



1 FRONT MATTER

- 2 Title
- 3 Mid-Holocene Antarctic sea-ice increase driven by marine ice sheet retreat
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1. ABSTRACT

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

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2. Introduction

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

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3. MATERIALS AND METHODS



60 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 continental shelves of Antarctica, and (ii) the subsequent development of large ice shelf cavities during 62 the Holocene. Both factors would significantly alter water mass formation on Antarctica's continental 63 shelves, which today are major source regions of Antarctic Bottom Water (AABW) and Antarctic 64 Surface Water (AASW). These interrelated processes are underrepresented in coupled ocean-atmosphere 65 models which currently do not simulate the timing, magnitude and rapid onset of the Neoglacial 66 (Supplementary Materials). 67 68 Integrated Ocean Drilling Program (IODP) Expedition 318 cored a 171 m thick deposit of laminated 69 diatomaceous ooze at Site U1357 offshore Adélie Land (Fig. 1), deposited over the past 11,400 years. 70 Here, we present a new Holocene record of glacial meltwater, sedimentary input and local sea ice 71 concentrations from Site U1357 using compound-specific hydrogen isotopes of fatty acid biomarkers 72 $(\delta^2 H_{FA})$, terrigenous grain size, biogenic silica accumulation, highly-branched isoprenoid alkenes (HBIs) 73 74 and Ba/Ti ratios (Fig. 2 and S4). 75 We interpret $\delta^2 H_{FA}$ (Fig. 2) fluctuations in Adélie Drift sediments as a record of meltwater input from 76 isotopically-depleted glacial ice (Supplementary Materials). Antarctic glacial ice is highly depleted in ²H 77 compared to ocean water, thus creating highly contrasting end-member values for the two major H 78 79 source pools. Grain size (sand and mud percentage and sorting), natural gamma radiation (NGR) and terrigenous and biosiliceous mass accumulation rates (MARs) reflect changing sediment delivery either 80 driven via local glacial meltwater discharge or advection of suspended sediment by oceanic currents. 81 The diene/triene HBI ratio is used as a proxy for coastal sea ice presence (Massé et al., 2011). Ba/Ti 82 83 enrichment is considered to reflect enhanced primary productivity. These records allow a unique opportunity to reconstruct the magnitude of the coupled response of the ocean and ice sheet during the 84 85 Neoglacial transition. Details on all proxies and associated uncertainties can be found in Section 4.2 and in the Supplementary Materials. 86 87

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3.1 Organic geochemical analyses

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

97 Two-thirds of the total lipid extract were sent to the University of Glasgow, UK and separated over an

aminopropyl silica gel column where the total neutral fraction was eluted with 4ml of DCM/ isopropanol

99 (1:1 v/v), and the total acid fractions were eluted into an 8ml vial with 4% acetic acid in ethyl-ether

solution (Huang et al., 1999). Derivatisation to Fatty Acid Methyl Esters was achieved by adding 200 µl

of MeOH containing 14% v/v Boron triflouride to the 8ml vial containing the TAF. The vial was seal

and placed in the drying cabinet at 70°C for one hour. The MeOH was dried under N₂ and the FAMES

were recovered and cleaned up by eluting through a pre-cleaned 3cm silica gel column (60 A; 35-70)

with 4ml of hexane and 4ml of DCM (containing the FAMES). The recovered FAMES fraction was

split 50:50 for compound specific carbon and hydrogen analysis, respectively. δ^2 H values indicate

depletion against the international standards: Vienna Pee Dee Belemnite (V-PDB) is the standard for

 δ 13C and Vienna Standard Mean Ocean Water (V-SMOW) for δ ²H.

109 Compound specific hydrogen isotope analyses of FAMES was performed at the Institute of Low

Temperature Science, Hokkaido University. δ^2 H values were obtained using a CS-IRMS system with a

HP 6890 gas chromatograph and a ThermoQuest Finnigan MAT Delta Plus XL mass spectrometer.

Separation of the FAMES was achieved with a HP-5 MS fused silica capillary column (30 m x 0.32 mm

i.d., film thickness of 0.25 µm) with a cooled on-column injector. An n-alkane and a reference gas

whose isotopic values were known was co-injected with the samples as an internal isotopic standard for

 δ^2 H. A laboratory standard (Mix F8 of FAMES from Indiana University) containing C₁₀–C₃₀ FAMES

was analyzed daily to check the accuracy and the drift of the instrument and to normalize the data to the

SMOW/SLAP isotopic scale. The H³⁺ factor was measured every three days.

3.2 Inorganic geochemical analysis and electronic microscopy

- 120 Major element concentrations were obtained using X-Ray Fluorescence Scanner on 412 analyses
- measured directly over undisturbed sediment sections. The bulk major element composition included in

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122 this study was measured between sections U1357B-1H-2 to U1357-19H-5 continuously each 50 cm. We 123 used an Avaatech X-ray fluorescence (XRF-Scanner) core scanner at the IODP-Core Repository/Texas A&M University laboratories (USA) during December 2010. Non-destructive XRF core-scanning 124 measurements were performed over 1 cm² area with slit size of 10 mm, a current of 0.8 mA and 125 sampling time of 45 seconds at 10 kV in order to measure the relative content of titanium (Ti) and 126 127 barium (Ba). 128 Field emission scanning electron microscopy (FESEM) images and corresponding spectrum were 129 obtained with an AURIGA FIB-FESEM Carl Zeiss SMT at Centro de Instrumentación Científica, 130 131 Granada University, Spain 132 3.3 Grain size analyses 133 A total of 341 samples were prepared for grain size analysis. Samples were treated for removal of 134 135 biogenic opal with a 1M sodium hydroxide NaOH solution and incubated in a water bath at 80°C for 24 hours. This procedure was repeated twice due to an incomplete dissolution of diatoms observed in smear 136 137 slides. The samples were then treated with H₂O₂ to remove organic material at 80°C for 24 hours. Samples were measured using a Beckman Coulter LS 13 320 Laser Diffraction Particle Size Analyser 138 (LPSA). The LPSA has a relatively narrow range of optimum obscuration which is determined by the 139 sample surface area, in turn determined by sample concentration and sample distribution. Prior to grain 140 size analysis, ~30 mL of 0.5 g/L Calgon (sodium hexametaphosphate) was added to the samples, and 141 sonicated and stirred in order to disperse the grains and prevent clumping. The range in sample mass for 142 most of the post-treatment samples varied from ~0.035-0.8 grams. Random biases propagating through 143 this process cannot be ruled out, especially when taking account of susceptibility of grains <10 µm to 144 clump(McCave et al., 1995) and random cohesion of grains due to any remaining organic content. The 145 146 aqueous liquid module in the LPSA also does not accurately record the <2 μm clay that may compromise a significant part of the size spectrum in glacial environments (McCave et al., 1995; 147 McCave and Hall, 2006). Given these considerations, subsamples were taken from a total of 84 samples 148 to test reproducibility of the data relating to sub-sampling biases, with a least squares regression 149 showing a high reproducibility with an r² value of 0.744. An additional 12 samples were sub-sampled 150 before the chemical treatment in order to test the reproducibility of the treatment methodology, with a 151 least squares regression showing a high reproducibility with an r² value of 0.752.

