Hydrological variations of the intermediate water masses of 1 the western Mediterranean Sea during the past 20 ka inferred 2 from neodymium isotopic composition in foraminifera and 3 cold-water corals 4

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29 Abstract. We present the neodymium isotopic composition (ɛNd) of mixed planktonic foraminifera species from 30 a sediment core collected at 622 m water depth in the Balearic Sea, as well as ENd of scleractinian cold-water 31 corals (CWC; Madrepora oculata, Lophelia pertusa) retrieved at 280-414 m water depth in the Alboran Sea and 32 the south Sardinian continental margin. The aim is to constrain hydrological variations at intermediate depths in 33 the western Mediterranean Sea during the last 20 kyr. Planktonic (Globigerina bulloides) and benthic 34 (*Cibicidoides pachyderma*) for a minifera were also analyzed for stable oxygen (δ^{18} O) and carbon (δ^{13} C) isotopes. 35 The foraminiferal and coral ENd values from the Balearic and Alboran Sea are comparable over the last ~13 kyr, 36 with mean values of -8.94 ± 0.26 (1 σ ; n=24) and -8.91 ± 0.18 (1 σ ; n=25), respectively. Before 13 ka BP, the 37 foraminiferal ENd values are slightly lower (-9.28±0.15) and tend to reflect a higher mixing between 38 intermediate and deep waters, which is characterized by more unradiogenic ENd values. The slight ENd increase after 13 ka BP is associated to a decoupling in the benthic foraminiferal δ^{13} C composition between intermediate 39 40 and deeper depths, which started at ~16 ka BP. This suggests an earlier stratification of the water masses and a

- 41 subsequent reduced contribution of unradiogenic ENd from deep waters. The CWC from the Sardinia Channel
- 42 show a much larger scattering of ε Nd values, from -8.66 \pm 0.30 to -5.99 \pm 0.50, and a lower average (-7.31 \pm 0.73;
- 43 n=19) compared to the CWC and foraminifera from the Alboran and Balearic Sea, indicative of intermediate
- 44 waters sourced from the Levantine basin. At the time of sapropel S1 deposition (10.2 to 6.4 ka), the ϵ Nd values
- 45 of the Sardinian CWC become more unradiogenic (-8.38±0.47; n=3 at ~8.7 ka BP), suggesting a significant
- 46 contribution of intermediate waters originated from the western basin. Accordingly, we propose that western

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47 Mediterranean intermediate waters replaced the Levantine Intermediate Water (LIW), which was strongly
48 reduced during the mid-sapropel (~8.7 ka BP). This observation supports a notable change of Mediterranean
49 circulation pattern centered on sapropel S1 that needs further investigations to be confirmed.

50 51

1. Introduction

52 The Mediterranean Sea is a mid-latitude semi-enclosed basin, characterized by evaporation exceeding 53 precipitation and river runoff, where the inflow of fresh and relatively warm surface Atlantic water is 54 transformed into saltier and cooler (i.e. denser) intermediate and deep waters. Several studies have demonstrated 55 that the Mediterranean thermohaline circulation was highly sensitive to both the rapid climatic changes 56 propagated into the basin from high latitudes of the Northern Hemisphere (Cacho et al., 1999, 2000, 2002; 57 Moreno et al., 2002, 2005; Paterne et al., 1999; Martrat et al., 2004; Sierro et al., 2005; Frigola et al., 2007, 58 2008) and orbitally-forced modifications of the eastern Mediterranean freshwater budget mainly driven by 59 monsoonal river runoff from the south (Rohling et al., 2002; 2004; Bahr et al., 2015). A link between the 60 intensification of the Mediterranean Outflow Water (MOW) and the intensity of the Atlantic Meridional 61 Overturning Circulation (AMOC) was proposed (Cacho et al., 1999, 2000, 2001; Bigg and Wadley, 2001; Sierro 62 et al., 2005; Voelker et al., 2006) and recently supported by new geochemical data in sediments of the Gulf of 63 Cádiz (Bahr et al., 2015). In particular, it has been suggested that the intensity of the MOW and, more generally, 64 the variations of the thermohaline circulation of the Mediterranean Sea could play a significant role in triggering 65 a switch from a weakened to an enhanced state of the AMOC through the injection of saline Mediterranean 66 waters in the intermediate North Atlantic at times of weak AMOC (Rogerson et al., 2006; Voelker et al., 2006; 67 Khélifi et al., 2009). The Mediterranean intermediate waters, notably the Levantine Intermediate Water (LIW), 68 which represent today up to 80 % in volume of the MOW (Kinder and Parilla, 1987) are considered an important 69 driver of MOW-derived salt into the North Atlantic. Furthermore, the LIW also plays a key role in controlling 70 the deep-sea ventilation of the Mediterranean basin, being strongly involved in the formation of deep waters in 71 the Aegean Sea, Adriatic Sea, Tyrrhenian Sea and Gulf of Lions (Millot and Taupier-Letage, 2005). It is 72 hypothesized that a reduction of intermediate and deep-water formation as a consequence of surface hydrological 73 changes in the eastern Mediterranean basin acted as a precondition for the sapropel S1 deposition by limiting the 74 oxygen supply to the bottom waters (De Lange et al., 2008; Rohling et al., 2015; Tachikawa et al., 2015). 75 Therefore, it is crucial to gain a more complete understanding of the variability of the Mediterranean 76 intermediate circulation in the past and its impact on the MOW outflow and, in general, on the Mediterranean 77 thermohaline circulation.

78 Previous studies have mainly focused on the glacial variability of the deep-water circulation in the western

79 Mediterranean basin (Cacho et al., 2000, 2006; Sierro et al., 2005; Frigola et al., 2007, 2008). During the Last

80 Glacial Maximum (LGM), strong deep-water convection took place in the Gulf of Lions, producing cold, well-

81 ventilated western Mediterranean Deep Water (WMDW) (Cacho et al., 2000, 2006; Sierro et al., 2005), while

82 the MOW flowed at greater depth in the Gulf of Cádiz (Rogerson et al., 2005; Schönfeld and Zahn, 2000). With

83 the onset of the Termination 1 (T1) at about 15 ka, the WMDW production declined until the transition to the

- 84 Holocene due to the rising sea level, with a relatively weak mode during the Heinrich Stadial 1 (HS1) and the
- 85 Younger Dryas (YD) (Sierro et al., 2005; Frigola et al., 2008), that led to the deposition of the Organic Rich
- 86 Layer 1 (ORL1; 14.5-8.2 ka BP; Cacho et al., 2002).

87 Because of the disappearance during the Early Holocene of specific epibenthic foraminiferal species, such as 88 *Cibicidoides* spp., which are commonly used for paleohydrological reconstructions, information about the 89 Holocene variability of the deep-water circulation in the western Mediterranean are relatively scarce and are 90 mainly based on grain size analysis and sediment geochemistry (e.g. Frigola et al., 2007). These authors have 91 identified four distinct phases representing different deep-water overturning conditions in the western 92 Mediterranean basin during the Holocene, as well as centennial- to millennial-scale abrupt events of overturning

93 reinforcement.

94 Faunal and stable isotope records from benthic foraminifera located at intermediate depths in the eastern basin 95 reveal uninterrupted well-ventilated LIW during the last glacial period and deglaciation (Kuhnt et al., 2008; 96 Schmiedl et al., 2010). Similarly, a grain-size record obtained from a sediment core collected within the LIW 97 depth range (~500 m water depth) at the east Corsica margin also documents enhanced bottom currents during 98 the glacial period and for specific time intervals during the deglaciation, such as HS1 and YD (Toucanne et al., 99 2012). The Early Holocene is characterized by a collapse of the LIW (Kuhnt et al., 2008; Schmiedl et al., 2010; 100 Toucanne et al., 2012) synchronous with the sapropel S1 deposition (10.2 - 6.4 cal ka BP; Mercone et al., 2000). 101 Proxies for deep-water conditions reveal the occurrence of episodes of deep-water overturning reinforcement in 102 the eastern Mediterranean basin at 8.2 ka BP (Rohling et al., 1997, 2015; Kuhnt et al., 2007; Abu-Zied et al., 103 2008, Siani et al., 2013; Tachikawa et al; 2015), responsible for the interruption of the sapropel S1 in the eastern

104 Mediterranean basin (Mercone et al., 2001; Rohling et al., 2015).

105 Additional insights into Mediterranean circulation changes may be obtained using radiogenic isotopes, such as 106 neodymium, that represent reliable tracers for constraining water-mass mixing and sources (Goldstein and 107 Hemming, 2003, and references therein). It has recently been shown that the neodymium (Nd) isotopic composition, expressed as $\epsilon Nd = ([(^{143}Nd/^{144}Nd)_{sample} / (^{143}Nd/^{144}Nd)_{CHUR}] - 1) \times 10000$ (CHUR: Chondritic 108 109 Uniform Reservoir [Jacobsen and Wasserburg, 1980]) of living and fossil scleractinian CWC faithfully traces 110 intermediate and deep-water mass provenance and mixing of the ocean (e.g. van de Flierdt et al., 2010; Colin et 111 al., 2010; López Correa et al., 2012; Monterro-Serrano et al., 2011, 2013; Copard et al., 2012). Differently from 112 the CWC, the ENd composition of fossil planktonic foraminifera is not related to the ambient seawater at 113 calcification depths but reflects the bottom and/or pore water ENd, due to the presence of authigenic Fe-Mn 114 coatings precipitated on their carbonate shell (Roberts et al., 2010; Elmore et al., 2011; Piotrowski et al., 2012; 115 Tachikawa et al., 2014; Wu et al., 2015). Therefore, the ENd composition of planktonic foraminiferal tests can be 116 used as a useful tracer of deep-water circulation changes in the past, although the effect of pore water on

117 for a miniferal εNd values could potentially complicate the interpretation (Tachikawa et al., 2014).

