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Absence of a strong, deep-reaching Antarctic Circumpolar

2 Current zonal flow across the Tasmanian Gateway during

3 the Oligocene to early Miocene.

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21 ABSTRACT

The vigorous eastward flow of the Antarctic Circumpolar Current (ACC) connects all major ocean basins and plays a prominent role in the transport of heat, carbon and nutrients around the globe. However, the establishment of a deep circumpolar flow, similar to the present-day ACC, remains controversial thereby obscuring our 26 understanding of its climatic impact. Deciphering the chemical composition of 27 Circumpolar Deep Water (CDW) within the ACC can provide critical insights about its 28 development and evolution. Here we present new fossil fish teeth/bone debris neodymium 29 isotope (ϵ_{Nd}) records from Deep Sea Drilling Project (DSDP) Sites 278 and 274 in the 30 southwest Pacific Ocean, with the aim to trace changes in deep water masses across the Tasmanian Gateway between the early Oligocene and early Miocene (~ 33-22 Ma). Site 31 32 274 provides the first Nd isotope record proximal to the Ross Sea during the Oligocene (33.5-23.4 Ma). Its Nd isotope composition shows excursions to very radiogenic values, 33 34 $\varepsilon_{Nd(t)}$ = -3.1 and $\varepsilon_{Nd(t)}$ -3.7, at 33.5 Ma and 23.8 Ma, respectively, in response to major 35 steps in Antarctic ice sheet expansion. A shift to lower, more unradiogenic $\epsilon_{Nd(t)}$ values between 29.7 and 29.1Ma is linked to an increased influence of proto-CDW upwelling at 36 37 the site. In contrast, the Nd isotope record from Site 278 in the southern Emerald Basin 38 shows little variability ($\varepsilon_{Nd(t)}$ = -6.0 to -6.7) throughout the Oligocene and early Miocene (30.9-21.8 Ma). Comparison with published data north of the ACC path, demonstrates 39 40 the presence of two deep water masses in the South Pacific prior to the inferred onset of the ACC (33-30 Ma), one occupying depths between ~2500 and 3000 m ($\varepsilon_{Nd(t)}$ ~-3 to -5) 41 42 and a deep/bottom water mass (> 3000 m) with a more unradiogenic Nd isotope 43 composition ($\epsilon_{Nd(t)=} \sim -6$). Site 278 located close to the proto-polar front (proto-PF) 44 indicates that following the inferred onset of the ACC, deep waters bathing the southern 45 Emerald Basin remained more radiogenic in the Southwest Pacific compared to sites along the proto-PF in the South Atlantic and Indian Ocean ($\varepsilon_{Nd(t)} \sim -8.1$). This indicates 46 47 a provinciality in Nd isotope compositions of deep waters along the proto-PF across the 48 Tasmanian Gateway. Our data are incompatible with the existence of a modern-like homogenous (lateral and vertical) Nd isotope composition of CDW along the main flow 49 50 path of the ACC in all oceanic basins in the Oligocene to early Miocene. We attribute

distinct Nd isotope compositions of deep waters across the Tasmanian Gateway to reflect a less deep reaching and weaker ACC (proto-ACC) than today. Our findings suggest that the modern strong and deep-reaching ACC flow must have been developed at a later point in the Neogene.

55

Keywords: ACC, CDW, neodymium isotope ratios, deep ocean circulation, Tasmanian
Gateway, Oligocene-early Miocene

58 1. INTRODUCTION

59 The Antarctic Circumpolar Current (ACC) is the world's largest and strongest ocean current, transporting ~136-162 x 10^6 m³ s⁻¹ of water along a ~20,000 km long path around 60 61 Antarctica (e.g., Rintoul et al., 2014). The absence of continental barriers around 60°S permits 62 the vigorous eastward flow of the ACC, which is driven by the strong westerly winds 63 (westerlies) and buoyancy forcing (Rintoul, 2018). Most of the circumpolar flow of the ACC takes place along the Polar and the Subantarctic Fronts (PF and SAF, respectively), which 64 extend from the surface to the seafloor (i.e. deep-reaching), connecting the Atlantic, Pacific 65 66 and Indian Oceans, thus actively modulating the global ocean circulation (Orsi et al., 1995; 67 Sokolov and Rintoul, 2007; Rintoul, 2018). This interbasin connection of the ACC is a critical 68 feature of the modern global overturning circulation, which carries heat, carbon and nutrients 69 around the globe. It modulates the exchange of heat, moisture and carbon dioxide (CO₂) 70 between the ocean and the atmosphere and controls local and global marine primary productivity (Rintoul, 2018). 71

Despite the critical role of today's strong deep-reaching ACC flow in the global ocean circulation and Earth's climate, there is still an ongoing debate regarding the timing of its onset and evolution towards a modern-like configuration throughout the Cenozoic period. Major 75 changes in paleogeography and seafloor paleobathymetry related to tectonic activity through 76 time, as well as changes in climate are likely to have controlled the initiation and evolution of 77 the ACC during the Cenozoic. For example, it is widely accepted that the ACC only initiated 78 after the opening of the two last major land barriers within its circum-Antarctic flow, the Drake 79 Passage and the Tasmanian Gateway, and their respective deepening to bathyal depths (>2000 80 m). Dating the time when the Drake Passage reached this depth is controversial, spanning from 81 the middle Eocene to the late Miocene (see Dalziel, 2014 for discussion). In contrast, the 82 opening of the Tasmanian Gateway is better constrained (Stickley et al, 2004).

83 A Southern Ocean with closed tectonic gateways featured clockwise gyres in the South 84 Pacific and South Indian/Atlantic Ocean basins (Huber et al., 2004). Between ~49 Ma and 35.5 85 Ma, a shallow current flowing westward across the Tasmanian Gateway (from the Pacific to 86 the Indian Ocean) was proposed based on marine microfossils, organic geochemical records 87 and model simulations (Huber et al., 2004; Bijl et al., 2013). Initial opening was followed by 88 intensified deepening of the seaway between ~35.5 and 30.2 Ma as indicated by the analysis 89 of sedimentological, micropaleontological and paleomagnetic data (Stickley et al., 2004). 90 Chemical water mass reconstructions suggest a westward flowing (i.e., from the Pacific to the 91 Indian Ocean) deep water current connecting both sides of the northern parts of the Tasmanian 92 Gateway between 32 and 30 Ma (Scher et al., 2015). A deep eastward flowing water connection 93 between the Indian and Pacific sectors of the Southern Ocean, was only established after 30 94 Ma, probably due to alignment of the northern part of the Tasmanian Gateway within the 95 latitudinal band of the westerlies (Scher et al., 2015). However, micropaleontological and 96 paleotemperature records suggest weak latitudinal paleotemperature gradients and warm 97 conditions prevailed in the Southern Ocean until at least the mid-Miocene (Bijl et al., 2018; 98 Hartman et al., 2018; Salabarnada et al., 2018; Sangiorgi et al., 2018). Thus, the characteristics 99 of this nascent ACC remain poorly understood.

100 One possible way to reconstruct past connectivity along the ACC is to track the 101 evolution of Circumpolar Deep Water (CDW), using neodymium (Nd) isotope compositions (¹⁴³Nd/¹⁴⁴Nd ratio) of ambient seawater in fossil fish teeth and bones (e.g., Wright et al., 2018). 102 103 Circumpolar Deep Water is the most voluminous water mass in the Southern Ocean, occupying 104 the shallow to deep layers of the ACC in the Southern Ocean (Orsi et al., 1995). Today, CDW 105 has a vertically and horizontally homogenous Nd isotope composition, probably due to the 106 strong vertical and horizontal mixing along the frontal systems (e.g., Stichel et al., 2012; 107 Lambelet et al., 2018). These findings indicate that the evolution of the Nd isotope composition 108 of CDW in the past may provide important insights into interbasinal connections/circulation in 109 the Southern Ocean.

110 Neodymium is incorporated into fish teeth at the sediment-water interface at the time 111 of deposition, remineralisation and burial (Martin and Haley, 2000). Post-burial alteration is 112 generally negligible relative to the early diagenetic rare earth element (REE) uptake, allowing 113 retention of the Nd isotope composition of bottom water signals (e.g., Martin and Haley, 2000). 114 Neodymium is a lithogenic element, introduced to the ocean at its interfaces with the solid 115 earth (e.g., van Flierdt et al., 2016). Water masses forming in the different oceanic basins are 116 distinct in their Nd isotope fingerprint, due to the distribution of geological ages and lithologies 117 around the globe, with younger volcanogenic material yielding more radiogenic (higher) Nd 118 isotope compositions, and older cratonic rocks being characterised by less radiogenic (lower) Nd isotope compositions. ε_{Nd} denotes the deviation of a measured ¹⁴³Nd/¹⁴⁴Nd ratio from the 119 120 chondritic uniform reservoir in parts per 10,000 (Jacobsen and Wasserburg, 1980). Because 121 seawater Nd has a relatively short residence time in the ocean (~500 to 1,000 years) (Tachikawa 122 et al., 2003), Nd can be carried from one ocean basin to another, and hence trace the provenance 123 of water masses.

