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# Radiogenic isotopic and clay mineralogical signatures of terrigenous particles as water mass tracers: new insights into South Atlantic deep circulation during the Last Termination.

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#### 14 Abstract

15 The past evolution of the Southern Ocean, one of the major components of the climatic system, is still a matter of debate. This study provides new insights into the deep 16 17 Southern Ocean circulation based on the radiogenic isotopes and clay mineralogical signature 18 of the terrigenous fractions transported by the main deep water masses to sediments recovered 19 in core MD07-3076Q from the central South Atlantic. This approach successfully permits: (1) provenance identification of the various grain-size fractions (clay, cohesive silt and 20 21 sortable silt); (2) assignment of each grain-size fraction to a specific water-mass; (3) 22 reconstruction of past changes in the main deep water-mass pathways. These data document 23 the evolution of deep-water masses in the South Atlantic Ocean during the last deglaciation. The Antarctic Bottom Water (AABW) speed and northward extension were maximum at the 24

25 end of the Last Glacial Maximum (LGM), associated with strong bottom water production in 26 the Weddell Sea, together with a vigorous Lower Circumpolar Deep Water (LCDW). In 27 contrast the North Atlantic Deep Water (NADW) circulation was weaker than today. The 28 onset of the deglaciation (from 17.5 ka to 15 ka, ~Heinrich Stadial 1, HS 1) was marked by 29 weakening and southerly retreat of the AABW and by an increase of mixing between AABW 30 and LCDW. The speed of the AABW remained at its lowest during the Bølling Allerød (B/A) 31 and the Younger Dryas (YD), and the LCDW slowed and retreated to the south, while the NADW progressively migrated southward, deepened, and strengthened between the 32 33 beginning of the Bølling Allerød and the Holocene (from ~15 to 10 ka).

#### 34 Keywords

Last Deglaciation; Paleoceanography; Southern Ocean; South Atlantic; Radiogenic
 isotopes; Clay mineralogy; Grain size distribution.

#### 37 I. Introduction

38 The Southern Ocean (SO) is a key area for understanding the present and past carbon 39 cycle. Direct observations indicate that the SO is a sink of atmospheric CO<sub>2</sub> today (Le Quéré 40 et al., 2007; Sallée et al., 2012) while it was likely a source of CO<sub>2</sub> to the atmosphere during 41 the deglaciation (Bouttes et al., 2012; Jaccard et al., 2013). Upwelling of CO<sub>2</sub>-rich deep water 42 likely explains most of the CO<sub>2</sub> rise observed during Heinrich Stadials (HS; Ahn and Brook, 43 2008; Anderson et al., 2009; Fischer et al., 2010; Skinner et al., 2010; Menviel et al., 2014, 44 2017). Several other processes occurring in the Southern Ocean have also been invoked to explain the deglacial atmospheric CO<sub>2</sub> rise. For example, reduction of biological pump due to 45 reduced iron fertilization (Watson et al., 2000; Wolff et al., 2006; Fischer et al., 2010; 46 47 Martinez-Garcia et al., 2014); reduced CO<sub>2</sub> solubility due to increased temperature of the 48 ocean (Köhler and Fischer, 2006; Fischer et al., 2010); reduction of sea ice extent (Fischer et

49 al., 2010). As a consequence, the organization and extension of the Southern Ocean deep 50 water-masses (i.e., Atlantic Meridional Overturning Circulation, AMOC; Skinner et al., 2010; 51 Shakun et al., 2012; Toggweiler et al., 2006) were studied, mainly using paleo-hydrological proxies ( $\delta^{18}$ O,  $\delta^{13}$ C,  $\epsilon_{Nd}$  of sea water,  $\Delta^{13}$ C; Barker et al., 2009; Vasquez-Riveiros et al., 2010; 52 Skinner et al., 2010, 2013, 2014; Waelbroeck et al., 2011; Gottschalk et al., 2015b). By 53 54 contrast, the terrigenous particle signal transported by these deep water-masses in the sub-55 Antarctic Atlantic Ocean remain poorly studied, even though locally it was successfully used 56 to reconstruct the deep ocean circulation dynamical component (Diekmann et al., 2000 -57 Scotia Sea; Walter et al., 2000 - Scotia Sea and southwest South Atlantic; Kuhn and 58 Diekmann, 2002 – Cape Basin). In this study, we aim to reconstruct the past evolution of 59 deep-water masses (i.e. contribution to sedimentation, vertical and horizontal extensions, and speeds) during the last deglaciation using the signal carried in the terrigenous particles 60 61 transported by these water masses. Sediment grain size distribution allows identifying the 62 contribution of distinct oceanic currents (Weltje and Prins, 2007; Weltje, 2012), and reconstructing the speeds of the main deep water-masses (McCave and Hall, 2006; McCave 63 64 and Andrew, 2019). Grain-size distribution displays distinct modes (Weltje, 2012) that can be 65 attributed to specific physical transport processes (e.g., wind, water currents, ice) and/or transport energy. Because grain-size distribution reflects both carrying vectors (e.g., wind, 66 67 water currents, ice) and flow speed, several studies focused on the respective isotopic 68 composition of distinct grain-size fractions in order to track distinct provenance patterns 69 (Meyer et al., 2011; Bayon et al., 2015; Beny et al., 2018). Similarly, clay minerals are useful 70 tracers of provenance, erosion balance, weathering, and transportation pattern of fine-grained sediments (Moriarty et al., 1977; Petschick et al., 1996; Kuhn and Diekmann, 2002; Montero-71 72 Serrano et al., 2010). Finally, radiogenic isotope compositions of sediments can be used to

decipher the different provenances of terrigenous particles and determine their respective
contributions through time (Walter et al., 2000; Bayon et al., 2009).

75 This study focuses on the South Atlantic Ocean, where northern water-masses (North Atlantic Deep Water, NADW) and southern ones (Lower Circumpolar Deep Water, LCDW 76 77 and Antarctic Bottom Water, AABW) mix, because this area is key for understanding the atmosphere-ocean exchanges that control CO<sub>2</sub> sequestration/outgassing. The reorganization of 78 79 these deep water-masses during the deglaciation is still a subject of debate and their impact on 80 deglaciation is not resolved (Barker et al., 2009; Skinner et al., 2010, 2013; Waelbroeck et al., 81 2011; Adkins, 2013; Menviel et al., 2014, 2015a,b, 2018). Recent studies of the last 82 deglaciation ventilation events in the South Atlantic Ocean demonstrated an apparent synergy 83 between physical and biological processes (Gottschalk et al., 2016), whereas chemical and 84 physical processes appear to be decoupled (Roberts et al., 2016). These contrasting results 85 highlight the lack of constraints on the dynamical aspect of the water-masses mixing. In this 86 paper, we combined grain-size distribution, clay mineralogy, and radiogenic isotope (Sr-Nd-87 Pb) analyses (on separated grain-size fractions) in order to investigate the evolution 88 (horizontal and vertical extensions, pathways, speed) of deep-water-masses in the South 89 Atlantic Ocean during the last termination.

#### 90 II. General setting



Figure 1: Simplified geological map of the study area. Light blue text names riverine
systems, dark blue text names seas, black text names oceanic basins, and white abbreviations
names areas. In South America: Guyana Highlands -GH-, Brazilian Highlands -BH-,
Argentinian Pampas -Pam-, Patagonia -Pat-; in Africa: Namibian deserts-Na-; in Antarctica:
Ellsworth Mountains -EM-, Antarctic Peninsula -AntP-, Pensacola Mountains -PM-, Berkner
Islands -BI-, Shackleton Range -SR-, Coats Land -CL-, Dronning Maud Land -DML-. Clay

98 mineral inputs to the South Atlantic Ocean derived from the studies of Petschick et al.
99 (1996); Diekmann et al. (2000); Walter et al. (2000); Guyot et al. (2007); Bayon et al.
100 (2016); and Khondoker et al. (2018).

101 A. Deep water-masses

102 Modern South Atlantic deep circulation is characterized by the incursion of the 103 NADW into the Southern Ocean, where it mixes/interacts with both the AABW and the 104 Circumpolar Deep Water (CDW; figure 1). The southward NADW mainly flows as Deep 105 Western Boundary Current along the South American Margin before leaving the continental 106 shelf and flowing eastward around 45°S (Reid, 1989; Larqué et al., 1997; Stramma and 107 England, 1999). A branch of the NADW crosses the Atlantic Ocean at the equator before to 108 flow southward along the African margin and to reach the Indian Ocean via the Good Hope 109 Cape (Larqué et al., 1997). In the Southern Ocean, NADW flows between two parts of the 110 CDW: the Upper Circumpolar Deep Water (UCDW) flows above, whereas the Lower 111 Circumpolar Deep Water flows underneath. The northern extension of the UCDW is very 112 asymmetric with larger extent in the eastern part of the South Atlantic (Larqué et al., 1997). 113 On the west side of the Mid-Atlantic Ridge (MAR), the LCDW penetrates into the Argentine 114 Basin via the Drake Passage and flows northward into the Brazil Basin (Speer and Zend, 115 1993). On the east of the MAR, the LCDW is a major oceanic feature in the Cape Basin, 116 where it becomes diluted in the Angola Basin (Larqué et al., 1997). The lower part of the 117 LCDW is currently mixed with the upper part of the AABW (Orsi et al., 1999) that is 118 produced heterogeneously along the Antarctic Shelf (Foster and Carmack, 1975; Orsi et al., 119 1999). Foster and Camarck (1975) demonstrated the formation of a very cold type of bottom 120 water in the vicinity of the shelf break (the Polynyas mode) between 29 and 40°W and along 121 the Antarctic Peninsula's tip, between 35 and 55°W and north of 65°S. Today this production 122 occurs mainly in winter when seawater is close to freezing and becomes saltier due to sea ice

123 formation (Foster and Camarck, 1975; Orsi et al., 1999), which increases its density and 124 promotes convection. This mode is called the Polynyas mode. However, in the Weddell Sea, a 125 second mode of bottom water formation occurs: the Ice Shelf Water mode, which corresponds 126 to bottom water formation under the Antarctic ice shelves due to super-cooling (Krueger et al. 127 and references therein, 2012). The AABW is very thick in the Weddell-Enderby Basin (more 128 than 4000 m; Orsi et al., 1999) and is partially exported to the Argentine Basin via the eastern 129 Scotia Sea (east and west of the South Sandwich Islands) toward the Brazil Basin via the 130 Vema Channel, and to the Agulhas and Crozet basins (Stramma and England, 1999). The 131 AABW is a dominant oceanic feature south of 50°S although its' northern influence extents to 132 around 10°S (Reid et al., 1989).

