

RESEARCH ARTICLE

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Key Points:

- Hydrological changes in the intermediate water of the southern Gulf of Cadiz over the past 40 ka
- Rapid changes of AAIW northward penetration tracked by Neodymium isotopes
- Neodymium isotopic composition of the Mediterranean end-member over the past 45 ka

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## South Atlantic intermediate water advances into the North-east Atlantic with reduced Atlantic meridional overturning circulation during the last glacial period

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**Abstract** The Nd isotopic composition ( $\epsilon_{\text{Nd}}$ ) of seawater and cold-water coral (CWC) samples from the Gulf of Cádiz and the Alboran Sea, at a depth of 280–827 m were investigated in order to constrain mid-depth water mass dynamics within the Gulf of Cádiz over the past 40 ka.  $\epsilon_{\text{Nd}}$  of glacial and Holocene CWC from the Alboran Sea and the northern Gulf of Cádiz reveals relatively constant values (−8.6 to −9.0 and −9.5 to −10.4, respectively). Such values are similar to those of the surrounding present-day middepth waters from the Mediterranean Outflow Water (MOW;  $\epsilon_{\text{Nd}} \sim -9.4$ ) and Mediterranean Sea Water (MSW;  $\epsilon_{\text{Nd}} \sim -9.9$ ). In contrast, glacial  $\epsilon_{\text{Nd}}$  values for CWC collected at thermocline depth (550–827 m) in the southern Gulf of Cádiz display a higher average value ( $-8.9 \pm 0.4$ ) compared to the present-day value ( $-11.7 \pm 0.3$ ). This implies a higher relative contribution of water masses of Mediterranean (MSW) or South Atlantic origin (East Antarctic Intermediate Water, EAAIW). Our study has produced the first evidence of significant radiogenic  $\epsilon_{\text{Nd}}$  values ( $\sim -8$ ) at 19, 23–24, and 27 ka, which are coeval with increasing iceberg discharges and a weakening of Atlantic Meridional Overturning Circulation (AMOC). Since MOW  $\epsilon_{\text{Nd}}$  values remained stable during the last glacial period, it is suggested that these radiogenic  $\epsilon_{\text{Nd}}$  values most likely reflect an enhanced northward propagation of glacial EAAIW into the eastern Atlantic Basin.