118 In the Mediterranean Sea, modern seawater ENd values display a large range from ~-11 to ~-5, and a clear

119 vertical and longitudinal gradient, with more radiogenic values encountered in the eastern basin and typically at

120 intermediate and deeper depths (Spivack and Wasserburg 1988; Henry et al., 1994; Tachikawa et al., 2004;

121 Vance et al., 2004). Considering this large ENd contrast, ENd recorded in fossil CWC and planktonic

122 foraminifera from the Mediterranean offers great potential to trace intermediate and deep-water mass exchange

between the two basins, especially during periods devoid of key epibenthic foraminifera, such as the sapropel S1

124 or ORL1 events.

125 Here, the εNd of planktonic foraminifera from a sediment core collected in the Balearic Sea and CWC samples

126 from the Alboran Sea and the Sardinia Channel was investigated to establish past changes of the seawater εNd at

127 intermediate depths and constrain hydrological variations of the LIW during the last ~20 kyr. The ɛNd values

128 have been combined with stable oxygen (δ^{18} O) and carbon (δ^{13} C) isotope measurements of benthic (*Cibicidoides*

129 pachyderma) and planktonic (Globigerina bulloides) foraminifera and sea-surface temperature estimates by

130 modern analogue technique (MAT). Results reveal significant ε Nd variations at intermediate depths in the

131 western basin interpreted as a drastic reduction of the hydrological exchanges between the western and eastern

132 Mediterranean Sea and the subsequent higher proportion of intermediate water produced in the Gulf of Lions

- 133 during the time interval corresponding to the sapropel S1 deposition.
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2. Seawater ENd distribution in the Mediterranean Sea

137 The Atlantic Water (AW) enters the Mediterranean Sea as surface inflow through the Strait of Gibraltar with an 138 unradiogenic ENd signature of ~-9.7 in the strait (Tachikawa et al., 2004) and ~-10.4 in the Alboran Sea 139 (Tachikawa et al., 2004, Spivack and Wasserburg, 1988) for depths shallower than 50 m. During its eastward 140 flowing, AW mixes with upwelled Mediterranean Intermediate Water forming the Modified Atlantic Water 141 (MAW) that spreads within the basin (Millot and Taupier-Letage, 2005) (Fig.1). The surface water ENd values 142 (shallower than 50 m) range from -9.8 to -8.8 in the western Mediterranean basin (Henry et al., 1994; Montagna 143 et al., in prep) and -9.3 to -4.2 in the eastern basin, with seawater off the Nile delta showing the most radiogenic 144 values (Tackikawa et al., 2004; Vance et al., 2004; Montagna et al., in prep). The surface waters in the eastern 145 Mediterranean basin become denser due to strong mixing and evaporation caused by cold and dry air masses 146 flowing over the Cyprus-Rhodes area in winter, and eventually sink leading to the formation of LIW 147 (Ovchinnikov, 1984; Lascaratos et al., 1993, 1998; Malanotte-Rizzoli et al., 1999; Pinardi and Masetti, 2000). 148 The LIW spreads throughout the entire Mediterranean basin at depths between ~150-200 m and ~600-700 m, 149 and is characterized by more radiogenic ε Nd values ranging from -7.9 to -4.8 (average value $\pm 1\sigma$: -6.6 ± 1) in 150 the eastern basin and from -10.4 to -7.58 (-8.7 \pm 0.9) in the western basin (Henry et al., 1994; Tachikawa et al., 151 2004; Vance et al., 2004; Montagna et al., in prep). The LIW acquires its ENd signature mainly from the partial 152 dissolution of Nile River particles (Tachikawa et al., 2004), which have an average isotopic composition of -3.25 153 (Weldeab et al., 2002), and the mixing along its path with overlying and underlying water masses with different 154 ENd signatures. The LIW finally enters the Atlantic Ocean at intermediate depths through the Strait of Gibraltar 155 with an average ε Nd value of -9.2 \pm 0.2 (Tachikawa et al., 2004; Montagna et al., in prep). 156 The WMDW is formed in the Gulf of Lions due to winter cooling and evaporation followed by mixing between 157 the relative fresh surface water and the saline LIW and spreads into the Balearic basin and Tyrrhenian Sea 158 between ~2000 m and 3000 m (Millot, 1999; Schroeder et al., 2013) (Fig. 1). The WMDW is characterized by an

159 average ε Nd value of -9.4 \pm 0.9 (Henry et al., 1994; Tachikawa et al., 2004; Montagna et al., in prep). Between

160 the WDMW and the LIW (from ~700 to 2000 m), the Tyrrhenian Deep Water (TDW) has been found (Millot et

- al., 2006), which is produced by the mixing between WMDW and Eastern Mediterranean Deep Water (EMDW)
- that cascades in the Tyrrhenian Sea after entering from the Strait of Sicily (Millot, 1999, 2009; Astraldi et al.,

163 2001). The TDW has an average ϵ Nd value of -8.1 \pm 0.5 (Montagna et al., in prep).

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- 165 **3.** Material and methods
- 166 3.1. Cold-water coral and foraminifera samples

167 Forty-four CWC samples belonging to the species Lophelia pertusa and Madrepora oculata collected from the 168 Alboran Sea and the Sardinia Channel were selected for this study (Fig. 1). Nineteen fragments were collected at 169 various core depths from a coral-bearing sediment core (RECORD 23; 38°42.18' N; 08°54.75' E; Fig. 1) 170 retrieved from 414 m water depth in the "Sardinian Cold-Water Coral Province" (Taviani et al., 2015) during the 171 R/V Urania cruise "RECORD" in 2013. The core contains well-preserved fragments of M. oculata and L. 172 pertusa embedded in a brownish muddy to silty carbonate-rich sediment. The Sardinian CWC samples were 173 used for U-series dating and Nd isotopic composition measurements. For the southern Alboran Sea, twenty-five 174 CWC samples were collected at water depths between 280 and 442 m in the "eastern Melilla Coral Province" 175 (Fig. 1) during the R/V Poseidon cruise "POS-385" in 2009 (Hebbeln et al. 2009). Eleven samples were 176 collected at the surface of two coral mounds (New Mound and Horse Mound) and three coral ridges (Brittlestar 177 ridges I, II and III), using a box corer and a remotely operated vehicle (ROV). In addition, fourteen CWC 178 samples were collected from various core depths of three coral-bearing sediment cores (GeoB13728, 13729 and 179 13730) retrieved from the Brittlestar ridge I. Details on the location of surface samples and cores collected in the 180 southern Alboran Sea and details on the radiocarbon ages obtained from these coral samples are reported in Fink 181 et al. (2013). Like the CWC sample set from the Sardinia Channel, the dated Alboran CWC samples were also 182 used for further Nd isotopic composition analyses in this study.

In addition, a deep-sea sediment core (barren of any CWC fragments) was recovered southwest of the Balearic Sea at 622 m water depth during the R/V Le Suroît cruise "PALEOCINAT II" in 1992 (SU92-33; 35°25.38' N; 0°33.86' E; Fig. 1). The core unit, which consists of 2.1 m of grey to brown carbonaceous clays, was subsampled continuously at 5-10 cm intervals for a total number of 24 samples used for δ^{18} O, δ^{13} C and ϵ Nd analyzes.

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189 3.2. Analytical procedures on cold-water coral samples

3.2.1. U/Th dating

191 The nineteen CWC samples collected from the sediment core RECORD 23 (Sardinia Channel) were analysed for uranium and thorium isotopes to obtain absolute dating using a Thermo ScientificTM Neptune^{Plus} MC-ICPMS 192 193 installed at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). Prior 194 to analysis, the samples were carefully cleaned using a small diamond blade to remove any visible contamination 195 and sediment-filled cavities. The fragments were examined under a binocular microscope to ensure against the 196 presence of bioeroded zones and finally crushed into a coarse-grained powder with an agate mortar and pestle. 197 The powders (~60-100 mg) were transferred to acid cleaned Teflon beakers, ultrasonicated in MilliQ water, 198 leached with 0.1N HCl for ~ 15 s and finally rinsed twice with MilliQ water. The physically and chemically 199 cleaned samples were dissolved in 3-4 ml dilute HCl (~10%) and mixed with an internal triple spike with known 200 concentrations of ²²⁹Th, ²³³U and ²³⁶U, calibrated against a Harwell Uraninite solution (HU-1) assumed to be at 201 secular equilibrium. The solutions were evaporated to dryness at 70°C, redissolved in 0.6 ml 3N HNO₃ and then 202 loaded into 500 µl columns packed with Eichrom UTEVA resin to isolate uranium and thorium from the other 203 major and trace elements of the carbonate matrix. The U and Th separation and purification followed a procedure slightly modified from Douville et al. (2010). The U and Th isotopes were determined following the 204 205 protocol recently revisited at LSCE (Pons-Branchu et al., 2014). The ²³⁰Th/U ages were calculated from measured atomic ratios through iterative age estimation (Ludwig and Titterington, 1994), using the ²³⁰Th, ²³⁴U 206

- 207 and 238 U decay constants of Cheng et al. (2013) and Jaffey et al. (1971). Due to the low 232 Th concentration (< 1 208 ng/g; see Table 1), no correction was applied for the non-radiogenic 230 Th fraction.
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3.2.2 Nd isotopic composition analyses on cold-water coral fragments

211 Sub-samples of the CWC fragments from the Sardinia Channel used for U-series dating in this study (Table 1) as 212 well as sub-samples of the twenty-five CWC fragments originating from the Alboran Sea, which were already 213 radiocarbon-dated by Fink et al. (2013) (Table 2), were used for further Nd isotopic composition analyses. The 214 fragments (350 to 600 mg) were subjected to a mechanical and chemical cleaning procedure. The visible 215 contaminations, such as Fe-Mn coatings and detrital particles, were carefully removed from the inner and 216 outermost surfaces of the coral skeletons using a small diamond blade. The physically cleaned fragments were 217 ultrasonicated for 10 min with 0.1 N ultra-clean HCl, followed by several MilliQ water rinses and finally 218 dissolved in 2.5 N ultraclean HNO₃. Nd was separated from the carbonate matrix using Eichrom TRU and LN 219 resins, following the analytical procedure described in detail in Copard et al. (2010).

