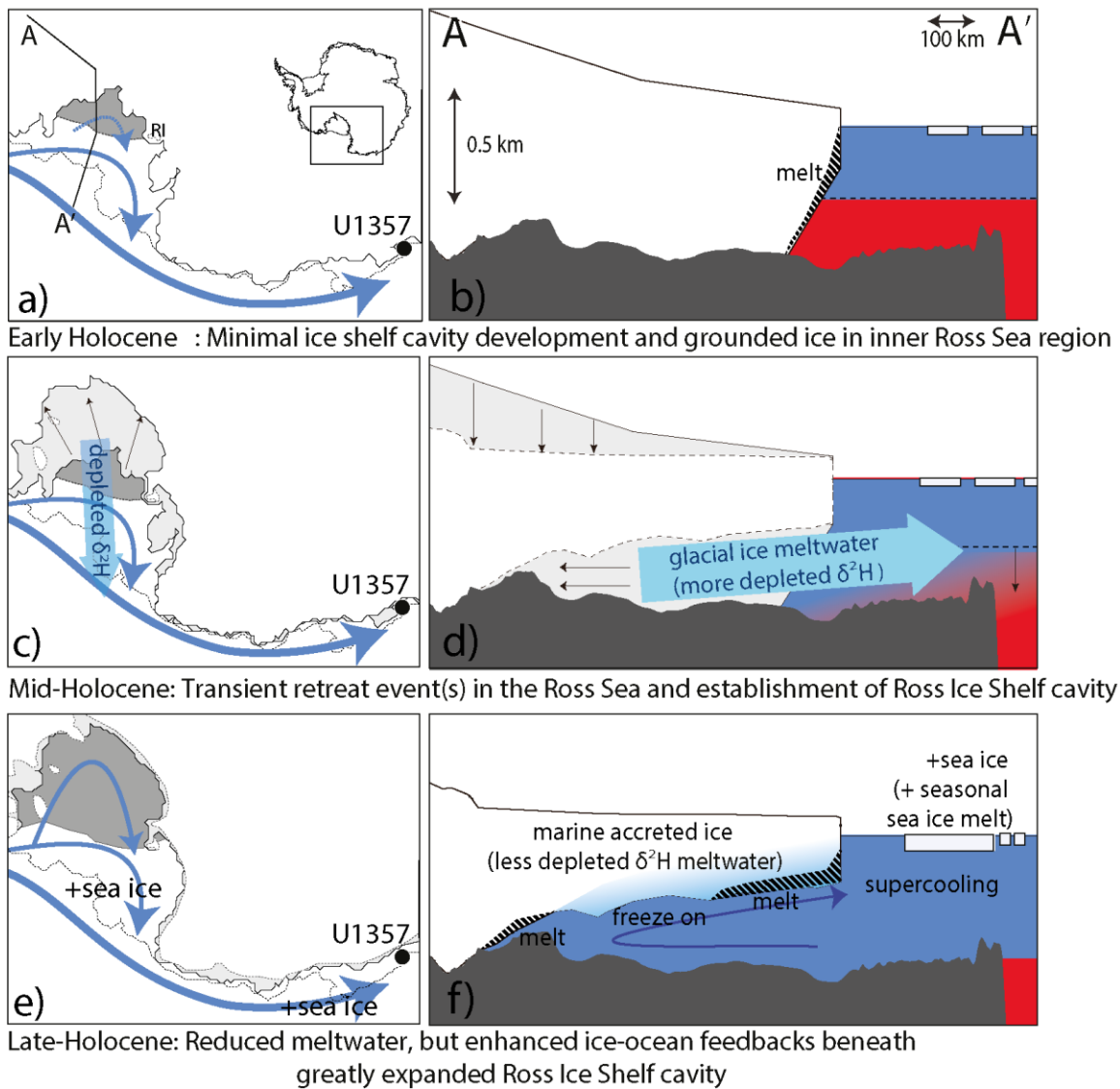


Figure 5 Holocene Adélie Land proxy records from IODP Site U1357 a) C₁₈ fatty acid $\delta^2\text{H}$ (errors bars based on replicate analyses), heavy line is a robust locally weighted scatterplot smoothing (rlowss) b) Natural Gamma Radiation, heavy line is a rlowss c) grain sorting (μm) calculated following Folk and Ward (1957), heavy line is a rlowss d) Percentage of biogenic silica (BSi), heavy line is a rlowss; e) Mass accumulation rates of biogenic (green line) and terrigenous (purple line) material f) Percentage of mud, heavy line is a rlowss.



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851 **Figure 6: Conceptual model of evolving Holocene glacial and oceanographic conditions in the Ross**
 852 **Sea region.** Panels on the left show modelled grounding line positions (McKay *et al.*, 2016), and
 853 proposed circulation of surface and sub-ice shelf circulating waters (light blue arrows). Panels on the
 854 right show cross sections of the Ross Ice Shelf (RIS) and ice-ocean interactions. Dark blue = cool
 855 surface waters, Red = warm subsurface waters. a) The grounding line in Adélie Land is near its modern
 856 location, but near Ross Island (RI) in the Ross Sea, and ice shelf cavity (dark grey shading) is reduced in
 857 size relative to today (McKay *et al.*, 2016). b) Continental shelf profile A-A' (panel a) shows a Ross Sea
 858 grounding line in a mid-continental shelf location in close proximity to the RIS calving line (McKay *et*
 859 *al.*, 2016), with subsurface warming on the continental shelf triggering WAIS deglaciation (Hillenbrand

860 *et al.*, 2017). c) Most grounding line retreat south of RI occurred between 9 and 4.5 ka (light grey
861 shading with black arrows represents area of retreat over this period), proposed to be the consequence of
862 marine ice sheet instability, but the ice shelf calving line remained near its present position (McKay *et*
863 *al.*, 2016; Spector *et al.*, 2017). d) Grounding line retreat and ice shelf thinning released meltwater with
864 negative $\delta^2\text{H}$ into the surface waters. Increasing ice shelf-oceanic interactions with the development of
865 the ice shelf cavity (dark grey) led to enhanced Antarctic Surface Water formation; f) Minimal
866 grounding line retreat has occurred since 4.5 ka, and the RIS supercools AASW leading to enhanced
867 sea-ice formation despite reduced glacial meltwater flux. Seasonal sea ice meltwater further freshens and
868 cools AASW. Increased production of AASW on the continental shelf leads to isopycnal deepening
869 (dotted line) and limits flow onto the continental shelf slowing further grounding line retreat. However,
870 as the ice shelf is near steady state mass balance and there is a component of marine accreted ice at the
871 base of the ice shelf (Rignot *et al.*, 2013), the strength of the $\delta^2\text{H}$ signal is reduced relative to periods of
872 mass balance loss.

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