124 Here we present new Oligocene to lower Miocene neodymium isotope data obtained 125 from fossil fish teeth and bones (fish debris) in sediments recovered by the Deep Sea Drilling 126 Project (DSDP) at Sites 278 and 274. Site 278 is strategically located within the main path of 127 the modern ACC in the southern Emerald Basin (Kennett et al., 1975; Rintoul et al., 2014), 128 while Site 274 is situated proximal to the Ross Sea in the Adare Basin (Figs. 1A; 1B; S1) 129 (Hayes et al, 1975). Our new data provide critical insights into the chemical evolution of CDW 130 in the Southwest Pacific in the time period following the initial (northern) opening of the 131 Tasmanian Gateway and leading up to the development of the modern-like ACC.

132 2. Modern oceanographic setting of drill sites DSDP 274 and 278

133 To study the history of the ACC, and particularly the chemical composition of deep 134 waters through the Oligocene to early Miocene, we used sediments recovered from Hole 278 135 (Kennett et al., 1975). Site 278 (56°33.42'S, 160°04.29'E, 3675 m water depth) is located on a 136 pelagic contourite drift in the southern Emerald Basin (Figs. 1A; 1B) (Kennett et al., 1975). At 137 present, the site is located close to the northern branch of the Polar Front (PF) and within the 138 main pathway of the ACC (Sokolov and Rintoul, 2007; Rintoul et al., 2014). As the ACC enters 139 the Southwest Pacific through the Tasmanian Gateway, its flow is constrained by the 140 Macquarie Ridge, which extends from depths of 5000 m to an average depth of 1500 m between 141 47°S and 56°S (Rintoul et al., 2014). The ACC passes over and through narrow gaps of the 142 Macquarie Ridge and south of the ridge crest around 56°S, before reaching the southern 143 Emerald Basin (Rintoul et al., 2014). From there, the ACC flows around the southern edge of 144 the Campbell Plateau and to the Bounty Trough, reinforcing the Deep Western Boundary 145 Current (DWBC). South of the Bounty Trough (46°S) the ACC turns to the east and continues its journey across the South Pacific, while the DWBC continues north towards the equator in 146 147 the Pacific Ocean basin (Carter and McCave 1997). The ACC carries CDW eastwards from the Southeast Indian Ocean into the Southwest Pacific Ocean. In the modern southern Emerald
Basin, CDW extends to the seafloor, bathing Site 278 (Figs. 1B; S1-S2) (Rintoul et al., 2014;
Lambelet et al., 2018).

Site 274 (68°59.81'S, 173°25.64'E, 3305 m water depth) was drilled on the lower 151 152 continental rise, 250 km north-northeast of Cape Adare (Hayes et al., 1975) (Fig. 1B). It is 153 currently situated south of the Southern ACC Front (SACCF) (Figs. 1B; S1) (Sokolov and 154 Rintoul, 2007) and within the main outflow path of bottom water formed in the western Ross 155 Sea (Gordon et al., 2009). At a depth of 3305 m, the main modern water mass encountered is 156 Ross Sea Bottom Water (RSBW) (Fig. 1B; S1). Today, most of the dense shelf water on the 157 Ross Sea shelf resides in the Joides and Drygalski troughs, from where it is exported as RSBW 158 to the abyssal plains (Fig. 1B) (Gordon et al., 2009). In contrast to CDW, which can be found 159 all around Antarctica, bottom topography restricts the pathway of the Antarctic Bottom Water 160 (AABW), including RSBW, in the Southern Ocean (Orsi et al., 1995). Only a small portion of 161 RSBW formed in the western Ross Sea flows northward from its outlets, with most of it being 162 diverted westward against the continental slope into the deep Australian-Antarctic Basin (Gordon et al., 2009). Through diapycnal mixing with overlying CDW, the hydrographic 163 164 properties of RSBW quickly erode along its westward pathway and are only detectable to ~140°E along the Adélie-Wilkes Coast (Rintoul, 1998). In contrast, there is a larger fraction of 165 166 RSBW detected north of the Ross Sea, but this variety of RSBW forms predominantly in the 167 central Ross Sea and is channelled to the continental slope by the Glomar Challenger trough 168 and into the Hillary Canyon. Much of this bottom water enters the southeast Pacific Basin (Fig. 169 **1B**) (Gordon et al., 2009).

170

FIGURE 1

171 **2.1** Modern Nd isotope signatures of deep and bottom waters around the Tasmanian

172 Gateway

173 Globally, North Atlantic Deep Water (NADW) and Pacific Deep Water (PDW) 174 constitute water mass endmembers in terms of their Nd isotope composition. Modern NADW 175 is characterised by relatively negative ε_{Nd} values ($\varepsilon_{Nd} \sim -12$ to 13) (e.g., Lambelet et al., 2016), 176 resulting from the weathering of old cratonic rocks surrounding the North Atlantic. Pacific 177 Deep Water, in contrast, carries less negative (i.e. more radiogenic) ε_{Nd} values of ~ -4 (e.g., 178 Amakawa et al., 2009), due to the influence of weathering of younger volcanogenic material 179 around the North Pacific. Modern CDW in the Southern Ocean is characterised by a relatively 180 homogenous Nd isotope composition along the flow path of the ACC across all ocean basins 181 and with depth ($\varepsilon_{Nd} = -8.3 \pm 1.5$; n = 158) (Figs. 2A) (Lambelet et al., 2018 and references 182 therein). In the western part of the Tasmanian Gateway along the ~140°E meridian, CDW 183 exhibits ε_{Nd} values of -8.6 \pm 0.4 (n=4) around 2500-4300 m (Lambelet et al., 2018). In the 184 eastern part of the gateway, in the Macquarie Ridge region, close to Site 278, CDW has ENd 185 values of -9.0 ± 0.2 and -9.2 ± 0.2 between 3201 and 4183 m, respectively (Figs. 2A; S2) (Lambelet et al., 2018). Ross Sea Bottom Water exhibits a more radiogenic Nd isotope 186 187 signature than CDW, probably acquired from Ross Sea shelf sediments and mixing with shelf waters ($\epsilon_{Nd} = -6.5$ to -7.5; Rickli et al., 2014; Basak et al., 2015) (Fig. 2B). 188

189

FIGURE 2

190 3. MATERIALS AND METHODS

191 **3.1 Study sites, age models, and paleodepths**

192 Our study focuses on the lower part of Hole 278 at DSDP Hole 278 (cores 34-26, 429-193 329 meters below sea floor (mbsf)). Sediments from this interval consist of (i) siliceous 194 nannofossil chalk (429-386 mbsf), and (ii) detrital and nannofossil-bearing radiolarian-diatom 195 ooze, alternating with siliceous oozes (386-329 mbsf) (Kennett et al., 1975). A new age model 196 for the studied sections of Site 278 has been established based on the integration of new 197 magnetostratigraphic data with revised diatom, radiolarian and calcareous nannofossil 198 biostratigraphy, calibrated to the Geological Time Scale (GPTS) 2012 (Gradstein et al., 2012) 199 (Fig. 3; Supplementary Text; Tables S1-S4).

200 For DSDP Site 274, we focus on two distinct intervals (408.5-313.5 mbsf and 199.5-201 180.5 mbsf). Shipboard core descriptions report that sediments between 408.5 and 328 mbsf 202 are composed of silty claystones with local presence of chert layers. Sediments between 323 203 and 180.5 mbsf consist of diatom-detrital silty clay and minor silty clay diatom ooze (Hayes et 204 al., 1975). Based on the initial age model, sediments between 408.5 and 180.5 mbsf are early 205 to late Oligocene in age (Hayes et al., 1975). New dinocyst constraints combined with new 206 magnetostratigraphic data and revised diatom biostratigraphy (updated from Hayes et al. 207 (1975)) calibrated to GPTS 2012 (Gradstein et al., 2012) provide an updated age model for the 208 studied sediments at Site 274 from ~33.7 to 23.3 Ma (Fig. S4) (Hoem et al., 2021).

209 We reconstructed the paleoposition of Sites 278 and 274 using the G-plates geodynamic 210 modeling (http://www.gplates.org; Müller et al., 2018), utilizing the plate circuit of Müller et 211 al. (2016). Paleodepths for the sites were obtained from the paleo-bathymetry grids generated 212 by the Earthbyte group (Scotese and Wright, 2018), incorporating the reconstructed 213 paleolocation of the two sites (Fig. S5). Our reconstructions show that Site 278 migrated 214 northwards from ~62.84°S (~30 Ma) to 61.4°S (~23 Ma); its paleodepth varied between 3500 215 and4000 m throughout the Oligocene and early Miocene. Site 274 has migrated north from a 216 position of ~70.22°S in the early Oligocene (~33 Ma) to ~69.57°S at the end of the Oligocene217 Miocene transition (~23 Ma) and remained at ~2500-3000 m water depth throughout the218 Oligocene.