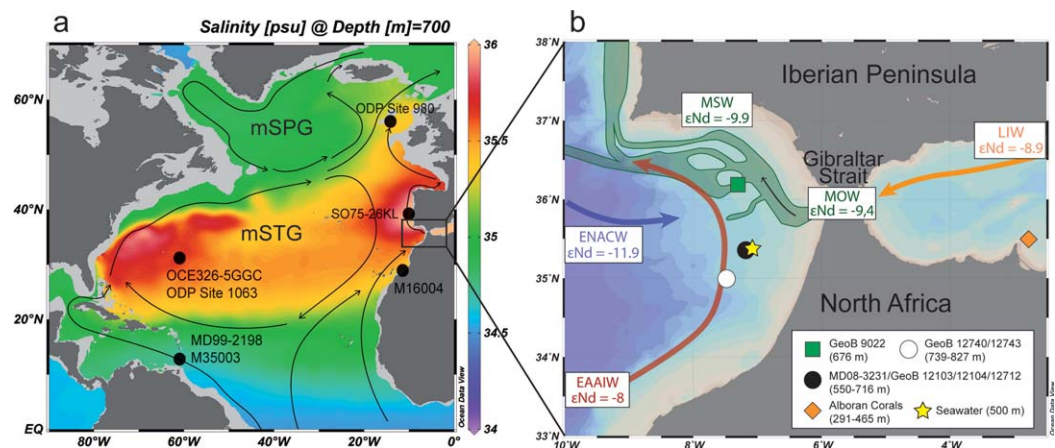
### 1. Introduction

The variability of the Atlantic Meridional Overturning Circulation (AMOC) affects the climate through the redistribution of heat and salinity from low to high latitudes by surface and intermediate currents, and the capacity of the ocean to store CO<sub>2</sub> [Bryden and Imawaki, 2001; Ganopolski and Rahmstorf, 2001; Sabine *et al.*, 2004]. It has been shown that the overturning circulation has been slowdown during the glacial period and was associated with the formation of shallower northern-sourced water (Glacial North Atlantic Intermediate Water, GNAIW) [Boyle and Keigwin, 1987; Zahn *et al.*, 1987; Ganopolski and Rahmstorf, 2001; McManus *et al.*, 2004; Böhm *et al.*, 2015] and the filling of a large part of the deep North Atlantic with southern-sourced water [Lynch-Stieglitz *et al.*, 2007; Roberts *et al.*, 2010]. The AMOC is known to have been particularly sluggish around the catastrophic iceberg discharge of the coldest Heinrich stadials (HS) [Bond *et al.*, 1993] and the Dansgaard-Oeschger stadials (D-O stadials) [Dansgaard *et al.*, 1993]. These slowdowns in the AMOC were associated with a reduction in North Atlantic Deep Water (NADW) formation, which was most likely driven by enhanced freshwater discharge from collapsing northern hemisphere ice sheets [e.g., Vidal *et al.*, 1997; Labeyrie *et al.*, 1999; Curry *et al.*, 1999; Elliot *et al.*, 2002; McManus *et al.*, 2004; Böhm *et al.*, 2015; Barker *et al.*, 2015] and which led to a northward penetration of deep and intermediate water masses from the southern ocean [Oppo and Lehman, 1993; Sarin *et al.*, 1994; Elliot *et al.*, 2002; Lynch-Stieglitz *et al.*, 2007; Roberts *et al.*, 2010; Böhm *et al.*, 2015].

Today, middepth water masses play an important role as intermittent carbon and heat storage reservoirs [Bower *et al.*, 2002] that sequester significant quantities of anthropogenic CO<sub>2</sub> [Sabine *et al.*, 2004]. However, little is known about the glacial and deglacial intermediate depth circulation patterns. Despite receiving increased attention over recent decades [Pahnke and Zahn, 2005; Pahnke *et al.*, 2008; Came *et al.*, 2008; Basak *et al.*, 2010; Xie *et al.*, 2012; Pena *et al.*, 2013; Huang *et al.*, 2014], the penetration of the Antarctic Intermediate Waters (AAIW) into the North Atlantic Ocean, associated with elevated Heinrich stadial fresh-water inputs, is still poorly constrained. Conflicting results have been obtained for the northward penetration of the AAIW into the western subtropical North Atlantic during the last glacial period. Indeed, while several studies have documented a greater northward penetration of AAIW into the western subtropical North Atlantic at times of iceberg discharge and reduced AMOC [e.g., Zahn and Stüber, 2002; Pahnke *et al.*, 2008], other studies propose a reduced northward flow of AAIW during the same intervals [Came *et al.*, 2008; Xie *et al.*, 2012; Huang *et al.*, 2014]. Based on evidence for enhanced northward propagation of the AAIW in the Pacific Ocean during the same cold events [Pahnke and Zahn, 2005; Basak *et al.*, 2010], it has been suggested that the flow of AAIW could have been rerouted into the Pacific Ocean instead of the North Atlantic [Huang *et al.*, 2014]. Nevertheless, several studies investigating the northward penetration of East Antarctic Intermediate Water (EAAIW) have indicated that this water mass reached the midlatitude northeast Atlantic (30°N) during glacial periods [Willamowski and Zahn, 2000; Montero-Serrano *et al.*, 2011]. The Gulf of Cádiz is a crucial area for studying middepth Atlantic hydrology as these studies have proposed that it delineates a hydrological front during glacial periods where mixing of northern (Eastern North Atlantic Central Water, ENACW) and southern-sourced (EAAIW) intermediate water masses occurred. In addition, the northern Gulf of Cádiz is also on the pathway of the Mediterranean Outflow Water (MOW). It has been suggested that variability in MOW during the last glacial period may have played a significant role in triggering a switch from a stadial to an interstadial mode by injecting high-salinity waters into the Northern North Atlantic at a time of weak AMOC [Johnson, 1997; Voelker *et al.*, 2006] and when MOW outflow was stronger [Voelker *et al.*, 2006; Toucanne *et al.*, 2007; Bahr *et al.*, 2015].