220 The ¹⁴³Nd/¹⁴⁴Nd ratios of all purified Nd fractions were analyzed using the ThermoScientific Neptune^{Plus} Multi-221 Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) hosted at LSCE. The mass-222 fractionation correction was made by normalizing ¹⁴⁶Nd/¹⁴⁴Nd to 0.7219 and applying an exponential law. During each analytical session, samples were systematically bracketed with analyses of JNdi-1 and La Jolla 223 224 standard solutions, which are characterised by accepted values of 0.512115±0.000006 (Tanaka et al., 2000) and 225 0.511855±0.000007 (Lugmair et al., 1983), respectively. Standard JNdi-1 and La Jolla solutions were analysed 226 at concentrations similar to those of the samples (5-10 ppb) and all the measurements affected by instrumental 227 bias were corrected, when necessary, using La Jolla standard. The external reproducibility (2σ) for time resolved 228 measurement, deduced from repeated analyses of La Jolla and JNdi-1 standards, ranged from 0.1 to 0.5 ENd 229 units for the different analytical sessions. The analytical error for each sample analysis was taken as the external 230 reproducibility of the La Jolla standard for each session. Concentrations of Nd blanks were negligible compared 231 to the amount of Nd of CWC investigated in this study.

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233 3.3. Analyses on sediment of core SU92-33

3.3.1. Radiocarbon dating

Radiocarbon dating was measured at UMS-ARTEMIS (Pelletron 3MV) AMS (CNRS-CEA Saclay, France). Seven AMS radiocarbon (¹⁴C) dating were performed in core SU92-33 on well-preserved calcareous tests of the planktonic foraminifera *G. bulloides* in the size fraction >150 μ m (Table 3). The age model for the core was derived from the calibrated planktonic ages by applying a mean reservoir effect of ~400 years (Siani et al., 2000, 2001). All ¹⁴C ages were converted to calendar years (cal. yr BP, BP = AD 1950) by using the INTCAL13 calibration data set (Reimer et al., 2013) and the CALIB 7.0 program (Stuiver and Reimer, 1993).

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242 *3.3.2. Stable isotopes*

Stable oxygen (δ^{18} O) and carbon (δ^{13} C) isotope measurements were performed in core SU92-33 on wellpreserved (clean and intact) samples of the planktonic foraminifera *G. bulloides* (250-315 µm fraction) and the epibenthic foraminifera *C. pachyderma* (250-315 µm fraction) using a Finnigan MAT-253 mass spectrometer at the State Key Laboratory of Marine Geology (Tongji University). Both δ^{18} O and δ^{13} C values are presented relative to the Pee Dee Belemnite (PDB) scale by comparison with the National Bureau of Standards (NBS) 18

248 and 19. The mean external reproducibility was checked by replicate analyses of laboratory standards and is better 249 than $\pm 0.07\%$ (1 σ) for δ^{18} O and $\pm 0.04\%$ for δ^{13} C.

250 *3.3.3 Nd isotope measurements on planktonic foraminifera*

251 Approximately 25 mg of mixed planktonic foraminifera species were picked from the >63 µm size fraction of 252 each sample already used for stable isotope measurements (Table 4). The samples were gently crushed between 253 glass slides under the microscope to ensure that all chambers were open, and ultrasonicated with MilliQ water. 254 Samples were allowed to settle between ultrasonication steps before removing the supernatant. Each sample was 255 rinsed thoroughly with MilliQ water until the solution was clear and free of clay. The cleaned samples were 256 dissolved in 1N acetic acid and finally centrifuged to ensure that all residual particles were removed, following 257 the procedure described in Roberts et al. (2010). Nd was separated following the analytical procedure reported in 258 Wu et al. (2015). For details on the measurement of Nd isotopes see the section above.

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260 *3.3.4. Modern analogue technique (MAT)*

261 The palaeo-sea surface temperatures (SST) were estimated using the modern analogue technique (MAT) 262 (Hutson, 1980; Prell, 1985), implemented by Kallel et al. (1997) for the Mediterranean Sea. This method directly 263 measures the difference between the faunal composition of a fossil sample with a modern database, and it 264 identifies the best modern analogues for each fossil assemblage (Prell, 1985). Reliability of SST reconstructions 265 is estimated using a square chord distance test (dissimilarity coefficient), which represents the mean degree of 266 similarity between the sample and the best 10 modern analogues. When the dissimilarity coefficient is lower than 267 0.25, the reconstruction is considered to be of good quality (Overpeck et al., 1985; Kallel et al., 1997). For core 268 SU92-33, good dissimilarity coefficients are <0.2, with an average value of ~ 0.13 (varying between 0.07 and 269 0.19; Fig. 2a). The calculated mean standard deviation of SST estimates observed in core MD90-917 are ~ 1.5 270 $^{\circ}$ C from the late glacial period to the Younger Dryas and ~ 1.2 $^{\circ}$ C for the Holocene.

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4. Results

4.1. Cold-water corals

274 The good state of preservation for the CWC samples from the Sardinia Channel (RECORD 23; Fig. 1) is attested by their initial δ^{234} U values (Table 1), which is in the range of the modern seawater value (146.8±0.1; Andersen 275 276 et al., 2010). If the uncertainty of the δ^{234} U_i is taken into account, all the values fulfill the so-called "strict" ± 4 277 ‰ reliability criterion and the U/Th ages can be considered strictly reliable. The coral ages range from 278 0.091±0.011 to 10.904±0.042 ka BP (Table 1), and reveal three distinct clusters of coral age distribution during 279 the Holocene representing periods of sustained coral occurrence. These periods coincide with the Early Holocene 280 encompassing a 700-years-lasting time interval from ~10.9 to 10.2 ka BP, the very late Early Holocene at ~8.7 281 ka BP, and the Late Holocene starting at ~1.5 ka BP (Table 1).

Radiocarbon ages obtained for CWC samples collected in the Alboran Sea were published by Fink et al. (2013)

283 (Table 2). They also document three periods of sustained CWC occurrence coinciding with the Bølling–Allerød

284 (B-A) interstadial (13.5–12.9 cal ka BP), the Early Holocene (11.2–9.8 cal ka BP) and the Mid- to Late Holocene

285 (5.4–0.3 cal ka BP).

286 The εNd record obtained from the CWC samples from the Alboran Sea displays a narrow range from -9.22±0.30

287 to -8.59 \pm 0.3, which is comparable to the ϵ Nd record of the planktonic foraminifera from the Balearic Sea over

288 the last 13.5 kyr (Table 2, Fig. 3b). Most of the CWC εNd values are similar within error and the record does not

reveal any clear difference over the last ~13.5 kyr.

290 On the contrary, the CWC samples from the Sardinia Channel display a relatively large ɛNd range, with values

 $291 \qquad \text{varying from -} 5.99 \pm 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene, and values as low as -} 8.66 \pm 0.30 \text{ Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ to -} 7.75 \pm 0.10 \text{ during the Early and Late Holocene} + 0.50 \text{ during the Early and Late Holocene} + 0$

- during the the mid-sapropel S1 deposition (S1a) at ~8.7 ka BP (Table 1, Fig. 3c).
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295 *4.2 Core SU92-33*

The stratigraphy of core SU92-33 was derived from the δ^{18} O variations of the planktonic foraminifera *G. bulloides* (Fig. 2b). The last glacial/interglacial transition and the Holocene encompasses the upper 2.1 m of the core (Fig. 2b). The δ^{18} O record of *G. bulloides* shows higher values (~3.5 ‰) during the late glacial compared to the Holocene (from ~1.5 to 0.8 ‰) exhibiting a pattern similar to those observed in nearby deep-sea cores from the Western Mediterranean Sea (Sierro et al., 2005; Melki et al., 2009).

301 The age model for the upper 1.2 m of the core SU92-33 was based on 7 AMS-¹⁴C age measurements and a 302 linear interpolation between these ages (Table 3, Fig. 2). For the lower portion of the core, a control point was 303 established at the onset of the last deglaciation, which is coeval in the western and central Mediterranean Sea at 304 ~17 cal ka BP (Sierro et al., 2005; Melki et al., 2009; Siani et al., 2001). Overall, the upper 2.1 m of core SU92-305 33 spans the last 19 kyr, with an estimated average sedimentation rate ranging from ~15 cm ka⁻¹ during the 306 deglaciation to ~10 cm ka⁻¹ during the Holocene.

307 April-May SST reconstruction was derived from MAT to define the main climatic events recorded in 308 core SU92-33 during the last 19 kyr. SST vary from 8.5°C to 17.5°C with high amplitude variability over the last 309 19 kyr BP (Fig. 2a). The LGM (19-18 ka BP) is characterized by SST values centered at around 12°C. Then, a 310 progressive decrease of ~4°C between 17.8 ka and 16 ka marks the Henrich Stadial 1 (HS1) (Fig. 2a). A 311 warming phase (~14°C) between 14.5 ka BP and 13.8 ka BP coincides with the B-A interstadial and is followed 312 by a cooling (~11°C) between 13.1 ka BP and 11.8 ka BP largely corresponding to the YD (Fig. 2a). During the 313 Holocene, SST show mainly values of ~16°C, with one exception between 7 ka BP and 6 ka BP pointing to an 314 abrupt cooling of $\sim 3^{\circ}$ C (Fig. 2a). From the late glacial to the Holocene, SST variations show a similar pattern to 315 that previously observed in the Gulf of Lions and Tyrrhenian Sea (Kallel et al., 1997; Melki et al., 2009) as well 316 as in the Alboran Sea (Martrat et al., 2014; Rodrigo-Gámiz et al., 2014). They are globally synchronous for the 317 main climatic transitions to the well dated South Adriatic Sea core MD90-917 (Siani et al., 2004) confirming the 318 robustness of the SU92-33 age model (Fig. 2a).