219 **3.2 Neodymium isotope analyses**

220 Fish teeth and bones (hereafter termed fish debris) were handpicked from the $> 63 \ \mu m$ 221 sediment fractions isolated by wet sieving. A total number of 32 samples were selected for fish 222 debris Nd isotope analyses (20 samples from Site 278 and 12 samples from Site 274) (Table 223 S5). All samples were treated with ultraclean 18 M Ω water (Milli-Q water) and methanol to 224 remove debris from surfaces and cavities following Martin and Haley (2000) in the MAGIC 225 laboratories at Imperial College London (see also Huck et al., 2017). Cleaned fish debris 226 samples were subsequently transferred into cleaned microcentrifuge tubes and dissolved 227 overnight in 50 µL of 2M HCl. Dissolved fish debris were loaded on Biorad cation exchange 228 resin (200-400 µm mesh) to separate the REEs from the sample matrix and Eichrom Ln-Spec 229 resin (50-100 µm bead size) to separate Nd from the other REEs.

230 Furthermore, seven samples were selected (two from Site 278 and five from Site 274; 231 Table S5) to determine the detrital Nd isotope composition in order to evaluate potential 232 contribution of the detrital sediments to the porewaters or overlying bottom water signature. Samples were dried and gently homogenised using mortar and pestle. Approximately 500 mg 233 234 of homogenised material was subjected to a carbonate leaching procedure to remove biogenic 235 carbonate using 30 ml of 1.5% buffered acetic acid (modified from Biscaye (1965)). Due to 236 the significant carbonate content of the samples from Site 278 (38 to 43 %), approximately 1.5 237 g of dried sample and 70-75 ml of 1.5% buffered acetic acid were used. Exchangeable ions 238 were subsequently removed using 10 ml 1 M MgCl₂ solution. In a third step, ferromanganese oxides and oxyhydroxides were removed using a weak reductive solution of 0.005 M 239 Hydroxylamine Hydrochloride (NH₂OH) 1.5% acetic acid and 0.03 M Na₂-EDTA for one 240

241 hour, followed by a stronger leaching step utilising 0.05 M NH₂OH for 17 hours. 50 mg of 242 leached and water washed detrital sediment was subsequently dried, weighted and digested on a hotplate using a mixture of 1 ml of concentrated HNO₃, 0.8 ml HClO₄ and 2 ml HF. The 243 244 detrital samples were processed using the same ion chromatography as the fish debris samples. 245 Neodymium isotope ratios of fish debris and detrital sediment samples were determined 246 on a high-resolution Nu Plasma multiple collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at Imperial College London, operated in static mode. Instrumental 247 mass bias was corrected for using a ¹⁴⁶Nd/¹⁴⁴Nd ratio of 0.7219. All reported ¹⁴³Nd/¹⁴⁴Nd ratios 248 249 are corrected to a nominal JNdi value of 0.512115 (Tanaka et al., 2000) using bracketing 250 standards. JNd_i standards were also used to monitor external reproducibility, and accuracy was 251 evaluated by processing USGS BCR-2 rock standards alongside samples, which yielded average 143 Nd/ 144 Nd ratios of 0.512636 ± 0.000008 (n=23; 2s.d.), in agreement with the 252 253 published BCR-2 143 Nd/ 144 Nd ratio of 0.512638 ± 0.000015 (Weis et al., 2006) (Table S5).

3.3 Rare earth element analyses

To correct for the decay of ¹⁴⁷Sm to ¹⁴⁴Nd within the fish debris over time, we used the 255 256 average Sm and Nd concentrations measured for two fish debris samples from Site 278 $(^{147}\text{Sm}/^{144}\text{Nd ratios of } 0.1275 \text{ and } 0.1256; ^{147}\text{Sm}/^{144}\text{Nd}_{average} = 0.1266)$ and three fish debris 257 samples from Site 274 (147 Sm/ 144 Nd ratios of 0.1532, 0.1921, and 0.2662; 147 Sm/ 144 Nd_{average} = 258 259 0.2038). The range of Sm/Nd ratios reported here are consistent with values from Oligocene to 260 Miocene fish debris material elsewhere (Huck et al., 2017; Wright et al., 2018; Evangelinos et al., 2020). Corrections for in situ decay of ¹⁴⁷Sm amounted to 0.19 to 0.28 ENd units for Site 278 261 262 and 0.02 to 0.14 ENd units for Site 274; (t) denotes age-corrected samples (Table S5).

263 To investigate whether the Nd in the fish debris is of authigenic nature (i.e. seawater-264 derived), the full suite of REE concentrations were determined on one fish debris sample from 265 Site 278 and three fish debris samples from Site 274 (Table S6). Rare earth element analysis 266 was performed at the Open University using an Agilent Technologies 8800 Triple-Quad 267 Inductively Coupled Plasma-Mass Spectrometer (ICP-MS). Analyses were standardized via a 268 suite of seven synthetic multi-element standards made up with certified plasma standard 269 solutions. An intermediate synthetic multi-element monitor standard, a fossil bone standard (Chavagnac et al. 2007) and a 2% HNO₃ blank were run every 5th sample to monitor instrument 270 271 drift and precision. Precision was generally better than $\pm 2\%$ (1 s.d.). Both oxide interferences 272 (CeO^+/CeO^-) (<0.5%) and doubly charged species (Ce⁺⁺/Ce⁺) (<1.2%) were kept low. All REE 273 data were normalised to Post Archean Shale (PAAS) concentrations (Taylor and McLennan, 274 1985).

275 **4. RESULTS**

276 4.1 Age model DSDP Site 278

277 An age-depth model for the Oligocene and lower Miocene intervals of DSDP Site 278 278 is developed from the integration of new magnetostratigraphic data, calcareous nannofossil, 279 marine diatom, and radiolarian biostratigraphy, calibrated using the Geological Time Scale (GPTS) 2012 (Gradstein et al., 2012) (Figs. 3; Supplementary Text; Tables S1-S4). We also 280 281 present an alternate age model that fit with the constraints of the data (for details see 282 supplementary material). The stratigraphic section cored at Site 278 from the bottom of the 283 sedimentary section in core 34 at ~429 mbsf up to sample 278-21R-1, 50 cm at ~282 mbsf 284 spans the time interval from ~ 31 to 17.7 Ma, with one unconformity noted between samples 285 278-31R-3, 35 cm and 278-31R-2, 139 cm (398.85 – 399.89 mbf), which removed a ~2.4 m.y. time interval from 28.2 to 25.8 Ma. The top of this interval is bounded by an unconformity 286 287 between samples 278-21R-1, 50 cm and 278-20R-6, 50 cm (281.5 mbsf), which removed the 288 time interval 17.6 to 15.5 Ma (Fig. 3). Studied sediments (cores 34R-26R) are dated between ~31 and 21.7 Ma, similar to the initial shipboard ages (Fig. 3), suggesting upper Oligocene to
lower Miocene sediments (Kennett et al., 1975).

291

FIGURE 3

292 **4.2 Fish debris Nd isotope compositions**

293	Fish debris Nd isotope data from Site 278 show little variability, ranging from $\varepsilon_{Nd(t)} = -$
294	6.0 ± 0.2 to 6.7 ± 0.2 from the early Oligocene to the early Miocene (~ 30.9-21.8 Ma) (Fig. 4;
295	Fig. S6A; Table S5). The average $\varepsilon_{Nd(t)}$ throughout the record is -6.4 ± 0.2 (n=20). In contrast,
296	fish debris Nd isotope data from Site 274 show more variability, and reach from maximum
297	values of -3.1 \pm 0.3 at 33.5 Ma and -3.7 \pm 0.3 at 23.8 Ma to relative minima of -5.0 \pm 0.1, at
298	~33.2-29.7 Ma, -6.2 \pm 0 at ~29 Ma, -7.3 \pm 0.3 and -6.8 \pm 0.2 at ~24.3 and 24.0 Ma, respectively
299	(Fig. 4; Fig. S6B; Table S5). The late Oligocene time period features a pronounced excursion
300	to more radiogenic $\varepsilon_{Nd(t)}$ values by ~3 epsilon units between 24.0 and 23.8 Ma.

301 4.3 Bulk sediment Nd isotope compositions

The detrital samples at Site 278 exhibit $\varepsilon_{Nd(t)}$ values of -10.4 ± 0.3 (~ 30.5 Ma) and -7.9 ± 0.3 (~ 21.8 Ma), respectively (Fig. 4; Fig. S6A; Table S5). Lower Oligocene detrital sediment Nd isotope compositions from Site 274 exhibit $\varepsilon_{Nd(t)}$ values of -4.2 ± 0.3 (~33.5 Ma), -7.6 ± 0.3 (~33.2 Ma) and -8.5 ± 0.3 (~29 Ma). Two additional late Oligocene detrital sediment samples yielded $\varepsilon_{Nd(t)}$ values of -10.3 ± 0.3 and -8.5 ± 0.3 at ~24.0 Ma and 23.8 Ma, respectively (Fig. 4; Fig. S6B; Table S5).