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3.4 HBIs

Highly branched isoprenoids (HBI) alkenes were extracted at Laboratoire d'Océanographie et du 155 Climat: Experimentations et Approches Numériques (LOCEAN), separately from the fatty acids, using a 156 mixture of 9mL CH₂Cl₂/MeOH (2:1, v:v) to which internal standards were added and applying several 157 sonication and centrifugation steps in order to extract properly the selected compounds (Etourneau et al., 158 2013). After drying with N₂ at 35°C, the total lipid extract was fractionated over a silica column into an 159 apolar and a polar fraction using 3 mL hexane and 6 mL CH₂Cl₂/MeOH (1:1, v:v), respectively. HBIs 160 were obtained from the apolar fraction by the fractionation over a silica column using hexane as eluent 161 following the procedures reported by Belt et al. (2007; Massé et al., 2011). After removing the solvent 162 with N₂ at 35°C, elemental sulfur was removed using the TBA (Tetrabutylammonium) sulfite method 163 (Jensen et al., 1977; Riis and Babel, 1999). The obtained hydrocarbon fraction was analyzed within an 164 Agilent 7890A gas chomatograph (GC) fitted with 30 m fused silica Agilent J&C GC column (0.25 mm 165 166 i.d., 0.25 µm film thickness), coupled to an Agilent 5975C Series mass selective detector (MSD). Spectra were collected using the Agilent MS-Chemstation software. Individual HBIs were identified on 167 168 the basis of comparison between their GC retention times and mass spectra with those of previously authenticated HBIs (Johns et al., 1999) using the Mass Hunter software. Values are expressed as 169 concentration relative to the internal standard. 170

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3.5 Biogenic silica

- 173 Biogenic silica concentrations (wt% BSi) were measured on 349 discrete samples using a molybdate
- blue spectrophotometric method modified from (Strickland and Parsons, 1970; DeMaster, 1981).
- Analytical runs included replicates from the previous sample group and from within the run, and each
- run was controlled by 10 standards and a blank with dissolved silica concentrations ranging from 0 μM
- 177 to 1200 μM. For each analysis, ~7 mg of dry, homogenized sediment was leached in 0.1M NaOH at
- 178 85°C, and sequential aliquots were collected after 2, 3, and 4 hours. Following addition of reagents,
- absorbance of the 812 nm wavelength was measured using a Shimadzu UV-1800 spectrophotometer.
- 180 Dissolved silica concentration of each unknown was calculated using the standard curve, and data from
- the three sampling hours were regressed following the method of DeMaster (1981) to calculate wt%
- BSi. In our U1357B samples, wt% BSi ranges from maximum of ~60% in early and mid-Holocene light

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laminae to a minimum of 31% in late Holocene dark laminae. The average standard deviation of replicate measurements is 0.5%.

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3.6 Model simulations

All numerical calculations were performed using the Massachusetts Institute of Technology general 187 circulation model (MITgcm) (Marshall et al., 1997); a three-dimensional, ocean sea-ice, hydrostatic, 188 primitive equation model. The experiments presented here were integrated on a global domain projected 189 onto a cube-sphere grid to permit a relatively even grid spacing and to avoid polar singularities (Adcroft 190 et al., 2004; Condron and Winsor, 2012). The ocean grid had a mean, eddy-permitting, horizontal grid 191 spacing of 1/6° (18-km) with 50 vertical levels ranging in thickness from 10m near the surface to 192 approximately 450m at the maximum model depth. The ocean model is coupled to a sea-ice model in 193 which ice motion is driven by forces generated by the wind, ocean, Coriolis force, and surface elevation 194 of the ocean, while internal ice stresses are calculated using a viscous-plastic (VP) rheology, as 195 196 described in Zhang and Hibler (1997). In all experiments, the numerical model is configured to simulate present-day (modern) conditions: Atmospheric forcings (wind, radiation, rain, humidity etc.) are 197 prescribed using 6-hourly climatological (1979-2000) data from the ERA-40 reanalysis product 198 produced by the European Centre for Medium-range Weather Forecasts and background rates of runoff 199 from the ice sheet to the ocean are based on the numerical ice sheet model of Pollard and Deconto 200 (2016) integrated over the same period (1979-2000). To study the pathway of meltwater in the ocean, 201 additional fresh (i.e. 0 psu) water was released into the surface layer of the ocean model at the grid 202 points closest to the front of the Ross Ice Shelf. Five different discharge experiments were performed by 203 releasing meltwater into this region at rates of 0.01 Sv (Sv = 10^6 m3/s), 0.05 Sv, 0.1 Sv, 0.5 Sv, and 1 Sv 204 for the entire duration of each experiment (~3.5 years). 205

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4. ENVIRONMENTAL SETTING AND INTERPRETATION OF PROXY DATA

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

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214215	4.1 The Adélie Drift
216	Site U1357 is located in the Dumont d'Urville Trough of the Adélie Basin, ca. 35 km offshore from
217	Adélie Land (66°24.7990'S, 140°25.5705'E; Fig 1). This is a >1000 m deep, glacially scoured
218	depression on the East Antarctic continental shelf, bounded to the east by the Adélie Bank. Further east
219	lays the Adélie Depression and the Mertz Bank, the latter located north of the Mertz Glacier floating ice
220	tongue. The Adélie Land region is dissected by several glaciers which could potentially contribute
221	terrigenous sediment into the coastal zone with the core site located 40 km to the north of the Astrolabe
222	Glacier, and ca. 75 and 300 km northwest of the Zélée and Mertz glaciers, respectively.
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224	The site itself is located within the Dumont d'Urville polynya (DDUP), which has a summer (winter)
225	extent of 13,020 km ² (920 km ²), but is also directly downwind and downcurrent of the much larger and
226	highly productive Mertz Glacier polynya (MGP) to the east, with a summer (winter) extent of 26,600
227	km² (591 km²) (Arrigo and van Dijken, 2003). The MGP forms as a result of reduced sea-ice westward
228	advection due to the presence of the Mertz Glacier Tongue (Massom et al., 2001) and strong katabatic
229	winds which blow off the Antarctic ice sheet with temperatures below -30°C (Bindoff et al., 2000).
230	Katabatic winds freeze the surface waters and blow newly formed ice away from the coast, making the
231	polynya an efficient sea-ice 'factory', with higher rates of sea-ice formation in comparison to non-
232	polynya ocean areas which undergo seasonal sea ice formation (Kusahara et al., 2010). The MGP
233	produces 1.3% of the total Southern Ocean sea ice volume despite occupying less than 0.1% of total
234	Antarctic sea ice extent (Marsland et al., 2004).
235	
236	As a result of the upwelling polynya environments, the area along the Adélie Coast is characterized by
237	extremely high primary productivity, with the water column known to host significant amounts of
238	phytoplankton, dominated by diatoms (Beans et al., 2008). The Mertz Glacier zone is generally
239	characterized by stratified waters in the summer, due to seasonal ice melt, with these conditions
240	corresponding to the highest phytoplankton biomass. The lack of ice cover means polynyas are the first
241	polar marine systems exposed to spring solar radiation, making them regions of enhanced biological
242	productivity compared to adjacent waters. A considerable amount of resultant sedimentation is focused
243	via the westward flowing currents from both of these polynyas within the deep, protected Adélie Basin,

