Reply to reviewer #1 comments:

Thank you very much for agreeing to review this paper and for your comments that have permitted to improve the quality of the manuscript.

Most of your comments have been taken into account in the revised version of the manuscript and all the proposed references have been added.

Please find below a point-by-point reply relative to your comments.

From the first part of the abstract, it seems that the neodymium isotopic composition of both mixed planktonic foraminifera and cold-water corals (CWC) have been investigated at the three selected locations. This could be made clear since CWC have been analyzed only at the Alboran Sea and the south Sardinian continental margin and foraminifera only at the Balearic basin. This aspect could be clarified, also explaining how the data have been integrated. The abstract includes the main implications of the study for hydrological variations during the deposition of the S1 sapropel but the data are also relevant to the deposition of the ORL1.

The abstract has been revised in order to make clear the different archives and locations of the study. We have also added a sentence to explain how the data have been integrated. However, the deposition of the ORL1 being not the aim of this paper, we do not conclude on potential implications about it.

The introduction could better highlight the aim of the work. Moreover, the classical references on Mediterranean climate variability are cited but more recent ones could also be included, for instance Martrat et al (2014) provide interesting high-resolution data on surface water variability of the Mediterranean Sea during the last two deglaciations, including the Holocene.

The introduction has been sharpened up following your recommendations and those of the reviewer #2. Martrat et al. (2014) has not been added in the introduction as we do not think that discussing SST variability in the introduction is relevant in our paper. However, this reference has been cited in other parts of the text.

In the material and methods section, though references are provided to get detailed information about CWC cores, additional information on core description could also be included in this paper to facilitate the whole picture of the analyzed materials. Similarly, a new core recovered in the Balearic Sea has been investigated but little is said about the description of the materials sampled except for barren of any CWC fragments. It is also mentioned that samples from this core have been used for multiproxy analyses but other than dating and estimation of SST by modern analogue techniques only neodymium and stable isotopes been analyzed so this could be better specified in section 3.1.

Additional information on the CWC and SU92.33 cores have now been included in the text. The term "multiproxy analyses" has been replaced by "$\delta^{18}O$, $\delta^{13}C$ and εNd analyzes".

Regarding the results section, there are three different subsections on core SU92- 33 that may be omitted and the results could be synthetized in just one as for CWC.

The subsections have been deleted and the results have been synthetized in two sections: CWC and core SU-92.33

Some general sentences referred to sedimentation rate as "the lowest values observed during the Holocene" could be more specific.

The sentence has been modified in order to better quantify the sedimentation rate.

In this section the information concerning the core MD90-917 is insufficient, it is cited as a well dated record but it is not clear if the references cited in the paragraph (line 294) are those providing the data included in Fig. 2a (in which a reference is not cited).

The reference for this core (Siani et al., 2004) has been integrated in the paragraph.

The discussion is relevant and highlights the most important aspects of the hydrological variations in the Mediterranean. However, some aspects could be further discussed as the role of the eolian input in the εNd variability and why it is not affected by changes in such input. Concerning this, some additional papers on eolian input could be considered, for instance Scheuvens et al. (2013) on bulk composition of northern African dust or Rodrigo-Gamiz et al (2015) on terrigenous input provenance in the western Mediterranean.

The role of the eolian input had been partially discussed in the text as we mentioned the papers by Arsouze et al. (2009) and Bout-Roumazeilles et al. (2013). However, following your recommendations, we have added an additional part of the discussion based on the paper by Rodrigo-Gámiz et al. (2015) on the terrigenous input provenance in the western Mediterranean.

In the submitted version of the manuscript, we had cited individual references (Grousset et al., 1992, 1998; Grousset and Biscaye, 2005) that are included in the synthesis paper by Scheuvens et al. (2013). In the revised version, we have decided to remove those references and only cite "see synthesis in Scheuvens et al., 2013" to make it clear.

Also regarding the Nile discharge, some other recent papers could be considered as Hennekam et al (2014).

This reference has been added to the text of the revised version.

In general, the results on SST are not sufficiently compared with other SST records, see for instance the previously mentioned paper from Martrat et al (2014) and also some recent papers on sea surface temperature variations in the western Mediterranean sea over the last 20 kyr (Rodrigo-Gamiz et al., 2014).

The SU92-33 SST record is now compared to SST reconstructions reported in Martrat et al. (2014) and Rodrigo-Gámiz et al. (2014).

It is also concluded that 18O and 13C values indicate a stratification of the water masses after 16 cal ka BP, but why the data are supporting this conclusion could be further explained in the conclusions section. The implications of the obtained results for the deposition of the ORL1 could also be included in this section.

The deposition of the ORL1 being not the aim of this paper, we do not conclude on potential implications about it.

Reply to reviewer #2 comments:

Thank you very much for agreeing to review this paper and for your comments that have improved the quality of the manuscript.

Most of your comments have been taken into account in the revised version of the manuscript.

Please find below a point-by-point reply relative to your comments.

Introduction. The Introduction could and should be improved and sharpened up (and the same may apply to the discussion). For example (Lines 57-65), the authors seem to build their rationale on the (potential) influence of the Mediterranean thermohaline circulation on the AMOC. But this is not the only reason for better characterising the patterns or variability and the drivers of the thermohaline circulation in this basin. The authors could also (or first) more clearly illustrate the importance of the Mediterranean circulation (an notably of the Levantine Intermediate Waters) for the deep-sea ventilation during the formation of organic-rich deposits (sapropels) across the basin (e.g., De Lange et al., 2008 – Nature Geoscience; Rohling et al., 2015 – Earth-Science Reviews and many others) and/or the more recent evidence of a link between Mediterranean circulation changes and positive phases of the North Atlantic Oscillation (e.g., Incarbona et al., 2016 – Scientific Reports). This would make the introduction section better suited for Climate of the Past by making a more convincing case for the wide relevance of studies like the one by Dubois-Dauphin et al. to the palaeoceanography of the Mediterranean Sea and more generally to our community.

The introduction has been modified by integrating the importance of intermediate and deep water circulation during the formations of organic rich deposits. However, the evidence of a link between Mediterranean circulation changes and positive phases of the North Atlantic Oscillation has not been added as it is relevant only on a decadal timescale, which is not the target of our paper.

Sea Surface Temperature record. The uncertainties associated with the sea surface temperature (SST) reconstructions presented in the paper (Lines 247-255) should be quantitatively assessed. The authors state ': : :Reliability of SST reconstructions is estimated using a square chord distance test (dissimilarity coefficient), which represents the mean degree of similarity between the sample and the best 10 modern analogues. When the dissimilarity coefficient is lower than 0.25, the reconstruction is considered to be of good quality: : :''. This is a merely qualitative statement; the associated with the SST record presented in the manuscript should instead be quantified.

The uncertainties associated with SST reconstruction have been plotted on figures 2 and 3. Additional information has also been added in the Material and methods section in order to better quantify the SST reconstruction.

Data analysis. I think data generated by Dubois-Dauphin et al. are of high quality, but I also think that their analysis and presentation could and should be improved. For example, could the records in Figure 3b be stacked? This would highlight the main trends in the data and help the reader to easily follow the interpretation presented by the authors (at the moment also because of a 'wordy' and fairly unfocused discussion this is not the case). Even better, a Monte Carlo analysis of the data in which both uncertainties in the neodymium isotopes and in the chronology are considered would considerably strengthen the data analysis, allow

more quantitative arguments, and make this a key example fo the use of neodymium isotopes to address palaeocirculation problems.