- The δ^{18} O and δ^{13} C records obtained from the benthic foraminifera *C. pachyderma* display significant variations at millennial time scales (Figs. 2c and 2d). The δ^{18} O values decrease steadily from ~4.5 ‰ during the LGM to ~1.5 ‰ during the Holocene, without showing any significant excursion during HS1 and the YD events (Fig.
- 322 2c), in agreement with results obtained from the neighbor core MD99-2343 (Sierro et al., 2005).
- 323 The δ^{13} C record of *C. pachyderma* shows a decreasing trend since the LGM with a low variability from ~1.6 ‰
- 324 to ~0.6 ‰ (Fig. 2d). The heaviest δ^{13} C values are related to the LGM (~1.6 ‰) while the lightest values (~0.6

- 325 ‰) characterize the Early Holocene and in particular the period corresponding to the sapropel S1 event in the326 eastern Mediterranean basin (Fig. 2d).
- 327 The ENd values of planktonic foraminifera of core SU92-33 from the Balearic Sea vary within a relatively
- 328 narrow range between -9.50±0.30 and -8.61±0.30, with an average value of -9.06±0.28 (Table 2, Fig. 3b). The
- 329 record shows a slight increasing trend since the LGM, with the more unradiogenic values (average -9.28±0.15;
- n=7) being observed in the oldest part of the record (between 18 and 13.5 ka BP), whereas Holocene values are
- 331 generally more radiogenic (average -8.84±0.22; n=17) (Fig. 3b).
- 332333

5. Discussion

Overall, the CWC and foraminiferal ɛNd values measured in this study point to a pronounced dispersion at intermediate depth in terms of absolute values and variability in Nd isotopes during the Holocene between the Alboran and Balearic Seas and the Sardinia Channel. Furthermore, the foraminiferal ɛNd record reveals an evolution towards more radiogenic values at intermediate water depth in the Balearic Sea over the last ~19 kyr (Fig. 3).

339 A prerequisite to properly interpret such ε Nd differences and variations through time consists in characterizing 340 first the present-day ε Nd of the main water-mass end-members flowing in the western Mediterranean basin. It is 341 also necessary to evaluate the temporal changes in ε Nd of the end-members since the LGM, and assess the 342 potential influences of lithogenic Nd input and regional exchange between the continental margins and seawater 343 ("boundary exchange"; Lacan and Jeandel, 2001, 2005) on the ε Nd values of intermediate water masses.

344 During its westward flow, the LIW continuously mixes with surrounding waters with different ENd signatures 345 lying above and below. For the western Mediterranean basin, these water masses are the MAW/Western 346 Intermediate Water (WIW) and the TDW/WMDW, respectively. Accordingly, a well-defined and gradual ɛNd 347 gradient exists at intermediate depth between the eastern and western Mediterranean basins, with LIW values 348 becoming progressively more unradiogenic towards the Strait of Gibraltar, from -4.8±0.2 at 227 m in the 349 Levantine basin to -10.4±0.2 at 200 m in the Alboran Sea (Tachikawa et al., 2004). Such an ENd pattern implies 350 an effective vertical mixing with more unradiogenetic water masses along the E-W LIW trajectory ruling out 351 severe isotopic modifications of the LIW due to the local exchange between the continental margins and 352 seawater. Unfortunately, no information exists on the potential temporal variability in ENd of the Mediterranean

353 water-mass end-members since the LGM.

354 It has been demonstrated that eolian dust input can modify the surface and sub-surface ENd distribution of the 355 ocean in some areas (Arsouze et al., 2009). The last glacial period was associated with an aridification of North 356 Africa (Sarnthein et al., 1981; Hooghiemstra et al., 1987; Moreno et al., 2002; Wienberg et al., 2010) and higher 357 fluxes of Saharan dust to the NE tropical Atlantic (Itambi et al., 2009) and the western Mediterranean Sea 358 characterized by unradiogenic ε Nd values (between -11±0.4 and -14±0.4;; see synthesis in Scheuvens et al., 359 2013). Bout-Roumazeilles et al. (2013) documented a dominant role of eolian supply in the Siculo-Tunisian 360 Strait during the last 20 ka, with the exception of a significant riverine contribution (from the Nile River) and a 361 strong reduction of eolian input during the sapropel S1 event. Such variations in the eolian input to the 362 Mediterranean Sea are not associated to a significant change in the seawater ENd record obtained for the Balearic 363 Sea (core SU92-33) during the sapropel S1 event (Fig. 3). Furthermore, the ENd signature of the CWC from the 364 Sardinia Channel (core RECORD 23) shifts to more unradiogenic values (-8.66±0.30) during the sapropel S1

- 365 event, which is opposite to what expected if it was related to a strong reduction of eolian sediment input. In a
- 366 recent study, Rodrigo-Gámiz et al. (2015) have documented variations in the terrigenous provenance from a
- 367 sediment record in the Alboran Sea (core 293G; 36°10.414'N, 2°45.280'W, 1840 m water depth) since the
- 368 LGM. Radiogenic isotopes (Sr, Nd, Pb) point to changes from North African dominated sources during the
- 369 glacial period to European dominated source during the Holocene. Nevertheless, the major Sr-Nd-Pb excursions
- documented by Rodrigo-Gámiz et al. (2015) and dated at ca. 11.5, 10.2, 8.9-8.7, 5.6, 2.2 and 1.1. ka cal BP do
- 371 not seem to affect the ϵ Nd values of our foraminifera and coral records.
- Taken together, these results suggest that changes of eolian dust input since the LGM were not responsible forthe observed ɛNd variability at intermediate water depths.
- 374 Consequently, assuming that the Nd isotopic budget of the western Mediterranean Sea has not been strongly 375 modified since the LGM, the reconstructed variations of the E-W gradient of εNd values in the western 376 Mediterranean Sea for the past and notably during the sapropel S1 event (Fig. 3) are indicative of a major 377 reorganization of intermediate water circulation.
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- 379

5.1 Hydrological changes in the Alboran and Balearic Seas since the LGM

- 380 The range in ε Nd for the CWC from the Alboran Sea (from -9.22±0.30 to -8.8.59±0.30; Table 2) is very close to 381 the one obtained for the planktonic foraminifera from the Balearic Sea (from -9.50 ± 0.30 to -8.61 ± 0.30 ; Table 4, 382 Fig. 3c), suggesting that both sites are influenced by the same intermediate water masses at least for the last 13.5 383 kyr BP. Today, LIW occupies a depth range between ~200 and ~700 m in the western Mediterranean basin 384 (Millot, 1999; Sparnocchia et al., 1999). More specifically, the salinity maximum corresponding to the core of 385 LIW is found at around 400 m in the Alboran Sea (Millot, 2009) and up to 550 m in the Balearic Sea (López-386 Jurado et al., 2008). The youngest CWC sample collected in the Alboran Sea with a rather "recent" age of 0.34 387 cal ka BP (Fink et al. 2013) displays an ENd value of -8.59±0.30 (Table 2) that is similar to the present-day value 388 of the LIW at the same site (-8.3 ± 0.2) (Dubois-Dauphin et al., submitted) and is significantly different from the 389 WMDW ϵ Nd signature in the Alboran Sea (-10.7 \pm 0.2, 1270 m water depth; Tachikawa et al., 2004). Considering 390 the intermediate depth range of the studied CWC and foraminifera samples, we can reasonably assume that 391 samples from both sites, in the Balearic Sea (622 m water depth) and in the Alboran Sea (280 to 442 m water 392 depth), record ENd variations of the LIW. The ENd record obtained from planktonic foraminifera generally 393 displays more unradiogenic and homogenous values before ~13 cal ka BP (range from -9.46 to -9.12) compared 394 to the most recent part of the record (range from -9.50 to -8.61), with the highest value of -8.61 ± 0.3 in the Early 395 and Late Holocene.
- 396 The SST record displays values centered at around 12°C during the LGM with a subsequent rapid SST decrease 397 towards 9°C , highlighting the onset of the HS1 (Fig. 2a). These values are well comparable to recent high-
- resolution SST data obtained in the Alboran Sea (Martrat et al., 2014; Rodrigo-Gámiz et al., 2014).
- 399 The δ^{18} O record obtained on *G. bulloides* indicates an abrupt 1‰ excursion towards lighter values centered at
- 400 about 16 cal ka BP (Table 4), synchronous with the HS1 (Fig. 2b), which is similar to the δ^{18} O shift reported by
- 401 Sierro et al. (2005) for a core collected at 2391 m water depth NE of the Balearic Islands (MD99-2343; Fig. 1).
- 402 As the Heinrich events over the last glacial period are characterized by colder and fresher surface water in the
- 403 Alboran Sea (Cacho et al., 1999; Pérez-Folgado et al., 2003; Martrat et al., 2004, 2014; Rodrigo-Gámiz et al.,
- 404 2014) and dry climate on land over the western Mediterranean Sea (Allen et al., 1999; Combourieu-Nebout et