308 **4.4 Fish debris REE patterns**

Five fish debris samples from Site 278 and Site 274 yielded middle-REE enriched
patterns (Fig. 5; Table S6). Only one sample from section 278-33R-6 showed a negative Ce

- anomaly, which together with the middle-REE enriched patterns represent a diagnostic feature
- 312 of seawater-derived Nd in fish teeth samples (Scher et al., 2011).
- 313
 FIGURE 4

 314
 FIGURE 5

315 **5. DISCUSSION**

316 5.1 Reliability of Nd isotopes from Sites 278 and 274

Results of Oligocene to Miocene-aged fish debris samples at Site 278 (paleodepth: 318 3500-4000 m) are more radiogenic (less negative) in their Nd isotope composition ($\epsilon_{Nd(t)} = -6.0$ to -6.7) than modern CDW in the south Emerald Basin ($\epsilon_{Nd} = \sim -9$) at ~3200-4200 m water depth (Lambelet et al., 2018). Results for Site 274 ($\epsilon_{Nd(t)} = -3.1$ to -7.3) show generally more radiogenic values compared to ambient RSBW ($\epsilon_{Nd} = -6.5$ to -7.5; Rickli et al., 2014; Basak et al., 2015) (Fig. 4). The first question to address is if the fish debris record a purely authigenic signal.

324 Site 278 is located near the Macquarie Ridge, a relict volcanic mid-ocean ridge, 325 characterised by basalts and peridotites with an ε_{Nd} value of +7 to +11 (Dijkstra et al., 2009; 326 Conway et al., 2012). Interaction of volcanic sediments and/or hydrothermal products with 327 ambient seawater could have altered the ε_{Nd} values at Site 278 towards more radiogenic values. 328 However, hydrothermal activity has been shown to be a sink rather than a source of Nd near 329 ridges (Stichel et al., 2018). Moreover, fish teeth records from the Kerguelen Plateau show no 330 changes in their Nd isotopic composition associated with the local volcanic activity during the 331 timing of the northern Kerguelen Plateau emplacement (Wright et al., 2018). Volcanic glass 332 and rock fragments have been reported from close to the base of siliceous nannofossil chalk at Site 278 (Kennett et al., 1975) (~31-28.2 Ma, based on our new age model). However, the lack
of any significant variability in our fish debris Nd record makes it difficult to reconcile any
major volcanic contribution to the porewater and hence to the fish debris Nd isotope signature
(Figs. 4; S7A).

337 To further evaluate the potential interaction between detrital sediments and pore 338 water/seawater Nd isotope composition, we can use the results from two detrital sediment 339 samples from the siliceous chalk unit (section 33R-6) and from the nannofossil-bearing diatom 340 ooze unit (section 26R-2) at Site 278. The detrital samples exhibit $\varepsilon_{Nd(t)}$ values of -10.4 \pm 0.3 341 and -7.9 ± 0.3 , respectively (Fig. 4; Table S5), within the range of terrigenous inputs from 342 Paleozoic granites and metasedimentary rocks ($\varepsilon_{Nd(t)}$ -4.9 to -13.9) of the South Island, New 343 Zealand (Pickett and Wasserburg 1989). This observation is in line with the general deep water 344 flow in the area, suggesting that terrigenous sediments in the Southern Emerald Basin are 345 predominantly derived from the erosion of the New Zealand's Southern Alps via the Solander 346 Channel (Carter and McCave, 1997). Importantly, the measured detrital sediment composition 347 is more unradiogenic (i.e. more negative ε_{Nd} values) than the almost contemporaneous fish 348 debris signatures and hence cannot be responsible for the observed more radiogenic values. In 349 addition, REE patterns from a fish debris sample from section 33R-6 (~30.5 Ma) exhibit a 350 pattern enriched in middle-REEs and a prominent negative cerium (Ce) anomaly, which 351 represent diagnostic features of uncontaminated seawater origin in the fish debris (Fig. 5; Table 352 S6) (Scher et al., 2011; Huck et al., 2016, 2017). The fish debris sample from section 26R-2 353 (21.8 Ma) exhibit also a typical middle-REEs enriched pattern but records a slightly positive 354 Ce anomaly (Fig. 5). Positive Ce anomalies in fish debris have been linked to anoxic seawater 355 and/or pore water conditions, resulting in remobilization of REEs from authigenic or organic 356 matter coatings (Elderfield and Pagett, 1986; Freslon et al., 2014; Huck et al., 2016). High biogenic productivity at Site 278 associated with proto-polar front upwelling processes
(Kennett et al., 1975) may have led to anoxic conditions at the seafloor.

359 Similar to Site 278, bulk sediment results from Site 274 exhibit a less radiogenic (i.e. 360 lower) Nd isotope fingerprint than fish debris when comparing results for the same samples 361 and samples taken very close to each other. The difference in $\varepsilon_{Nd(t)}$ for three pairs of fish debris-362 detrital sediment samples analyzed is -4.9, -3.5 and -2.4 ENd units, while adjacent samples show 363 differences in $\varepsilon_{Nd(t)}$ of -2.5 and -1.1 (Figs. 4; S7B; Table S5). This pronounced difference 364 between the $\varepsilon_{Nd(t)}$ values of the fish debris and the detrital sediment samples at Site 274 strongly 365 supports the absence of significant exchange between the detrital sediments and the fish debris. 366 Overall, the more radiogenic $\varepsilon_{Nd(t)}$ values of the fish debris, compared to the more unradiogenic 367 $\varepsilon_{Nd(t)}$ values of the detrital samples, argues against a contribution of Nd from the detrital 368 sediments.

369 Furthermore, REE patterns of three fish debris samples at Site 274 yielded middle-REE 370 bulge patterns, including a slightly positive Ce anomaly (Figs. 5; Table S6). The close 371 proximity of Site 274 to the Antarctic continental margin and the high biogenic fraction in the 372 sediment (Hayes et al., 1975) may have caused the observed positive Ce anomaly resulting 373 from increased weathering inputs and/or remobilized REEs from authigenic or organic matter 374 coatings due to anoxic conditions in the pore waters (Elderfield and Pagett, 1986; Freslon et 375 al., 2014; Huck et al., 2016). Further work is required to fully understand the positive Ce 376 anomaly in our fish debris samples. Nevertheless, REE patterns from Site 278 and 274 are 377 similar to REE patterns from Eocene-Oligocene fish debris records from around the Tasmanian 378 Gateway, which have been interpreted to preserve a predominantly seawater signature, despite 379 positive Ce anomalies (Huck et al., 2016, 2017). We therefore suggest an authigenic nature of 380 the fish debris record from Sites 278 and 274 and use our new data to explore the chemical fingerprint of proto-CDW and RSBW across the Tasmanian Gateway, and its implications forthe ACC evolution.

5.2 Early Oligocene epoch (33 -30 Ma): Prior to the inferred onset of the ACC throughflow via the Tasmanian Gateway

Neodymium isotope values from our deeper Site 278 (paleodepth: ~4000 m) between 385 386 31 and 30 Ma, indicate that the southern Emerald Basin was bathed by an unradiogenic deep water mass ($\varepsilon_{Nd(t)} = \sim -6.4$) (Figs. 4; 6). These values converge with published Nd isotope 387 388 records from the deep South Pacific (Site U1370, paleodepth ~5000 m, Site 596 paleodepth ~5000 m and Site 323, paleodepth ~4000 m; $\varepsilon_{Nd(t)} = \sim -6.3$) (Thomas et al. 2014; McKinley et 389 390 al., 2019), suggesting that abyssal sites in the South Pacific were likely influenced by a 391 common deep/bottom water mass, such as South Pacific Deep Water (SPDW) (Fig. 6). 392 Temporal and spatial Nd isotope patterns and modelling results show that SPDW was a 393 prominent and persistent water mass of the deep (≥ 3500 m) South Pacific (between at least 394 ~30°S-63°S) from 70 to 20 Ma, characterised by $\varepsilon_{Nd(t)}$ of ~ -6 (Thomas et al. 2014; McKinley 395 et al., 2019; Sarkar et al., 2019). We therefore suggest that Site 278 (paleolocation: ~63°S) was 396 bathed by SPDW between 33 and 30 Ma.

397 Neodymium isotope records consistent with model results indicate bimodal mode of deep ocean circulation in the Pacific Ocean from ~70 to 25 Ma (Scher, 2014; Thomas et al., 398 2014; Scher et al., 2015; Huck et al., 2017; McKinley et al., 2019; Sarkar et al., 2019). 399 400 Convection of SPDW occurred in the South Pacific (likely in the Ross Sea region), while North Pacific Deep Water (NPDW) formation occurred in the North Pacific. Our ENd(t) values from 401 402 Site 274, located proximal to the Ross Sea, however, range from -3.1 to -5 between 33.5 and 403 29.7 Ma and albeit of low resolution are more radiogenic (less negative), compared to both the 404 present-day RSBW Nd isotopic composition (ε_{Nd} = -6.4 to -7.5; Rickli et al., 2014; Basak et al.,