Although both sites in the Balearic and Alboran Sea are likely bathed by the same water mass (LIW), εNd records are based on different archives (i.e. cold-water corals and planktonic foraminifera). Furthermore, the age model is different as core SU92-33 is based on ^{14}C measurements while CWC are dated by the U-Th method.
On the other hand, data obtained from CWC from the Sardinia Channel display only specific time slices instead of a continuous record over time.
For these reasons, we do not think that a Monte Carlo analysis and/or a stacked record would be relevant for this study.

Data interpretation. I wonder if the data presented can be so unequivocally interpreted as a reduction of Levantine Intermediate Water (formation? circulation?) during the deposition of sapropel S1 to the extent of arguing for a circulation reversal (which most quantitative analyses so far suggest to be highly unlikely). A possibility that the data cannot rule out is that the Levantine Intermediate Water shoaled rather than weakened and the core sites were bathed by a water mass with a different isotopic fingerprint (e.g., the western Mediterranean intermediate waters proposed by the authors) because of this shoaling.

This alternative hypothesis is now presented at the end of the discussion.

Minor Points
Lines 36-39: text is not very clear; I would recommend rewriting this bit.

The sentence has been slightly rephrased.

Lines 272-283: I think this section can be moved to the methods and merged with sections 3.3.

This section has been re-organised following also recommendations of the reviewer #1

Lines 483-484: What do the authors mean by 'intensity changes'?

We mean changes in LIW production (enhanced or reduced). The sentence has been slightly modified to make it clear.

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

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Abstract. We present Tthe neodymium isotopic composition (εNd) of mixed planktonic foraminifera species, from a sediment core collected at 622 m water depth in the Balearic Sea, and as well as εNd of scleractinian cold-water corals (CWC; *Madrepora oculata, Lophelia pertusa*) collected retrieved at 280 620 414 m water depth in the Balearic Sea, the Alboran Sea and south Sardinian continental margin. The aim wasis to investigated to constrain hydrological variations at intermediate depths in the western Mediterranean Sea during the last 20 kyra. Planktonic (*Globigerina bulloides*) and benthic (*Cibicidoides pachyderma*) foraminifera are also analyzed for stable oxygen ($\delta^{18}O$) and carbon ($\delta^{13}C$) isotopes. The foraminiferal and coral εNd values from the Balearic Sea and the Alboran Sea are comparable over the past last ~13 kyra, with mean values of -8.94±0.26 (1σ; n=24) and -8.91±0.18 (1σ; n=25), respectively. Before 13 ka BP, the foraminiferal εNd values are slightly lower (-9.28±0.15) and tend to reflect a higher mixing between intermediate and deep waters, which is characterized by more unradiogenic εNd values. The slight εNd increase after 13 ka BP is associated to a marked differencedecoupling in the benthic foraminiferal $\delta^{13}C$ composition of between intermediate and deeper depths, which started at ~16 ka BP. This suggests an earlier stratification of the water masses and a subsequent reduced contribution of unradiogenic εNd from deep waters. The CWC from the Sardinia Channel show a much larger scattering of εNd values, from -8.66±0.30 to -5.99±0.50, and a lower average (-7.31±0.73; n=19) compared to the CWC and foraminifera from the Alboran Sea and Balearic Sea, indicative of intermediate waters sourced from the Levantine basin. At the time of sapropel S1 deposition (10.2 to 6.4 ka), the εNd values of the Sardinian CWC become more unradiogenic (-8.38±0.47; n=3 at ~8.7 ka BP), suggesting a significant contribution of

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

1. Introduction

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

Previous studies have mainly focused on the glacial variability of the deep-water circulation in the western Mediterranean basin (Cacho et al., 2000, 2006; Sierro et al., 2005; Frigola et al., 2007, 2008). During the Last Glacial Maximum (LGM), strong deep-water convection took place in the Gulf of Lions, producing cold, well-ventilated western Mediterranean Deep Water (WMDW) (Cacho et al., 2000, 2006; Sierro et al., 2005), while the MOW flowed at greater depth in the Gulf of Cádiz (Rogerson et al., 2005; Schönfeld and Zahn, 2000). With the onset of the Termination 1 (T1) at about 15 ka, the WMDW production declined until the ~~transition to the~~ Holocene due to the rising sea level, with a relatively weak mode during the Heinrich Stadial 1 (HS1) and the

87 Younger Dryas (YD) (Sierro et al., 2005; Frigola et al., 2008), that led to the deposition of the Organic Rich

88 Layer 1 (ORL1; 14.5-8.2 ka BP; Cacho et al., 2002).

89 Because of the disappearance during the Early Holocene of specific epibenthic foraminiferal species, such as

90 *Cibicidoides* spp., which are commonly used for paleohydrological reconstructions, information about the

91 Holocene variability of the deep-water circulation in the western Mediterranean are relatively scarce and are

92 mainly based on grain size analysis and sediment geochemistry (e.g. Frigola et al., 2007). These authors have

93 identified four distinct phases representing different deep-water overturning conditions in the western

94 Mediterranean basin during the Holocene, as well as centennial- to millennial-scale abrupt events of overturning

95 reinforcement.

96 Faunal and stable isotope records from benthic foraminifera located at intermediate depths in the eastern basin

97 reveal ~~uninterrupted~~ well-ventilated LIW during the last glacial period and deglaciation (Kuhnt et al., 2008;

98 Schmiedl et al., 2010). Similarly, ~~A~~a grain-size record obtained from a sediment core collected within the LIW

99 depth range (~500 m water depth) at the east Corsica margin also ~~reveals~~ documents enhanced bottom currents

100 during the glacial period and for specific time intervals ~~during~~ the deglaciation, such as HS1 and ~~the~~ YD

101 (Toucanne et al., 2012). The Early Holocene is characterized by a collapse of the LIW (Kuhnt et al., 2008;

102 Schmiedl et al., 2010; Toucanne et al., 2012) synchronous with the sapropel S1 deposition (10.2 – 6.4 cal ka BP;

103 Mercone et al., 2000). Proxies for deep-water conditions reveal the occurrence of episodes of deep-water

104 overturning reinforcement in the eastern Mediterranean basin at 8.2 ka BP (Rohling et al., 1997, 2015; Kuhnt et

105 al., 2007; Abu-Zied et al., 2008, Siani et al., 2013; Tachikawa et al; 2015), responsible for the interruption of the

106 sapropel S1 in the eastern Mediterranean basin (Mercone et al., 2001; Rohling et al., 2015).

107 Additional insights into Mediterranean circulation changes may be ~~obtained~~ using radiogenic isotopes, such as

108 neodymium, that represent reliable tracers for constraining water-mass mixing and sources (Goldstein and

109 Hemming, 2003, and references therein).

110 It has recently been shown that the neodymium (Nd) isotopic composition, expressed as $\varepsilon Nd =$

111 $([(^{143}Nd/^{144}Nd)_{sample} /(^{143}Nd/^{144}Nd)_{CHUR}] - 1) \times 10000$ (CHUR: Chondritic Uniform Reservoir [Jacobsen and

112 Wasserburg , 1980]) of living and fossil scleractinian CWC faithfully traces intermediate and deep-water mass

113 provenance and mixing of the ocean (e.g. van de Flierdt et al., 2010; Colin et al., 2010; López Correa et al.,

114 2012; Monterro-Serrano et al., 2011, 2013; Copard et al., 2012). Differently from the CWC, the εNd

115 composition of fossil planktonic foraminifera is not related to the ambient seawater at calcification depths but

116 reflects the bottom and/or pore water εNd, due to the presence of authigenic Fe-Mn coatings precipitated on their

117 carbonate shell (Roberts et al., 2010; Elmore et al., 2011; Piotrowski et al., 2012; Tachikawa et al., 2014; Wu et

118 al., 2015). Therefore, the εNd composition of planktonic foraminiferal tests can be used as a useful tracer of

119 deep-water circulation changes in the past, although the effect of pore water on foraminiferal εNd values could

120 potentially complicate the interpretation (Tachikawa et al., 2014).