- 405 al., 2002; Sanchez Goni et al., 2002; Bartov et al., 2003), lighter δ^{18} O values of planktonic *G. bulloides* are 406 thought to be the result of the inflow of freshwater derived from the melting of icebergs in the Atlantic Ocean 407 into the Mediterranean Sea (Sierro et al., 2005; Rogerson et al., 2008).
- 408 During this time interval, the δ^{13} C record of *C. pachyderma* from the Balearic Sea (core SU92-33) displays a
- 409 decreasing δ^{13} C trend after ~16 cal ka BP (from 1.4 ‰ to 0.9 ‰; Table 4; Fig. 4a). Moreover, the δ^{13} C record
- 410 obtained on benthic foraminifera *C. pachyderma* from the deep Balearic Sea (core MD99-2343) reveals similar
- 411 δ^{13} C values before ~16 cal ka BP suggesting well-mixed and ventilated water masses during the LGM and the
- 412 onset of the deglaciation (Sierro et al., 2005).
- The slightly lower foraminiferal ε Nd values before ~13 cal ka BP could reflect a stronger influence of water masses deriving from the Gulf of Lions as WMDW (ε Nd: -9.4±0.9; Henry et al., 1994; Tachikawa et al., 2004; Montagna et al., in prep). This is in agreement with ε Nd results obtained by Jiménez-Espejo et al. (2015) from planktonic foraminifera collected from deep-water sites (1989 m and 2382 m) in the Alboran Sea (Fig. 4c). Jiménez-Espejo et al. (2015) documented lower ε Nd values (ranging from -10.14±0.27 to -9.58±0.22) during the LGM, suggesting an intense deep-water formation. This is also associated to an enhanced activity of the deeper branch of the MOW in the Gulf of Cádiz (Rogerson et al., 2005; Voelker et al., 2006) linked to the active
- 420 production of the WMDW in the Gulf of Lions during the LGM (Jiménez-Espejo et al., 2015).
- 421 The end of the HS1 (14.7 cal ka BP) is concurrent with the onset of the B-A warm interval characterized by 422 increased SST up to 14°C in the Balearic Sea (SU92-33: Fig. 3a), also identified for various sites in the 423 Mediterranean Sea (Cacho et al., 1999; Martrat et al., 2004, 2014; Essallami et al., 2007; Rodrigo-Gámiz et al., 424 2014). The B-A interval is associated with the so-called melt-water pulse 1A (e.g. Weaver et al., 2003) occurring 425 at around 14.5 cal ka BP. This led to a rapid sea-level rise of about 20 m in less than 500 years and large 426 freshwater discharges in the Atlantic Ocean due to the melting of continental ice sheets (Deschamps et al., 2012), 427 resulting in an enhanced Atlantic inflow across the Strait of Gibraltar. Synchronously, cosmogenic dating of 428 Alpine glacier retreat throughout the western Mediterranean hinterland suggests maximum retreat rates (Ivy-429 Ochs et al., 2007; Kelly et al., 2006). Overall, these events are responsible for freshening Mediterranean waters 430 and reduced surface water density, and hence, weakened ventilation of intermediate (Toucanne et al., 2012) and 431 deep-water masses (Cacho et al., 2000; Sierro et al., 2005). Similarly, lower benthic δ^{13} C values obtained for the 432 Balearic Sea (Fig. 4a) point to less ventilated intermediate water relative to the late glacial. In addition, a 433 decoupling in the benthic δ^{13} C values is observed between deep (MD99-2343) and intermediate (core SU92-33) 434 waters after ~16 cal ka BP (Sierro et al. 2005), suggesting an enhanced stratification of the waters masses (Fig. 435 4a). At this time, the shallowest ENd record from the deep Alboran Sea (core 300G) shifted towards more 436 radiogenic values, while the deepest one (core 304G) remained close to the LGM values (Jimenez-Espejo et al., 437 2015) (Fig. 4c). Furthermore, results from the UP10 fraction (particles > 10 μ m) of the MD99-2343 sediment 438 core (Fig. 4d) indicate a declining bottom-current velocity at 15 ka BP (Frigola et al., 2008). Rogerson et al. 439 (2008) have hypothesized that during deglacial periods the sinking depth of dense waters produced in the Gulf of 440 Lions was shallower resulting in new intermediate water (WIW) rather than new deep-water (WMDW) as 441 observed today during mild winters (Millot, 1999; Schott et al., 1996). Therefore, intermediate depths of the 442 Balearic Sea could have been isolated from the deep-water with the onset of the T1 (at ~15 ka BP). The reduced 443 convection in the deep western Mediterranean Sea together with the shoaling of the nutricline (Rogerson et al., 444 2008) led to the deposition of the ORL 1 (14.5 to 8.2 ka B.P; Cacho et al., 2002) and dysoxic conditions below

2000 m in agreement with the absence of epibenthic foraminifera such as *C. pachyderma* after 11 cal ka BP in
MD99-2343 (Sierro et al., 2005) (Fig. 4a).

After 13.5 ka BP, planktonic foraminifera εNd values from the Balearic Sea (core SU92-33) become more
radiogenic and are in the range of CWC εNd values from the Alboran Sea (Fig. 4b). These values may reveal a
stronger influence of the LIW in the Balearic Sea during the Younger Dryas, as also supported by the sortable

450 silt record from the Tyrrhenian Sea (Toucanne et al., 2012) (Fig. 4e). Deeper depths of the Alboran Sea also

451 record a stronger influence of the LIW with an ε Nd value of -9.1 \pm 0.4 (Jimenez-Espejo et al., 2015). In addition, 452 a concomitant activation of the upper MOW branch, as reconstructed from higher values of Zr/Al ratio in

a concomitant activation of the upper MOW branch, as reconstructed from higher values of Zr/Al ratio in
sediments of the Gulf of Cádiz, can be related to the enhanced LIW flow in the western Mediterranean Sea (Fig.
454 4f) (Bahr et al., 2015).

The time of sapropel S1 deposition (10.2 - 6.4 ka) is characterized by a weakening or a shutdown of intermediate- and deep-water formation in the eastern Mediterranean basin (Rossignol-Strick et al., 1982; Cramp and O'Sullivan, 1999; Emeis et al., 2000; Rohling et al., 2015). At this time, planktonic foraminifera ϵ Nd values from intermediate water depths in the Balearic Sea (core SU92-33) remain high (between -9.15 \pm 0.3 and -8.61 \pm 0.3) (Fig. 4b). On the other hand, the deeper Alboran Sea provides a value of -9.8 \pm 0.3 pointing to a stronger contribution of WMDW (Jimenez-Espejo et al., 2015), coeval with the recovery of deep-water activity from core MD99-2343 (Frigola et al., 2008).

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5.2 Hydrological changes in the Sardinia Channel during the Holocene

464 The present-day hydrographic structure of the Sardinia Channel is characterized by four water masses, with the 465 surface, intermediate and deep-water masses being represented by MAW, LIW and TDW/WMDW, respectively 466 (Astraldi et al., 2002a; Millot and Taupier-Lepage, 2005). In addition, the WIW, flowing between the MAW and 467 the LIW, has also been observed along the Channel (Sammari et al., 1999). The core of the LIW is located at 468 400-450 m water depth in the Tyrrhenian Sea (Hopkins, 1988; Astraldi et al., 2002b), which is the depth range of 469 CWC samples from the Sardinia Channel (RECORD 23; 414 m) (Taviani et al., 2015). The youngest CWC 470 sample dated at ~0.1 ka BP has an ϵ Nd value of -7.70 \pm 0.10 (Table 1, Fig. 5), which is similar within error to the 471 value obtained from a seawater sample collected at 451 m close to the coral sampling location (-8.0±0.4; 472 Montagna et al., in prep).

473 The CWC dating from the Sardinia Channel shows three distinct periods of sustained coral occurrence in this 474 area during the Holocene, with each displaying a large variability in ENd values. CWC from the Early Holocene 475 (10.9-10.2 ka BP) and the Late Holocene (<1.5 ka BP) exhibit similar ranges of ENd values (ranging from -476 5.99±0.50 to -7.75±0.20; Table 1, Fig 5c). Such variations are within the present-day ENd range being 477 characteristic for intermediate waters in the eastern Mediterranean Sea (-6.6±1.0; Tachikawa et al., 2004; Vance 478 et al., 2004). However, the CWC ENd values are more radiogenic than those observed at mid-depth in the 479 present-day western basin (ranging from -10.4 ± 0.2 to -7.58 ± 0.47 ; Henry et al., 1994; Tachikawa et al., 2004; 480 Montagna et al., in prep), suggesting a stronger LIW component in the Sardinia Channel during the Early and 481 Late Holocene. The Sardinian CWC ENd variability also reflects the sensitivity of the LIW to changes in the 482 eastern basin such as rapid variability of the Nile River flood discharge (Revel et al., 2014; 2015; Weldeab et al., 483 2014) or a modification through time in the proportion between the LIW and the Cretan Intermediate Water 484 (CIW). Today, the intermediate water outflowing from the Strait of Sicily is composed by ~66 to 75 % of LIW

485 and 33 to 25 % of CIW (Manca et al., 2006; Millot, 2014). As the CIW is formed in the Aegean Sea, this 486 intermediate water mass is generally more radiogenic than LIW (Tachikawa et al., 2004; Montagna et al., in 487 prep). Following this hypothesis, a modification of the mixing proportion between the CIW and the LIW may 488 potentially explain values as radiogenic as about -6 in the Sardinia Channel during the Early and Late Holocene 489 (Fig. 5c). However, a stronger LIW and/or a CIW contribution cannot be responsible for ENd values as low as -490 8.66±0.30 observed during the sapropel S1 event at 8.7 ka BP (Table 1, Fig. 5c). Considering that such 491 unradiogenic value is not observed at intermediate depth in the modern eastern Mediterranean basin, the most 492 plausible hypothesis suggested here is that the CWC were influenced by a higher contribution of intermediate 493 water from the western basin.

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5.3 Hydrological implications for the intermediate water masses of the western Mediterranean Sea

496 The ε Nd records of the Balearic Sea, Alboran Sea and Sardinia Channel document a temporal variability of the 497 east-west gradient in the western Mediterranean basin during the Holocene. The magnitude of the gradient 498 ranges from ~1.5 to ~3 ε units during the Early and Late Holocene and it is strongly reduced at 8.7 ka BP, 499 coinciding with the sapropel S1 event affecting the eastern Mediterranean basin (Fig. 5). Such variations could 500 be the result of a modification of the Nd isotopic composition of intermediate water masses due to changes of the 501 LIW production through time and a higher contribution of the western-sourced intermediate water towards the 502 Sardinia Channel coinciding with the sapropel S1 event.