133 The characteristics of deep water masses were not constant through time and the evolution of the AMOC is controversial (Weber and Drijfhout, 2007; Vasquez Riveiros et al., 134 2010; Waelbroeck et al., 2011; Howe et al., 2016). Some studies have suggested that during 135 136 the Last Glacial Maximum (LGM) the NADW was as strong as today but shallower (Curry 137 and Oppo, 2005; Gherardi et al., 2009). Other studies suggest that it was weaker and 138 shallower (Menviel et al., 2012; Howe et al., 2016). The NADW southern extension seems 139 limited during glacial times, whereas southern-born water-masses extended further north 140 (Kuhn and Diekmann, 2002; Skinner et al., 2013). Similarly, modifications of the Antarctic 141 Circumpolar Current (ACC, including the CDW) during the last climatic cycle are still 142 debated. Some studies reported severe modifications of the ACC intensity in the Atlantic 143 (Noble et al., 2012) and Indian (Dezileau et al., 2000; Mazaud et al., 2010) sectors of the 144 Southern Ocean: the activity of the ACC increased during the LGM, probably linked to 145 enhanced westerly winds (Dezileau et al., 2000; Walter et al., 2000; Mazaud et al., 2010). In 146 contrast, other studies support insignificant modification of the ACC in the Atlantic sector of 147 the Southern Ocean (Gersonde et al., 2003) and Drake Passage (McCave et al., 2014). Finally,

the evolution of the AABW is also uncertain. Krueger et al. (2012) suggests that the production of AABW was similar during glacial and interglacial, but dropped during terminations (including the last deglaciation). In addition, Menviel et al. (2017) suggest that the "AABW" (corresponding to CDW and AABW in our study) was weaker during the LGM than today. In contrast, other studies suggest enhanced northward flow of AABW via deep western boundary currents in all three major ocean (Pacific: Hall et al., 2001; Govin et al., 2009; Indian: Govin et al., 2009; Atlantic: Govin et al., 2009; Spooner et al., 2018).

155 Any modifications of the AMOC have a potential effect on carbon storage/outgassing 156 that is tightly controlled by ocean stratification/ventilation, itself resulting from contrasting 157 physical and chemical properties of the water-masses. High ocean stratification – as during 158 the last glacial – inhibits vertical mixing and isolates the deep ocean from the atmosphere. In 159 contrast, ventilation events - breakdown of the stratification that occurred during the last 160 deglaciation – allow  $CO_2$  to upwell from the deep ocean and to be rapidly transferred to the 161 atmosphere (Toggweiler et al., 2006; Barker et al., 2010; Skinner et al., 2010, 2013, 2014; 162 Burke and Robinson, 2012).

163

#### B. Geological setting

164 The Atlantic sector of the Southern Ocean may receive sediments from Antarctica, the 165 Scotia Sea area (i.e., Scotia Arc, southern Patagonia, and northern Antarctic Peninsula) South 166 America, and Africa. Here, we present the general geological structure of these areas, and 167 their respective isotopic signatures (figure 1).

Antarctica corresponds to two distinct geological domains: East and West Antarctica. West Antarctica includes the Ellsworth Mountains (EM), the Antarctic Peninsula (Ant P), the Scotia Arc region (SCA). The geology of the Ellsworth Mountains (figure 1) is poorly known due to the lack of outcrop. These mountains are composed of Precambrian rocks, corresponding mainly to granodiorite orthogneiss intruded by granitic and pegmatitic sheets

173 (Tingey et al., 1991). The isotopic composition of the area is not known, but the age of the 174 rock suggests radiogenic Sr and Pb isotope ratios, and unradiogenic  $\varepsilon_{Nd}$  (figure 1). This area 175 mostly delivers illite to the sea (Petschick et al., 1996; figure 1). The Antarctic Peninsula, 176 composed of alkaline and calcalkaline magmatic rocks associated with deformed sediments of 177 similar composition (Anderson, 1965; Tingey et al., 1991), is a potential source of terrigenous 178 sediment for CDW and AABW. The Antarctic Peninsula is characterized by radiogenic  $\varepsilon_{Nd}$ , 179 and low Sr and high Pb isotope ratios (figure 1). Clay minerals from this area mostly 180 correspond to well-crystallized smectite (crystallinity around 1.5, Petschick et al., 1996; 181 figure 1), and chlorite. The Scotia Arc islands rocks are similar to those of the Antarctic 182 Peninsula but with slightly more radiogenic Nd, and slightly less radiogenic Sr and Pb (figure 183 1). This area exports exclusively well-crystallized smectite to the ocean (crystallinity lower 184 than one; Petschick et al., 1996).

185 The East Antarctic domain is an old shield with rocks > 0.9 Ga. Rocks from the 186 Pensacola Mountains (PM) to the Shackleton Range (SR) represent the Ross Orogen (i.e., 187 Transantarctic Mountains) and are mainly composed of varied metamorphic rocks aged from 188 the Late Proterozoic to Late Paleozoic (Barrett, 1991; Laird, 1991; Tingey, 1991; Hauptvogel 189 and Passchier and references therein, 2012). The isotopic composition of the area is 190 characterized by unradiogenic Nd, with intermediate to high Sr isotope ratios (figure 1). The 191 Pb isotope composition unknown, as is the case for most of East Antarctica rocks. The few 192 studies reporting Pb isotope ratios have been carried out on feldspars only. The area releases mostly illite and chlorite (Petschick et al., 1996). Smectite is also exported from the Berkner 193 194 Island, but is found only locally (Petschik et al., 1996). To the east, the Grenville Belts (920 to 195 1330 Ma) outcrop on the Coats Land (CL) and western Dronning Maud Land (WDML) with 196 a wide range of gneissic and clastic sediments (Capurro, 1955; Anderson, 1965; Tingey et al., 197 1991) with unradiogenic Nd, and radiogenic Sr isotope ratios (figure 1). Clay minerals

produced in this area are mainly chlorite and illite (Petschick et al., 1996). In the northernmost part of the Dronning Maud Land, the old Grunehogna Craton is composed mainly of Archean granitoids (Tingey et al., 1991; Mieth and Jokat, 2014; Pierce et al., 2014) with very unradiogenic Nd ( $\varepsilon_{Nd}$ << -20) and radiogenic Sr ratios (figure 1). The clay minerals generated in that area are mainly chlorite and illite (Petschick et al., 1996).

203 South America can be divided into two blocks: the recent Andean Orogen, and the old 204 South American Shield. In the Atlantic Sector of the Southern Ocean, the Andean Orogen 205 mostly corresponds to Patagonia (Pat). The geology and the isotopic composition of this 206 region is similar with the Antarctic Peninsula: radiogenic Nd, unradiogenic Sr and Pb isotope ratios, but with higher <sup>207</sup>Pb/<sup>204</sup>Pb than the Antarctic Peninsula (figure 1; Olivero and 207 Martinioni, 2001; Savago et al., 2001 and references therein; Smith et al., 2003; Khondoker et 208 209 al., 2018). Even if the climate is slightly more humid than in the Antarctic Peninsula, 210 Patagonia also produces mainly well-crystallized smectite (Petschick et al., 1996; Desiage et 211 al., 2018; Khondoker et al., 2018).