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

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

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

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

125 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 ~~and~~ or ORL1 events.

Here, the εNd of planktonic foraminifera from a sediment core collected in the Balearic Sea and CWC samples from the Alboran Sea and the Sardinia Channel was investigated to establish past changes of the seawater εNd ~~values~~ at intermediate depths and constrain hydrological variations of the LIW during the ~~past~~ last ~20 kyr~~a~~. The εNd values have been combined with stable oxygen ($\delta^{18}O$) and carbon ($\delta^{13}C$) isotope measurements of benthic (*Cibicidoides pachyderma*) and planktonic (*Globigerina bulloides*) foraminifera and sea-surface temperature estimates by modern analogue technique (MAT). Results reveal significant εNd ~~changes~~ variations ~~of the E-W gradient of εNd values for the LIW of~~ at intermediate depths in the western basin interpreted ~~by~~ as a drastic reduction of the hydrological exchanges between the western and eastern Mediterranean Sea and the subsequent higher proportion of intermediate water produced in the Gulf of Lions during the time interval corresponding to the sapropel S1 deposition.

2. Seawater εNd distribution in the Mediterranean Sea

The Atlantic Water (AW) enters the Mediterranean Sea as surface inflow through the Strait of Gibraltar with an unradiogenic εNd signature of ~-9.7 in the strait (Tachikawa et al., 2004) and ~-10.4 in the Alboran Sea (Tachikawa et al., 2004, Spivack and Wasserburg, 1988) for depths shallower than 50 m. During its eastward flowing, AW mixes with upwelled Mediterranean Intermediate Water forming the Modified Atlantic Water (MAW) that spreads within the basin (Millot and Taupier-Letage, 2005) (Fig.1). The surface water εNd values (shallower than 50 m) range from -9.8 to -8.8 in the western Mediterranean basin (Henry et al., 1994; Montagna et al., in prep) and -9.3 to -4.2 in the eastern basin, with seawater off the Nile delta showing the most radiogenic values (Tackikawa et al., 2004; Vance et al., 2004; Montagna et al., in prep). The surface waters in the eastern Mediterranean basin become denser due to strong mixing and evaporation caused by cold and dry air masses flowing over the Cyprus-Rhodes area in winter, and eventually sink leading to the formation of LIW (Ovchinnikov, 1984; Lascaratos et al., 1993, 1998; Malanotte-Rizzoli et al., 1999; Pinardi and Masetti, 2000). The LIW spreads throughout the entire Mediterranean basin at depths between ~150-200 m and ~600-700 m, and is characterized by more radiogenic εNd values ranging from -7.9 to -4.8 (average value ± 1σ: -6.6 ± 1) in the eastern basin and from -10.4 to -7.58 (-8.7 ± 0.9) in the western basin (Henry et al., 1994; Tachikawa et al., 2004; Vance et al., 2004; Montagna et al., in prep). The LIW acquires its εNd signature mainly from the partial dissolution of Nile River particles (Tachikawa et al., 2004), which have an average isotopic composition of -3.25 (Weldeab et al., 2002), and the mixing along its path with overlying and underlying water masses with different εNd signatures. The LIW finally enters the Atlantic Ocean at intermediate depths through the Strait of Gibraltar with an average εNd value of -9.2 ± 0.2 (Tachikawa et al., 2004; Montagna et al., in prep).

The WMDW is formed in the Gulf of Lions due to winter cooling and evaporation followed by mixing between ~~the relative fresh~~ surface water, and the ,saline LIW and spreads into the Balearic basin and Tyrrhenian Sea between ~2000 m and 3000 m (Millot, 1999; Schroeder et al., 2013) (Fig. 1). The WMDW is characterized by an average εNd value of -9.4 ± 0.9 (Henry et al., 1994; Tachikawa et al., 2004; Montagna et al., in prep). Between the WDMW and the LIW (from ~700 to 2000 m), the Tyrrhenian Deep Water (TDW) has been found (Millot et

166 al., 2006), which is produced by the mixing between WMDW and Eastern Mediterranean Deep Water (EMDW)
167 that cascades in the Tyrrhenian Sea after entering ~~from~~ the Strait of Sicily (Millot, 1999, 2009; Astraldi et al.,
168 2001). The TDW has an average εNd value of -8.1 ± 0.5 (Montagna et al., in prep).

170 **3. Material and methods**

171 *3.1. Cold-water coral and foraminifera samples*

172 Forty-four CWC samples belonging to the species *Lophelia pertusa* and *Madrepora oculata* collected from the
173 Alboran Sea and the Sardinia Channel were selected for this study (Fig. 1). Nineteen fragments were collected at
174 various core depths from a coral-bearing sediment core (RECORD 23; 38°42.18' N; 08°54.75' E; Fig. 1)
175 retrieved from 414 m water depth in the "Sardinian Cold-Water Coral Province" (Taviani et al., 2015) during the
176 R/V Urania cruise "RECORD" in 2013. The core contains well-preserved fragments of *M. oculata* and *L.*
177 *pertusa* embedded in a brownish muddy to silty carbonate-rich sediment. The Sardinian CWC samples were
178 used for U-series dating and Nd isotopic composition measurements. For the southern Alboran Sea, twenty-five
179 CWC samples were collected at water depths between 280 and 442 m in the "eastern Melilla Coral Province"
180 (Fig. 1) during the R/V Poseidon cruise "POS-385" in 2009 (Hebbeln et al, 2009). Eleven samples were
181 collected at the surface of two coral mounds (New Mound and Horse Mound) and three coral ridges (Brittlestar
182 ridges I, II and III), using a box corer and a remotely operated vehicle (ROV). In addition, fourteen CWC
183 samples were collected from various core depths of three coral-bearing sediment cores (GeoB13728, 13729 and
184 13730) retrieved from the Brittlestar ridge I. Details on the location of surface samples and cores collected in the
185 southern Alboran Sea and details on the radiocarbon ages obtained from these coral samples are reported in Fink
186 et al. (2013). Like the CWC sample set from the Sardinia Channel, the dated Alboran CWC samples were also
187 used for further Nd isotopic composition analyses in this study.

188 In addition, a deep-sea sediment core (barren of any CWC fragments) was recovered ~~in the~~ southwest of the
189 Balearic Sea at 622 m water depth during the R/V Le Suroît cruise "PALEOCINAT II" in 1992 (SU92-33;
190 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,
191 ~~and~~ was ~~-~~sub-sampled continuously at 5-10 cm intervals ~~for the upper 2.1 m~~ for a total number of 24 samples
192 used for $\delta^{18}O$, $\delta^{13}C$ and εNd ~~further multi-proxy~~ analyzes.