405 2015) and the deep record from Site 278 (Figs. 4; 6). The average $\varepsilon_{Nd(t)}$ value of ~ -4.6 for the 406 early Oligocene (~ 33.5-29.7 Ma) at Site 274 (paleodepth: ~ 2500-3000 m) overlaps with 407 published data from the East Tasman Plateau (Site 1172; average $\varepsilon_{Nd(t)} = -4.2$ paleodepth: ~ 408 2400 m) and Hikurangi Plateau (Site 1124; average $\varepsilon_{Nd(t)} = \sim -4.6$; paleodepth: ~ 3000 m) (Scher 409 et al., 2015; Sarkar et al., 2019) (Fig. 6). Similarities in Nd isotope values and paleodepths 410 between the Antarctic margin record from Site 274 and the published records for the northern 411 parts of the Tasmanian Gateway (Sites 1172 and 1124; Scher et al. 2015), suggest that these 412 sites may have been influenced by a common water mass, occupying water depths between ~ 413 2500and 3000 m. This more radiogenic signature is akin to northern sources Equatorial Pacific 414 Deep Water (Fig. 6) (Scher, 2014), which originated in the deep Equatorial Pacific, where 415 NPDW and SPDW mixed (Thomas et al., 2014). Neodymium isotope data from Sites 1172 and 416 1124 suggest that Equatorial Pacific water was present at 2500-3000 m, reaching at least to 417 southern latitudes of 60°S-65°S between 36 and 30 Ma (Scher et al., 2015; Sarkar et al., 2019). 418 We here suggest that water from a northern source with a more Equatorial Pacific-like Nd 419 isotope signature reached to the Adare Basin (Site 274, ~70°S) between 33 and 30 Ma. 420

420 Our results point to the presence of two distinct deep water masses in the late 421 Eocene/early Oligocene South Pacific: one occupying depths between ~2500 and 3000 m with 422 Nd composition ($\varepsilon_{Nd(t)} = \sim -3$ to -5) and a second deep/bottom water mass (> 3000 m) with more 423 unradiogenic Nd composition ($\varepsilon_{Nd(t)} = \sim -6$) (Fig. 6).

424

FIGURE 6

425 5.3 Early Oligocene to early Miocene epoch (30-22 Ma): After the inferred onset of the
426 ACC throughflow via the Tasmanian Gateway

427 Neodymium isotope values at Site 274 show a shift towards less radiogenic values 428 $(\varepsilon_{Nd(t)} = -6.2 \pm 0.3)$ between 29.7 and 29.1 Ma. Slightly less radiogenic $\varepsilon_{Nd(t)}$ values are observed 429 around 24.3 Ma and 24.0 Ma (-7.3 \pm 0.25 and -6.8 \pm 0.15, respectively) (Figs. 4; 6), resembling 430 modern RSBW values (ε_{Nd} = -6.4 to -7.5; Rickli et al., 2014; Basak et al., 2015). The shift 431 between 29.7 and 29.1 Ma at Site 274 coincides with the shift observed in the two Nd isotope 432 records from the East Tasman Plateau (Site 1172 $\varepsilon_{Nd(t)}$ = -4.8 to -6.8) and the Hikurangi Plateau 433 (Site 1124 $\varepsilon_{Nd(t)}$ = -5.6 to -6.2) between 29.6 and 29 Ma, respectively (Scher et al., 2015) (Fig. 434 6). This latter shift has been linked to the northward migration and alignment of the Tasmanian Gateway with the westerly winds (Scher et al., 2015). The change in the $\varepsilon_{Nd(t)}$ values at Site 435 436 274 between 29.7 and 29.1 Ma does not coincide with any significant known change in the 437 Antarctic ice sheet that would have influenced the sediment input around the Ross Sea. Instead, 438 given that Site 274 was located south of the proto-PF during this period (Nelson and Cooke, 439 2001; Scher et al., 2015), we posit that the shift to more unradiogenic Nd contribution is 440 probably due to the onset of proto-CDW upwelling reaching the Adare Basin and Site 274.

441 Site 278, on the other hand, was located close to the proto-PF and proto-SAF between 442 30 Ma and 22 Ma and thus in the main pathway of the proto-ACC and proto-CDW (Nelson 443 and Cooke, 2001), explaining its invariant Nd isotope fingerprint (average $\varepsilon_{Nd(t)} = -6.4$) 444 throughout the early Oligocene to early Miocene (29.7-22 Ma) (Figs. 4; 6). These values are 445 matched by the two published Nd isotope records (Sites 1172 and 1124; $\varepsilon_{Nd(t)} \sim -6.5$) from the 446 eastern side of Tasmanian Gateway after the onset of ACC influence at this northern location 447 (Scher et al., 2015) (Fig. 5). All three records show more radiogenic Nd isotope compositions 448 than modern CDW in the Emerald Basin ($\varepsilon_{Nd} = -9$; Lambelet et al., 2018) (Figs. 2A; S2). They 449 furthermore differ from records to the west of the Tasmanian Gateway. Throughout the 450 Oligocene (30-23 Ma) Nd isotope records along the proto-PF in the South Atlantic and the 451 South Indian Ocean are characterised by proto-CDW with a Nd isotope composition between 452 ~ -7 and -9 (Site 689: Scher and Martin, 2004; Site 748 and 744: Wright et al., 2018; Site 269: 453 Evangelinos et al., 2020) (Figs. 6; 7). Such values are similar to modern CDW (ε_{Nd} = -8.3 ± 1.5, 454 n=158) (Lambelet et al., 2018) (Fig. 6).

455 Comparison between our new and previously published data along the proto-ACC 456 pathway around the Tasmanian Gateway therefore reveals a provinciality in seawater Nd 457 isotopes with more radiogenic proto-CDW ($\varepsilon_{Nd(t)} = -6$ to -7.5) on the Pacific side of the Tasmanian Gateway, and less radiogenic proto-CDW ($\varepsilon_{Nd(t)}$ = -7 to -9) in the Indian and Atlantic 458 459 sectors of the Southern Ocean (Figs. 6; 7). This observation implies an absence of a 460 homogenous deep water mass across the Tasmanian Gateway during the Oligocene and early 461 Miocene, and suggests a more complex evolution of the deep ocean circulation across the 462 gateway than previously assumed.

463 We propose that the provinciality in water mass signatures across the gateway was due 464 to a less deep reaching and probably also weaker proto-ACC flow during the Oligocene to early 465 Miocene (Fig. 7). A shallower and weaker ACC flow entering the South Pacific would have 466 increased the amount of time that Pacific deep water could have mixed with the proto-CDW, 467 allowing for acquisition of a more radiogenic isotopic composition of proto-CDW. These 468 inferences are consistent with numerical simulations showing limited throughflow of the proto-469 ACC due to Australasian paleogeography during the Oligocene (Hill et al., 2013) and weaker 470 global overturning circulation due to weaker westerly winds from the early to mid-Miocene 471 (Herold et al, 2012).

472 Recently collected sedimentological data, microfossil assemblages and past 473 reconstructions of sea surface temperatures from the Antarctic-Australian Basin and around 474 the Tasmanian Gateway, also infer a weaker proto-ACC frontal system, characterised by 475 intrusions of warm waters from northern latitudes from the Oligocene to the middle Miocene, 476 weak latitudinal paleotemperature gradients and absence of strong sea ice seasonality (34-11) Ma) (Bijl et al., 2018; Hartman et al., 2018; Salabarnada et al., 2018; Sangiorgi et al., 2018;
Evangelinos et al., 2020; Hoem et al., 2021). The question remains when the modern-like ACC
was established. Our data suggest that this occurred during the Neogene, but future work is
necessary to address the timing of its establishment and the wider implications of a weaker
Oligocene to Miocene ACC.

482

FIGURE 7

483 **5.4 Evidence of a glacial weathering event during the latest Oligocene (~25 Ma)**

484 The most pronounced ε_{Nd} excursion observed at Site 274 (from -6.8 ± 0.3 to -3.7 ± 0.3) 485 between 24.0 and 23.8 Ma is a peculiar feature of our new record and may reflect a local change 486 in seawater composition (Figs. 4; 6). Although only represented by a single data point, another 487 radiogenic isotope excursion is hinted at ~ 33.5 Ma ($\varepsilon_{Nd(t)} = -3.1 \pm 0.3$; Figs. 4; 6). Considering 488 the relatively invariant Nd isotope compositions of Oligocene Pacific deep water (Scher, 2014; 489 McKinley et al., 2019) and proto-CDW (this study; Wright et al., 2018; Evangelinos et al., 490 2020) during the Oligocene, we consider two potential causes for the radiogenic excursion at 491 Site 274: (i) increased boundary exchange/volcanic activity, and (ii) pulses in Antarctic ice 492 advance leading to increased weathering inputs to local seawater.

Volcanic activity and deposition of material from the McMurdo volcanic group (ε_{Nd} > 0; see references in Cook et al., 2013) have been reported to occur predominantly after 24 Ma (Roberts et al., 2013). In addition, there is little physical evidence of volcanic contributions to the sediments at Site 274 (Hayes et al., 1975), an observation that is confirmed by our measured detrital sediment $\varepsilon_{Nd(t)}$ values of -8.5 ± 0.3 (Fig. 4). Furthermore, REE patterns indicate that fish debris predominantly preserve a seawater signature at 23.8 Ma (Fig. 5). We therefore 499 exclude the possibility of boundary exchange/volcanic activity as a cause for the positive Nd500 isotope excursion.