212 The South American Shield is composed of several cratonic areas separated by vast 213 ancient orogenic belts. (Cordani and Sato, 1999; Engler, 2009; Oyhantçabal and Siegesmund, 214 2011; Siegesmund et al., 2011; de Alkmim, 2015). Cratonic areas are mainly composed of 215 granitoids and a wide range of metamorphic rocks, while orogenic belts are composed of a 216 large variety of metamorphic rocks with high original composition (from the granite to the 217 ophiolite and the juvenile magmatic arc; Almeida et al., 1981; Barros et al., 1982; Moura and Gaudette, 1993; Neves, 2003; de Alkmim, 2015; Nogueira et al., 2007; Pimentel et al., 2011; 218 219 Fuck et al., 2014; McGee et al., 2015; D'el-Rey Silva et al., 2016). The overall isotopic 220 composition of the South American Shield is radiogenic Sr and Pb, and very unradiogenic Nd 221 isotope ratios (figure 1). South of the South American Shield, a large tholeiitic-basaltic 222 province is known as the Parana basalt (Waichel et al., 2012; Milani and Ramos, 2017). The composition of the Parana basalts differs from the South American Shield with radiogenic Nd,
 and very low <sup>87</sup>Sr/<sup>86</sup>Sr ratio (<0.5; Tapani Ramo et al., 2016).</li>

225 This South American Shield is drained by important riverine systems such as the 226 Amazon and Parana rivers. The Amazon drains sediments from the Andes, as well as the 227 South American Shield. The South American Shield is characterized by rocks with 228 unradiogenic Nd and radiogenic Sr and Pb (figure 1). The clay mineral exported to the ocean 229 is mainly kaolinite (Petschick et al., 1996; Guyot et al., 2007; Bayon et al., 2015). However, 230 sediments are mainly exported northwards toward the Caribbean Sea. The Parana River drains 231 the southern part of the South American Shield, the Parana basalts, and the north of 232 Patagonia. Its isotopic composition has intermediate Nd, Sr, and Pb isotope ratios (figure 1). 233 Clay minerals transported via this river are illite (~60%; Petschick et al., 1996; Campodonico 234 et al., 2016), kaolinite, and smectite. Other smaller rivers drain the south of Brazil, and mainly 235 deliver sediments characterized by unradiogenic Nd, and radiogenic Sr and Pb isotope ratios, 236 containing mainly smectite derived from the Parana basalts and kaolinite (figure 1; Petschick 237 et al., 1996). On the other hand, Patagonia and Pampas are dry and windy areas (Clapperton, 238 1993). Consequently, part of the detrital particles is exported to the ocean via aeolian 239 processes as loess (Mathias et al., 2014). The overall isotopic composition of this smectite-240 rich loess is radiogenic Nd, unradiogenic Pb and Sr ratios. According to Mathias et al. (2014), 241 the sediment delivery to the southwest Atlantic is currently dominated by loess, and by the 242 Parana River sediments.

A large proportion of the African continent comprises Archean Cratons (western Africa, Congo), and rocks related to the Pan African Orogen. The cratons are composed of gneisses while the Pan African Orogen is mainly composed of flysch deposits associated with volcanic formations, and intruded by granitoids. The overall geochemical composition of Africa is intermediate to very unradiogenic Nd, and intermediate to very radiogenic Sr and Pb isotope ratios (figure 1). Clay minerals formed in the Congo depression are mainly poorly
crystallized smectite and kaolinite, whereas the Namibian deserts release illite-rich loess
(Bremner and Willis, 1993; Petschick et al., 1996; Bayon et al., 2015; figure 1).

251 III. Material and methods

#### A. Material

This study is based on sediment samples from core MD07-3076Q (44°9.2'S, 253 254 14°13.7'W) recovered by the French Vessel Marion Dufresne in 2007 on the eastern flank of 255 the Mid-Atlantic Ridge at 3770 m depth (figure 1). The core chronology is based on 256 planktonic foraminifer radiocarbon dates (30 measurements for the studied interval) and 257 stratigraphic control points (Skinner et al., 2010, 2013, 2014; Gottschalk et al, 2015b). Today, 258 the site is bathed in the LCDW. From the seafloor to the surface, the water masses observed 259 in the water column above the study site are the LCDW, NADW, UCDW, and intermediate 260 and surface waters. In nearby deeper oceanic basins, the AABW is also found, and it is 261 thought that particles transported by this bottom water can reach our site by advection and 262 mixing with the LCDW.

263

#### B. Sample preparation and analytical procedure

All samples were first decarbonated using 0.1N HCl and then deflocculated by repeated washing using pure water.

266

#### 1. Grain-size distribution

Grain-size distribution was measured on carbonate-free sediment using a Malvern Mastersizer 2000 laser (0.02-2000  $\mu$ m) following standard protocols (Montero-Serrano et al., 2009). The clay-size fraction corresponds to the proportion (in volume) of particles with a diameter smaller than 2  $\mu$ m, the cohesive silts correspond to the 2 to 10  $\mu$ m diameter

particles, the sortable silts correspond to the 10 to 63 µm diameter particles, and the sand
fraction to the particles coarser than 63 µm.

273

#### 2. Clay mineralogy

274 The clay-size fraction ( $< 2\mu m$ ) was isolated by differential settling according to 275 Stoke's Law. This fraction was then concentrated by centrifugation (40 minutes, 3500 rotation 276 per minute) and an aliquot was placed on glass slides. X-ray diagrams were obtained using a 277 Bruker D4Endeavor (standard 30 kV and 30 mA). Each sample is run three times between 278 2.49 and  $32.5^{\circ}2\theta$ : (1) dried sample, (2) glycolated sample (12 hours in ethylene glycol), and 279 (3) heated sample (490°C for 2 hours). The proportion of each clay mineral (smectite, 280 chlorite, illite, and kaolinite) is determined using the main X-ray diffraction peaks (layer and 281 interlayer) on X-ray spectra according to their crystallographic characteristics (Brown, 1980). Smectite is characterized by a main peak at 14-15 Å on the air-dried samples that expands at 282 283 17 Å after ethylene glycol saturation and collapses at 10 Å after heating. Illite is characterized 284 by a main peak at 10 Å on the three runs. Kaolinite and chlorite are measured based on their 285 main peak at 7-7.2 Å and further distinguished using pseudo-void deconvolution of the 286 kaolinite/chlorite doublet at 3.54/3.58 Å. All measurements and semi-quantitative estimations 287 are processed using the McDiff software (Petschick, 2000). The index of Esquevin, (Esquevin, 1969) corresponds to the ratio between the intensity of the peak of illite at 5 Å and at 10 Å 288 289 (illite  $5\text{\AA}/10\text{\AA}$ ). In this study, the crystallinity of illite and smectite has been measured using 290 the method of integral breath to enable comparison with the work of Petschik et al. (1996) in 291 the South Atlantic. High crystallinity (high values of the index) corresponds to badly 292 crystallized mineral.

293

#### 3. Isotope Analysis

The grain-size distribution recognized three distribution modes. These three grain-size fractions were then isolated in order to examine their provenance. The clay-size fraction (0 $2 \mu m$ ) was isolated by repeated extractions by settling. The coarse fraction (20-40  $\mu m$  in the old section of the core before 11.5 ka cal BP, noted hereafter as ka, and 20-32  $\mu m$  after 11.5 ka) was isolated by sieving the clay-free residue. The 2-20  $\mu m$  grain-size fraction is thus the residue of clay separation and coarse fraction sieving. An aliquot of 10 to 150 mg (depending of the sediment quantities available) was then used for chemical procedures.

301 Chemical and isotopic measurements were performed at the Faculty of Science of the Vrije Universiteit, Amsterdam. Sediment was digested in closed Savillex<sup>TM</sup> Teflon beakers 302 303 using a mixture of concentrated HF and HNO<sub>3</sub> ( $\sim 2/3$  and 1/3, respectively) for a few days at 304 120°C. After digestion, samples were dried out and re-dissolved in ~6.5N HCl to remove 305 fluorides. Nd, Sr, and Pb were purified using conventional ion chromatography. Pb isotopes 306 were processed first and purified using AG-X8 200-400 mesh resin. Light rare earth elements (LREE) were isolated using true-spec resin medium (Eichrom<sup>TM</sup>, 100-150 µm mesh size) and 307 308 Nd was eluted from the LREE fraction using LN-resin (Eichrom<sup>TM</sup>, 50-100 µm mesh size). Sr 309 was recovered using Sr resin column. Nd and Pb measurements were performed by bracketing 310 using respectively CIGO and NBS981 standards on Thermo Scientific Neptune multi-311 collector inductive coupled plasma-mass spectrometer (MC-ICPMS), whereas Sr 312 measurements were performed on a thermal ionization multi-collector-MS (TIMS). Due to 313 relatively low content of Nd in some of the coarse grain-size fraction (20-32/40µm), Nd 314 measurements of these fractions were also performed by TIMS.

#### 315 IV. Results



Figure 2: Grain-size results from core MD07-3076Q. (a) and (b) Variation of the coarse and fine grain size mode values, respectively, through time. Black lines represent the average value for both grain-size modes while dashed lines represent their standard deviation; (b) typical grain-size distribution curve from the core MD07-3076Q. Two grain-size modes can be identified from the grain-size distribution curve. YD: Younger Dryas, BA: Bølling Allerød, HS: Heinrich Stadial; LGM: Last Glacial Maxima.

The grain-size distribution is bimodal throughout the core (figure 2) with a coarse mode ranging in the sortable silts fraction (from 23.6 and 36.2  $\mu$ m, average: 30.4, standard deviation: 2.48; table 1) and a fine mode ranging in the cohesive silts fraction (from 5.6 and 9.3  $\mu$ m, average: 6.8, standard deviation: 0.8; table 1). The sortable silt (10-63  $\mu$ m) fraction is dominant (41 to 52% in volume of the total terrigenous fraction, average: 47%, standard deviation: 2.4; table 1) and the cohesive silts (2-10  $\mu$ m) represent 35 to 45% (average: 39%, standard deviation: 2.4; table 1). The clay (<2  $\mu$ m) and sand fractions (>63  $\mu$ m) represent

331	respectively 2 to 5% (average: 3.4%, standard deviation: 0.7; table 1) and 1 to 6% (average:
332	3.0, standard deviation: 1.0; table 1) of the total terrigenous fraction (table 1).

In details, the mean of the coarse mode (hereafter mentioned as the sortable silt mode) is slightly coarser during the LGM (figure 2.a), and decreases during the deglaciation and reaches its minimum values after the Younger Dryas (YD). The average size of the fine mode (hereafter mentioned as the cohesive silt mode) is maximum at the end of the LGM and displays its lowest values during the deglaciation (figure 2.b).