194 *3.2. Analytical procedures on cold-water coral samples*

195   *3.2.1. U/Th dating*

196 The nineteen CWC samples collected from the sediment core RECORD 23 (Sardinia Channel) were analysed for
197 uranium and thorium isotopes to obtain absolute dating using a Thermo Scientific[TM] Neptune[Plus] MC-ICPMS
198 installed at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). Prior
199 to analysis, the samples were carefully cleaned using a small diamond blade to remove any visible contamination
200 and sediment-filled cavities. The fragments were examined under a binocular microscope to ensure against the
201 presence of bioeroded zones and finally crushed into a coarse-grained powder with an agate mortar and pestle.
202 The powders (~60-100 mg) were transferred to acid cleaned Teflon beakers, ultrasonicated in MilliQ water,
203 leached with 0.1N HCl for ~ 15 s and finally rinsed twice with MilliQ water. The physically and chemically
204 cleaned samples were dissolved in 3-4 ml dilute HCl (~10%) and mixed with an internal triple spike with known
205 concentrations of ^{229}Th, ^{233}U and ^{236}U, calibrated against a Harwell Uraninite solution (HU-1) assumed to be at

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5

206 secular equilibrium. The solutions were evaporated to dryness at 70°C, redissolved in 0.6 ml 3N HNO_3 and then
207 loaded into 500 µl columns packed with Eichrom UTEVA resin to isolate uranium and thorium from the other
208 major and trace elements of the carbonate matrix. The U and Th separation and purification followed a
209 procedure slightly modified from Douville et al. (2010). The U and Th isotopes were determined following the
210 protocol recently revisited at LSCE (Pons-Branchu et al., 2014). The ^{230}Th/U ages were calculated from
211 measured atomic ratios through iterative age estimation (Ludwig and Titterington, 1994), using the ^{230}Th, ^{234}U
212 and ^{238}U decay constants of Cheng et al. (2013) and Jaffey et al. (1971). Due to the low ^{232}Th concentration (< 1
213 ng/g; see Table 1), no correction was applied for the non-radiogenic ^{230}Th fraction.
214
215   *3.2.2 Nd isotopic composition analyses on cold-water coral fragments*
216 Sub-samples of the CWC fragments from the Sardinia Channel used for U-series dating in this study (Table 1) as
217 well as sub-samples of the twenty-five CWC fragments originating from the Alboran Sea, which were already
218 radiocarbon-dated by Fink et al. (2013) (Table 2), were used for further Nd isotopic composition analyses. The
219 fragments (350 to 600 mg) were subjected to a mechanical and chemical cleaning procedure. The visible
220 contaminations, such as Fe-Mn coatings and detrital particles, were carefully removed from the inner and
221 outermost surfaces of the coral skeletons using a small diamond blade. The physically cleaned fragments were
222 ultrasonicated for 10 min with 0.1 N ultra-clean HCl, followed by several MilliQ water rinses and finally
223 dissolved in 2.5 N ultraclean HNO_3. Nd was separated from the carbonate matrix using Eichrom TRU and LN
224 resins, following the analytical procedure described in detail in Copard et al. (2010).
225 The ^{143}Nd/^{144}Nd ratios of all purified Nd fractions were analyzed using the ThermoScientific Neptune^{Plus} Multi-
226 Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) hosted at LSCE. The mass-
227 fractionation correction was made by normalizing ^{146}Nd/^{144}Nd to 0.7219 and applying an exponential law.
228 During each analytical session, samples were systematically bracketed with analyses of JNdi-1 and La Jolla
229 standard solutions, which are characterised by accepted values of 0.512115±0.000006 (Tanaka et al., 2000) and
230 0.511855±0.000007 (Lugmair et al., 1983), respectively. Standard JNdi-1 and La Jolla solutions were analysed
231 at concentrations similar to those of the samples (5-10 ppb) and all the measurements affected by instrumental
232 bias were corrected, when necessary, using La Jolla standard. The external reproducibility (2σ) for time resolved
233 measurement, deduced from repeated analyses of La Jolla and JNdi-1 standards, ranged from 0.1 to 0.5 εNd
234 units for the different analytical sessions. The analytical error for each sample analysis was taken as the external
235 reproducibility of the La Jolla standard for each session. Concentrations of Nd blanks were negligible compared
236 to the amount of Nd of CWC investigated in this study.
237
238 **3.3. Analyses on sediment of core SU92-33**
239   *3.3.1. Radiocarbon dating*
240 Radiocarbon dating was measured at UMS-ARTEMIS (Pelletron 3MV) AMS (CNRS-CEA Saclay, France).
241 Seven AMS radiocarbon (^{14}C) dating were performed in core SU92-33 on well-preserved calcareous tests of the
242 planktonic foraminifera *G. bulloides* in the size fraction >150 µm (Table 3). The age model for the core was
243 derived from the calibrated planktonic ages by applying a mean reservoir effect of ~400 years (Siani et al., 2000,
244 2001). All ^{14}C ages were converted to calendar years (cal. yr BP, BP = AD 1950) by using the INTCAL13
245 calibration data set (Reimer et al., 2013) and the CALIB 7.0 program (Stuiver and Reimer, 1993).

3.3.2. Stable isotopes

Stable oxygen (δ^{18}O) and carbon (δ^{13}C) isotope measurements were performed in core SU92-33 on well-preserved (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 and 19. The mean external reproducibility was checked by replicate analyses of laboratory standards and is better than ±0.07‰ (1σ) for δ^{18}O and ±0.04‰ for δ^{13}C.

3.3.3 Nd isotope measurements on planktonic foraminifera

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

3.3.4. Modern analogue technique (MAT)

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

4. Results

4.1. Cold-water coral~~s ages~~

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 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 ‰ reliability criterion and the U/Th ages can be considered strictly reliable. The coral ages range from 0.091±0.011 to 10.904±0.042 ka BP (Table 1), and reveal three distinct clusters of coral age distribution during the Holocene representing periods of sustained coral occurrence. These periods coincide with the Early Holocene

286 encompassing a 700-years-lasting time interval from ~10.9 to 10.2 ka BP, the very late Early Holocene at ~8.7
287 ka BP, and the Late Holocene starting at ~1.5 ka BP (Table 1).
288 Radiocarbon ages obtained for CWC samples collected in the Alboran Sea were published by Fink et al. (2013)
289 (Table 2). They also document three periods of sustained CWC occurrence coinciding with the Bølling–Allerød
290 (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
291 (5.4–0.3 cal ka BP).
292 The εNd record obtained from~~or~~ the CWC samples from the Alboran Sea displays a narrow range from -
293 9.22±0.30 to -8.59±0.3, which is comparable to the εNd record ~~obtained on~~of the planktonic foraminifera from
294 the Balearic Sea over the last 13.5 kyr~~a~~ (Table 2, Fig. 3b). Most of the CWC εNd values are similar within error
295 and the record does not reveal any clear difference over the last ~13.5 kyr~~a~~.
296 On the contrary, ~~Finally, t~~the CWC samples from the Sardinia Channel display a relatively large εNd range, with
297 values ~~values ranging~~varying from -5.99±0.50 to -7.75±0.10 during the Early and Late Holocene, and values as
298 low as -8.66±0.30 during the the mid-sapropel S1 deposition (S1a) at ~~(~~~8.7 ka BP~~)~~ (Table 1, Fig. 3c).

301 **4.2 ~~Chronological framework for core~~Core SU92-33**
302 The stratigraphy of core SU92-33 was derived from the δ^{18}O variations of the planktonic foraminifera
303 *G. bulloides* (Fig. 2b). The last glacial/interglacial transition and the Holocene encompasses the upper 2.1 m of
304 the core (Fig. 2b). The δ^{18}O record of *G. bulloides* shows higher values (~3.5 ‰) during the late glacial
305 compared to the Holocene (from ~1.5 to 0.8 ‰) exhibiting a pattern similar to those observed in nearby deep-sea
306 cores from the Western Mediterranean Sea (Sierro et al., 2005; Melki et al., 2009).
307 The age model for the upper 1.2 m of the core ~~of core~~ SU92-33 ~~is~~was based on 7 AMS-^{14}C age
308 measurements ~~for the upper 1.2 m of the core~~ and a ~~by a~~ linear interpolation between these ages (Table 3, Fig. 2).
309 For the lower portion of the core~~Below~~, a control point ~~has been~~was established ~~for~~at the onset of the last
310 deglaciation, which is~~that presents a~~ coeval ~~age~~ in the western and central Mediterranean Sea at ~~~about~~17 cal
311 ka BP (Sierro et al., 2005; Melki et al., 2009; Siani et al., 2001). Overall, ~~T~~the upper 2.1 m of core SU92-33
312 spans the last 19 kyr~~a~~, with an estimated average sedimentation rate ranging from ~15 cm ka^{-1} during the
313 deglaciation to ~10 cm ka^{-1} ~~between 9 to 15 cm ka^{-1}, with the lowest values observed~~ during the Holocene.