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245 2011). 246 Although biogenic and terrigenous sediment is interpreted to be sourced locally in the Adélie Land 247 region, the mass accumulation rate of these sediments in this drift is associated with the intensity of 248 westward flowing currents (S2.2). Critically, these westward currents also act to transport water masses 249 from further afield, and Site U1357 is directly oceanographically downstream of the Ross Sea, meaning 250 the continental shelf in this region receives significant Antarctic Surface Water (ASSW) transported by 251 the Polar Easterlies and the Antarctic Slope Current (ASC) from the Ross Sea embayment (Fig 3). Thus, 252 changes in the surface waters of the Ross Sea influence Site U1357. Whitworth et al. (1998) confirm the 253 continuity of the westward flowing ASC between the Ross Sea and the Wilkes Land margin. This flow 254 is largely associated with the Antarctic Slope Front, which reflects the strong density contrast between 255 AASW and Circumpolar Deep Water (CDW). McCartney and Donohue (2007) estimate that the 256 257 transport in the westward ASC, which links the Ross Sea to the Wilkes Land margin, reaches 76 Sv (106 m³ s⁻¹). This contributes to a cyclonic gyre, which together with the ASC dominate the circulation at Site 258 U1357. The gyre transports around 35 Sv, and comes mainly from the Ross Sea region, with a lesser 259 contribution from a westward flow associated with the Antarctic Circumpolar Current. 260 While several small glaciers within Adélie Land may contribute meltwater to the site, the region is also 261 likely to be influenced significantly by changes in Ross Sea waters. Freshwater release simulations from 262 the Ross Ice Shelf (RIS) confirm this oceanographic continuity between the Ross Sea and the Wilkes 263 region (Fig 3). Five simulations with fluxes from 0.01 to 1 Sv released from the edge of the RIS all 264 indicate that meltwater is almost completely entrained within the westward coastal surface current and 265 reaches Site U1357 within 4 months to 1 year (Fig 4). These fluxes cover a wide range of meltwater 266 inputs and show a strong linear relationship with salinity at the core site (Fig. 4). This suggests that the 267 magnitude of the signal recorded at Site U1357 is directly related to the magnitude of meltwater. 268 Local processes do also play a critical role in this region. For example, episodic calving events of the 269 Mertz Glacier tongue release fast ice over the drill site and create strong surface water stratification, 270 cutting off local AABW production (Campagne et al., 2015). Although appearing to be only a local 271 272 process, there is still a regional (Ross Sea) influence, as this fast ice that builds up behind the Mertz Glacier is formed by the freezing of fresher AASW transported from the Ross Sea (Fig 3). Thus, 273

resulting in a remarkably high sedimentation rate of ca. 1.5-2 cm year⁻¹ at Site U1357 (Escutia et al.,

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conditions in the Ross Sea, such as the melting of isotopically depleted glacial ice, would influence both the isotopic composition and amount of this sea ice. 4.2 Site specific interpretation of $\delta^2 H_{FA}$ as a glacial meltwater proxy 4.2.1. Source of fatty acids To best interpret the hydrogen isotope signal recorded by the C₁₈ FA, it is important to determine the most likely source these compounds are derived from, and thus the habitat in which they are produced. The C₁₈ FA, however, is known to be produced by a wide range of organisms and so we cannot preclude the possibility of multiple sources, especially in a highly diverse and productive region such as the surface waters of offshore Adélie Land. However, we can attempt to determine the most dominant producer(s), which will help us understand the main signal being recorded by the isotopes. An analysis of the FAs within eight classes of microalgae by Dalsgaard et al. (2003) (compiling results from multiple studies) showed Cryptophyceae, Chlorophyceae, Prasinophyceae and Prymnesiophyceae to be the most dominant producers of total C₁₈ FAs. The Bacillariophyceae class, on the other hand, which includes the diatoms, were found to produce only minor amounts of C₁₈ FA, instead synthesizing abundant C_{16:1} FAs. Thus, despite the water column offshore Adélie Land being dominated by diatoms, these are unlikely to be a major source of the C₁₈ FA within U1357B (Beans et al., 2008; Riaux-Gobin et al., 2011). Of the four microalgae classes dominating C₁₈ production (Dalsgaard et al., 2003), species from the Chlorophyceae and Prymnesiophyceae classes have been observed within surface waters offshore Adélie Land after spring sea-ice break-up (Riaux-Gobin et al., 2011). Here, Phaeocystis antarctica of the Prymnesiophytes was found to dominate the surface water phytoplankton community (representing 16% of the phytoplankton assemblage), whereas Cryptophyceae spp. were found in only minor abundances (Riaux-Gobin et al., 2011). In the Antarctic, Phaeocystis is thought to be the most dominant producer of C_{18} FAs (Dalsgaard et al., 2003), and thus is likely to be a key producer of the C_{18} FA in U1357B samples.

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304 To investigate this further, we measured compound-specific carbon isotopes of the C_{18} FAs in U1357B samples, which gives an average δ^{13} C value of -29.8 \pm 1.0 % (n=85). Budge *et al.* (2008) measured a 305 similar δ^{13} C value of -30.7 \pm 0.8% from C₁₆ FAs derived from Arctic pelagic phytoplankton, while sea 306 ice algae and higher trophic level organisms all had much higher δ¹³C values (sea ice algae having 307 values of -24.0 \pm 2.4‰). Assuming similar values apply for the C_{18} FA and for organisms within the 308 water column at our site, this suggests that our C₁₈ FA is predominantly derived from pelagic 309 phytoplankton. 310 311 Furthermore, δ^{13} C measurements of suspended particulate organic matter (SPOM) near Prydz Bay, East 312 Antarctica by Kopczynska et al. (1995) showed that sites with high δ^{13} C SPOM values (-20.1 to -313 22.4‰) were characterized by diatoms and large heterotrophic dinoflagellates, whereas the lowest δ^{13} C 314 SPOM values (-29.7 to -31.85%) were associated with *Phaeocystis*, naked flagellates and autotrophic 315 dinoflagellates. Wong and Sackett (1978) measured the carbon isotope fractionation of seventeen 316 species of marine phytoplankton and showed that Haptophyceae (of which *Phaeocystis* belongs) had the 317 largest fractionation of -35.5%. 318 319 Therefore, based on the known producers of C₁₈ FAs, observed phytoplankton assemblages within 320 modern surface waters offshore Adélie Land, and the δ^{13} C value of C₁₈ FAs in U1357B samples, as 321 discussed above, we argue that the C_{18} FA here is predominantly produced by *Phaeocystis* (most likely 322 P. antarctica), but with potential minor inputs from other algal species such as Cryptophytes or diatoms. 323 324 Phaeocystis antarctica is a major phytoplankton species within the Antarctic, dominating spring 325 phytoplankton blooms, particularly in the Ross Sea (DiTullio et al., 2000; Schoemann et al., 2005). It is 326 327 known to exist both within sea ice and in open water (Tang et al., 2008) and has been observed in 328 surface waters in great abundance following spring sea-ice break-up, at both coastal and offshore sites in Adélie Land (Riaux-Gobin et al., 2011). 329 330 Although a large proportion of organic matter produced in the surface water is recycled in the upper 331 332 water column, the small fraction which is deposited in the sediment reaches the sea floor through large particles sinking from above as "marine snow". This export production includes large algal cells, fecal 333 pellets, zooplankton carcasses and molts, and amorphous aggregates (Mayer, 1993). In the Ross Sea, 334