- 503 The LIW acquires its radiogenic ENd in the Mediterranean Levantine basin mainly from Nd exchange between 504 seawater and lithogenic particles originating mainly from Nile River (Tachikawa et al., 2004). A higher sediment 505 supply from the Nile River starting at ~15 ka BP was documented by a shift to more radiogenic ENd values of 506 the terrigenous fraction obtained from a sediment core having been influenced by the Nile River discharge 507 (Revel et al., 2015) (Fig. 5e). Others studies pointed to a gradual enhanced Nile River runoff as soon as 14.8 ka 508 BP and a peak of Nile discharge from 9.7 to 8.4 ka recorded by large increase in sedimentation rate from 9.7 to 509 8.4 ka (>120 cm/ka) (Revel et al., 2015; Weldeab et al., 2014; Castaneda et al., 2016). Similarly, enhanced Nile 510 discharge at ~9.5 cal kyr B.P was inferred based on δ^{18} O in planktonic foraminifera from a sediment core in the 511 southeast Levantine Basin (PS009PC (32°07.7'N, 34°24.4'E; 552 m water depth) (Hennekam et al., 2014). This 512 increasing contribution of the Nile River to the eastern Mediterranean basin has been related to the African 513 Humid Period (14.8–5.5 ka BP; Shanahan et al., 2015), which in turn was linked to the precessional increase in 514 Northern Hemisphere insolation during low eccentricity (deMenocal et al., 2000; Barker et al., 2004; Garcin et 515 al., 2009). An increasing amount of radiogenic sediments dominated by the Blue/Atbara Nile River contribution 516 (Revel et al., 2014) could have modified the ENd of surface water towards more radiogenic values (Revel et al., 517 in prep). Indeed, planktonic foraminifera ENd values as high as ~ -3 have been documented in the eastern 518 Levantine Basin (ODP site 967; 34°04.27'N, 32°43.53'E; 2553 m water depth) during the sapropel S1 event as a 519 result of enhanced Nile flooding (Scrivner et al., 2004). The radiogenic signature was likely transferred to 520 intermediate depth as a consequence of the LIW formation in the Rhodes Gyre, and it might have been 521 propagated westwards towards the Sardinia Channel. 522 Therefore, considering the more unradiogenic value of the CWC samples from the Sardinia Channel during the
- sapropel S1a event, it is very unlikely that eastern-sourced water flowed at intermediate depth towards the Sardinia Channel. A possible explanation could be the replacement of the radiogenic LIW that was no longer

- 525 produced in the eastern basin (Rohling, 1994) by less radiogenic western intermediate water (possibly WIW).
- 526 Such a scenario could even support previous hypotheses that invoke a potential circulation reversal in the eastern
- 527 Mediterranean from anti-estuarine to estuarine during sapropel formation (Huang and Stanley, 1972; Calvert,
- 528 1983; Sarmiento et al., 1988; Buckley and Johnson, 1988; Thunell and Williams, 1989). An alternative
- 529 hypothesis would be that reduced surface water densities in the eastern Mediterranean during sapropel S1
- 530 resulted in the LIW sinking to shallower depths than at present. As a result of this shoaling, CWC from the
- 531 Sardinia Channel would have been bathed by underlying Western Intermediate Water during the sapropel S1a
- 532 event.
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534 6. Conclusions

- 535 The foraminiferal ENd record from intermediate depths in the Balearic Sea reveals a relatively narrow range of
- 536 ENd values varying between -9.50 and -8.61 since the LGM (~20 ka). Between 18 and 13.5 cal ka BP, the more
- 537 unradiogenic ε Nd values support a vigorous deep overturning in the Gulf of Lions while δ^{18} O and δ^{13} C values
- 538 indicate a stratification of the water masses after 16 cal ka BP. The stratification together with a decrease of the
- 539 deep-water intensity led to more radiogenic values after ~13 cal ka BP. The foraminiferal ɛNd record, supported
- 540 by ENd values from CWC in the Alboran Sea, shows only minor changes in neodymium isotopes from 13.5 cal
- 541 ka BP to 0.34 cal ka BP, suggesting that the westernmost part of the western Mediterranean basin is not very
- 542 sensitive to hydrological variations of the LIW.
- 543 On the contrary, CWC located at the depth of the LIW in the Sardinia Channel exhibit large ENd variations 544 (between -7.75±0.10 and -5.99±0.50) during the Holocene, suggesting either the role of the Nile River in 545 changing the ENd of the LIW in the eastern Mediterranean basin or a variable LIW/CIW mixing of the water 546 outflowing from the Strait of Sicily. At the time of the sapropel S1 event at ~8.7 ka BP, CWC display a shift 547 toward lower values (-8.66 ± 0.30), similar to those obtained at intermediate depths in the westernmost part of the 548 western basin. This suggests that western-sourced intermediate water likely filled mid-depth of the southern 549 Sardinia, replacing LIW that was no longer produced (or heavily reduced) in the eastern basin. These results 550 could potentially support a reversal of the Mediterranean circulation, although this assumption needs further 551 investigation to be confirmed.
- 552

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 971 Geosystems, 16(11), 3883–3904, doi:10.1002/2015GC005871, 2015.
- 972 **Table captions**
- 973

974 Table 1. U-series ages and εNd values obtained for cold-water coral samples collected from sediment core RECORD 23975 (Sardinia Channel).

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977 Table 2. εNd values obtained for cold-water corals from the southern Alboran Sea. The AMS ¹⁴C ages published by Fink et
978 al. (2013) are also reported as Median probability age (ka BP).

- Table 3. AMS ¹⁴C ages of samples of the planktonic foraminifer *G. bulloides* from 'off-mound' sediment core SU92-33. The
 AMS ¹⁴C ages were corrected for ¹³C and a mean reservoir age of 400 yrs, and were converted into calendar years using the
 INTCAL13 calibration data set (Reimer et al., 2013) and the CALIB 7.0 program (Struiver et al., 2005).
- 983

Table 4. Multiproxy data obtained for the upper 2.1 m of sediment core SU92-33 (Balearic Sea). Stable oxygen and carbon isotopes were measured on benthic (*C. pachyderma*) and planktonic (*G. bulloides*) foraminifera; εNd values were obtained on mixed planktonic foraminifera samples. The age results from a combination of 7 AMS-¹⁴C age measurements for the upper 1.2 m of the core and by a linear interpolation between these ages as well as the $\delta^{18}O$ variations of the planktonic foraminifera *G. bulloides*.

- 989
- 990 Figure captions

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992 Figure 1. Map of the western Mediterranean Sea showing the locations of samples investigated in this study. Yellow dot 993 indicates the sampling location of the sediment core from the Balearic Sea (SU92-33); yellow stars indicate the locations of 994 the CWC-bearing cores from the Sardinia Channel (RECORD 23) and the southern Alboran Sea (for further details on the 995 CWC from the Alboran Sea refer also to Fink et al., 2013). The cores discussed in this paper (Gulf of Cádiz: IODP site 996 U1387, Balearic Sea: MD09-2343, northern Tyrrhenian Sea: MD01-2472, Adriatic Sea: MD90-917) are indicated by black 997 dots, and seawater stations are marked by open squares. Arrows represent the main oceanographic currents. The black line 998 shows the general trajectory of the Modified Atlantic Water (MAW) flowing at the surface from the Atlantic Ocean toward 999 the western and eastern Mediterranean. The orange line represents the Levantine Intermediate Water (LIW) originating from 1000 the eastern basin. The black dashed line shows the trajectory of the Western Mediterranean Deep Water (WMDW) flowing 1001 from the Gulf of Lions toward the Strait of Gibraltar.

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Figure 2. (a) Sea Surface Temperature (SST) records of cores SU92-33 (red line) and MD90-917 (green line; Siani et al., 2004), (b) δ^{18} O record obtained on planktonic foraminifer *G. bulloides* for core SU92-33, (c) δ^{18} O record obtained on benthic foraminifer *C. pachyderma* for core SU92-33, (d) δ^{13} C record obtained on benthic foraminifer *C. pachyderma* for core SU92-33. LGM: Last Glacial Maximum; HS1: Heinrich Stadial 1; B-A: Bølling-Allerød; YD: Younger Dryas. Black triangles indicate AMS ¹⁴C age control points.

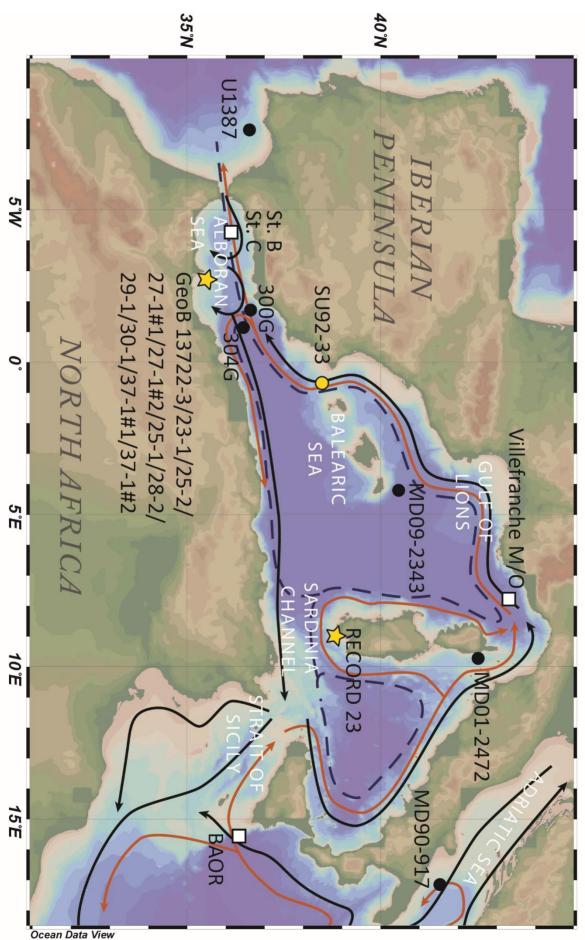
1008

Figure 3. (a) Sea Surface Temperature (SST) record of core SU92-33 (red line), (b) εNd records obtained on mixed
 planktonic foraminifers from core SU92-33 (open circles) and from cold-water coral fragments collected in the Alboran Sea
 (red squares), (c) εNd values of cold-water corals from core RECORD 23 (Sardinia Channel).