315 **4.3 SST reconstructions of core SU92-33**
316 April-May SST reconstruction was derived from MAT to define the main climatic events recorded in
317 core SU92-33 during the last 19 kyr~~a~~. SST~~s~~ vary from 8.5°C to 17.5°C with high amplitude variability over the
318 last 19 k~~a~~yr BP (Fig. 2a). The LGM (19-18 ka BP) is characterized by SST values centered at around 12°C.
319 Then, a progressive decrease of ~4°C between 17.8 ka and~~to~~ 16 ka marks the Henrich Stadial 1 (HS1) (Fig. 2a).
320 A warming phase (~14°C) between 14.5 ka BP and 13.8 ka BP coincides with the B-A interstadial and is
321 followed by a cooling (~11°C) between 13.1 ka BP and 11.8 ka BP largely corresponding to the YD (Fig. 2a).
322 During the Holocene, SST~~s~~ show mainly values of ~16°C, with one exception between 7 ka BP and 6 ka BP
323 pointing to an abrupt cooling of ~3°C (Fig. 2a). From the late glacial to the Holocene, SST variations show a
324 similar pattern to that previously observed in the Gulf of Lions and Tyrrhenian Sea (Kallel et al., 1997; Melki et
325 al., 2009) as well as in the Alboran Sea (Martrat et al., 2014; Rodrigo-Gámiz et al., 2014)~~ and~~. They are globally

synchronous for the main climatic transitions to the well dated South Adriatic Sea core MD90-917 (Siani et al., 2004) confirming the robustness of the SU92-33 age model (Fig. 2a).

4.4 Benthic stable oxygen and carbon isotope records of core SU92-33

The $\delta^{18}O$ and $\delta^{13}C$ records obtained from the benthic foraminifera *C. pachyderma* display significant variations at millennial time scales (Figs. 2c and 2d). The $\delta^{18}O$ 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. 2c), in agreement with results obtained fromor the neighbor core MD99-2343 (Sierro et al., 2005). The $\delta^{13}C$ record obtained fromof *C. pachyderma* shows a decreasing trend since the LGM with a low variability from ~1.6 ‰ to ~0.6 ‰ (Fig. 2d). The heaviest $\delta^{13}C$ values are related to the LGM (~1.6 ‰) while the lightest values (~0.6 ‰) characterize the Early Holocene and in particular the period corresponding to the sapropel S1 event in the eastern Mediterranean basin (Fig. 2d). The

4.5 Nd isotopic composition of planktonic foraminifera and cold-water corals

εNd values of planktonic foraminifera of core SU92-33 collected from the Balearic Sea vary within a relatively 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 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 generally more radiogenic (average -8.84±0.22; n=17) (Fig. 3b).

The εNd record obtained for the CWC samples from the Alboran Sea displays a narrow range from -9.22±0.30 to -8.59±0.3, which is comparable to the εNd record obtained on planktonic foraminifera from the Balearic Sea over the last 13.5 ka (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 ka.

Finally, the CWC samples from the Sardinia Channel display εNd values ranging from -5.99±0.50 to -7.75±0.10 during the Early and Late Holocene, and values as low as -8.66±0.30 during the the mid sapropel S1 deposition (S1a) (~8.7 ka BP) (Table 1, Fig. 3c).

5. Discussion

As first observationsOverall, the CWC and foraminiferal εNd values measured forin this study indicate 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. In additionFurthermore, the foraminiferal εNd record reveals an evolution towards more radiogenic values at intermediate water depth in the Balearic Sea over the last ~19 kyra (Fig. 3).

A prerequisite to properly interpret such εNd values differences and variations through time consists in characterizing first the present-day εNd of the main water-mass end-members circulating flowing in the western Mediterranean basin. This is possibleIt is also necessary toby evaluateing the temporal changes in εNd of the end-members since the LGM, and assessing the potential influences of lithogenic Nd input and regional exchange between the continental margins and seawater ("boundary exchange"; Lacan and Jeandel, 2001, 2005) on the εNd values of intermediate water masses.

9

365 During its westward flow, the LIW continuously mixes with surrounding waters with different εNd signatures

366 lying above and below. For the western Mediterranean basin, these water masses are the MAW/Western

367 Intermediate Water (WIW) and the TDW/WMDW, respectively. Accordingly, a well-defined and gradual εNd

368 gradient exists at intermediate depth between the eastern and western Mediterranean basins, with LIW values

369 becoming progressively more unradiogenic towards the Strait of Gibraltar, from -4.8±0.2 at 227 m in the

370 Levantine basin to -10.4±0.2 at 200 m in the Alboran Sea (Tachikawa et al., 2004). Such an εNd pattern implies

371 an effective vertical mixing with more unradiogenetic water masses along the E-W LIW trajectory ruling out

372 severe isotopic modifications of the LIW due to the local exchange between the continental margins and

373 seawater. Unfortunately, no information exists on the potential temporal variability in εNd of the Mediterranean

374 water-mass end-members since the LGM.

375 It has been demonstrated that eolian dust input can modify the surface and sub-surface εNd distribution of the

376 ocean in some areas (Arsouze et al., 2009). The last glacial period was associated with an aridification of North

377 Africa (Sarnthein et al., 1981; Hooghiemstra et al., 1987; Moreno et al., 2002; Wienberg et al., 2010) and higher

378 fluxes of Saharan dust to the NE tropical Atlantic (Itambi et al., 2009) and the western Mediterranean Sea

379 characterized by unradiogenic εNd values (between -1011±0.4 and -1714±0.4; Grousset et al., 1992, 1998;

380 Grousset and Biscaye, 2005; see synthesis in Scheuvens et al., 2013). Bout-Roumazeilles et al. (2013)

381 documented a dominant role of eolian supply in the Siculo-Tunisian Strait during the last 20 ka, with the

382 exception of a significant riverine contribution (from the Nile River) and a strong reduction of eolian input

383 during the sapropel S1 event. Such variations in the eolian input to the Mediterranean Sea are not associated to a

384 significant change in the seawater εNd record obtained for the Balearic Sea (core SU92-33) during the sapropel

385 S1 event (Fig. 3). Furthermore, the εNd signature of the CWC from the Sardinia Channel (core RECORD 23)

386 shifts to more unradiogenic values (-8.66±0.30) during the sapropel S1 event, which is opposite to what expected

387 if it was related to a strong reduction of eolian sediment input. In a recent study, addition, Rodrigo-Gámiz et al.

388 (2015) have documented variations in the terrigenous provenance from a sediment record in the Alboran Sea

389 (core 293G; 36°10.414'N, 2°45.280'W, 1840 m water depth) since the LGM. Radiogenic isotopes (Sr, Nd, Pb)

390 point to changes from North African dominated sources during the glacial period to European dominated source

391 during the Holocene. Nevertheless, the major Sr-Nd-Pb excursions documented by Rodrigo-Gámiz et al. (2015)

392 and dated at ca. 11.5, 10.2, 8.9-8.7, 5.6, 2.2 and 1.1. ka cal BP do not seem to affect the εNd values of our

393 foraminifera and coral records.