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aggregates of P. antarctica, have been observed to sink at speeds of more than 200 m day⁻¹, meaning 335 they could reach deep water very quickly (Asper and Smith, 1999). In this way, a proportion of the lipid 336 content of *P. antarctica* and other algae is transported and sequestered in the sediments. 337 338 Initial diagenesis is characterized by the preferential degradation of more labile organic compounds e.g. 339 sugars, proteins, amino acids. Proportionally, lipids are relatively recalcitrant compared to other 340 biological components and thus are more likely to be preserved as molecular biomarkers on geological 341 timescales, even where the rest of the organism may be completely degraded. The final proportion of 342 lipids that are preserved within sediments are affected by factors including the export production, O₂ 343 content, residence time in the water column and at the sediment/water interface before deposition, 344 molecular reactivity, formation of macromolecular complexes, adsorption to mineral surfaces and 345 bioturbation (Meyers and Ishiwatari, 1993; Killops and Killops, 2004). Within lacustrine sediments, a 346 significant shift in FA distribution has been shown to occur younger than 100 years due to early 347 348 diagenesis, after which the FA distribution remains relatively unaffected by diagenesis (Matsuda, 1978), thus major changes are assumed to reflect primary environmental signals on longer timescales such as in 349 350 our Holocene record. Due to the hyperproductivity of the surface waters offshore Adélie land, we assume the dominant inputs of the C₁₈ FA are from algal sources in overlying waters and upcurrent 351 regions. Allochthonous inputs e.g. long-range aeolian transport of plant material are assumed to be 352 minimal. 353

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4.2.2. Interpretation of hydrogen isotopes

Compound-specific H isotopes of algal biomarkers are a well-used climate proxy in sediments 356 throughout the Cenozoic (e.g. Pagani et al., 2006; Feakins et al., 2012). Although diagenetic alteration, 357 including H-exchange, is possible within sedimentary archives, this has shown to be minimal in 358 sediments younger than 20 Ma (Sessions et al., 2004). Furthermore, if H-exchange had occurred, we 359 would expect $\delta^2 H$ values between different FA chain lengths and closely spaced samples to be driven 360 towards homogeneity, yet large variability remains, suggesting this is not the case. Thus, we are 361 confident that our measured H isotopes are indicating a primary signal throughout the Holocene. 362

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The $\delta^2 H$ value preserved in biomarkers is known to be correlated, but offset, with the $\delta^2 H$ of the water 364 from which the hydrogen was derived. Measured $\delta^2 H$ can therefore be described as a function of either 365 the δ^2 H of the water source, or the fractionation occurring between source water and the lipid $(\epsilon_{l/w})$ (i.e. 366 367 vital effects), in which various environmental factors play a part (Sachse et al., 2012). 368 The main environmental factors controlling $\epsilon_{l/w}$ are salinity and temperature, with which $\delta^2 H$ increases 369 by 1-4‰ per increase in practical salinity unit (psu) (Schouten et al., 2006; Sachse et al., 2012) and 370 decreases by 2-4% per degree C increase (Zhang et al., 2009), respectively. The $\delta^2 H_{FA}$ record from Site 371 U1357 displays an absolute range of ca. 123‰, and millennial to centennial scale variability with an 372 amplitude of ca. 50%, throughout the core. This would imply extremely large and pervasive variations 373 in temperature (up to ca. 60°C) and salinity (up to 123 psu) if fractionation driven by either of these 374 375 factors were the main control. One study has shown the salinity of present day Adélie shelf waters to vary between 34 and 34.8 psu (Bindoff et al., 2000), while tetraether-lipid based subsurface (50-200 m) 376 temperature estimates from nearby Site MD03-2601 (about 50 km west of Site U1357) range from -0.17 377 to 5.35°C over the Holocene (Kim et al., 2010). Therefore, fractionation changes driven by temperature 378 or salinity cannot be invoked as a major control on $\delta^2 H_{FA}$ in the Holocene. 379 380 Thus, the most parsimonious explanation relates to changes in $\delta^2 H_{FA}$ of the water source (Sachse *et al.*, 381 2012). In the Adélie Basin, the most apparent controls on this are advection, upwelling or inputs of 382 isotopically depleted glacial meltwater. The $\delta^2 H_{FA}$ value within Antarctic glaciers is highly depleted 383 relative to sea water due to the Rayleigh distillation process, leading to highly negative isotope values 384 for precipitation over the continent. 385 386 The glacial meltwater originating from the Ross Ice Shelf is likely to combine ice precipitated 387 throughout the Holocene and glacial period, and from both the East and West Antarctic Ice Sheets. 388 However, as noted by Shackleton and Kennett (1975) in their first oxygen isotope record of the 389 Cenozoic (see their Fig. 6), most of the ice that melts around the margin has been coastally precipitated 390 (due to higher accumulation rates). Since ice precipitated further inland has a greater residence time 391 (Shackleton and Kennett, 1975) and significantly lower accumulation rates it will contribute 392 significantly less to this signal. Thus, the ice that contributed to a marine-based ice sheet collapse along 393 this margin is best represented by average values of coastal ice dome records at a similar latitude to that 394

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395 which melted since the LGM (such as TALDICE and Siple Dome) than more southerly locations. Glacial to Holocene $\delta^2 H_{FA}$ values from TALDICE, located on the western edge of the Ross Sea in the 396 East Antarctic, for example, vary between -276.2 and -330.3% (Steig et al., 1998) (converted from δ^{18} O 397 values following the global meteoric water line (GMWL): $\delta^2 H_{FA} = 8.13 \ (\delta^{18}O) + 10.8$), while values 398 from Siple Dome on the eastern edge of the Ross Sea in the West Antarctic, vary from ca. -200 to -399 400 293‰ (Brook et al., 2005). Taking the average of these values as a rough estimate for the meltwater gives a δ^2 H value of ca. -275‰. We note our calculations are based on averages of set time periods, 401 which we expect would integrate ice of various ages - rather than extreme values which could relate to 402 specific melt events of ice or biases to certain ages/regions. This seems reasonable - the isotopic signal 403 of coastal surface waters masses advected from the RIS to the Adélie land (as illustrated in Fig. 3 and 4) 404 405 must integrate a range of source areas across the RIS and from the coast around to Adélie Land. 406 In comparison to the highly negative glacial ice isotope composition, sea surface water δ^{18} O 407 measurements taken near the Mertz Glacier offshore Adélie Land (140-150°E) in summer 2000-2001 408 ranged between -0.47 and 0.05% (Jacobs et al., 2004), equivalent to δ^2 H values of 6.9 to 11.2% 409 410 (average = 9‰) following the GMWL. Thus, the two major hydrogen source pools (RIS glacial ice and ocean water) have highly contrasting isotope values, meaning inputs of upstream glacial ice could have a 411 large effect on surface water δ^2 H values in the Adélie Land region. 412 413 Taking the average glacial meltwater $\delta^2 H$ value as -275% and the average modern Adélie surface water 414 δ^2 H value of 9‰ as end-members, and assuming a biosynthetic offset between the FA and sea water of 415 173‰ (see below), we can use a simple mixing model to estimate the percentage of glacial meltwater 416 required in the surface waters to change the $\delta^2 H_{FA}$ value to those recorded in U1357B samples. The 417 most negative values occur during the early Holocene, 11.4 – 8.2 ka, averaging -214.2‰ (n=18) which, 418 converted to a surface water value of -41%, requires 17.6% of the surface water to be comprised of 419 glacial meltwater. During this time, we argue that large volumes of meltwater were reaching the core 420 site as local glaciers retreated, leading to intense surface-water stratification. Thus, a relatively high 421 percentage of meltwater in the Adélie Land surface waters seems reasonable. During the mid-Holocene 422 423 (5-4 ka), the average δ^2 H_{FA} is very similar (-213.9\%, n=7), requiring 17.2\% of the surface water to be derived from glacial meltwater. During this time, we argue for the dominant meltwater source as coming 424