Cold-water corals (CWCs) have been shown to act as useful archives for identifying rapid changes in the dynamics of intermediate water masses [Lutringer *et al.*, 2005; van de Flierdt *et al.*, 2010; Copard *et al.*, 2010, 2011; Montero-Serrano *et al.*, 2013]. Their aragonitic skeleton can be precisely dated thanks to U-series disequilibrium methods [Adkins *et al.*, 1998; Cheng *et al.*, 2000; Douville *et al.*, 2010]. In addition, it has been shown that the Nd isotopic composition, expressed as  $\epsilon\text{Nd} = \left( \frac{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{sample}}}{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}} - 1 \right) \times 10,000$  (CHUR: Chondritic Uniform Reservoir) [Jacobsen and Wasserburg, 1980], of living CWC from the Strait of Gibraltar to the Norwegian Sea faithfully traces local seawater  $\epsilon\text{Nd}$  and middepth water mass provenance for the Northeast Atlantic [Colin *et al.*, 2010; Copard *et al.*, 2010, 2012; Montero-Serrano *et al.*, 2013]. For the Northeast Atlantic Ocean, the northern-sourced waters, which correspond to a mixing between Labrador slope waters and the north eastward-spreading, middepth waters (forming the middepth Subpolar Gyre or mSPG), carry  $\epsilon\text{Nd}$  values between  $-13$  and  $-15$  [Piepgras and Wasserburg, 1987; Lacan and Jeandel, 2004]. Conversely, the  $\epsilon\text{Nd}$  values of southern-sourced waters (notably, Antarctic bottom water—AABW and AAIW) range between  $-7$  and  $-9$  in the Southern Ocean [Jeandel, 1993; Stichel *et al.*, 2012]. Thus, northern and southern-sourced waters can be distinguished by studying  $\epsilon\text{Nd}$  values along a north-south basin-scale transect [Von Blanckenburg, 1999]. In the middepth ocean, the only way to alter the initial Nd isotopic composition of water masses is to add Nd with a different isotopic composition through riverine and eolian inputs, boundary exchange, and reversible scavenging or by the mixing of isotopically different water masses [e.g., Lacan and Jeandel, 2005]. Away from major lithogenic Nd sources,  $\epsilon\text{Nd}$  can be considered as a quasi-conservative water mass tracer in the ocean with a residence time of  $\sim 600$ – $1000$  years [Tachikawa *et al.*, 2003].

In the Northeast Atlantic, framework-forming CWCs are fairly abundant although their spatial distribution varies over time [e.g., Schröder-Ritzrau *et al.*, 2005; Wienberg *et al.*, 2010; Frank *et al.*, 2011; Eisele *et al.*, 2011]. Here, we present the first submillennial  $\epsilon\text{Nd}$  record for the last glacial period (MIS 2 from 40 to 15 ka) derived from CWC from the southern Gulf of Cádiz. This record was compared to ambient seawater  $\epsilon\text{Nd}$  in order to assess the glacial to present day variability of middepth water  $\epsilon\text{Nd}$ . CWC from the Alboran Sea have also been investigated for the first time in order to establish the Nd isotopic composition of the MOW during glacial and interglacial periods. Our objective is to track rapid (centennial-scale) changes in water mass provenance and to evaluate whether the circulation of water masses at middepths in the eastern



**Figure 1.** (a) Salinity (psu) contour map of the Atlantic Ocean at 700 m water depth (Ocean Data View; WOA 2013 data set). Black dots indicate the locations of cores discussed in this paper. Arrows show the general surface and intermediate-water mass circulation of the Atlantic Ocean. mSPG: middepth Subpolar Gyre; mSTG: middepth Subtropical Gyre. (b) Locations of cold-water corals and seawater station studied here. Simplified distribution of intermediate water mass circulations and their Nd isotopic composition within the Gulf of Cádiz are also reported.

North Atlantic has changed significantly during abrupt climate variations over the last glacial period. Our results also provide the opportunity to constrain the variability of the northward penetration of EAAIW, which was associated with abrupt variations in the AMOC.