394 Thus, allTaken together, these results suggest that changes of eolian dust input since the LGM were not

395 responsible for the observed εNd variability at intermediate water depths.

396 Consequently, assuming that the Nd isotopic budget of the western Mediterranean Sea has not been strongly

397 modified since the LGM, the reconstructed variations of the E-W gradient of εNd values in the western

398 Mediterranean Sea for the past and notably during the sapropel S1 event (Fig. 3) are indicative of a major

399 reorganization of intermediate water circulation.

400

401 *5.1 Hydrological changes in the Alboran and Balearic Seas since the LGM*

402 The range in εNd for the CWC from the Alboran Sea (from -9.22±0.30 to -8.8.59±0.30; Table 2) is very close to

403 the one obtained for the planktonic foraminifera from the Balearic Sea (from -9.50±0.30 to -8.61±0.30; Table 4,

404 Fig. 3c), suggesting that both sites are influenced by the same intermediate water masses at least for the last 13.5

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10

405   kyra BP. Today, LIW occupies a depth range between ~200 and ~700 m in the western Mediterranean basin

406   (Millot, 1999; Sparnocchia et al., 1999). More specifically, the salinity maximum corresponding to the core of

407   LIW is found at around 400 m in the Alboran Sea (Millot, 2009) and up to 550 m in the Balearic Sea (López-

408   Jurado et al., 2008). The youngest CWC sample collected in the Alboran Sea with a rather "recent" age of 0.34

409   cal ka BP (Fink et al. 2013) displays an εNd value of -8.59±0.30 (Table 2) that is similar to the present-day value

410   of the LIW at the same site (-8.3±0.2) (Dubois-Dauphin et al., submitted) and is significantly different from the

411   WMDW εNd signature in the Alboran Sea (-10.7±0.2, 1270 m water depth; Tachikawa et al., 2004). Considering

412   the intermediate depth range of the studied CWC and foraminifera samples, we can reasonably assume that

413   samples from both sites, in the Balearic Sea (622 m water depth) and in the Alboran Sea (280 to 442 m water

414   depth), record εNd variations of the LIW. The εNd record obtained from~~on~~ planktonic foraminifera generally

415   displays more unradiogenic and homogenous values before ~13 cal ka BP (range from~~:~~ -9.46 to -9.12) compared

416   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

417   and Late Holocene.

418   The SST record displays values centered at around 12°C during the LGM with a subsequent rapid SST decrease

419   towards 9°C , highlighting the onset of the HS1 (Fig. 2a). These values are well comparable to recent high-

420   resolution SST data obtained in the Alboran Sea (Martrat et al., 2014; Rodrigo-Gámiz et al., 2014).

421   The δ^{18}O record obtained on *G. bulloides* indicates an abrupt 1‰ excursion towards lighter values centered at

422   about 16 cal ka BP (Table 4), synchronous with the HS1 (Fig. 2b), which is similar to the δ^{18}O shift reported by

423   Sierro et al. (2005) for a core collected at 2391 m water depth NE of the Balearic Islands (MD99-2343; Fig. 1).

424   As the Heinrich events over the last glacial period are characterized by colder and fresher surface water in the

425   Alboran Sea (Cacho et al., 1999; Pérez-Folgado et al., 2003; Martrat et al., 2004, 2014; Rodrigo-Gámiz et al.,

426   2014) and dry climate on land over the western Mediterranean Sea (Allen et al., 1999; Combourieu-Nebout et

427   al., 2002; Sanchez Goni et al., 2002; Bartov et al., 2003), lighter δ^{18}O values of planktonic *G. bulloides* are

428   thought to be the result of the inflow of freshwater derived from the melting of icebergs in the Atlantic Ocean

429   into the Mediterranean Sea (Sierro et al., 2005; Rogerson et al., 2008).

430   During this time interval, the δ^{13}C record of *C. pachyderma* from the Balearic Sea (core SU92-33) displays a

431   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

432   obtained on benthic foraminifera *C. pachyderma* from the deep Balearic Sea (core MD99-2343) reveals similar

433   δ^{13}C values before ~16 cal ka BP suggesting well-mixed and ventilated water masses during the LGM and the

434   onset of the deglaciation (Sierro et al., 2005).

435   The slightly lower foraminiferal εNd values before ~13 cal ka BP could reflect a stronger influence of water

436   masses deriving from the Gulf of Lions as WMDW (εNd: -9.4±0.9; Henry et al., 1994; Tachikawa et al., 2004;

437   Montagna et al., in prep). This is in agreement with εNd results obtained by Jiménez-Espejo et al. (2015) from

438   planktonic foraminifera collected from deep-water sites (1989 m and 2382 m) in the Alboran Sea (Fig. 4c).

439   Jiménez-Espejo et al. (2015) documented lower εNd values (ranging from -10.14±0.27 to -9.58±0.22) during the

440   LGM, suggesting an intense deep-water formation. This is also associated to an enhanced activity of the deeper

441   branch of the MOW in the Gulf of Cádiz (Rogerson et al., 2005; Voelker et al., 2006) linked to the active

442   production of the WMDW in the Gulf of Lions during the LGM (Jiménez-Espejo et al., 2015).

443   The end of the HS1 (14.7 cal ka BP) is concurrent with the onset of the B-A warm interval characterized by

444   increased SST up to ~~(14°C)~~ in the Balearic Sea (SU92-33; Fig. 3a), also identified for various sites in the

Mediterranean Sea (Cacho et al., 1999; Martrat et al., 2004, 2014; Essallami et al., 2007; Rodrigo-Gámiz et al., 2014)., in agreement with the SST record obtained for the Balearic Sea (SU92-33: Fig. 3a). The B-A interval is associated withto the so-called melt-water pulse 1A (e.g. Weaver et al., 2003) occurring 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 freshwater discharges in the Atlantic Ocean due to the melting of continental ice sheets (Deschamps et al., 2012), resulting in an enhanced Atlantic inflow across the Strait of Gibraltar. Synchronously, cosmogenic dating of Alpine glacier retreat throughout the western Mediterranean hinterland suggests maximum retreat rates (Ivy-Ochs et al., 2007; Kelly et al., 2006). Overall, these events are responsible for freshening Mediterranean waters and reduced surface water density, and hence, weakened ventilation of intermediate (Toucanne et al., 2012) and deep-water masses (Cacho et al., 2000; Sierro et al., 2005). Similarly, lower benthic δ^{13}C values obtained for the Balearic Sea (Fig. 4a) point to less ventilated intermediate water relative to the late glacial. In addition, a decoupling in the benthic δ^{13}C values is observed between deep (MD99-2343) and intermediate (core SU92-33) waters after ~16 cal ka BP (Sierro et al. 2005), suggesting an enhanced stratification of the waters masses (Fig. 4a). At this time, the shallowest εNd record from the deep Alboran Sea (core 300G) shifted towards more radiogenic values, while the deepest one (core 304G) remained close to the LGM values (Jimenez-Espejo et al., 2015) (Fig. 4c). Furthermore, results from the UP10 fraction (particles > 10 μm) of the MD99-2343 sediment core (Fig. 4d), indicate a declining bottom-current velocity at 15 ka BP (Frigola et al., 2008). Rogerson et al. (2008) have hypothesized that during deglacial periods the sinking depth of dense waters produced in the Gulf of Lions was shallower resulting in new intermediate water (WIW) rather than new deep-water (WMDW) as observed today during mild winters (Millot, 1999; Schott et al., 1996). Therefore, intermediate depths of the Balearic Sea could have been isolated from the deep-water with the onset of the T1 (at ~15 ka BP). The reduced convection in the deep western Mediterranean Sea together with the shoaling of the nutricline (Rogerson et al., 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 silt record from the Tyrrhenian Sea (Toucanne et al., 2012) (Fig. 4e). Deeper depths of the Alboran Sea also record a stronger influence of the LIW with an εNd value of -9.1±0.4 (Jimenez-Espejo et al., 2015). In addition, 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. 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 εNd values from intermediate water depths in the Balearic Sea (core SU92-33) remain high (between -9.15±0.3 and -8.61±0.3) (Fig. 4b). On the other hand, the deeper Alboran Sea provides a value of -9.8±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).