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425 from the Ross Sea, and interpret this as a major period of glacial retreat (see section 5.2), during which 426 large volumes of meltwater are injected into the surface water and transported to the Adélie coast. In contrast, the most recent samples (last 0.5 ka, n=7), which includes the most positive value of the record, 427 has an average δ^2 H FA value of -174.5%. This brings the surface water value up to -1.5%, which 428 approaches modern measured values, and requires just 3.7% (e.g. well within uncertainties) of the 429 surface waters in the Adélie Land to be glacial meltwater. However, it is also possible that the meltwater 430 431 was dominated by more LGM-aged ice. In either case, perturbation of the exact isotopic values still indicate only significant changes in the flux of glacial meltwater can account for this signal. For 432 example, the use of -330% (LGM values) for the ice input gives an estimate of 3% of the surface water 433 being comprised of glacial meltwater for latest Holocene values, and 14.7% for pre 8 ka values. Taking -434 240% (Holocene values) for the ice input gives an estimate of 4% for latest Holocene values, and 20% 435 for pre 8 ka values). Thus even with changing isotopic values though the deglacial, this signal of 436 changing meltwater flux would still dominate. We note these are semi-quantitative estimates, as the 437 salinity and temperature fractionation could reduce these estimates further (but cannot account for the 438 whole signal). 439 440 Surface water δ¹⁸O values around Antarctica (below 60°S), measured between 1964 and 2006, ranged 441 from -8.52% to 0.42% (Schmidt et al., 1999), the most negative value having been measured proximal 442 to the George VI Ice Shelf edge, where high melt rates have been observed (Potter and Paren, 1985). If 443 converted to δ^2 H using the global meteoric water line, these values give a δ^2 H range of 83.4‰. Thus, 444 our absolute $\delta^2 H_{FA}$ range of 123% over the Holocene suggests a range of isotopically depleted 445 meltwater inputs to our core site over this time that are 1.5 times greater than that occurring in different 446 locations around the Antarctic in recent decades. This seems plausible seen as geological evidence 447 indicates large glacial retreat and ice mass loss occurred from the Ross Sea sector during the Holocene 448 (McKay et al., 2016), meaning resultant changes in surface water are likely to be greater in magnitude 449 than observed around the Antarctic in recent decades. This assumes a relatively constant value for the 450 451 isotopic composition of glacial meltwater, however, there is likely to be some variability due to the possibility of melting ice of different δ^2 H values. But, as discussed above, the meltwater is best 452 represented by the average values of the ice sheet, rather than extreme values, since it must (over the 453 broad expanse of the RIS) include an integrated signal, and thus the actual variation in meltwater $\delta^2 H$ 454 will be significantly within the range of the end-members. 455

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457 Although the biosynthetic fractionation of the C₁₈ FAs in U1357B is unknown, we assume that the offset with surface water remains relatively constant throughout the record. Sessions et al. (1999) showed the 458 biosynthetic fractionation of hydrogen isotopes in the C₁₈ FA from four different marine algae to range 459 from -189 to -157‰. If we take the average of these values of 173‰ and apply this as a biosynthetic 460 461 offset to the youngest samples in U1357B (last 0.5 ka, n=7), which includes the most positive value of the record, gives an average δ^2 H_{FA} value of -174.5‰. This brings the surface water value up to -1.5‰, 462 which approaches modern measured values (Jacobs et al., 2004). 463 464 Furthermore, it is interesting to note that the biosynthetic offsets measured by Sessions et al. (1999) for 465 the C_{18} FA from different algal species have a total δ^2 H range of 32‰. Although we cannot dismiss 466 changes in the relative contribution of C₁₈ from different species in U1357B samples (and thus different 467 biosynthetic fractionations), we argue this would only be a minor control on $\delta^2 H$ compared to other 468 influences. As a thought experiment, taking the above end-members for biosynthetic fractionation from 469 Sessions et al. (1999), even with a 100% change in C₁₈ producer to a different algal source, this could 470 only explain a quarter of the observed δ^2 H change (i.e. 32% of 123%). 471

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5. DISCUSSION

glaciers and ice-shelves.

479 The stratigraphy of U1357B is divided into three units: the lowermost 10 cm recovered Last Glacial Maximum (LGM) till (Unit III), overlain by 15 m of laminated mud-rich diatom oozes with ice rafted 480 481 debris (IRD) (Unit II), and the upper most 171 m (Unit I) consists of laminated diatom ooze with a general lack of IRD and a significant reduction in terrigenous sediment (Escutia et al., 2011). The 482 483 sedimentology and geometry of the drift prior to ~11.4 ka (Unit II) is consistent with the calving bay reentrant model (Domack et al., 2006; Leventer et al., 2006) (Fig. 1 and Supplementary Fig. S5; 484 485 Supplementary Materials), whereby LGM ice retreated in the deeper troughs while remaining grounded on shallower banks and ridges. Sediment laden meltwater and IRD content in Unit II (>11.4 ka) is thus 486

Therefore, we interpret the first order control on $\delta^2 H_{FA}$ at Site U1357 as inputs of isotopically depleted

glacial meltwater. Such inputs are, in turn, influenced by the mass balance of the proximal or up-current

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487 likely derived from local outlet glaciers. However, anomalously old radiocarbon ages due to glacial reworking precludes development of a reliable age model prior to the Holocene (Supplementary 488 489 Materials). 490 5.1 Early Holocene 491 492 The base of the drift deposit shows downlapping of material suggesting a supply from the south, indicating local focusing of meltwater and terrigenous material was the dominating influence until 11.4 493 ka (Supplementary Materials). This is overlain by onlapping strata (Unit I) with the drift forming an 494 east-west elongation on the northern flank of the Dumont d'Urville Trough, which is more consistent 495 with advection of material from the east than with delivery from local outlet glaciers to the south. Thus, 496 an increased meltwater influence from the Ross Sea is likely since this time (Supplementary Materials). 497 498 Due to the potential for competing sources of glacial meltwater in the earliest Holocene, we focus our 499 500 study on Unit I, where there is less influence of calving bay processes (Escutia et al., 2011). However, the earliest part of Unit I (11.4 to 8 ka BP), which includes the most negative $\delta^2 H_{FA}$ values, is 501 characterized by a very gradual upcore increase of sorting in the terrigenous sediment supply, decreasing 502 natural gamma ray (NGR) values (Supplementary Fig. S4) and a general lack of IRD. We conservatively 503 interpret this as potentially maintaining some local glacial meltwater input from local outlet glaciers in 504 505 the lowermost interval of Unit I. Nevertheless, this process was probably greatly reduced relative to Unit II deposition and it is likely much of this signal between 11.4 and 8 ka could still be derived from water 506 masses advecting to the site from the east (e.g. the Ross Sea). 507 508 509 This is supported by geological and cosmogenic evidence which demonstrates that the Wilkes Land margin of the East Antarctic, and also the Amundsen Sea margins, had retreated to their modern-day 510 positions by ~10 ka (Bentley et al., 2014; Mackintosh et al., 2014; Hillenbrand et al., 2017); 511 Supplementary Materials), thus these margins are unlikely to contribute large scale shifts in meltwater 512 513 fluxes to the Adélie Coast during most of the Holocene. Glacial retreat, however, persisted in the Ross Sea until at least 3 ka (Anderson et al., 2014; Spector et al., 2017) (Supplementary Materials) providing 514 a large upstream source of meltwater feeding into the Adélie Coast. We therefore interpret our meltwater 515 signal as being dominated by Ross Sea inputs since at least 8 ka, but potentially as early as 11.4 ka. 516 Furthermore, the retreat of grounded ice from the outer Ross Sea continental shelf was accompanied by 517

to the drift deposit.