Figure 3: Clay mineral data from core MD07-3076Q. Purple dots represent the
radiocarbon dates used for the age model. (a) relative proportion of smectite (%); (b)
smectite crystallinity (°2θ); (c) illite crystallinity (°2θ); (b) and (c) note that the lower is the
crystallinity index, the better the mineral is crystallized; (d) relative proportion of illite (%);
(e) relative proportion of chlorite (%); (f) relative proportion of kaolinite; (g)
kaolinite/chlorite ratio (K/C); (h) illite/smectite ratio (I/S).

345 **B**.

Clay Mineralogy

346 The clay mineral assemblage is composed of smectite ((S) 18-59%; figure 3.a), illite 347 ((I) 21-40%; figure 3.d), chlorite ((C) 12-33%; figure 3.e), and kaolinite ((K) 5-14%; figure 348 3.f) with higher smectite content at the end of the deglaciation, and higher illite, chlorite and 349 kaolinite contents during the LGM. The smectite crystallinity ranges from 0.89 and 1.45 350 (figure 3.b) and varies in phase with the smectite content with slightly higher variability 351 between 25 and 30 ka. The illite crystallinity varies between 0.27 and 0.42 (figure 3.c and 4) 352 without major variation. Clay mineral proportions are relatively stable during the LGM but 353 records a shift during the deglaciation with an increase of smectite compared to the other clay 354 minerals. The smectite crystallinity varies simultaneously with smectite proportion changes. 355 The K/C ratio is stable during the LGM, and increases during the deglaciation. The I/S ratio 356 presents two maxima at the beginning of the record and at the end of the LGM, sharply 357 decreases at the beginning of the HS 1, and steadily decreases during the deglaciation.



359 Crystallographic properties of illite from site MD07-3076Q (black dots) and Figure 4: comparison with properties of illite from potential sources to the South Atlantic basin. The 360 361 data used for comparison are from Petschick et al., (1996). Illite 5Å/10Å ratio gives information on the chemical properties of illite (Fe-Mg- vs Al-rich illite) while the 362 crystallinity gives clues on the quality of the mineral crystal (high crystallinity: badly 363 crystallized mineral). 364

C. 365

1.

#### Radiogenic Isotopes

Isotopic results are presented for the sortable silt mode fraction (20-32/40  $\mu$ m) and the 366 cohesive silt mode fraction (2–20  $\mu$ m) and for the clay-size fraction (<2  $\mu$ m) (tables 2 to 4). 367 368 Lead isotope ratios were only been measured on the clay-size fraction (table 2).

369

Clay-size fraction (<2  $\mu$ m)

370 The  $\varepsilon_{Nd}$  of the clay-size fraction ranges from -7.2 to -4.8. It is relatively stable during the last glacial ( $\leq \pm 0.5 \epsilon_{Nd}$ ), decreases by 2  $\epsilon$  units during the deglaciation and shows 371 minimum values at the end of the deglaciation (figure 5.c). The <sup>87</sup>Sr/<sup>86</sup>Sr ratio ranges from 372 373 0.711 to 0.712: it is rather high and stable ( $\leq 0.0003$ ), during the last glaciation and displays 374 a rapid decrease at ~15 ka. The ratio records a short rebound at 12.7 ka before reaching its

minimum values at the end of the deglaciation (0.7107; figure 5.d). The  ${}^{87}$ Sr/ ${}^{86}$ Sr ratio and the 375 376  $\varepsilon_{\rm Nd}$  show a similar temporal evolution (they both decrease during the deglaciation), which is 377 surprising as in nature they usually record opposite relationship. The three Pb isotope ratios 378 record their lowest values around 25 ka, and increase thereafter. This trend is more pronounced for the <sup>207</sup>Pb/<sup>204</sup>Pb ratio during the deglaciation. Some fluctuations of the three 379 380 ratios are observed during the deglaciation with higher ratios at climatic transitions 381 (LGM/Heinrich Stadial 1 -HS1-, HS1/Bølling-Allerød - B-A - , B-A/Younger Dryas -YD-, 382 Holocene onset), and low values during the middle of the HS1, the middle of the B-A, and the second half of the YD (except for the <sup>206</sup>Pb/<sup>204</sup>Pb). We note that the Sr isotope rebound 383 384 observed at 12.7 ka is also associated with variations Pb isotope ratios.

385

#### 2. Cohesive silt mode fraction (2-20 μm)

386 The  $\varepsilon_{Nd}$  of the 2-20 µm grain-size fraction (cohesive silt mode fraction) ranges from -387 5.5 to -3.9 with the lowest values at 13, 18, 23 and 29 ka (figure 5.b). The <sup>87</sup>Sr/<sup>86</sup>Sr ratios are 388 generally constant with higher values around 13, 19, and 24 ka (figure 5.f).

389

#### 3. Sortable silt mode fraction: (20-32/40 μm)

The  $\varepsilon_{Nd}$  of the sortable silt mode fraction ranges from -4.9 to -1.4. Values are relatively unchanging during the last glacial period (-2 ± 0.5  $\varepsilon_{Nd}$ ) but decreases after 18 ka and reaches a minimum (-5) at the end of the deglaciation (figure 5.a). The <sup>87</sup>Sr/<sup>86</sup>Sr ratio ranges from 0.708 and 0.710. <sup>87</sup>Sr/<sup>86</sup>Sr values are lower during the HS 2, at the beginning of the HS 1, and at the beginning of the YD, while the highest value is recorded at the B-A offset (figure 5.e).



Figure 5: Isotopic results from core MD07-3076Q. (a)  $\varepsilon_{Nd}$  from the sortable silt mode fraction (20-32/40 µm); (b)  $\varepsilon_{Nd}$  from the cohesive silt mode fraction (2-20 µm); (c)  $\varepsilon_{Nd}$  from the clay-size fraction (<2 µm); (d) <sup>87</sup>Sr/<sup>86</sup>Sr from the clay-size fraction (<2 µm); (f) <sup>87</sup>Sr/<sup>86</sup>Sr from the sortable silt mode fraction (20-32/40 µm); (g) <sup>87</sup>Sr/<sup>86</sup>Sr from the cohesive silt mode fraction (2-20 µm); (h) <sup>208</sup>Pb/<sup>204</sup>Pb, (i) <sup>207</sup>Pb/<sup>204</sup>Pb, and (j) <sup>206</sup>Pb/<sup>204</sup>Pb from the clay-size fraction (<2 µm).

- 403 V. Discussion
- 404

#### A. Sediment provenance

#### 405 1. Clay size fraction: an overall provenance signal

406 To determine the provenance of this grain-size fraction, both clay mineral proportions 407 and radiogenic isotopes have been used. Kaolinite and chlorite have distinct provenances: at 408 present, kaolinite is mainly found at low latitudes where chemical weathering is strong. 409 Brazilian rivers (Amazon and Parana), as well as equatorial and tropical African rivers are 410 characterized by high contents of kaolinite (figure 1). Inherited kaolinite is also found in East 411 Antarctica marine sediments (figure 1), but is transported by the westward Antarctic Coastal 412 Current and remains trapped in the Weddell-Enderby Basin due to the Weddell Gyre 413 circulation (Petschick et al., 1996) and this kaolinite is therefore unlikely to contribute to 414 sedimentation at the studied site. Consequently, kaolinite can be used to trace the low latitude 415 provenance. Conversely, presently, chlorite mainly characterizes high latitude regions, where 416 weak chemical weathering allows its preservation. It is abundant in Antarctica and the Scotia 417 Sea (figure 1). Consequently, chlorite can be considered a tracer of high latitude provenance. 418 Thus, the kaolinite/chlorite ratio (K/C) discriminates southern-born versus northern-born 419 material in the southern Atlantic (Petschick et al., 1996; Kuhn and Diekmann., 2002; Kruger 420 et al., 2008).

421 Illite provenance is more complex. It is very abundant in the Weddell Sea, Namibian 422 deserts, the Parana River (60% of the delivered sediment), and it is also present, although at 423 lower concentrations (<30%), in the Scotia Sea (figure 1). To decipher the provenance of illite, we used the chemical (5/10Å ratio, i.e., Fe-Mg vs. Al rich illite) and crystallographic 424 properties (crystallinity) of this mineral. Data are compared with the 5/10Å ratio and the 425 crystallinity of illite of potential illite sources from the database of Petschick et al. (1996) 426 427 (figure 4). This comparison demonstrates that the illite from core MD07-3076Q is similar to 428 illite from the Weddell and Scotia seas while the chemical and crystallographic properties of 429 the illite recovered at site MD07-3076Q is not compatible with a Namibian source. In the 430 Scotia Sea, illite represents less than 30% of the total clay mineral fraction (from 10-30%; Petschick et al., 1996) and represents a mix of illite coming from the Weddell Sea embayment 431 432 via the AABW, and illite from the Pacific sector of Antarctica delivered by the CDW 433 (Diekmann et al., 2000; Walter et al., 2000; Petschick et al., 1996). Importantly, in the 434 Weddell Sea, illite represents at least 50% of the total clay mineral fraction (more than 80% in 435 some areas; Petschick et al., 1996). Consequently, the Weddell Sea embayment is considered 436 as the major, source of illite for our studied site.