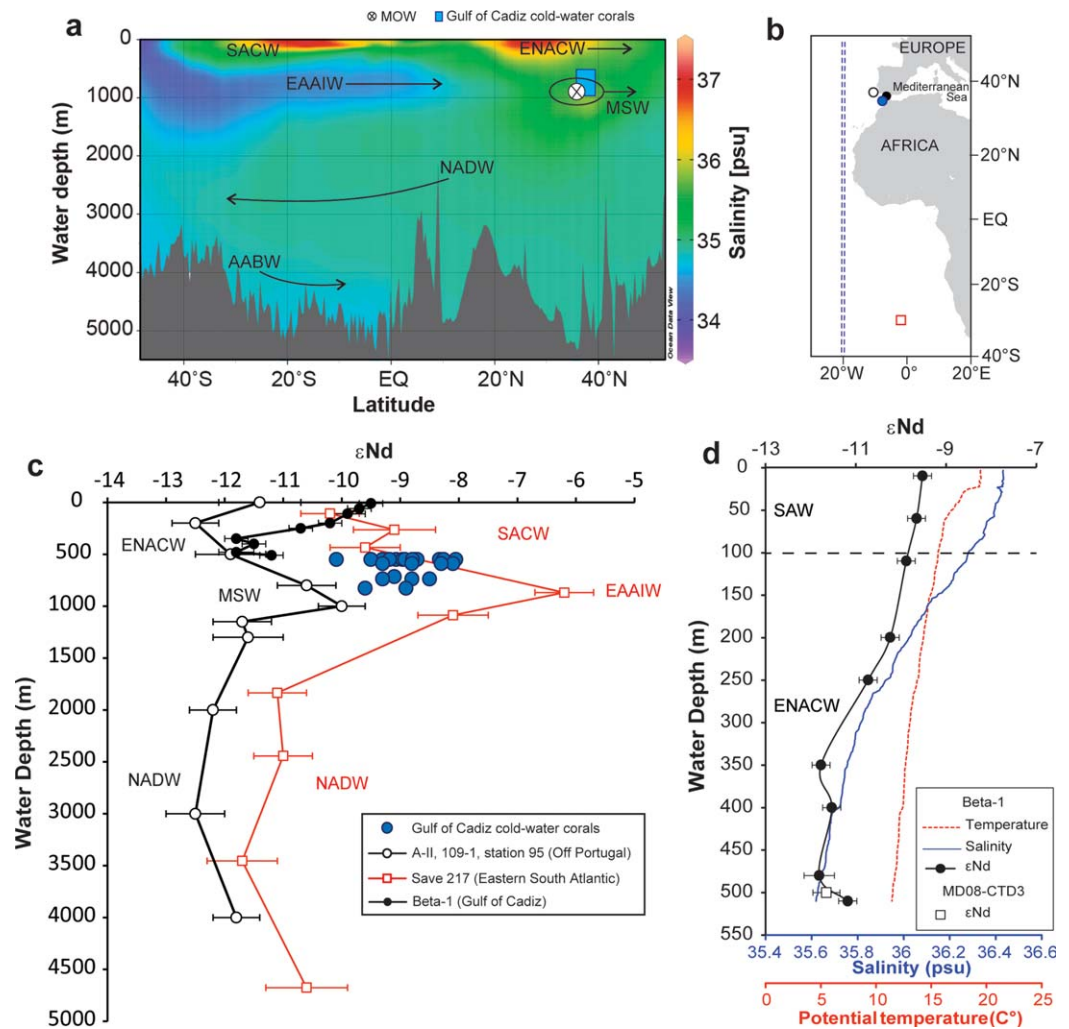
## 2. Hydrological Setting

In the Gulf of Cádiz, the surface water, termed Surface Atlantic Water (SAW;  $T > 16^{\circ}\text{C}$ ,  $S \sim 36.4$ ) [Criado-Aldeanueva *et al.*, 2006], corresponds to the upper 100 m water depth and has an  $\epsilon\text{Nd}$  value of  $-11.4 \pm 0.7$  [Piepgras and Wasserburg, 1983]. Below the SAW, the Eastern North Atlantic Central Water (ENACW) flows from west to east between depths of 100 and 600 m (Figures 1 and 2). It is characterized by salinity values of between 35.3 and 36.6, temperatures ranging from 11 to  $16^{\circ}\text{C}$ , and  $\epsilon\text{Nd}$  of  $-11.9 \pm 0.3$  [Piepgras and Wasserburg, 1983; Criado-Aldeanueva *et al.*, 2006; Louarn and Morin, 2011; Copard *et al.*, 2011]. At the Strait of Gibraltar, two layers of water can be distinguished by a sharp density gradient: an eastward flow of low-density Atlantic inflow overlies the more saline westward outflow of Mediterranean water corresponding to the MOW. At the outflow, the MOW is characterized by high salinities ( $S > 36.9$ ), temperatures ranging from  $13.22$  to  $13.76^{\circ}\text{C}$ , a density  $> 28.8 \text{ kg}\cdot\text{m}^{-3}$ , and  $\epsilon\text{Nd}$  of  $-9.4 \pm 0.2$  [Tachikawa *et al.*, 2004; Spivack and Wasserburg, 1988]. The MOW is greatly modified by vertical mixing with the overlying ENACW during its overflow and is thus called Mediterranean Sea Water (MSW) in the Gulf of Cádiz. To the north of the Gulf of Cádiz, MSW is generally characterized by a salinity value of  $> 36.2$  and an  $\epsilon\text{Nd}$  value of  $-9.9 \pm 0.4$  [Piepgras and Wasserburg, 1983]. Two layers of MSW can be distinguished, an upper layer flowing between 500 and 800 m, and a lower layer occurring at depths from 800 to 1200 m [Hernández-Molina *et al.*, 2006; Louarn and Morin, 2011]. The MSW mainly flows north-westwards along the topographic boundary (Figure 1) [Hernandez-Molina *et al.*, 2014].