501 A more promising connection can be made with the timing of a major expansion of ice 502 from Antarctica. The Eocene-Oligocene transition is well documented as a major ice advance 503 on Antarctica (e.g., Zachos et al., 1992; Galeotti et al., 2016). A seawater expression of 504 weathering inputs from this ice expansion has been documented in the Prydz Bay area, where 505 late Eocene and Eocene Oligocene Transition (EOT) fish teeth show a shift in seawater 506 composition consistent with increased Antarctic weathering inputs (Scher et al., 2011; Bohaty 507 et al., 2012; Wright et al., 2018). Even though only documented by a single data point, the very 508 radiogenic $\varepsilon_{Nd(t)}$ value of -3.1 \pm 0.3, our Site 274 exhibits the most extreme Nd isotope signature 509 ~ 33.5 Ma (Figs. 4; 6). A shift towards more radiogenic values may indicate erosion and 510 delivery of more radiogenic material exposed upstream in the Transantarctic Mountains, such 511 as basalts and sills of the Ferrar Large Igneous Province (ENd: -3.5 to -6.9; see Cook et al., 2013 512 for a summary) or rocks like the Granite Harbour Intrusives in the Gabbro Hills ($\epsilon_{Nd} = -2.3 \pm$ 513 1.6, n=4; Borg et al., 1990).

514 Interestingly, our data clearly define a second major seawater Nd isotope excursion 515 between 24.0 and 23.8 Ma, which appears to be closely associated with one of the most 516 significant climate transitions since the EOT resulting in a major Antarctic ice sheet expansion (~24.5-24 Ma; Levy et al., 2019). Kulhanek et al. (2019) described the first occurrence of ice 517 518 proximal glaciomarine sediments from DSDP Site 270 on the shelf of the Ross Sea at this time. 519 In addition disconformities are observed at another Ross Sea drill site proximal to the 520 Transantarctic Mountains (CRP-2/2A; Naish et al., 2008), and in seismic data (Sorlien et al., 521 2007), which are consistent with the first major expansion of marine ice sheets across the Ross 522 Sea continental shelf (see also Levy et al., 2019). We consider it very likely that large changes 523 in the Antarctic cryosphere, i.e. large ice advance in this case, would have changed significantly the erosional input from Antarctica to the Ross Sea margin, and hence would have imprinted aradiogenic Nd isotope signal in local seawater.

526

527 6. CONCLUSIONS

528 New Oligocene to Miocene seawater Nd isotope data from Site 274 in the Adare Basin, Ross 529 Sea, reveal two major shifts in seawater chemistry in response to major steps in Antarctic ice 530 sheet expansion at the Eocene/Oligocene boundary (33.7 Ma) and between ~24.5 and 24.0 Ma. 531 In contrast, Site 278 in the southern Emerald Basin shows an invariant Nd isotopic composition 532 $(\varepsilon_{Nd(t)} = -6.0 \text{ to } -6.7)$ throughout the Oligocene and early Miocene (31-22 Ma). Comparison 533 with previously published data suggests the presence of two deep water masses in the South 534 Pacific prior to the inferred onset of the ACC (34-30 Ma), one occupying depths between 535 ~2500 and 3000 m with Nd composition $\epsilon_{Nd(t)=}$ -3 to -5 and a deep/bottom water mass (>3000 536 m) with more unradiogenic Nd composition ($\varepsilon_{Nd(t)} \sim -6$). Following the onset of the ACC and 537 the opening of the Tasman Gateway (30-22 Ma), the seawater Nd isotope record at Site 278, 538 remained more radiogenic than the proto-CDW in the South Atlantic and Indian Ocean. We 539 attribute the provinciality in the Nd isotope composition of deep waters across the Tasmanian 540 Gateway to a weaker, less deep reaching ACC (proto-ACC) than today. Our finding implies 541 that the modern strong and deep-reaching ACC, which creates a near homogenous Southern 542 Ocean Nd isotope fingerprint must developed at a later point in the Neogene.

543

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545

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558	
559	Data availability
560	The datasets to this article are available in the Supplement.
561	
562	Appendix A. Supplementary data: Supplementary figures and tables
563	
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- 791
- 792 List of Figures



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795 Figure 1. Overview map of modern Southern Ocean configuration (Ocean Data View, 796 (Schlitzer, 2016; version 4.79; http://odv.awi.de). A: Antarctic Circumpolar Current (ACC) 797 frontal system, study sites (DSDP Sites 274 and 278) (yellow stars) and sites referred to in this study (black squares). SAF: Subantarctic front (black dashed line), PF: Polar front (black line), 798 799 SACCF: Southern Antarctic Circumpolar Current front (black dotted line) adapted from Orsi 800 et al. (1995). B: Major modern deep ocean water circulation paths across the Tasmanian 801 Gateway adapted from Sokolov and Rintoul. (2007) and Gordon et al. (2009). The main 802 transport of Circumpolar Deep Water along the ACC takes place around the PF and SAF, Ross 803 Sea bottom water (RSBW) (purple) feeds Antarctic Bottom Water. Deep Western Boundary Current (DWBC) (black arrows), MR: Macquarie Ridge, EB: Emerald Basin, ETP: East 804 805 Tasman Plateau, CP: Campbell Plateau, BT: Bounty Trough, CR: Chatham Rise, HP: 806 Hikurangi Plateau, CA: Cape Adare, JB: Joides Trough, DB: Drygalski Trough, GCT: Glomar 807 Challenger Trough.



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811 Figure 2. Modern neodymium (Nd) isotope composition for deep and bottom waters in the Southern Ocean (between 0°E and 110°W). Figures are made with Ocean Data View (Schlitzer, 812 813 2016; version 4.79; <u>http://odv.awi.de</u>). (A): Nd isotope composition of Circumpolar Deep 814 Water (CDW). (B): Nd isotope composition of Antarctic Bottom Water (AABW) (Stichel et al., 2012; Rickli et al., 2014; Garcia-Solsona et al., 2014; Basak et al., 2015; Lambelet et al., 815 816 2018; Amakawa et al., 2019). Antarctic Circumpolar Current (ACC) fronts (mean positions) adapted from Sokolov and Rintoul, 2007. SAF (blue), PF (red), SACCF (black), MR: 817 818 Macquarie Ridge, WL: Wilkes Land, KP: Kerguelen Plateau



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Figure 3. Revised age-depth model for Site 278. Stratigraphy of Site 278, from left to right: depth in meters below sea floor (mbsf), core recovery, lithologic log based on initial shipboard sedimentological descriptions and local polarity based on inclination values. Grey vertical stripes mark the normal polarity zones of the geomagnetic polarity timescale (normal=black, reversed=white) (Gradstein et al., 2012). Black/red crosses= magnetostratigraphic constraints,

green triangles= diatom biostratigraphic constraints, blue triangles = calcareous nannofossil biostratigraphic ranges, orange triangles= radiolarian biostratigraphic constraints. FAD=first appearance datum; LAD= last appearance datum . Horizontal error bars on biostratigraphic events indicate total age range for the given datum. Vertical error bars indicate the depth range for the given datum. Datum and biozone labels correspond to those in Tables: S1, S2, S3 and S4. The back line following the different constraints shows our preferred chronology, and in red a possible alternate.



Figure 4. Fish debris $\varepsilon_{Nd(t)}$ records generated in this study from DSDP Sites 274 (purple circles) and 278 (red diamonds). Detrital sediment $\varepsilon_{Nd(t)}$ values from DSDP Sites 274 (purple open circles) and 278 (red open diamonds). Present-day Ross Sea Bottom Water (RSBW) ε_{Nd} endmember ranges shown in magenta (Rickli et al., 2014; Basak et al., 2015). Present-day Circumpolar Deep Water (CDW) ε_{Nd} endmember ranges shown in blue (Lambelet et al., 2018 and references therein).



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Figure 5. Rare earth element (REE) patterns normalised to Post Archean Shale (PAAS)
concentrations (Taylor and McLennon, 1985) for DSDP Sites 278 and 274.



Figure 6. Comparison of fish debris ε_{Nd} records generated in this study from DSDP Sites 274
and 278 with those derived from sites along the proto-CDW path in the Atlantic, Indian and
Pacific sectors of the Southern Ocean. Atlantic/Indian Ocean locations include ODP Site 689
(Maud Rise; Scher and Martin, 2004), ODP Sites 748 and 744 (Kerguelen Plateau; Wright et
al., 2018), and DSDP Site 269 (off Adélie Coast; Evangelinos et al., 2020) (light blue shading).
ODP Site 1124 (Hikurangi Plateau) and ODP Site 1172 (East Tasmanian Plateau; Scher et al.,

857 2015) are combined in the green shading. Pacific Equatorial Age Transect (PEAT) Nd isotope 858 results from the Equatorial Pacific are based on IODP Sites U1331, U1332, U1333, U1334, 859 and U1335 (Scher, 2014) (red shading). The only data shown as individual symbols other than 860 our new results are South Pacific DSDP Site 596, IODP Site U1370 (McKinley et al., 2019) 861 and ODP Site 323 (Thomas et al., 2014). Blue shaded ranges refer to the present-day Nd 862 signature of the CDW and magenta shaded rangers refer to the present-day Nd isotopic composition of the RSBW (Rickli et al., 2014; Basak et al., 2015; Lambelet et al., 2018 and 863 864 references therein).