437 Smectite has several potential provenances: the Angola Basin, southern South 438 America, Scotia Sea Islands, and Antarctic Peninsula. The crystallinity of this mineral was 439 measured in order to decipher its provenance. The well-crystallized smectite (low 440 crystallinity; figure 3) is compatible with a provenance from southern South America 441 (Pampas and Patagonia), Scotia Arc, or Antarctic Peninsula, which are all characterized by 442 crystallinity lower than 1.5 (Petschick et al., 1996; figure 3). It rules out a contribution of 443 poorly crystallized smectite (crystallinity > 2; Petschick et al., 1996) from Angola. In detail, 444 the crystallinity of the smectite from our studied site likely corresponds to a mix between two 445 types of sources: (1) Patagonia and Antarctic Peninsula with a crystallinity of 1.5; and (2)

Scotia Arc with a crystallinity lower than 1.0. Interestingly, the proportion and crystallinity of smectite record very similar variations that suggest a dominant and rather continuous flux of smectite from the Scotia Arc (low crystallinity -  $\sim$ 1.1) during the last glacial period, with a supplementary supply of less well crystallized smectite (slightly higher crystallinity – 1.5;  $\sim$ 20%) from Patagonia or Antarctic Peninsula during the deglaciation (supplementary figures S1 and S2)

In summary, both the proportions of the clay minerals and their crystallographic properties suggest that kaolinite mainly derives from low latitude areas, chlorite originates from Antarctica and the Scotia Sea, while illite mainly derives from the Weddell Sea embayment and smectite is derived from several sources (Scotia Arc, Patagonia, and Antarctic Peninsula). Moreover, mineralogical and crystallographic properties exclude Africa as a source of clay minerals consistent with the dominant westward oceanic circulation in the ACC (figure 1).



 $^{87}$ Sr/ $^{86}$ Sr- $\varepsilon_{Nd}$  diagram of MD07-3076Q sediments and of potential sources of Figure 6: 460 461 sediments for the South Atlantic Basin. Boxes representing the reference data delimit the first 462 and the third quartile. The median values are represented by horizontal and vertical lines attached to the boxes for the  $\varepsilon_{Nd}$  and the  ${}^{87}Sr/{}^{86}Sr$  ratios, respectively. The short strokes at the 463 end of these lines represent the  $10^{th}$  and  $90^{th}$  percentile values. Accordingly, the box represent 464 465 50% of values found for each potential provenance, and 80% is represented with the lines. Data sources: -South American Shield- Allegre et al. (1996); Basile et al. (1997); Walter et 466 al. (2000); de Mahiques (2008); Jeandel et al. (2007), and references therein; Mallmann et 467 468 al. (2007); Ganade de Araujo et al. (2014); Fuck et al. (2014); -Patagonia, Pampas, Southern 469 Andes- Stern et al. (1990); Grousset et al. (1992); Basile et al. (1997); Walter et al. (2000); Smith et al. (2003); Jeandel et al. (2007), and references therein; Khondoker et al. (2018); -470 471 Scotia Arc and Ridge- Jeandel et al. (2007); Harrison et al. (2003);-Antarctic Peninsula-

472 Hole el al. (1993); Kosler et al. (2009); Roy et al. (2007); Lee et al. (2005); -East Antarctica473 DePaolo et al. (1982); Luttinen et al. (1998, 2010); Roy et al. (2007); Will et al. (2010).

474 The coupled  $\varepsilon_{Nd}$  - <sup>87</sup>Sr/ <sup>86</sup>Sr isotope variation of the clay-size fraction (figure 6) reveal 475 a mixed provenance between young sources around the Scotia Sea (Scotia Arc, Antarctic 476 Peninsula, Patagonia) and southern South America (Patagonia and Pampas), and an old source 477 from East Antarctica or the South American Shield (i.e., Brazil and Parana River).



Figure 7: Pb isotope composition of core MD07-3076Q and of potential sources of
sediments. The reference data used for comparison are from Fretzdorff et al. (2002), -Scotia
Ridge-; Khondoker et al. (2018), -Patagonia-; Cohen and O'Nions (1982); Barreiro (1983), Scotia Arc-; Lee et al. (2005), Kosler et al. (2009), Flowerdew et al. (2012) -Antarctic
Peninsula-, de Mahiques et al. (2007), -South American Shield-; Flowerdew et al., (2012), Eastern Weddell Sea-.

# The coherent Pb isotope variations in figure 7 support this interpretation recording a mix between the Antarctic Peninsula, Scotia Arc, and South American Shield. However, since

487 very few data are available for the Pb isotope compositions of East Antarctica, a contribution 488 from this area cannot be excluded. The three isotopic systems suggest an increased 489 contribution of older material during the deglaciation compared with the LGM. Based solely 490 on Nd and Pb isotope data, it is rather difficult to distinguish South America and East 491 Antarctica as the old end-members during the deglaciation. However, the increase of the K/C 492 ratio during the deglaciation supports the hypothesis of enhanced relative contribution from 493 the South American Shield.

494

#### 2. Cohesive silt mode fraction (2-20 μm)

Both  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\epsilon_{Nd}$  of the cohesive silt mode fraction record limited variation 495 496 throughout the deglaciation (figure 5). The isotopic results indicate that the provenance is 497 located in Patagonia and in the Antarctic Peninsula, with an additional contribution of old 498 source (South American Shield and/or East Antarctica). These results suggest that no major 499 modification of provenance occurred during the deglaciation. Strontium and Nd isotope ratios 500 do not allow discrimination of the source of old material for the cohesive silt mode fraction. 501 The Nd isotope ratios suggest on average a higher radiogenic contribution for cohesive silts 502 compared with the clay-size fraction (figure 6).

503

#### 3. Sortable silt mode fraction ( $20-32/40 \mu m$ )

Both Sr and Nd isotope ratios of the sortable silt during the LGM record one dominant provenance: the Scotia Sea area (Patagonia, Scotia Arc, Antarctic Peninsula; figure 5). During the deglaciation, an important shift to less radiogenic  $\varepsilon_{Nd}$  values (4 units) is observed implying an increased contribution of an older end-member such as East Antarctica and/or the South American Shield. However, because East Antarctica and the South American shield have similar Sr and Nd isotope composition, it is not possible to distinguish them with these isotopes (figure 6). Consequently, investigating the main transportation patterns is necessary 511 to identify the source of old material for both the cohesive (2-20  $\mu$ m) and the sortable silt 512 mode fractions (20-32/40  $\mu$ m).

513 In summary, (figure 6) all three grain-size fractions compositions are located between 514 two end-members: the Scotia Sea-southern South America area (Antarctic Peninsula, Scotia 515 Arc and Ridge, Patagonia, Pampas, Southern Andes), and crustal shields (East Antarctica, 516 South American Shield). The clay-size fraction appears the most crustal-like, whereas the 517 sortable silt mode presents the most mantle-like composition. The 2-20 µm grain-size fraction presents the lowest <sup>87</sup>Sr/<sup>86</sup>Sr, meaning that the difference with the other grain size fraction is 518 519 related to distinct provenances rather than grain size effect. Both the clay size and the sortable 520 silt mode fractions display an increase of the crustal end-member. In the clay size fraction, 521 clay mineralogy enabled the identification of this old crustal source (i.e., the South American Shield). 522

523

#### B. Transportation pattern and palaeoclimatic implications

The provenances of the sediments for the three distinct grain-size fractions have been identified (or partially identified) in the previous section using their mineralogical and geochemical characteristics. Now, we will focus on identifying their transportation patterns and vectors from the source to the sedimentation site.

Several studies propose that aeolian processes may lead to significant particle transfers from Patagonia into the central Atlantic sector of the Southern Ocean (Hegner et al., 2007; Desiage et al., 2018). The aeolian contribution of loess from Patagonia was enhanced during the LGM as far as Antarctica (Petit et al., 1990; Grousset et al., 1992; Diekmann, 2007; Lambert et al., 2008). Although this hypothesis could be supported by the radiogenic Nd signal observed in both the clay-size and the sortable silt mode fraction during the LGM, the mineralogical of MD07-3076Q sediments is not in agreement with an enhanced contribution from Patagonia. The dominant clay mineral in Patagonia is smectite (up to 95%), and the contribution of Patagonian smectite in our clay-size fraction is lowest during the LGM (figure 3). Consequently, direct aeolian inputs cannot be significant at our site, and oceanic currents appear to be the main contributors to sedimentation (Moriarty, 1977; Petschick et al., 1996; Kuhn and Diekmann, 2002). This means that any aeolian particles from Patagonia delivered to the southwest Atlantic (Mathias et al., 2014) likely reach our site via oceanic transport processes.

542 The size of particles transported by water flow is directly related to the flow energy, 543 with size increasing with the flow speed. This property was used in previous studies to track 544 the temporal evolution of paleo-current flow (e.g. McCave and Hall, 2006; McCave and 545 Andrews, 2019). Below we show that the different grain-size modes not only correspond to 546 distinct flow speeds, but specific water mass at site MD07-3076Q. Consequently, grain-size 547 modes (2-20 µm and 20-32/40 µm) can be associated to specific deep-water-masses.

548 The clay-size fraction (i.e., the  $< 2 \mu m$  particles) with very high specific surface 549 readily floats and hence can undergo long-range transportation (>500 km). Consequently, 550 particles within this grain-size fraction can be used to record the relative contribution of the 551 main deep-water masses.