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518 the growth of a significant floating ice shelf (which was not the case in the Amundsen Sea or proximal 519 East Antarctic coast) (Bentley et al., 2014). 520 An overall trend to more positive $\delta^2 H_{FA}$ values, from the most negative value of the record at ~9.6 ka, to 521 ~8 ka indicates decreasing meltwater (Fig. 2a), thus suggesting a gradually diminished input from either 522 local outlet glaciers or the Ross Sea. This is associated with an increase in MARs, between 10 and 8 ka, 523 524 and is tentatively interpreted to represent the final retreat of residual ice from local bathymetric highs allowing more material to advect into the drift (Supplementary Fig. S4). Although there is millennial 525 scale variability, MARs remain relatively high until 4.5 ka. However, δ²H_{FA} and MARs show greater 526 coherence at the millennial-scale after 7 ka BP, suggesting that increased fluxes of glacial meltwater 527 broadly corresponded to stronger easterly currents, which advected biogenic and terrigenous material 528 into the drift. 529 530 5.2 Middle Holocene 531 532 A negative excursion in $\delta^2 H_{FA}$ starting from 6 ka and culminating at 4.5 ka is interpreted to record a period of enhanced glacial meltwater flux to the site relating to a final retreat phase of the major ice 533 534 sheet grounding line in the Ross Sea embayment (Fig. 5). A marked enrichment of Ba/Ti ratios also occurs at 4.5 ka, reaching values of 36.1, on a background of baseline fluctuations between 0.1 and 2.7 535 (Fig. 2g), which suggests enhanced primary productivity, potentially driven by meltwater-induced 536 stratification. Ongoing Holocene retreat in the Ross Sea is interpreted to be primarily the consequence of 537 538 marine ice sheet instability processes resulting from the overdeepened continental shelf in that sector 539 (McKay et al., 2016). 540 The $\delta^2 H_{FA}$ peak at 4.5 ka in U1357 coincides directly with a rapid shift in HBI biomarker ratios at the 541 site, as well as sea ice proxies recorded in nearby site MD03-2601, in the Ross embayment (Taylor 542 Dome ice core on a revised age model) (Steig et al., 1998; Baggenstos et al., 2018) and Prydz Bay 543 544 (JPC24) (Denis et al., 2010) (Fig. 2), reflecting a widespread increase in coastal sea-ice concentration and duration. We interpret decreasing MAR and finer-grained terrigenous content (e.g. increased mud 545 percent) at Site U1357 after 4.5 ka (Supplementary Fig. S4) to also be a consequence of increased 546 547 coastal sea ice, reducing wind stress on the ocean surface and limiting the easterly advection of detritus

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550 Coastal sea-ice concentration and duration remain high throughout the rest of the Holocene (this study and Steig et al., 1998; Crosta et al., 2008; Denis et al., 2010), compared to the period before 4.5 ka, 551 despite a decrease in glacial meltwater flux to the U1357 site. In addition, meltwater input prior to 4.5 ka 552 does not have a major influence on sea ice extent. Thus, an increase in meltwater flux cannot explain the 553 Neoglacial intensification of sea ice at ~4.5 ka. Here, we propose that greater coastal sea ice cover since 554 4.5 ka is related to the development of a large ice-shelf cavity in the Ross Sea as the ice sheet retreats 555 (Fig. 5), which pervasively modified ice shelf-ocean interactions and increased sea ice production. 556 Models suggest a large cavity on the continental shelf increases contact between basal-ice and 557 circulating ocean water, driving the formation of a cool, fresh water mass feeding into the AASW, 558 stabilizing the water column and enhancing the production of sea ice (Hellmer, 2004) (Fig. 5). However, 559 under small cavities such as in the modern Amundsen Sea influenced by warm-water incursions, ice 560 shelf melting results in an "ice pump" enhancement of sub-ice shelf circulation. This increases flow of 561 562 warm Circumpolar Deep Water (CDW) under the ice shelf that is 100-500 times the rate of melt, and this volume of water does not allow for supercooling. Small cavity ice shelf outflows are therefore warm 563 and act to restrict sea ice at the ice shelf front (Jourdain et al., 2017). Thus, during the Holocene, the size 564 of the cavity must have reached a threshold after which this positive warming feedback switched to a 565 negative feedback. We argue that such a tipping point takes place at 4.5 ka BP, when our proxy data 566 suggest meltwater peaks, and would explain why the increase in sea-ice concentration appears rapid and 567 only occurs at the peak of the meltwater input, and not during its prior increase, or previous meltwater 568 inputs (Fig. 2a-g). 569 570 Although the glacial meltwater volume is greatly reduced after 4.5 ka BP, the volume of Ice Shelf Water 571 (ISW) produced beneath the modern RIS is estimated at 0.86 Sv-1.6 Sv (Holland et al., 2003; Smethie 572 573 and Jacobs, 2005). We note that ISW is not glacial meltwater, but it is defined as a supercooled water mass formed through interaction with the base of the RIS, but once formed acts to modify other water 574 masses in the Ross Sea. A significant proportion of ISW is high salinity and is thus advected northwards 575 at intermediate waters depth to ultimately form AABW. However, a significant volume of ISW is lower 576 577 salinity and buoyant, due to development of frazil ice, and acts to mixes with surface waters (Robinson et al., 2014). Currently, a 0.4 Sv plume of ISW in the western margin of the Ross Ice Shelf (Robinson et 578 al., 2014) is directly delivered to the surface resulting in enhanced sea ice production, while seasonal 579

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melt of this enhanced sea ice further acts to cool and freshen surface waters. Although unrealistic in the context of a post-LGM meltwater flux from the Ross Sea alone, the larger meltwater release scenarios in our simulations (0.5 to 1 Sv) show the potential pathways that a cool, fresher surface water mass collecting and forming on the broad Ross Sea continental shelf would follow (Fig. 3). These waters are transported in easterly coastal currents to the Weddell Sea and the Antarctic Peninsula. This eventually retroflects to join the Antarctic Circumpolar Current (Fig. 3b), and thus has potential for cooling and freshening in the South Atlantic far offshore, as the ice shelf cavity increased in the Ross Sea. Indeed, offshore site ODP 1094 records increased lithics in the South Atlantic after 4.5 ka (Fig. 2f), relative to the period before, suggested to have been predominantly transported by sea ice indicating a cooling in sea surface temperatures and increase in sea-ice extent in the South Atlantic at this time (Nielsen *et al.*, 2007). However, it also is feasible that this circum-Antarctic cooling signal indicates similar melt processes may have been occurring in the Weddell Sea at ~4.5 ka, as suggested by cosmogenic nuclide data (Hein *et al.*, 2016).