485

5.2 Hydrological changes in the Sardinia Channel during the Holocene

486

487 The present-day hydrographic structure of the Sardinia Channel is characterized by four water masses, with the
488 surface, intermediate and deep-water masses being represented by MAW, LIW and TDW/WMDW, respectively
489 (Astraldi et al., 2002a; Millot and Taupier-Lepage, 2005). In addition, the WIW, flowing between the MAW and
490 the LIW, has also been observed along the Channel (Sammari et al., 1999). The core of the LIW is located at
491 400-450 m water depth in the Tyrrhenian Sea (Hopkins, 1988; Astraldi et al., 2002b), which is the depth range of
492 CWC samples from the Sardinia Channel (RECORD 23; 414 m) (Taviani et al., 2015). The youngest CWC
493 sample dated at ~0.1 ka BP has an εNd value of -7.70±0.10 (Table 1, Fig. 5), which is similar within error to the
494 value obtained from a seawater sample collected at 451 m close to the coral sampling location (-8.0±0.4;
495 Montagna et al., in prep).
496 The CWC dating from the Sardinia Channel shows three distinct periods of sustained coral occurrence in this
497 area during the Holocene, with each displaying a large variability in εNd values. CWC from the Early Holocene
498 (10.9-10.2 ka BP) and the Late Holocene (<1.5 ka BP) exhibit similar ranges of εNd values (ranging from -
499 5.99±0.50 to -7.75±0.20; Table 1, Fig 5c). Such variations are within the present-day εNd range being
500 characteristic for intermediate waters in the eastern Mediterranean Sea (-6.6±1.0; Tachikawa et al., 2004; Vance
501 et al., 2004). However, the CWC εNd values are more radiogenic than those observed at mid-depth in the
502 present-day western basin (ranging from -10.4±0.2 to -7.58 ±0.47; Henry et al., 1994; Tachikawa et al., 2004;
503 Montagna et al., in prep), suggesting a stronger LIW component in the Sardinia Channel during the Early and
504 Late Holocene. The Sardinian CWC εNd variability also reflects the sensitivity of the LIW to changes in the
505 eastern basin such as rapid variability of the Nile River flood discharge (Revel et al., 2014; 2015; Weldeab et al.,
506 2014) or a modification through time in the proportion between the LIW and the Cretan Intermediate Water
507 (CIW). Today, the intermediate water outflowing from the Strait of Sicily is composed by ~66 to 75 % of LIW
508 and 33 to 25 % of CIW (Manca et al., 2006; Millot, 2014). As the CIW is formed in the Aegean Sea, this
509 intermediate water mass is generally more radiogenic than LIW (Tachikawa et al., 2004; Montagna et al., in
510 prep). Following this hypothesis, a modification of the mixing proportion between the CIW and the LIW may
511 potentially explain values as radiogenic as about -6 in the Sardinia Channel during the Early and Late Holocene
512 (Fig. 5c). However, a stronger LIW and/or a CIW contribution cannot be responsible for εNd values as low as -
513 8.66±0.30 observed during the sapropel S1 event at 8.7 ka BP (Table 1, Fig. 5c). Considering that such
514 unradiogenic value is not observed at intermediate depth in the modern eastern Mediterranean basin, the most
515 plausible hypothesis suggested here is that the CWC were ~~bathed~~ influenced by a higher contribution of
516 intermediate water from the western basin ~~in intermediate waters~~ ~~which were more marked by the western~~
517 ~~basin.~~

518

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

519

520 The εNd records of the Balearic Sea, Alboran Sea and Sardinia Channel document a temporal variability of the
521 east-west gradient in the western Mediterranean basin during the Holocene. The magnitude of the gradient
522 ranges from ~1.5 to ~3 ε units during the Early and Late Holocene and it is strongly reduced at 8.7 ka BP
523 coinciding with the sapropel S1 event affecting the eastern Mediterranean basin (Fig. 5). Such variations could

be the result of a modification of the Nd isotopic composition of intermediate water masses due to ~~intensity~~
changes of the ~~the~~ LIW production through time and a higher contribution of the western-sourced intermediate
water towards the Sardinia Channel coinciding with the sapropel S1 event.

The LIW acquires its radiogenic εNd in the Mediterranean Levantine basin mainly from Nd exchange between
seawater and lithogenic particles originating mainly from Nile River (Tachikawa et al., 2004). A higher sediment
supply from the Nile River starting at ~15 ka BP was documented by a shift to more radiogenic εNd values of
the terrigenous fraction obtained from a sediment core having been influenced by the Nile River discharge
(Revel et al., 2015) (Fig. 5e). ~~However, O~~others studies pointed to a gradual enhanced Nile River runoff as soon
as 14.8 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 8.4 ka (>120 cm/ka) (Revel et al., 2015; Weldeab et al., 2014; Castaneda et al., 2016). Similarly,
enhanced Nile discharge at ~9.5 cal kyr B.P was inferred based on $\delta^{18}O$ in planktonic foraminifera from a
sediment core in the southeast Levantine Basin (PS009PC (32°07.7′N, 34°24.4′E; 552 m water depth)
(Hennekam et al., 2014). Th~~is~~e increasing~~e in~~ contribution of the Nile River ~~discharge~~ to the eastern
Mediterranean basin has been related to the African Humid Period (14.8–5.5 ka BP; Shanahan et al., 2015),
which in turn was linked to the precessional increase in Northern Hemisphere insolation during low eccentricity
(deMenocal et al., 2000; Barker et al., 2004; Garcin et al., 2009). An increasing amount of radiogenic sediments
dominated by the Blue/Atbara Nile River contribution (Revel et al., 2014) could have modified the εNd of
surface water towards more radiogenic values (Revel et al., ~~in prep~~). Indeed, planktonic foraminifera εNd values
as high as ~ -3 have been documented in the eastern Levantine Basin (ODP site 967; 34°04.27′N, 32°43.53′E;
2553 m water depth) during the sapropel S1 event as a result of enhanced Nile flooding (Scrivner et al., 2004).
Th~~is~~e radiogenic signature was likely transferred to intermediate depth as a consequence of the LIW formation in
the Rhodes Gyre, and it might have been propagated westwards towards the Sardinia Channel.