552

#### 1. Clay-size fraction: the distant terrigenous signal

The NADW carries fine suspended particles from as far as the Labrador Sea. However, this signal is probably overprinted by the supply of sediments from more proximal sources such as Africa or South America. The western branch of the NADW (NADW-W; i.e., Deep Western Boundary Current) can transport sediments from Brazil, Parana River, Pampas, and Patagonia (figure 1). Both mineralogical and geochemical compositions of the particles transported by the western branch of the NADW are likely controlled by the southern extension of the water-mass. Currently the western branch drifts away from the South

560 American coast around 45°S. This southern extension likely changed during the last glacial 561 period and deglaciation (Skinner et al., 2010, 2014; Vasquez-Riveiros et al., 2010; Waelbroeck et al., 2011; Gottschalk et al., 2015a,b; Howe et al., 2016). Depending on its 562 563 southern extent, the NADW is thus able to carry either mostly kaolinite (equatorial Brazil), or 564 kaolinite and smectite (southern Brazil), or illite (Parana River basin), or well-crystallized 565 smectite (crystallinity ~1.5; Patagonia). As shown above, the  $\varepsilon_{Nd}$  signature of these sediments 566 is more radiogenic when the NADW extends further south (-20 in equatorial Brazil, to ~0 in 567 Patagonia) while the Pb and Sr yield a slightly less radiogenic signature (figures 6 and 7).

568 The CDW transports sediments from the Scotia Sea area, including Scotia Arc, 569 Patagonia, and the Antarctic Peninsula, as well as from the Weddell Sea when mixing occurs 570 between the lower part of the LCDW and AABW. Then particles transported by the CDW likely record a younger isotopic signature (i.e., more radiogenic  $\varepsilon_{Nd}$  and unradiogenic Sr and 571 572 Pb isotopes), and a mineralogical composition dominated by smectite and chlorite, while 573 additional contribution of old material (unradiogenic  $\varepsilon_{Nd}$  and radiogenic Sr and Pb isotopes) 574 and illite is possible when the LCDW and AABW mix together. In summary, illite is 575 considered as injected into the sedimentary system via the AABW through mixing processes 576 between LCDW and AABW while smectite and chlorite are likely transported by the CDW.

577 The AABW in the Atlantic sector mainly forms in the Weddell Sea embayment (Orsi 578 et al., 1999), and thus, must carries sediments from its drainage basin (East Antarctica, 579 Antarctic Peninsula), but may also transport sediment winnowed on its pathway such as the 580 Scotia Arc or southern Patagonia in the Argentine Basin. The main clay minerals in the 581 Weddell Sea embayment are illite associated with chlorite. Smectite may be injected in the 582 AABW at the vicinity of the Scotia Arc. Kaolinite is almost absent in both the Weddell Sea 583 embayment and the Scotia Sea area (Patagonia, Antarctic Peninsula and Scotia Arc) and is unlikely to be transported by the AABW. The isotopic composition of fine particles 584

transported by the AABW ranges between two end-members: the very old sediments from East Antarctica ( ${}^{87}$ Sr/ ${}^{86}$ Sr > 0.72;  $\epsilon_{Nd}$  < -20), and younger material from the Antarctic Peninsula and the Scotia Arc ( ${}^{87}$ Sr/ ${}^{86}$ Sr ~ 0.70;  $\epsilon_{Nd}$  > -5; intermediate Pb isotope ratios). Consequently, the composition of particles transported by the AABW flowing out of the Weddell Sea is intermediate, with  $\epsilon_{Nd}$  around -9 (Walter et al., 2000), and is potentially enriched in young material when entering the Scotia Sea.

The above observations are summarized in table 5, in which specific isotopic and mineralogical signatures are associated to each deep water mass. It is clear that the K/C ratio as well as the  $\varepsilon_{Nd}$  composition reflect northern- versus southern-born water mass contributions (i.e., NADW versus CDW and AABW). Similarly, smectite and illite are transported by distinct water-masses and the illite/smectite ratio (I/S) can efficiently distinguish the AABW from the other water-masses. Thus, this ratio will further be used as a tool for estimating the AABW contribution through time.

598 The illite/smectite and kaolinite/smectite ratios have been plotted in figure 8. The three 599 end-members for AABW, CDW and NADW are plotted according to the clay composition of 600 their suspended loads. Figure 8 highlights how the relative contribution of the three water 601 masses (i.e., AABW, CDW, and NADW) to clay deposition has changed with time since the 602 LGM. To interpret this figure it is important to keep in mind that changes observed in the 603 contribution one deep water mass are relative compared to the other water masses. To 604 construct figure 8.b, the proportion of clay particles deposited by each deep water mass was 605 calculated as described below.

End-member deep water mass compositions were calculated from the average composition of deep water-masses shown in figure 8.a. Weights are computed as the inverse of the distance between the position of water-mass end-member  $(x_w;y_w)$  with  $x_w=K/C_w$  and

 $y_w = I/S_w$  and the position of the data point  $(x_d; y_d)$ , with  $x_d = K/C_d$  and  $y_d = I/S_d$  taken from the 609 graph (figure 8.a) for each water-mass:  $W_w = 1/[(x_d - x_w)^2 + (y_d - y_w)^2]^{1/2}$ . The position used for 610 611 the deep water masses are:  $xA_{ABW} = K/C_{AABW} = 0.3$ ;  $y_{AABW} = I/S_{AABW} = 2.5$ ;  $x_{CDW} = K/C_{CDW}$ 612 = 0.1;  $y_{CDW} = I/S_{CDW} = 1$ ;  $x_{NADW} = K/C_{NADW} = 0.9$ ;  $y_{NADW} = I/S_{NADW} = 1$ . The values used 613 for the NADW correspond to the mineralogical composition of the NADW during the LGM. 614 As a result, the contribution of NADW to the deposition of clay particles is underestimated 615  $(\sim 10\%)$  at the end of the deglaciation while the contribution of the CDW is overestimated 616 (~10%). This choice does not have a strong impact on the contribution of the AABW (less 617 than 4.5%). Second, the percentage of the contribution of each water mass is calculated by bringing the sum of the weights computed to 100: [NADW's contribution] =  $W_{NADW}/(W_{NADW})$ 618 619  $+ W_{CDW} + W_{AABW} x 100$ 

620 To test the validity of this method to reconstruct the evolution of the past deep 621 circulation, we compared our results with those of other water-masses proxies (e.g. seawater  $\varepsilon_{Nd}$  and  $\delta^{13}$ C). However, these latter methods proved incapable of distinguishing AABW from 622 CDW. Consequently, we based the comparison on the proportion of the NADW compared to 623 the southern sourced deep waters. Based on data from the same core, our estimates give 624 625 comparable results to these two proxies (see table 6). This suggests that our approach 626 successfully reconstructs the evolution of these three deep water masses. However, our 627 estimate of the proportion of the NADW is higher than the estimate of Howe et al. (2016) based on  $\varepsilon_{Nd}$ . This discrepancy can be explained by two factors. First, we were unable to 628 629 make corrections for concentration differences of particles in the three deep water masses 630 (i.e., quantity of clay particles per volume of water) due to the large uncertainties involved. In 631 addition, it is very likely that the NADW is more concentrated in clay particles than the AABW and CDW due to sediment source effect (e.g., dry East Antarctica below thick ice 632 633 sheets vs. humid South America highly drained by equatorial rivers). Consequently, even 634 though our tool successfully reconstruct the variations of the NADW, the contribution of 635 NADW is probably overestimated. In other words, our estimate of NADW contribution to the 636 clay deposition is likely higher than the proportion of NADW (in volume of water). Second, 637 the choice of the end member for the seawater  $\varepsilon_{Nd}$  of AABW+CDW made by Howe et al. 638 (2016) is debatable (-5.5 during the LGM, -8.5 during the Holocene). Skinner et al. (2013) 639 used a compositional end-member between -5 and -2, which yields a better match with the 640 isotopic composition of seawater Nd at our site (-5.5, Skinner et al., 2013). Therefore, the 641 estimate of Howe et al. (2016) appears to underestimate the contribution of NADW at our 642 site.

643 From 25 to 20 ka, the CDW is clearly dominant, with a contribution of 50% on 644 average, while the NADW and AABW deliver 35% and 15% of the clay fraction, 645 respectively. The highest contribution of the AABW is observed at the end of the LGM 646 between 20 and 17.5 ka (27% in average, but up to 52% at 18 ka). During this interval, the 647 NADW and CDW deliver respectively 34% and 39% on average. At the end of this period, 648 the mineralogical composition of the fine sediments deposited at site MD07-3076Q suggests 649 that the AABW was dominant. At the beginning of the deglaciation, between 17.5 and 15 ka 650 (i.e., during HS 1), the clay mineral composition of core MD07-3076Q indicates a decreased 651 relative contribution of the AABW end-member (15% on average) associated to an increase 652 of the relative contribution of the CDW (49%) end-member. This observation of an increase 653 of the CDW contribution is supported by the radiogenic  $\varepsilon_{Nd}$  of the clay-size fraction and a  $^{87}$ Sr/ $^{86}$ Sr close to the Scotia Arc composition (figures 5 and 6). 654



Figure 8: (a) Plot of kaolinite/chlorite vs illite/smectite ratios from core MD07-3076Q
from 25 to 10 ka. The composition of deep-water masses has been determined using the data
from Petschick et al. (1996). (b) Ternary diagram comparing the contribution of the three
deep water masses to the clay deposition at site MD07-3076Q.