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- 1013 Figure 4. (a) δ^{13} C records obtained on benthic foraminifer *C. pachyderma* for cores SU92-33 (red line) and MD99-2343
- 1014 (blue line; Sierro et al., 2005). (b) εNd records obtained on mixed planktonic foraminifers from core SU92-33 (open circles)
- 1015 and from cold-water coral fragments collected in the Alboran Sea (red squares). Modern ENd values for LIW (orange dashed
- 1016 line) and WMDW (blue dashed line) are also reported for comparison. (c) ENd values obtained for planktonic foraminifera
- 1017 with Fe-Mn coatings at sites 300G ($36^{\circ}21.532'$ N, $1^{\circ}47.507'$ W; 1860 m; open dots) and 304G ($36^{\circ}19.873'$ N, $1^{\circ}31.631'$ W;
- 1018 2382 m; black dots) in Alboran Sea (Jimenez-Espejo et al., 2015). (d) UP10 fraction (>10 µm) from core MD99-2343
- 1019 (Frigola et al., 2008). (e) Sortable silt mean grain-size of core MD01-2472 (Toucanne et al., 2012). (f) Ln Zr/Al ratio at IODP
- 1020 site U1387 (36°48.3' N 7°43.1' W; 559 m) (Bahr et al., 2015).
- 1021
- **Figure 5.** (a) δ^{18} O record obtained on planktonic foraminifer *G. bulloides* for core SU92-33, (b) δ^{13} C records obtained on benthic foraminifer *C. pachyderma* for core SU92-33, (c) ϵ Nd values of cold-water corals from core RECORD 23 (Sardinia Channel), (d) ϵ Nd values records obtained on mixed planktonic foraminifera from core SU92-33 (open circles) and from cold-water coral fragments collected in the Alboran Sea (red squares), (e) ϵ Nd values obtained on terrigenous fraction of
- 1026 MS27PT located close the Nile River mouth in the eastern Mediterranean basin (Revel et al., 2015).
- 1027