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5.3 What Drove the Neoglacial Transition?

595 Our observed coastal sea-ice increase is part of a widespread transition to Neoglacial conditions both globally and at high southern latitudes (Kim et al., 2002; Masson-Delmotte et al., 2011; Marcott et al., 596 2013; Solomina et al., 2015). However, most current climate models do not simulate this cooling trend, 597 resulting in a significant data-model mismatch (Liu et al., 2014) (Supplementary Fig. S7). Marine ice 598 sheet retreat along the entire Pacific margin of West Antarctic has previously been proposed to be 599 triggered by enhanced wind-driven incursions of warm CDW onto the continental shelves in the early 600 601 Holocene (Hillenbrand et al., 2017), with continued retreat in the Ross Sea being the consequence of the overdeepened continental shelf and marine ice sheet instability processes (McKay et al., 2016). 602 However, we propose that a series of negative feedbacks associated with this retreat and RIS cavity 603 604 expansion occurred in the mid-Holocene, with similar processes possibly occurring in the Weddell Sea, 605 leading to the onset and continuation of Neoglacial conditions. Widespread albedo changes associated with increased coastal sea ice would have amplified regional cooling trends (Masson-Delmotte et al., 606 2011), whilst increased stratification resulting from seasonal sea-ice melt and increased ISW production 607 608 drove the deepening of the fresher water surface isopycnal at the continental shelf break. Grounding line retreat creates new space for continental shelf water masses to form, while ice shelf cavity expansion 609 promotes supercooling and freshening of AASW. Thus, as seasonal sea ice melt and ice shelf 610

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611 supercooling processes played a greater role in enhancing AASW production on the continental shelf, 612 they would have acted to restrict warmer subsurface water transport onto the continental shelf (Smith Jr. et al., 2012) (Fig. 5). Furthermore, the Neoglacial sea-ice increase itself may have been associated with 613 a stabilising feedback mechanism (which also is not resolved in ice-ocean models) through its role in 614 dampening ocean-induced wave flexural stresses at ice shelf margins, reducing their vulnerability to 615 616 catastrophic collapse (Massom et al., 2018). We suggest that some combination of the above processes slowed the rate of Ross Sea grounding line retreat (Supplementary Materials) and reduced basal ice shelf 617 melt as indicated by a trend towards more positive $\delta^2 H_{FA}$ values in U1357 between 4.5 and 3 ka (Fig. 618 2a). Furthermore, large Antarctic ice shelves currently have large zones of marine accreted ice resulting 619 from supercooling (Rignot et al., 2013), thus the signature of $\delta^2 H_{FA}$ is anticipated to become more 620 positive as the ice shelf approaches a steady state of mass balance, relative to the thinning phases when 621 basal melt rates exceed that of accretion. The stabilization of $\delta^2 H_{FA}$ values observed at 3 ka in U1357 622 suggests the Ross Ice Shelf has maintained a relatively steady state of mass balance since this time. 623 624 A recent study implies an increase in katabatic winds since at least 3.6 ka in the Ross Sea (Mezgec et al., 625 2017) (Supplementary Materials), leading to enhanced polynya activity. During colder Antarctic 626 627 climates, increased latitudinal temperature gradients enhanced katabatic winds in the Ross Sea (Rhodes 628 et al., 2012). We interpret this katabatic wind and polynya activity signal to be a response to the preceding Neoglacial cooling at 4.5 ka and evolution of the modern ocean-ice shelf connectivity, which 629 our data suggest was primarily driven by ice shelf cavity expansion. Furthermore, this transition takes 630 631 place on the background of declining winter insolation (Berger and Loutre, 1991) which would have acted to further enhance and maintain these changes. This insolation decline has previously been 632 hypothesised as a driver of the Neoglacial increase in coastal sea ice (Denis et al., 2010), however this 633 monotonic decrease contrasts with the markedly rapid increase in sea ice observed in many records (Fig 634 2). Our mechanism of ice shelf cavity expansion, reaching a threshold that promoted significant 635 supercooling of continental shelf water masses, reconciles both the rapidity and timing of Neoglacial 636 onset in the middle Holocene. 637 638

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particular the incorporation of evolving ice shelf cavities, with Trace-21k for example, instead

6. Conclusions and Implications for Antarctic Climate, Sea-Ice and Ice Shelf Behaviour

The lack of these coupled ice-ocean processes is apparent in recent Earth system model experiments, in





simulating a decrease in Antarctic sea-ice extent and thickness after 5 ka (Supplementary Fig. S7). These model outputs are in direct contrast to multiple lines of proxy data in this study and previous work (Steig *et al.*, 1998; Crosta *et al.*, 2008; Denis *et al.*, 2010). Consequently, our results provide insights into the magnitude of this data-model mismatch, as well as a mechanism for rapid sea-ice change and grounding line stabilisation on the background of a warming climate (Liu *et al.*, 2014), both on modern and Holocene time scales. Better representation of the role of evolving ice shelf cavities on oceanic water mass evolution and sea-ice dynamics, which our data indicate acted as a strong negative feedback, will be fundamental to understanding the oceanographic and glaciological implications of future ice shelf loss in the Antarctic.

Figures

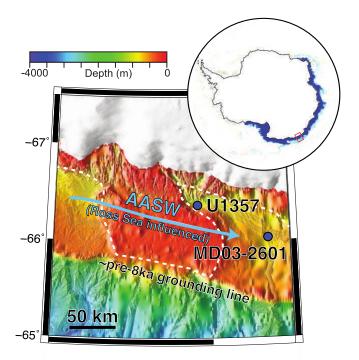


Figure 1: Location of Sites U1357 and MD03-2601 (blue dots). The ice sheet grounding line formed a

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calving-bay environment (dashed white line) prior to 11.4 ka, but since at least 8.2 ka Antarctic Surface
Water flow is largely advected from the Ross Sea (blue line). Inset map: pathway of freshwater (dark
blue) after 1 year of 1 Sv meltwater released from along the edge of the Ross Ice Shelf in a model
simulation (Supplementary Materials).