660 From 15 ka on, the clay mineral composition at the study site records an increasing 661 contribution of the NADW until the end of the deglaciation (at least 47% for the interval 10-662 12 ka, figure 8). Compared with the LGM this suggests enhanced contribution of the NADW 663 with reduced contributions from the CDW (40% for the interval 10-12 ka) and AABW (13% 664 for the interval 10-12 ka). These results are strongly supported by the sharp decrease in  $\varepsilon_{Nd}$ and by the concomitant increase of <sup>208</sup>Pb/<sup>204</sup>Pb and <sup>207</sup>Pb/<sup>204</sup>Pb ratios from the clay size 665 666 fraction (figures 5 and 7). This corroborates the conclusions based on the *cibicides*  $\delta^{13}$ C 667 measured in the same core (figure 9.e, Waelbroeck et al., 2011). In addition, increasing inputs 668 of less crystallized smectite (slightly higher crystallinity: ~1.5, supplementary figure S1) from 669 Patagonia support the southward penetration of the NADW in the southern Atlantic Ocean, 670 consistent with an increasing contribution of the NADW-W to sedimentation at site MD07-671 3076Q. Interestingly, the transition between B/A (that starts simultaneously with the southern 672 Hemisphere cooling, the Antarctic Cold Reversal) and the YD appears to have no effect on 673 the relative contribution of deep water masses and shows a continuous increase of the relative 674 contribution of the NADW during these two periods.

Overall, the clay size fraction demonstrates that the contribution of the three deep water-masses changed through time. An increase of contribution of the AABW is recorded at the end of the LGM and is followed by a two-step deglaciation. First, an important reduction of the AABW is observed at the HS 1 onset. Second, an increase in the NADW contribution compared to the CDW is observed during the B/A and YD.

#### 2. Cohesive silt mode fraction (2-20 μm): an AABW signal

681 According to our geochemical data, the provenance of the cohesive silt fraction is 682 dominated by sources from the Scotia Sea area, especially Patagonia and Antarctic Peninsula, 683 and by an old, unidentified, crustal source. According to the modern deep oceanic circulation, 684 only the CDW and AABW are able to transport sediment from the Scotia Sea to the studied site (figure 1). It was also likely the case during the LGM and deglaciation. The AABW and 685 686 CDW were likely spreading further north while the NADW was likely shallower, and 687 retreated to the north (Kuhn and Diekmann, 2002; Barker et al., 2010; Waelbroeck et al., 688 2011). These modifications do not affect the continental sources of terrigeneous sediment for 689 the CDW and AABW compared the present day, and the NADW is still unable to transport 690 sediments from the Scotia Sea. Consequently, it excludes the NADW as a transport medium 691 for the cohesive silt mode fraction to site MD07-3076Q. Therefore, because the NADW is 692 also the only water-mass able to transport sediment from the South American Shield to our 693 study site, it excludes the South American Shield as a source of sediment for the cohesive silt 694 mode fraction, and implies East Antarctica as the source of old material identified in the cohesive silt mode fraction. The terrigenous supplies deriving from East Antarctica/Scotia Sea 695 696 strongly suggest the AABW has been the dominant carrier of the 2-20 µm grain-size fraction 697 since the LGM. In addition, because the mean isotopic composition of this grain-size fraction 698 transported by the AABW is relatively constant through time (figure 6), the relative contributions of East Antarctica and Scotia Sea area seem to have remained largely 699 700 unchanged since the LGM.

701

#### 3. Sortable silt mode fraction (20-32/40 μm): A LCDW signal

A part of the particles transported by the AABW and the NADW are derived from old crustal sources (i.e., East Antarctica and South American Shield, respectively). Thence, these two deep-water masses cannot be the source of the sortable silt mode fraction during the 705 LGM. In contrast, the CDW is a good candidate to transport particles solely from the Scotia 706 Sea, as identified for the sortable silt mode fraction during the LGM. Consequently, the only 707 possible source of this grain size fraction is the CDW. During the deglaciation, the Nd isotope 708 composition of the sortable silt mode fraction became gradually less radiogenic while the <sup>87</sup>Sr/<sup>86</sup>Sr ratio fluctuated, suggesting a modification of provenances and/or an additional 709 710 contribution of less radiogenic Nd source. Such isotopic changes could be explained in two 711 ways; (1) a change in the CDW pathway; (2) the incorporation of terrigenous material by 712 mixing with other water-masses (such as AABW or NADW). The CDW slowed down and 713 migrated southward during the deglaciation in concert with the southward migration of the 714 subpolar and polar fronts (Howard and Prell, 1992; Anderson et al., 2009; Denton et al., 715 2010). However, such a change in the CDW pathway would have no effect on the terrigenous 716 sources of particles. In contrast, several studies proposed increased ventilation in the Southern Ocean, enabling the degassing of oceanic CO<sub>2</sub> to the atmosphere (Ahn and Brook, 2008; 717 718 Fischer et al., 2010; Skinner et al., 2010, 2013, 2014; Siani et al., 2013). Currently the base of 719 the LCDW is mixed with the AABW, which was apparently not the case during the LGM 720 (Skinner et al., 2010). The onset of AABW/LCDM mixing could explain the incorporation of 721 unradiogenic terrigenous material from the Weddell Sea embayment (i.e., East Antarctica) into the LCDW. This mechanism likely explains the shift in  $\varepsilon_{Nd}$  composition during the 722 723 deglaciation. This hypothesis is supported by the results of Walter et al. (2000) who reported 724 lower  $\varepsilon_{Nd}$  in terrigenous sediments in the southern Scotia Sea during the interglacial period 725 and interpreted this as higher contribution of sediment from the Weddell Sea embayment due 726 to the mixing of the AABW into the ACC. From 15 ka, the Nd isotope composition of the 727 sortable silt mode fraction is very close to the one from the cohesive silt mode fraction 728 transported by the AABW. It suggests that the sortable silt mode fraction is specifically 729 transported by the LCDW rather than by the entire CDW. This highlights the fact that the  $\varepsilon_{Nd}$ 

- variations from the sortable silt mode fraction provide a useful tool to study the mixing
- 731 between the AABW and LCDW.





733 Figure 9: Comparison between paleoceanographic proxies from core MD07-3076Q and 734 data produced in this study. Grey areas delimit the main climatic events and the red band 735 highlights the particularly strong AABW event. (a) size of the sortable silt mode (µm), a proxy 736 of the speed of the LCDW; (b) size of the cohesive silt mode (µm), a proxy of the speed of the 737 AABW; (c) bottom water ventilation age from site MD07-3076Q (data from Skinner et al., 2010); (d)  $\varepsilon_{Nd}$  from the sortable silt size fraction; (e) Cibicides  $\delta^{13}C$  from site MD07-3076Q 738 (Waelbroeck et al., 2011); (f)  $\varepsilon_{Nd}$  from the cohesive silt size fraction; (g) seawater  $\varepsilon_{Nd}$  from 739 740 site MD07-3076Q (Skinner et al., 2013); (h) proportion of clay particles delivered by the 741 *NADW* (see figure 8.b); (i) proportion of clay particles delivered by the CDW (see figure 8.b); 742 (*j*) proportion of clay particles delivered by the AABW (see figure 8.b). On curves (h), (i) and 743 (j), the red line represent values calculated using the Holocene end-member of the NADW (I/S 744 = 0.5), while the blue line correspond to the values calculated using the LGM end-member of 745 the NADW (I/S = 1).

746

#### 4. Paleoceanographic implication

747 Our results establish that different grain-size modes can be attributed to distinct water-748 masses and that variation of the particle size of the different grain-size modes (figure 2) can 749 be used to record the variations of flow speed of these water-masses as well as changes in 750 pathways/mixing. This method has the advantage of distinguishing the speed of each water mass instead of giving the average speed of all the water masses, as it is the case for the 751 752 sortable silt method (McCave and Hall, 2006). During the last glacial period, the large mean 753 particle diameter (>30 µm) of the sortable silt mode fraction reflects strong LCDW activity. 754 At the beginning of the deglaciation, the variation of the mean particle size suggests that the 755 LCDW speed slightly decreased, but remained high. Since ~15 ka, the LCDW speed has been 756 showing a gradual decrease. McCave et al. (2014) observed minor change in the Antarctic 757 Circumpolar Current (which includes the CDW) flow speed at the Drake Passage during the deglaciation. Our observation, however, is consistent with other studies in the south Indian sector of the Southern Ocean indicating that the Antarctic Circumpolar Current (which includes the CDW) flow speed was maximum during the LGM and decreased during the deglaciation (Mazaud et al., 2010), with a notable increase from ~15 ka on (Mazaud et al., in prep.). Altogether, these studies and our results suggest that the weakening of the ACC started at ~15 ka.

764 From ~26 to 21 ka, the low particle sizes recorded in the cohesive silt mode (figure 765 2.b, transported by the AABW) indicate slow AABW, which is consistent with the observed 766 moderate relative contribution of AABW in clay particles (figure 8 and 9). In this time 767 interval, one isolated high value is recorded at 24 ka and suggests that, temporarily, AABW 768 activity was stronger. However, no change is registered in the clay minerals (figure 3 and 9). 769 The grain-size data indicate that the AABW became more vigorous from 21 ka to reach its 770 strongest speed at the end of the LGM. This maximum speed is concomitant with the 771 strongest relative contribution of the AABW of the clay record (figures 3.h and 9), as shown 772 by the peak of the illite/smectite ratio (figure 3) illustrated on figure 8. Interestingly, this 773 AABW maximum preceded the HS 1, and both its speed and relative contribution fell at the 774 LGM/HS1 transition as defined by Missiaen et al. (2019). At the beginning of the 775 deglaciation, the abrupt decrease of the fine grain-size mode indicates a slowdown of the 776 AABW (figure 2). This is supported by a sharp decrease in the illite/smectite ratio in the clay-777 size fraction (figure 3), which also suggests an abrupt decrease of the relative contribution of 778 the AABW. At ~16 ka, a slowdown of the AABW is indicated by a second sharp decrease of 779 the cohesive silt mode (figure 2), which reaches its lowest values of the entire record, 780 indicating low AABW speed from 16 to 11.5 ka, when the AABW activity increased 781 suddenly and recovered the speed observed before 16 ka.