The presence of EAAIW corresponds to a local salinity minimum at a depth of roughly  $900 \pm 100$  m in the southern Gulf of Cádiz (Figure 2) [Cabeçadas *et al.*, 2002; Louarn and Morin, 2011]. Although it is present in significant proportions (about 75%), EAAIW cannot be traced on the basis of physical properties further north along the European margin [Louarn and Morin, 2011]. The EAAIW enters the Gulf of Cádiz in the south-western part of the basin and spreads cyclonically (Figure 1) [Louarn and Morin, 2011]. The EAAIW is formed at the polar front in the south-east Pacific and is characterized by  $\epsilon\text{Nd}$  values of between  $-7$  and  $-9$  [Jeandel, 1993; Stichel *et al.*, 2012].

## 3 Material and Methods

This study has investigated 35, well-preserved (fossil) CWC, of the species *Lophelia pertusa*, *Madrepora oculata*, *Desmophyllum dianthus*, and *Dendrophyllia alternata*, collected in several cores from the Gulf of Cádiz



**Figure 2.** (a) Section of salinity (psu) along the eastern Atlantic Ocean (see the dashed lines in Figure 2b) with labeled water masses. (b) Map with locations of cold-water corals and seawater stations reported in Figure 2c. (c) Blue dots represent the  $\epsilon\text{Nd}$  values for the CWC from the Gulf of Cádiz.  $\epsilon\text{Nd}$  profiles from three seawater stations are also represented for comparison. A-II 109-1, station 95 [Piepgras and Wasserburg, 1983]; Save 217 [Jeandel, 1993]; Beta-1 (this study). (d) Vertical profiles of potential temperature (red), salinity (blue), and  $\epsilon\text{Nd}$  (black dots) at Station Beta-1. The  $\epsilon\text{Nd}$  values of one seawater sample located at the bottom of station M08-CTD3 and at the same location as core MD08-3231 are reported for comparison (open square). EAAIW: East Antarctic Intermediate Water; AABW: Antarctic Bottom Water; NADW: North Atlantic Deep Water; ENACW: East North Atlantic Central Water; MSW: Mediterranean Sea Water; SACW: Southern Atlantic Central Water; and SAW: Surface Atlantic Water.