866 867 Figure 7. Map of neodymium (Nd) isotopic data discussed in the main text plotted against 868 paleo-latitude and longitude (upper panels) and paleo-depth (lower panels) from 33 Ma to 30 869 Ma and from 29 to 22 Ma. Plate tectonic reconstructions and paleolocation of study Sites 278 870 and 274, and other sites discussed in the main text were adapted from G-plates, based on the

global plate motion model from Muller et al. (2016). Continent with present-day coastlines (black), grey boundary around continents shows areas of non-oceanic crust (e.g. continental shelves). Circles represent site locations and are coloured based on their average $\varepsilon_{Nd(t)}$ values for each period considered. Grey band indicates the position of the Tasmanian Gateway (TG). 875

- 876 Supplementary Information
- 877 Supplementary text
- 878 Age model for Site 278
- 879

880 An age-depth model for the Oligocene and lower Miocene intervals of DSDP Site 278 881 is developed from the integration of new magnetostratigraphic data, calcareous nannofossil, 882 marine diatom, and radiolarian biostratigraphy, calibrated using the Geological Time Scale 883 (GPTS) 2012 (Gradstein et al., 2012) (Fig. 3; Tables S1-S4). The initial age model prepared by 884 the Shipboard Scientific Party on DSDP Leg 29 (Kennett et al., 1975) was of coarse resolution 885 due in part to the infancy of diatom and radiolarian biostratigraphy. A compilation of age models from Neogene intervals of DSDP drill holes presented by Lazarus et al. (1995) included 886 887 DSDP Site 278 and incorporated improved biostratigraphic age assignments as a result of 20 888 years of additional development and subsequent Ocean Drilling Program (ODP) drilling in the 889 Southern Ocean during ODP Legs 113, 114, 119, and 120. These later drilling efforts included 890 consistent integration of magnetic polarity stratigraphy in assessing the ages and correlation of 891 biostratigraphic datum levels. The compilation of Lazarus et al. (1995) provided the general 892 trend of age-depth for DSDP 278, but did not identify several short hiatuses of 2 - 4 m.y. 893 duration that are resolved in our analysis. We report here the age interpretation for the time 894 interval ~31 to ~18 Ma, and refer the reader to the report of Evangelinos et al. (in preparation) 895 for a revised age model for the time interval < 18 Ma at DSDP Site 278.

896 A total of 19 samples were examined for the identification of calcareous nannofossils. 897 Smear slides were prepared from unprocessed sediment and examined with a Leica DMRP 898 polarizing microscope at 1000 x magnification. Occasionally, x1250 and x1600 magnification was also used for the identification of small specimens. At least 3 mm² (ca. 200 visual fields at 899 900 x1000) were scanned to provide semi-quantitative values. To detect the presence of rare or very 901 rare specimens an additional two to three slide traverses were observed. The nannofossil 902 assemblages were used to define define the First and Last appearance datums (FAD and LAD) 903 Selected calcareous nannofossil bioevents were considered in moderate to well preserved 904 samples, Standard low to mid-latitude markers of the Martini (1971) and Okada and Bukry 905 (1980) were absent. We instead used the high southern latitude zonations developed by Wei, 906 1991 and 1992; Wei and Wise (1990, 1992); Monechi and Reale (1997); Watkins and Villa 907 (2000); Agnini et al. (2014). Biochronology was developed following Lourens et al. (2004) 908 and Raffi et al. (2006), using astronomically calibrated ages from magnetic reversals and 909 reference isotope stratigraphies, adjusted to the ATNTS2004 (Gradstein et al., 2004). Only 12 910 of these samples contained enough amounts of nannofossils to provide biostratigraphic 911 information (Table S1). In most of these samples calcareous nannofossils are abundant. The 912 lowest cores 278-34R to 31R consist of siliceous nannofossil chalk comprising 70 to 95% 913 calcareous nannofossils. Diatoms increase in number at the expense of calcareous nannofossils 914 in core 31R, and become the dominant sediment-forming microfossil in cores 30R and above. 915 Calcareous nannofossils are present in most intervals, but their abundance drops to trace 916 amounts or absent in some samples in cores 29R and 28R (Kennett et al., 1975).

Diatom biostratigraphic data for DSDP Site 278 were compiled from a variety of published sources, as well as new data collected for this study. Schrader (1976) provided the initial biostratigraphic characterization of 72 samples in this interval between samples 278-34R-3, 50 cm to 278-21R-1, 50 cm (his dataset continues up to core 278-9R-1, 50 cm), 921 reporting 58 diatom taxa in this lower interval. Weaver and Gombos (1981) compared the 922 stratigraphy of DSDP Sites 278 and 266 in an attempt to reconcile divergent records of diatom 923 ranges, with the resultant interpretation of a hiatus within core section 15R-2 in DSDP Site 278 924 that resulted in truncated ranges. Harwood (1982) examined 20 samples across the Oligocene-Miocene interval between samples 278-34R-1, 135 cm and 278-25R-2, 44 cm in the attempt to 925 926 apply the diatom zonation proposed by Gombos and Ciesielski (1983) from DSDP Leg 71 in 927 the South Atlantic to the DSDP Site 278 record, and documented the occurrence of 69 taxa. 928 Based on the stratigraphic abundance distribution of *Rocella* spp. taxa, Harwood (1982) 929 interpreted a hiatus between samples 278-31R-3, 139-141 cm and 278-3R1-2, 139-141 cm, 930 which was also noted in Fenner (1984), along with the ranges of 19 other diatoms in DSDP 931 Site 278. 35 new samples were examined by Harwood for the present study in the interval 932 between samples 278-34R-1, 142 cm and 278-26R1, 7 cm, wherein 112 diatom taxa were 933 noted, along with 17 other siliceous microfossil taxa (silicoflagellates, ebridians, endoskeletal 934 dinoflagellates, and a range of unrecorded chrysophycean cysts). The diatom and other 935 siliceous microfossil record require further documentation, as it is clear that the DSDP 278 936 record is an important reference section for diatom biostratigraphy. Schrader (1976) appears to 937 have examined strewn slides from chemically-treated sediment residues, whereas Harwood 938 (1982), following the methodology of Gombos and Ciesielski (1981), examined slides of size 939 fractions from $>37 \mu m$ and $<37 \mu m$, as well as raw strewn, chemically-treated sample residues. 940 Similarly, in preparation for the present study, diatom samples were sieved through a smaller, 941 20 um mesh sieve. Sieving results in the recovery of a greater number of whole and identifiable 942 taxa, and is responsible for the recording of numerous taxa not reported by Schrader (1976), 943 including many marker taxa used in the Gombos and Ciesielski (1983) zonation. In addition, 944 the recent compilation of biostratigraphic events developed for IODP Expedition 374 in the 945 Ross Sea (McKay et al., 2019) was consulted to provide ages for diatom events noted in this study. This compilation drew from output from the Constrained Analysis (CONOP) model
output associated with the study of ODP Site 744 as presented in Farmer (2011) and Florindo
et al. (2013), which builds upon the composite ordering of events and model age output for
Southern Ocean data sets presented by Cody et al. (2008, 2012) (Table S2).

Radiolarian biostratigraphic event data of first and last appearance datum levels are 950 951 interpreted from Table 1 of Petrushevskaya (1975). Taxon names presented in Table S3 reflect 952 those presented by Petrushevskaya, as well as names in current use. Age information and 953 taxonomic revisions are drawn from the works of Abelmann (1990), Caulet (1991), Takemura 954 (1992), and Takemura and Ling (1997), as well as from the age model output provided by the 955 CONOP analyses from ODP Site 744. Assignment of radiolarian datum events to magnetic 956 polarity chrons are drawn from the radiolarian range data of select taxa presented in Abelmann 957 (1990) from ODP Sites 689 and 690 in conjunction with age models for these sites provided 958 by Florindo and Roberts (2005).

959 In order to provide independent age constraints to biostratigraphy we studied the 960 magnetic stratigraphy of DSDP Site 278. The interpreted magnetic chrons are presented in 961 Table S4 and in Figures 3. Magnetic reversals in the samples were identified through stepwise 962 demagnetisation experiments by alternating fields (AF) using the 2G magnetometer (2G-963 SRM750 Superconducting Rock Magnetometer) at the Paleomagnetic Laboratory of Barcelona 964 (CCiTUB-CSIC). A total of 47 samples provided reliable inclinations. Most of the samples 965 were successfully demagnetized after 80 mT, although some of them likely with higher 966 coercivity minerals were demagnetized after 120-150 mT. Core orientation is not 967 reconstructed, and magnetic inclinations are used to determine magnetic polarities (Fig. S3A) 968 In general samples depict normal and reversed polarities (Fig. S3B). We have identified 7 969 reversal events shown on Figure 3 and Supplementary Table S4.

The stratigraphic section cored at DSDP Site 278 from the bottom of the sedimentary section in core 34R at ~429 mbsf up to sample 278-21R-1, 50 cm at ~282 mbsf spans the time interval from ~ 31 to 17.7 Ma, with one unconformity noted between samples 278-31R-3, 35 cm and 278-31R-2, 139 cm (398.85 – 399.89 mbf), which removed a 2.4 m.y. time interval from 28.2 to 25.8 Ma. The top of this interval is bounded by an unconformity between samples 278-21R-1, 50 cm and 278-20R-6, 50 cm (281.5 mbsf) which removed the time interval 17.6 to 15.5 Ma.