~~The Nile River runoff was also strongly enhanced during the sapropel S1 event (Revel et al., 2010; Weldeab et al., 2014; Revel et al., 2014). Based on δ¹⁸O ruber record from a site in the southeast Levantine Basin (PS009PC (32°07.7′N, 34°24.4′E; 552 m water depth), Hennekam et al. (2014) have documented a maximum Nile discharge at ~9.5 cal kyr B.P.~~ Scrivner et al. (2004) have reported very high foraminifera εNd values (-3 to -3.5)
~~corresponding to the sapropel S1 event in the eastern Levantine Basin (ODP site 967; 34°04.27′N, 32°43.53′E; 2553 m water depth), pointing to a maximum Nile discharge at this time.~~ ~~Hence~~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 produced in the eastern basin
(Rohling, 1994) by less radiogenic western intermediate water (possibly WIW). Such a scenario could even
support previous hypotheses ~~that invoke~~ a potential circulation reversal in the eastern Mediterranean from anti-
estuarine to estuarine during sapropel formation (Huang and Stanley, 1972; Calvert, 1983; Sarmiento et al.,
1988; Buckley and Johnson, 1988; Thunell and Williams, 1989). An alternative ~~explanation~~hypothesis would be
that reduced surface ~~-~~water densities in the eastern Mediterranean during sapropel S1 resulted in the LIW sinking
to shallower depths ~~during sapropel time~~ than at present. ~~In this case~~As a result of this shoaling, CWC from the
Sardinia Channel ~~during the sapropel S1a event~~ would have been bathed by underlying ~~w~~Western ~~i~~Intermediate
~~w~~Water ~~-~~during the sapropel S1a event.

6. Conclusions

The foraminiferal εNd record from ~~the~~ intermediate <u>depths in the</u> Balearic Sea reveals a relatively narrow range of εNd values varying between -9.50 and -8.61 since the LGM (~20 ka). Between 18 and 13.5 cal ka BP, the more unradiogenic εNd values support a vigorous deep overturning in the Gulf of Lions while δ^{18}O and δ^{13}C values indicate a stratification of the water masses after 16 cal ka BP. The stratification together with a decrease of the deep-water intensity led to more radiogenic values after ~13 cal ka BP. The <u>foraminiferal</u> εNd record ~~from planktonic foraminifera~~, ~~supplemented~~ <u>supported</u> by <u>εNd values from</u> CWC ~~from the intermediate depths of~~<u>in</u> the Alboran Sea, show<u>s</u> only minor changes in <u>neodymium isotopes</u> ~~εNd values~~ from 13.5 cal ka BP to 0.34 cal ka BP, suggesting that the westernmost part of the western Mediterranean basin is not very sensitive to hydrological variations of the LIW.

On the contrary, CWC located at the depth of the LIW in the Sardinia Channel ~~indicate~~ <u>exhibit</u> ~~high amplitude~~<u>large</u> <u>εNd</u> variations ~~of the εNd values~~ (between -7.75±0.10 and -5.99±0.50) during the Holocene, ~~which could highlight~~<u>suggesting</u> either the role of the Nile River in changing the εNd of the LIW in the eastern Mediterranean basin or a ~~different~~ <u>variable</u> LIW/CIW mixing of the water outflowing from the Strait of Sicily. ~~Coinciding~~ <u>At the time of the</u>~~with the~~ sapropel S1 event at ~8.7 ka BP, CWC display a shift toward lower values (-8.66±0.30), similar to those ~~obtained~~ <u>,</u> at intermediate depths in the westernmost part of the western basin. This suggests that western-sourced intermediate water likely filled mid-depth of the southern Sardinia, replacing LIW that was no longer produced (or heavily reduced) in the eastern basin. These results could potentially support a reversal of the Mediterranean circulation, although this assumption needs further investigation to be confirmed.

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1014 **Table captions**

1015

1016 **Table 1**. U-series ages and εNd values obtained for cold-water coral samples collected from sediment core RECORD 23
1017 (Sardinia Channel).

1018

1019 **Table 2**. εNd values obtained for cold-water corals from the southern Alboran Sea. The AMS ^{14}C ages published by Fink et
1020 al. (2013) are also reported as Median probability age (ka BP).

1021

1022 **Table 3**. AMS ^{14}C ages of samples of the planktonic foraminifer *G. bulloides* from 'off-mound' sediment core SU92-33. The
1023 AMS ^{14}C ages were corrected for ^{13}C and a mean reservoir age of 400 yrs, and were converted into calendar years using the
1024 INTCAL13 calibration data set (Reimer et al., 2013) and the CALIB 7.0 program (Struiver et al., 2005).

1025

1026 **Table 4**. Multiproxy data obtained for the upper 2.1 m of sediment core SU92-33 (Balearic Sea). Stable oxygen and carbon
1027 isotopes were measured on benthic (*C. pachyderma*) and planktonic (*G. bulloides*) foraminifera; εNd values were obtained on
1028 mixed planktonic foraminifera samples. The age results from a combination of 7 AMS-^{14}C age measurements for the upper
1029 1.2 m of the core and by a linear interpolation between these ages as well as the δ^{18}O variations of the planktonic
1030 foraminifera *G. bulloides.*

1031

1032 **Figure captions**

1033

1034 **Figure 1**. Map of the western Mediterranean Sea showing the locations of samples investigated in this study. Yellow dot
1035 indicates the sampling location of the sediment core from the Balearic Sea (SU92-33); yellow stars indicate the locations of
1036 the CWC-bearing cores from the Sardinia Channel (RECORD 23) and the southern Alboran Sea (for further details on the
1037 CWC from the Alboran Sea refer also to Fink et al., 2013). The cores discussed in this paper (Gulf of Cádiz: IODP site
1038 U1387, Balearic Sea: MD09-2343, northern Tyrrhenian Sea: MD01-2472, Adriatic Sea: MD90-917) are indicated by black
1039 dots, and seawater stations are marked by open squares. Arrows represent the main oceanographic currents. The black line
1040 shows the general trajectory of the Modified Atlantic Water (MAW) flowing at the surface from the Atlantic Ocean toward
1041 the western and eastern Mediterranean. The orange line represents the Levantine Intermediate Water (LIW) originating from
1042 the eastern basin. The black dashed line shows the trajectory of the Western Mediterranean Deep Water (WMDW) flowing
1043 from the Gulf of Lions toward the Strait of Gibraltar.

1044

1045 **Figure 2**. (a) Sea Surface Temperature (SST) records of cores SU92-33 (red line) and MD90-917 (green line; Siani et al.,
1046 2004), (b) δ^{18}O record obtained on planktonic foraminifer *G. bulloides* for core SU92-33, (c) δ^{18}O record obtained on benthic
1047 foraminifer *C. pachyderma* for core SU92-33, (d) δ^{13}C record obtained on benthic foraminifer *C. pachyderma* for core SU92-
1048 33. LGM: Last Glacial Maximum; HS1: Heinrich Stadial 1; B-A: Bølling-Allerød; YD: Younger Dryas. Black triangles
1049 indicate AMS ^{14}C age control points.

1050

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).

Figure 4. (a) δ^{13}C records obtained on benthic foraminifer *C. pachyderma* for cores SU92-33 (red line) and MD99-2343 (blue line; Sierro et al., 2005). (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). Modern εNd values for LIW (orange dashed line) and WMDW (blue dashed line) are also reported for comparison. (c) εNd values obtained for planktonic foraminifera with Fe-Mn coatings at sites 300G (36°21.532' N, 1°47.507' W; 1860 m; open dots) and 304G (36°19.873' N, 1°31.631' W; 2382 m; black dots) in Alboran Sea (Jimenez-Espejo et al., 2015). (d) UP10 fraction (>10 μm) from core MD99-2343 (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 site U1387 (36°48.3' N 7°43.1' W; 559 m) (Bahr et al., 2015).

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) εNd values of cold-water corals from core RECORD 23 (Sardinia Channel), (d) εNd 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) εNd values obtained on terrigenous fraction of MS27PT located close the Nile River mouth in the eastern Mediterranean basin (Revel et al., 2015).