and the Alboran Sea, (Figures 1 and Table 1). Gravity core MD08-3231 (35°18.87'N, 06°48.05'W; 550 m water depth; Figure 1) was retrieved on the Gamma mound from the Pen Duick Escarpment during the R/V Marion Dufresne “MD 169 MiCROSYSTEMS” expedition in July 2008 [Van Rooij et al., 2011]. Further CWCs were selected in the vicinity of core MD08-3231 in the southern Gulf of Cádiz, where corals span a depth range of 590–827 m (Figure 1). The northern part of the Gulf of Cádiz has been also investigated using core GeoB 9022, collected at a water depth of 676 m (Figure 1). Finally, eight CWC (*L. pertusa* and *M. oculata*) were retrieved from the Melilla Coral Province (Brittlestar Ridges I and III, New mound) in the southern Alboran Sea, at depths of between 280 and 465 m (Figure 1).

In addition, nine seawater samples were collected using 12 L Niskin bottles fitted on a CTD-Rosette sampler system at station Beta-1 during the EuroFLEETS cruise (June 2013) and at station MD08-CTD3 (35°18.87'N; 6°48.05'W) during the “MD 169 MiCROSYSTEMS” cruise (July 2008) (Table 2 and Figure 1). These samples were collected in order to assess seawater  $\epsilon\text{Nd}$  around the CWC reef investigated in the southern Gulf of Cádiz. Seawater samples were filtered on-board using a 0.45  $\mu\text{m}$  membrane (Millipore Corp.) and were

**Table 1.** Core Locations, Coral Species, Sample Depths, U-Series, and Radiocarbon Dates of Cold-Water Samples, and  $\epsilon\text{Nd}^a$