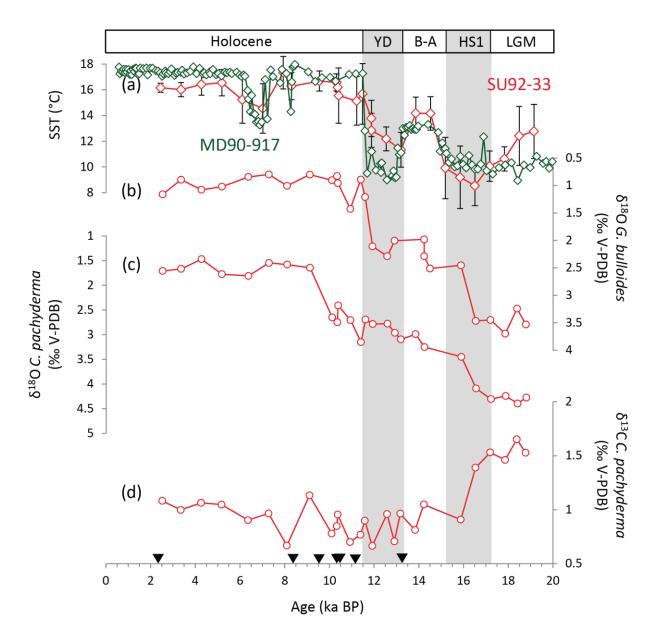


Figure 2

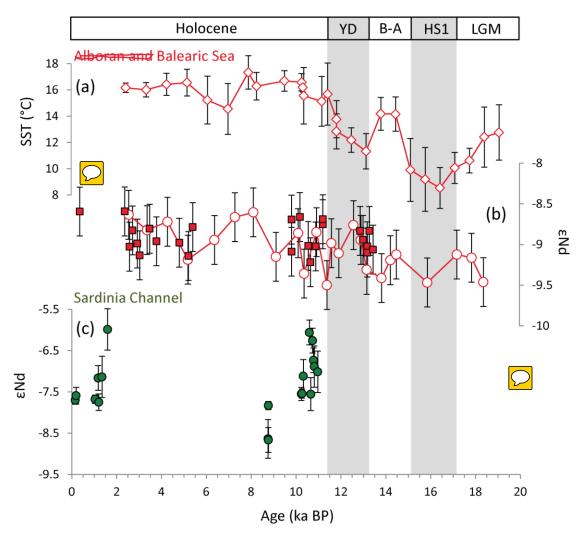


Figure 3

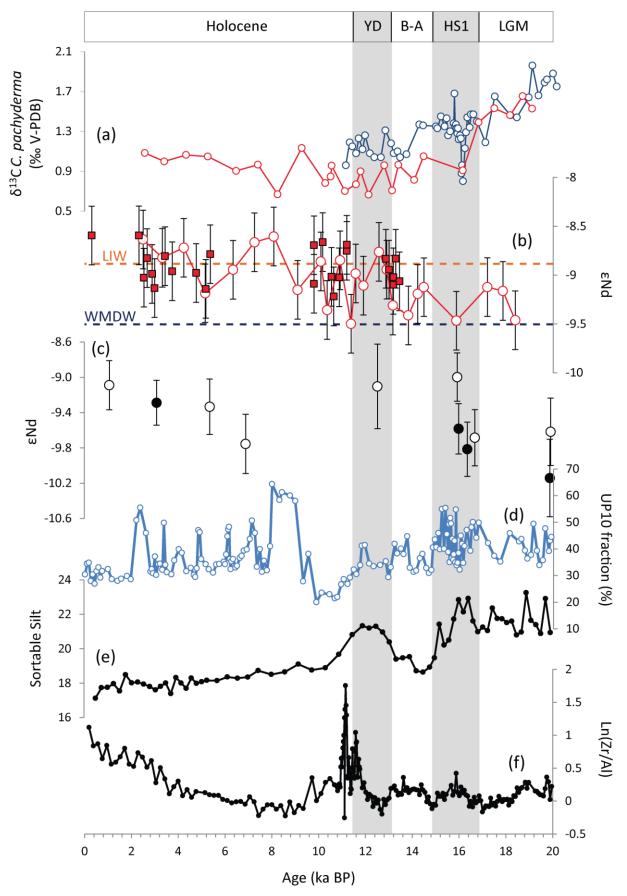


Figure 4

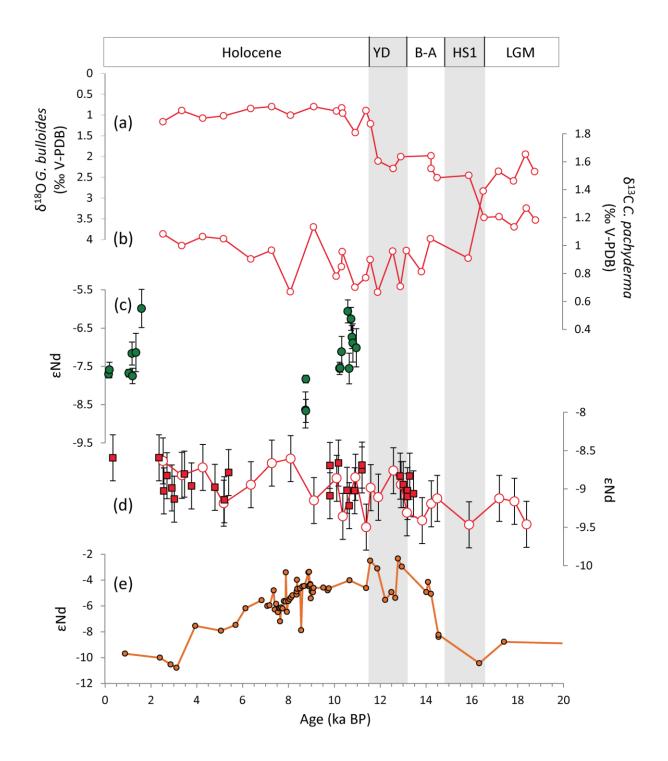


Figure 5

Sample ID	Depth in core (cm)	Corals species	²³⁸ U (µg/g)	²³² Th (ng/g)	δ ²³⁴ U _m (‰)	²³⁰ тһ/ ²³⁸ U	²³⁰ Th/ ²³² Th	Age (ka BP)	δ ²³⁴ U ₍₀₎ (‰)	¹⁴³ Nd/ ¹⁴⁴ Nd	٤Nd
RECORD_23_V	0-3,5	Madrepora oculata	3.31 ±0.005	0.68 ±0.014	151.85 ±1.7	0.00163 ±0.00011	25 ±1.7	0.091 ± 0.011	151.92 ±1.7 0	0.512243 ±0.000005	-7.70 ±0.10
RECORD_23_V	3-7	Madrepora oculata	3.23 ±0.002	0.52 ±0.001	147.11 ±0.6	0.00199 ±0.00006	38 ±1.1	0.127 ±0.006	147.19 ±0.6 0	0.512249 ±0.000010	-7.59 ±0.20
RECORD_23_V	7-10	Madrepora oculata	3.99 ±0.007	0.25 ±0.002	147.52 ±1.7	0.01227 ±0.00022	640 ±11.6	1.110 ± 0.023	148.01 ±1.7 0	148.01 ±1.7 0.512244 ±0.000015	-7.68 ±0.30
RECORD_23_V	8-10	Madrepora oculata	3.79 ±0.005	0.41 ± 0.001	147.77 ±0.7	0.01253 ±0.00007	350 ±2.0	1.135 ± 0.008	148.27 ±0.7 0	148.27 ±0.7 0.512271 ±0.000010	-7.16 ±0.20
RECORD_23_IV	6-9	Madrepora oculata	4.06 ±0.006	0.35 ±0.001	148.47 ±1.2	0.01366 ±0.00011	480 ±3.8	1.243 ± 0.012	149.02 ±1.2 0	0.512241 ±0.000010	-7.75 ±0.20
RECORD_23_IV	27-30	Madrepora oculata	4.06 ±0.003	1.09 ± 0.001	146.91 ± 1.3	0.01405 ±0.00013	159 ± 1.4	1.283 ± 0.014	147.47 ±1.3 0	147.47 ±1.3 0.512272 ±0.000026	-7.14 ±0.50
RECORD_23_IV	37-40	Madrepora oculata	3.52 ±0.005	0.08 ±0.000	148.25 ± 1.1	0.01663 ±0.00012	2308 ±16.4	1.529 ± 0.013	148.92 ±1.1 0	0.512331 ±0.000026	-5.99 ±0.50
RECORD_23_III	55-57	Madrepora oculata	3.63 ±0.002	0.27 ±0.000	145.30 ±0.7	0.08832 ±0.00020	3530 ±8.1	8.685 ±0.027	148.93 ±0.8 0	0.512195 ±0.000026	-8.64 ±0.50
RECORD_23_III	58-61	Madrepora oculata	4.24 ±0.004	0.36 ±0.001	146.71 ± 1.2	0.08859 ±0.00037	3336 ±14.0	8.702 ±0.048	150.39 ±1.2 0	0.512237 ±0.000010	-7.83 ±0.20
RECORD_23_III	63-66	Lophelia pertusa	4.15 ±0.005	0.42 ±0.002	147.19 ±0.8	0.08863 ±0.00054	2783 ±17.1	8.703 ±0.063	150.89 ±0.9 0	0.512194 ±0.000015	-8.66 ±0.30
RECORD_23_I	0-2	Lophelia pertusa	3.35 ±0.002	0.37 ±0.000	147.02 ±0.7	0.10283 ±0.00018	2788 ±4.8	10.173 ±0.025	151.34 ±0.7 0	0.512251 ±0.000010	-7.55 ±0.20
RECORD_23_II	62-65	Lophelia pertusa	3.27 ±0.003	0.39 ±0.002	144.75 ±1.2	0.10289 ±0.00061	2721 ±16.1	10.201 ±0.075	149.01 ±1.2 0	0.512251 ±0.000010	-7.54 ±0.20
RECORD_23_II	50-52	Lophelia pertusa	2.92 ±0.003	0.92 ±0.003	145.39 ± 1.6	0.10351 ±0.00061	1046 ±6.2	10.260 ±0.079	149.69 ±1.6 0	0.512273 ±0.000021	-7.12 ±0.40
RECORD_23_I	12-14	Lophelia pertusa	3.07 ±0.002	0.49 ±0.000	145.22 ±0.7	0.10609 ±0.00023	1971 ±4.3	10.531 ±0.031	149.64 ±0.7 0	0.512327 ±0.000015	-6.06 ±0.30
RECORD_23_I	5-7	Lophelia pertusa	3.50 ±0.002	0.42 ±0.000	146.35 ± 0.9	0.10677 ±0.00016	2654 ±4.0	10.591 ±0.025	150.82 ±0.9 0	0.512251 ±0.000021	-7.55 ±0.40
RECORD_23_II	94-98	Lophelia pertusa	3.14 ±0.003	0.62 ±0.002	146.42 ± 1.0	0.10755 ±0.00047	1737 ±7.6	10.672 ±0.059	150.94 ±1.0 0	0.512317 ±0.000015	-6.26 ±0.30
RECORD_23_I	15-17	Lophelia pertusa	3.40 ±0.003	0.46 ±0.000	146.01 ± 0.9	0.10790 ±0.00021	2409 ±4.6	10.713 ±0.031	150.53 ±0.9 0	0.512293 ±0.000015	-6.73 ±0.30
RECORD_23_II	96-100	Lophelia pertusa	3.61 ± 0.004	0.35 ±0.001	145.50 ±0.8	0.10821 ±0.00044	3579 ±14.7	10.750 ±0.055	150.02 ±0.8 0	0.512285 ±0.000026	-6.89 ±0.50
RECORD_23_II	93-95	Lophelia pertusa	3.19 ±0.003	0.24 ±0.000	143.33 ±0.8	0.10947 ±0.00032	4381 ±12.7	10.904 ±0.042	147.85 ±0.9 0	0.512279 ±0.000026	-7.01 ±0.50
Table 1											
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(cm)		Species	Water Depth (m)	Median probability age (ka BP)	¹⁴³ Nd/ ¹⁴⁴ Nd	εNd
GeoB 13727-1#1	Surface	Lophelia pertusa	363	0.339	0.512198 ±0.000015	-8.59 ±0.30
GeoB 13727-1#2	Surface	Madrepora oculata	353	2.351	0.512198 ±0.000015	-8.59 ±0.30
GeoB 13730-1	6	Lophelia pertusa	338	2.563	0.512175 ±0.000015	-9.03 ±0.30
GeoB 13728-1	Bulk (0-15)	Lophelia pertusa	343	2.698	0.512185 ±0.000015	-8.83 ±0.30
GeoB 13728-2	2	Lophelia pertusa	343	2.913	0.512177 ±0.000015	-8.99 ±0.30
GeoB 13722-3	Bulk (0-15)	Madrepora oculata	280	3.018	0.512170 ±0.000015	-9.13 ±0.30
GeoB 13722-3	Bulk (15-30)	Madrepora oculata	280	3.463	0.512186 ±0.000015	-8.81 ±0.30
GeoB 13735-1	Bulk (0-15)	Madrepora oculata	280	3.770	0.512179 ±0.000015	-8.96 ±0.30
GeoB 13723-1	Bulk (0-8)	Madrepora oculata	291	4.790	0.512178 ±0.000015	-8.98 ±0.30
GeoB 13725-2	Surface	Madrepora oculata	355	5.201	0.512169 ±0.000015	-9.14 ±0.30
GeoB 13723-1	Bulk (8-20)	Madrepora oculata	291	5.390	0.512187 ±0.000015	-8.79 ±0.30
GeoB 13729-1	2.5	Lophelia pertusa	442	9.810	0.512172 ±0.000015	-9.09 ±0.30
GeoB 13729-1	2.5	Lophelia pertusa	442	9.810	0.512193 ±0.000015	-8.69 ±0.30
GeoB 13729-1	49	Lophelia pertusa	442	10.181	0.512194 ±0.000015	-8.66 ±0.30
GeoB 13730-1	102	Lophelia pertusa	338	10.556	0.512176 ±0.000015	-9.02 ±0.30
GeoB 13730-1	194	Lophelia pertusa	338	10.652	0.512165 ±0.000015	-9.22 ±0.30
GeoB 13729-1	315	Lophelia pertusa	442	10.889	0.512176 ±0.000015	-9.02 ±0.30
GeoB 13729-1	375	Lophelia pertusa	442	11.206	0.512189 ±0.000015	-8.75 ±0.30
GeoB 13730-1	298	Lophelia pertusa	338	11.208	0.512193 ±0.000015	-8.69 ±0.30
GeoB 13728-2	191	Lophelia pertusa	343	12.874	0.512185 ±0.000015	-8.83 ±0.30
GeoB 13737-1#2	Surface	Lophelia pertusa	297	13.005	0.512180 ±0.000015	-8.94 ±0.30
GeoB 13728-2	295	Lophelia pertusa	364	13.194	0.512176 ±0.000015	-9.02 ±0.30
GeoB 13728-2	295	Lophelia pertusa	364	13.194	0.512171 ±0.000015	-9.10 ±0.30
GeoB 13730-1	427	Lophelia pertusa	338	13.291	0.512185 ±0.000015	-8.83 ±0.30
GeoB 13737-1#1	Surface	Lophelia pertusa	299	13.452	0.512174 ±0.000015	-9.06 ±0.30

Core	Depth in core (cm)	¹⁴ C-age (years)	±1σ (years)	Median probability age (ka BP)
SU92-33	0	2770	70	2437
SU92-33	64	7870	90	8280
SU92-33	70	8670	80	9528
SU92-33	74	9510	100	10295
SU92-33	84	9610	90	10389
SU92-33	90	10180	100	11192
SU92-33	120	11710	110	13172

Depth in core (cm)	Age (ka BP)	δ ¹³ C C. pachyderma (‰ VPDB)	δ ¹⁸ Ο C. pachyderma (‰ VPDB)	δ ¹³ C G. bulloides (‰ VPDB)	δ ¹⁸ Ο G. bulloides (‰ VPDB)	¹⁴³ Nd/ ¹⁴⁴ Nd	εN	ld
1	2.53	1.08	1.71	-0.6	1.16	0.512195 ±0.000015	-8.64	±0.30
10	3.35	1.00	1.67	-0.82	0.90	0.512186 ±0.000015	-8.82	±0.30
19.5	4.26	1.06	1.47	-0.55	1.08	0.512191 ±0.000015	-8.72	±0.30
29.5	5.18	1.05	1.78	-0.55	1.02	0.512167 ±0.000015	-9.19	±0.30
42.5	6.36	0.90	1.81	-0.91	0.84	0.512179 ±0.000015	-8.95	±0.30
52.5	7.28	0.97	1.55	-0.80	0.80	0.512194 ±0.000015	-8.66	±0.30
61.5	8.10	0.67	1.58	-0.95	1.01	0.512197 ±0.000015	-8.61	±0.30
67.5	9.11	1.13	1.65	-1.07	0.80	0.512169 ±0.000015	-9.15	±0.30
72.5	10.10	0.78	2.65	-1.27	0.91	0.512184 ±0.000015	-8.86	±0.30
77.5	10.33	0.85	2.75	-1.10	0.83		-	-
81.5	10.37	0.96	2.41	-1.21	0.96	0.512158 ±0.000015	-9.36	±0.30
87.5	10.92	0.70	2.71	-0.11	1.43	0.512184 ±0.000015	-8.85	±0.30
92.5	11.39	0.77	3.15	-1.00	0.89	0.512151 ±0.000015	-9.50	±0.30
95.5	11.59	0.90	2.69	-1.14	1.21	0.512178 ±0.000015	-8.98	±0.30
100.5	11.92	0.67	2.78	-0.44	2.11	0.512171 ±0.000015	-9.11	±0.30
110.5	12.58	0.96	2.78	-0.86	2.29	0.512189 ±0.000015	-8.76	±0.30
115.5	12.91	0.71	2.96	-0.54	2.01	0.512180 ±0.000015	-8.94	±0.30
119.5	13.17	0.96	3.09	-	-	0.512161 ±0.000015	-9.31	±0.30
129.5	13.83	0.81	2.99	-	-	0.512156 ±0.000015	-9.41	±0.30
135.5	14.23	1.05	3.25	-1.16	1.98	0.512167 ±0.000015	-9.19	±0.30
135.5	14.23	-	-	-0.94	2.29		-	-
139.5	14.49	-	-	-0.96	2.51	0.512170 ±0.000015	-9.12	±0.30
159.5	15.88	0.91	3.45	-0.81	2.45	0.512153 ±0.000015	-9.47	±0.30
169.5	16.54	1.39	4.09	-0.76	3.47		-	-
179.5	17.20	1.53	4.30	-0.98	3.45	0.512170 ±0.000015	-9.12	±0.30
190	17.86	1.46	4.24	-1.10	3.70	0.512168 ±0.000015	-9.16	±0.30
198	18.39	1.65	4.39	-1.24	3.24	0.512153 ±0.000015	-9.46	±0.30
206	18.78	1.53	4.28	-0.90	3.53		-	-