These variations of the AABW speed could indicate the transition from the Ice Shelf 782 783 Mode dominated bottom water formation at the end of the LGM to the Polynyas Mode 784 dominated bottom water formation during interglacial periods. The variations of the cohesive 785 silt mode support a strong AABW during the LGM and the early Holocene, and a minimum 786 flow during the deglaciation. This minimum flow would result from the reduction of bottom 787 water production via the Ice Shelf Mode and still low production via the Polynyas Mode. Our 788 results confirm the results of Krueger et al. (2012) who demonstrated the relation between the 789 flow speed changes in the Cape Basin and the higher AABW production rates and indicate 790 that the size of the cohesive silt mode is a reliable tracer of the AABW flow speed and hence 791 of AABW production rates in the Weddell Sea.

792 Overall, the variations of the AABW flow speed and ventilation age are similar, with 793 higher average speeds corresponding to the old ventilation ages. Taken together with the 794 reconstructed vigorous LCDW during the LGM (figure 9.a.), these observations suggest that 795 the deep Southern Ocean was poorly ventilated but with a vigorous circulation. It means that 796 both the AABW and LCDW remained isolated from the surface during the LGM. This 797 conclusion is consistent with our hypothesis that AABW was formed under ice shelves during 798 this period and that the major expansion of sea ice (Bianchi and Gersonde, 2004; Bostock et 799 al., 2012; Adkins 2013) kept the AABW isolated from the atmosphere during the LGM. In 800 addition, poorly ventilated deep Southern Ocean associated with a vigorous deep circulation 801 (i.e., high AABW and LCDW flow) suggests that a barrier existed between the deep ocean 802 and the surface, which is consistent with the study of Howe et al. (2016). In contrast, direct 803 comparison highlights the fact that the peaks of ventilation age and of the AABW flow are not 804 in phase. Indeed, the peaks of the AABW flow (figure 9.b) and of the AABW contribution to 805 clay deposition (figure 9.j) are synchronous with the first decrease in deep water ventilation 806 ages after the LGM and prior to HS1. Moreover, this well-defined change in the AABW

behavior is recorded by two distinct proxies (i.e., clay minerals and grain size distribution) 807 808 and in two distinct grain size fractions (clavs and silts). These synchronous changes are 809 associated with a drop in ventilation age prior to the HS 1. The variations in two grain size 810 fractions that are transported and deposited by distinct dynamic processes indicates a major 811 perturbation in Southern Ocean circulation. Interestingly, the increase in the AABW strength 812 associated to the early decrease in deep water ventilation age took place significantly earlier 813 than the changes recorded by the other proxies measured in core MD07-3076Q (i.e., sea water  $\varepsilon_{Nd}$  between 17.5 and 16 ka,  $\delta^{13}C$  at 14.5 ka,  $\delta^{18}O$  at 15 ka, LCDW flow speed at 15 ka, 814 815 NADW proportion at 15 ka) and also precedes the atmospheric CO<sub>2</sub> rise (Bereiter et al., 2015) 816 and the change in the opal flux (Anderson et al., 2009). For the first time, these data indicate a 817 change in the deep Southern Ocean circulation that is associated with ventilation change that 818 precedes the HS 1. This suggest that the identified large temporary (~1 kyr long) increase in 819 the AABW contribution and flow speed may have had a substantial impact on the deep 820 Southern Ocean ventilation, and by extension, on the deglacial CO<sub>2</sub> rise. Indeed, the 821 magnitude of the change in the AABW behavior has the potential to "break" the hypothetical 822 physical barrier between the deep Southern Ocean and the surface that was responsible of 823 poorly ventilated deep water during the LGM. This combination of events also explains the 824 unexpected early fall of the deep water ventilation age observed at ~18.2 ka.

#### 825 Conclusion

The mineralogical characteristics of the clay-size fraction provide new insights into the general thermohaline circulation in the southern South Atlantic. The mineralogical signal demonstrates strong influence of both the CDW and AABW during the last glacial period. Clay assemblages reveal a particularly high contribution of the AABW at the end of the LGM, which is interpreted as reflecting strong Ice Shelf Mode bottom water production in the Weddell Sea. The contribution of the CDW and AABW then progressively decreases during the deglaciation together with the growing influence of the NADW (figure 8). Indeed, clay size fraction data monitor the progressive incursion of NADW into the Southern Ocean during the deglaciation from ~15 ka (figure 8), as well as the synchronous southward retreat of the southern-born deep water-masses (i.e., CDW and AABW).

836 The isotopic compositions of the distinct grain size modes provides invaluable 837 information. The sortable silt mode fraction helps to trace the LCDW activity whereas the 838 cohesive silt mode fraction is associated with the AABW signal. Consequently, the temporal 839 evolution of flow speeds, pathways and interactions of these deep water-masses can be 840 determined in a very robust way. This study suggests: (1) vigorous LCDW during the LGM 841 and a marked weakening from ~15 ka; (2) strong AABW at the end of the LGM and during 842 the early Holocene, and weak AABW during the deglaciation between 16 and 11.5 ka. These 843 results suggest a decrease of AABW production rates during the deglaciation that could result 844 from the transition from strong Ice Shelf Mode bottom water production during the LGM to 845 stronger Polynyas Mode bottom water during the Holocene.

The strong LCDW speed observed during the LGM is associated with increased clay deposition by the CDW, and the reduction of LCDW speed since the B/A is accompanied by an increased clay contribution from the NADW. Similarly, the highest AABW speed recorded at the end of the LGM is concomitant with the highest relative contribution of the AABW to clay mineral deposition. Interestingly, this AABW maximum preceded the HS 1 and both its speed and contribution fell markedly at the LGM/HS 1 transition.

Finally, this study demonstrates that based on (1) the combination of isotopic and mineral investigation of the clay size fraction, (2) the combination of isotopic measurement of isolated grain size modes and of grain size distribution results, and (3) a complete synthesis of the potential sources of sediments, it is possible to obtain reliable information on the lithogenic material transported by each deep water mass. This approach allows efficient tracking of the evolution of distinct water-masses through time, since it helps to isolate the specific signal associated to each water-mass. Based on these observations, we conclude that the increase in the AABW strength at the end of the LGM may have had a substantial impact on the ventilation recovery of the deep Southern Ocean, and by extension, to the deglacial  $CO_2$  rise.

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### 1369 Supplementary figures



1371 SUPPLEMENTARY FIGURE S1: Crystallinity of smectite from site MD07-3076Q
1372 (black curve) and comparison with crystallinity of smectite from potential sources to the
1373 South Atlantic basin. The data used for comparison are from Petschick et al., 1996.



1375 SUPPLEMENTARY FIGURE S2: Contribution of the two types of smectite from site 1376 MD07-3076Q. The black line represent the total smectite content, the purple area represent 1377 the smectite content from the Scotia Arc, and the red area represent the smectite content from 1378 Patagonia. The proportion of the two types of smectite are calculated as followed:

1379

[%Patagonian smectite] = [%total smectite] ×  $\frac{(\text{sample crystallinity}] - (\text{Scotia Sea crystallinity}]}{(Patagonia crystallinity] - (\text{Scotia Sea crystallinity}]} ×100$ [%Scotia Sea smectite] = [%total smectite] - (%Patagonian smectite]



SUPPLEMENTARY FIGURE S3: Sedimentation rate from site MD07-3076Q
calculated from the age model published in Skinner et al. (2010, 2013, 2014) and Gottschalk
et al. (2015b).

# 1383 Tables

	average (10-30 ka)	LGM (25-17 ka)	Deglaciation (17-11.7 ka)	standard deviation (10-30 ka)
fine mode (μm)	6.8	7.2	6.3	0.8
coarse mode (μm)	30.4	31.3	30.4	2.5
clay fraction (%)	3.4	3.0	3.8	0.7
sortable silts (10-63 $\mu$ m;				
%)	47.0	47.2	46.2	2.4
cohesive silts (2-10 μm;				
%)	39.4	38.8	40.2	2.4
sand (%)	3.0	3.5	2.6	1.0
smectite (%)	30.7	26.7	35.7	7.9
illite (%)	33.5	34.7	31.7	3.3
chlorite (%)	25.1	27.2	22.3	4.2
kaolinite (%)	10.6	11.4	10.3	1.3
I/S	1.2	1.3	0.9	0.3
K/C	0.4	0.4	0.5	0.1
ε <sub>νd</sub> (<2 μm)	-5.8	-5.0	-6.0	0.8
$\epsilon_{Nd}$ (fine mode)	-4.6	-4.6	-4.7	0.4
$\epsilon_{Nd}$ (coase mode)	-3.3	-1.8	-3.7	1.2
<sup>87</sup> Sr/ <sup>86</sup> Sr (<2 μm)	0.71130	0.71144	0.71136	0.00036
<sup>87</sup> Sr/ <sup>86</sup> Sr (fine mode)	0.70795	0.70800	0.70798	0.00019
<sup>87</sup> Sr/ <sup>86</sup> Sr (coase mode)	0.70889	0.70859	0.70926	0.00058
<sup>208</sup> Pb/ <sup>204</sup> Pb (<2 μm)	38.774	38.775	38.785	0.012
<sup>207</sup> Pb/ <sup>204</sup> Pb (<2 μm)	15.644	15.639	15.645	0.008
<sup>206</sup> Pb/ <sup>204</sup> Pb (<2 μm)	18.752	18.749	18.758	0.030

1384

1385 TABLE 1: Average values of the different proxies for the entire record, the LGM, and

1386 the deglaciation.