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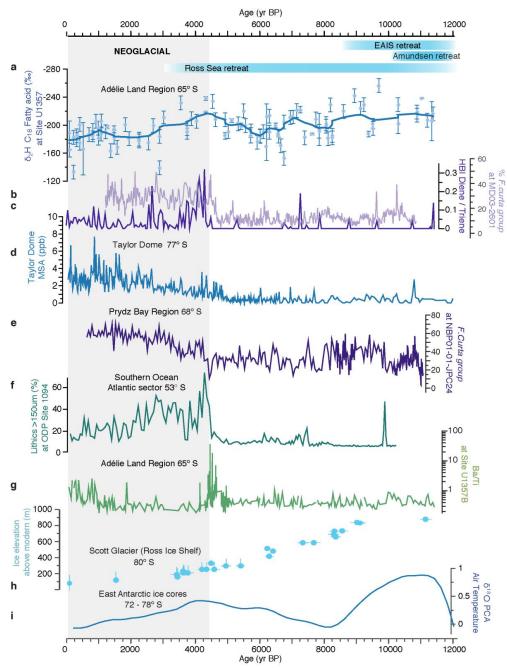


Figure 2: Holocene Adélie Land proxy records from IODP Site U1357 and other circum-Antarctic sites. Glacial retreat chronologies are shown as bars at the top as discussed in the text. a) δ^2 H C₁₈ fatty acid at Site U1357 (errors bars based on replicates), with robust locally weighted smoothing (rlowss). b)

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666	Fraguariopsis curta group (F. curta and F. cylinarus) relative abundance at MD03-2601, as a proxy of
667	sea-ice conditions (Crosta et al., 2008) c) di-unsaturated HBI (C25:2; Diene)/tri-unsaturated HBI isomer
668	(C _{25:3} ; Triene) ratio at Site U1357 d) Methanesulfonate (MSA) concentrations (ppb) from Taylor Dome
669	ice core e) F. curta group relative abundances in core NBP-01-JPC24 f) Coarse lithic (ice-rafted)
670	content at ODP 1094 (Nielsen et al., 2007) g) Ba/Ti (logarithmic scale) at Site U1357 h) 10 Be
671	cosmogenic nuclide ages from Scott Glacier in the SW Ross Ice Shelf region (Spector et al., 2017) i)
672	Temperature signal from principal component analyses of five $\delta^{18}O$ records in five East Antarctic ice
673	cores (Vostok, EPICA Dome C, EPICA Dronning Maud Land, Dome Fuji and Talos Dome) (Masson-
674	Delmotte et al., 2011).
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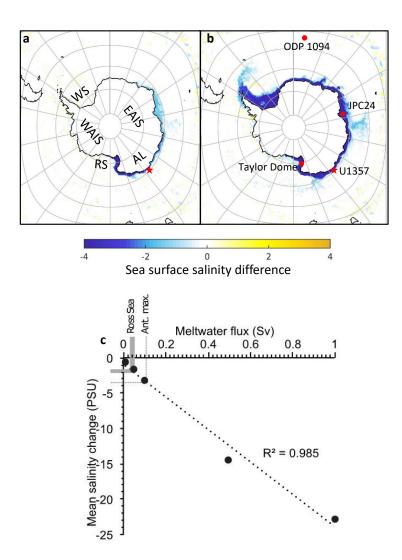


Figure 3: MITgcm simulations of meltwater release from along the edge of the Ross Ice Shelf. First two images show sea-surface salinity difference (in practical salinity units) after 3.5 model years resulting from meltwater release volumes of a) 0.1 Sv and b) 0.5 Sv. Red star indicates position of Site U1357 (this study) and red dots show positions of other core sites mentioned in this study where a Mid Holocene increase in sea ice and/or cooling is recorded: Taylor Dome (Steig *et al.*, 1998; Baggenstos *et al.*, 2018), JPC24 (Denis *et al.*, 2010) and ODP 1094 (Nielsen *et al.*, 2007). AL = Adélie Land, RS = Ross Sea, WS = Weddell Sea, EAIS = East Antarctic Ice Sheet, WAIS = West Antarctic Ice Sheet. c) Scatter plot of simulated meltwater flux (Sv) against mean salinity difference at U1357 core site. Grey





band indicates range of plausible Holocene to deglacial Ross Sea meltwater inputs. Dotted line indicates maximum Antarctic meltwater during the Holocene.

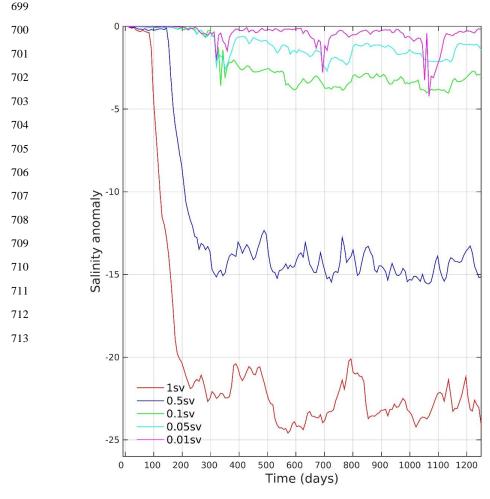
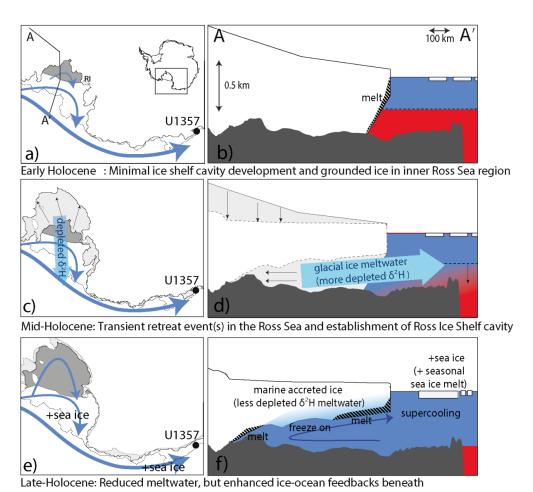


Figure 4 Simulated salinity anomalies over time at Site U1357 for the five meltwater release experiments.







greatly expanded Ross Ice Shelf cavity

Figure 5: Conceptual model of evolving Holocene glacial and oceanographic conditions in the Ross Sea region. Panels on the left show modelled grounding line positions (McKay *et al.*, 2016), and proposed circulation of surface and sub-ice shelf circulating waters (light blue arrows). Panels on the right show cross sections of the Ross Ice Shelf (RIS) and ice-ocean interactions. Dark blue = cool surface waters, Red = warm subsurface waters. a) The grounding line in Adélie Land is near its modern location, but near Ross Island (RI) in the Ross Sea, and ice shelf cavity (dark grey shading) is reduced in size relative to today (McKay *et al.*, 2016). b) Continental shelf profile A-A' (panel a) shows a Ross Sea grounding line in a mid-continental shelf location in close proximity to the RIS calving line (McKay *et al.*, 2016), with subsurface warming on the continental shelf triggering WAIS deglaciation (Hillenbrand *et al.*, 2017). c) Most grounding line retreat south of RI occurred between 9 and 4.5 ka (light grey

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725 shading with black arrows represents area of retreat over this period), proposed to be the consequence of 726 marine ice sheet instability, but the ice shelf calving line remained near its present position (McKay et al., 2016; Spector et al., 2017). d) Grounding line retreat and ice shelf thinning released meltwater with 727 negative δ^2 H into the surface waters. Increasing ice shelf-oceanic interactions with the development of 728 the ice shelf cavity (dark grey) led to enhanced Antarctic Surface Water formation; f) Minimal 729 grounding line retreat has occurred since 4.5 ka, and the RIS supercools AASW leading to enhanced 730 731 sea-ice formation despite reduced glacial meltwater flux. Seasonal sea ice meltwater further freshens and cools AASW. Increased production of AASW on the continental shelf leads to isopycnal deepening 732 (dotted line) and limits flow onto the continental shelf slowing further grounding line retreat. However, 733

as the ice shelf is near steady state mass balance and there is a component of marine accreted ice at the

base of the ice shelf (Rignot *et al.*, 2013), the strength of the δ^2 H signal is reduced relative to periods of mass balance loss.

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