977 Sediments between 396.95-428.45 mbsf (Cores 34R-31R) contain calcareous 978 nannofossils Reticulofenestra daviesii, Cyclicargolithus floridanus, Chiasmolithus altus, 979 Reticulofenestra bisect/stavensis, andCyclicargolithus abisectus. In this interval occasional 980 reworked taxa have been observed. The interval between 379.5 and 414.41 mbsf is assigned to 981 be between NP25 (23.13-26.84 Ma) and NP23 (29.62-32.02 Ma) due to the co-occurrence of 982 Chiasmolithus altus (absent above 379.5 mbsf) and Cyclicargolithus abisectus (regular above 983 410.41 mbsf). The interval below 422.74 mbsf is attributed to an interval between biozones NP 984 23-NP 16?, due to the absence of *Reticulofenestra umbilicus* and the consistent presence of Reticulofenestra bisecta/stavenis (Martini et al.1970; Watkins and Villa, 2000; Young et al., 985 986 2017, Kulhanek et al., 2019). The LAD of diatoms Pyxilla eocena and Hemiaulus rectus var. twista at 422.09 and 425.43 mbsf, respectively indicate an age near 30.8 to 30.5 Ma at the 987 988 bottom of DSDP 278. Recognition of the FAD of diatom Rocella vigilans var. A at 421.85 989 mbsf (30.2 Ma) and the FAD of radiolarian Cyrtocapsella longithorax at 417.45 mbsf (31.5 to 990 32.5 Ma) further constrains the age of core 33R, and allows assignment to the polarity Chrons 991 C12n and C11n (Fig. 3; Tables S2-S4). Accordingly, the magnetic reversal at 425.44 mbsf (N1) 992 is interpreted as the top of Chron C12n (30.59 Ma). Guided by the biostratigraphic datumsit is 993 possible to place the base of C11n.2n and top of C11n.1n (Fig. 3, N2; Tables S4). These tie-994 points and derived sedimentation rate can be extended complementing the age model up to

995 section 31R-3, where a cluster of diatom first appearance datum levels is noted, indicating the 996 presence of a hiatus. The FAD of diatoms 'Coscinodiscus' rhombicus (28.45 Ma), Azpeitia 997 gombosi (28.3 Ma), Rocella gelida (26.4 Ma) in a sample from 398.85 mbsf, the FAD of 998 diatoms Triceratium groningensis (27.2 Ma) and Rocella schraderi (~26.3 Ma) in sample from 999 398.39 mbsf, and the FAD of radiolarian Stylosphaera radiosa (~26.0 Ma) in a sample from 1000 ~397 mbsf identify this break in time. The LAD of diatom Rocella vigilans (25.4 Ma) in a 1001 samplefrom 398.39 mbsf identifies when sedimentation resumed after the erosion or non-1002 deposition associated with the hiatus (Fig. 3). Accordingly, the interval of normal polarity (N3) 1003 measured between 398.86 and 397.34 mbsf is interpreted to correlate to Chron C8n.2n (Table 1004 **S**4).

1005 A major change in the sedimentation occurs in core 30R (378.91-379-75mbsf) marked 1006 by the high abundance of siliceous microfossils (Kennett et al., 1975). This interval is 1007 characterized by the presence of calcareous nannofossils Reticulofenestra daviesii, 1008 Cyclicargolithus floridanus, Discoaster deflandrei, while regular taxa recorded above, such as 1009 Reticulofenestra bisecta/stavensis and Chiasmolithus altus are absent. This assemblage is 1010 assigned to zone NP25 (23.13-26.48 Ma) (Martini et al., 1970; Young et al., 2017), late 1011 Oligocene; however, due to the absence of characteristic marker species in the studied samples, 1012 we cannot discard an assignment of this interval to NP25-NN1. Despite the weak and low 1013 inclinations at this interval, we tentatively assign the normal polarity zone (N4) found at 377.94 1014 mbsf to Chron C7An, suggesting an age of 24.8 Ma. A long interval of normal polarity 1015 identified in five samples from 366.28 to 357.66 mbsf (N5; Table S4) is interpreted as 1016 correlative to Chron C7n.2n (Fig. 3). One sample of reversed polarity at 352.32 mbsf is here 1017 interpreted as Chron C6Cr.

1018The FAD of diatom Raphidodiscus marylandicus (22.7 Ma) is noted in a sample at1019347.39 mbsf and Thalassiosira spumellaroides (22.7 Ma) in a sample at 342 mbsf. These

1020 diatom assemblages together with the presence of the calcareous nannofossil Cyclicargolithus 1021 floridanus and absence of Reticulofenestra bisecta/stavensis and other markers recorded in the 1022 lower part of Site 278, support an assignment of this interval of DSDP 278 to be assigned to 1023 the interval to biozones NN1 (22.82-23.13 Ma) and NN2 (19-22.82 Ma) (Martini et al., 1970; 1024 Young et al., 2017; Kulhanek et al., 2019). This is consistent with the diatom record, although 1025 no precise markers were observed. The biostratigraphic constraints of cores 27R and 26R here 1026 guide the magnetostratigraphic correlation. The reversal at 337.76 mbsf (base of N7) is 1027 interpreted as the base of Subchron C6Bn.2n. Above, the reversed magnetozone is assigned to 1028 C6Bn.1r whilst the top of Chron C6Bn.1n is ascribed to the reversal occurring at 331.44 mbsf 1029 (Fig.3; Table S4). Because of the wide range provided by biostratigraphic constraints for the 1030 interval encompassing cores 29 to 26, an alternative magnetostratigraphic correlation is 1031 allowed. In such case, magnetozone N6 could correspond to Subchron C6Bn.2n and C6Bn.1n; 1032 N7 to C6An.2n; and N8 to C6An.1n. Nevertheless, we consider this correlation less likely as 1033 it implies a change in the sedimentation trend towards lower values, unlike the rest of our 1034 interpretation. In addition, the fit pattern between our local reversals and the GPTS is not as 1035 good as the one using our preferred model.

1036 Above the polarity reversals of core 26R, there is limited chronostratigraphic control 1037 up to cores 22R and 21R where numerous diatom events are noted. The LAD of Thalassiosira 1038 spumellaroides (18.5 Ma) noted in a sample at 294.5 mbsf and the FAD of Fragilariopsis 1039 pusilla (18.5 Ma) at 286.5 mbsf indicate the age at this level. Similarly, the close association 1040 of the FAD of Crucidenticula ikebei (18.0 Ma) at 282.66 mbsf and the LAD of Thalassiosira 1041 praefraga (17.4 - 17.9 Ma) at 285.25 mbsf provides age control for this interval. Between 1042 cores 21R and 20R, we interpret the existence of a hiatus based on the coincident FADs of 1043 several diatoms in the sample at 280.01 mbsf: Araniscus lewisianus (17.8 Ma), Denticulopsis

- 1044 maccollumii (16.7 Ma), Nitzschia grossepunctata (15.6 Ma), and radiolarian FAD of
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Eucyrtidium punctatum (17.3 Ma) in a sample at 281.60 mbsf. **Supplementary Figures**

$\begin{array}{c} 1048\\ 1049 \end{array}$

1050 Figure S1. Maps showing the location of the sites and the distribution of dissolved oxygen, 1051 temperature and salinity versus depth along the Tasmanian to Ross Sea transect. Data derived 1052 from HYDROGRAPHIC ATLAS OF THE SOUTHERN OCEAN (Olberns et al., 1992). Maps 1053 were produced using Ocean Data View (Schlitzer, 2016). UCDW= Upper Circumpolar Deep 1054 Water; LCDW= Lower Circumpolar Deep Water; RSBW= Ross Sea Bottom Water. 1055



Figure S2. Neodymium isotopic composition from station TAN0803 station 125 (Lambelet et al., 2018) versus depth. Map and section were created using Ocean Data View software
(Schlitzer, 2016). Antarctic Surface Water= AASW; UCDW= Upper Circumpolar Deep 1061 Water; LCDW= Lower Circumpolar Deep Water.



A) Demagnetization Diagrams (Alternating Field)









1067 1068

Figure S4. Age mode for Site 274 from Hoem et al. (2021). (a) Core numbers, core recovery and lithologic log are based on Hayes et al. (1975). (b) Magnetic correlation for Site 274 with comparison to Jovane et al. (2020) (dotted lines). LO= Last occurrence and FO= First occurrence (see Hoem et al. (2021) for details).





Figure S5. Paleobathymetry and paleotopography reconstructions of the Southern Ocean
derived from Paleo-digital elevation model by Scotese and Wright (2018). Reconstructions of
the paleolocation of study sites and sites discussed were adapted from G-plates based on Muller
et al., 2016 global plate motion model.



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Figure S6: Neodymium isotopic composition $(\varepsilon_{Nd(t)})$ of fish debris and detrital sediment samples from this study. **A:** Fish debris $\varepsilon_{Nd(t)}$ values (red diamonds) and detrital sediment $\varepsilon_{Nd(t)}$ values (red open diamonds) at Site 278. **B:** fossil fish debris $\varepsilon_{Nd(t)}$ values (purple circles) and detrital sediment $\varepsilon_{Nd(t)}$ values (purple open circles) at Site 274.

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