Depth in Core (cm)	Species	Age B.P. cal. (ka)	$\epsilon\text{Nd}$
Southern Gulf of Cadiz			
MD08-3231 (35°18.87'N 6°48.05'W, water depth 550 m)			
2.5	<i>L. pertusa</i>	14.44 ± 0.40 <sup>(1)</sup>	-10.1 ± 0.3
15	<i>L. pertusa</i>	20.72 ± 0.79	-9.5 ± 0.4
17.5	<i>L. pertusa</i>	21.65 ± 0.74 <sup>(1)</sup>	-9.1 ± 0.5
22.5	<i>L. pertusa</i>	19.53 ± 0.99	-8.3 ± 0.5
32.5	<i>L. pertusa</i>	25.08 ± 0.41 <sup>(1)</sup>	-9.2 ± 0.4
32.5	<i>L. pertusa</i>	25.08 ± 0.41 <sup>(1)</sup>	-8.8 ± 0.6
47.5	<i>L. pertusa</i>	24.18 ± 0.28 <sup>(1)</sup>	-8.1 ± 0.5
65	<i>L. pertusa</i>	27.05 ± 0.85 <sup>(1)</sup>	-8.3 ± 0.3
65	<i>L. pertusa</i>	27.05 ± 0.85 <sup>(1)</sup>	-8.7 ± 0.6
155	<i>L. pertusa</i>	26.97 ± 0.57	-9.3 ± 0.5
125	<i>L. pertusa</i>	28.31 ± 0.33 <sup>(1)</sup>	-9.0 ± 0.5
165	<i>L. pertusa</i>	28.33 ± 1.11	-8.8 ± 0.4
215	<i>L. pertusa</i>	35.02 ± 0.57 <sup>(1)</sup>	-8.9 ± 0.3
245	<i>L. pertusa</i>	36.70 ± 0.70 <sup>(1)</sup>	-9.2 ± 0.3
265	<i>L. pertusa</i>	34.17 ± 1.10	-9.2 ± 0.3
GeoB 12104 (35°21.99'N 06°51.90'W, water depth 590 m)			
8	<i>L. pertusa</i>	23.02 ± 0.40 <sup>(2)</sup>	-8.1 ± 0.5
GeoB 12103 (35°21.18'N 06°50.90'W, water depth 591 m)			
88	<i>L. pertusa</i>	25.45 ± 0.49 <sup>(2)</sup>	-9.3 ± 0.6
101	<i>D. dianthus</i>	24.80 ± 0.31 <sup>(2)</sup>	-8.8 ± 0.8
101	<i>D. dianthus</i>	24.80 ± 0.31 <sup>(2)</sup>	-8.3 ± 0.5
GeoB 12712 (35°22.27'N 06°54.19'W, water depth 716 m)			
8	<i>M. oculata</i>	34.57 ± 0.87 <sup>(1)</sup>	-9.1 ± 0.3
GeoB 12740 (35°00.02'N 07°04.47'W, water depth 739 m)			
2	<i>M. oculata</i>	14.34 ± 0.32 <sup>(1)</sup>	-9.3 ± 0.1 <sup>(4)</sup>
133	<i>M. oculata</i>	25.02 ± 0.44 <sup>(1)</sup>	-8.5 ± 0.3 <sup>(4)</sup>
209	<i>M. oculata</i>	32.73 ± 0.73 <sup>(1)</sup>	-8.8 ± 0.4 <sup>(4)</sup>
GeoB 12743 (35°03.47'N 07°08.78'W, water depth 827 m)			
5	<i>M. oculata</i>	27.52 ± 0.61 <sup>(1)</sup>	-9.6 ± 0.6
39	<i>M. oculata</i>	30.67 ± 0.41 <sup>(1)</sup>	-8.9 ± 0.2
Northern Gulf of Cadiz			
GeoB 9022 (36°10.98'N 07°18.36'W, water depth 676 m)			
Surface	<i>D. alternata</i>	0.582 ± 0.04	-10.4 ± 0.3
Surface	<i>L. pertusa</i>	31.19 ± 0.23 <sup>(2)</sup>	-9.9 ± 0.2 <sup>(4)</sup>
Surface	<i>M. oculata</i>	35.37 ± 0.28 <sup>(2)</sup>	-9.5 ± 0.1 <sup>(4)</sup>
Alboran Sea			
GeoB 13727 (35°26.16'N 02°30.84'W, water depth 363 m)			
Surface	<i>L. pertusa</i>	0.34 <sup>*(3)</sup>	-8.6 ± 0.3
GeoB 13728 (35°26.28'N 02°30.89'W, water depth 343 m)			
2	<i>L. pertusa</i>	2.90 <sup>*(3)</sup>	-9.0 ± 0.3
GeoB 13723 (35°21.05'N 02°34.82'W, water depth 291 m)			
14	<i>M. oculata</i>	5.40 <sup>*(3)</sup>	-8.8 ± 0.3
GeoB 13730 (35°26.20'N 02°30.87'W, water depth 338 m)			
102	<i>L. pertusa</i>	10.56 <sup>*(3)</sup>	-9.0 ± 0.3
GeoB 13737 (35°28.06'N 02°33.56'W, water depth 297 m)			
Surface	<i>L. pertusa</i>	13.01 <sup>*(3)</sup>	-8.9 ± 0.3
GP21 (35°26.498'N 2°31.004'W, water depth 465 m)			
15	<i>L. pertusa</i>	13.90 ± 0.06	-9.0 ± 0.3
211.5	<i>L. pertusa</i>	28.56 ± 0.09	-9.0 ± 0.3
278	<i>L. pertusa</i>	43.04 ± 0.12	-8.7 ± 0.3

<sup>a</sup>Calibrated AMS <sup>14</sup>C dates are indicated by an asterisk. Previously published dates are identified by the numbers 1 [Frank et al., 2011], 2 [Wienberg et al., 2010], and 3 [Fink et al., 2013]. Previously published  $\epsilon\text{Nd}$  values are indicated by the number 4 [Montero-Serrano et al., 2011].

acidified to a pH of less than 2 with suprapur 2N HCl immediately after collection, following GEOTRACES recommendations [van de Fliedert et al., 2012].

The cleaning procedure and chemical purification of Nd (using TRU-Spec and Ln-Spec resins) used here for CWC samples are described in detail in Copard et al. [2010] and Montero-Serrano et al. [2011]. The Nd isotopic composition of seawater was analyzed following the analytical procedures outlined in Copard et al. [2011] and Wu et al. [2015]. Briefly, REEs were preconcentrated from seawater samples of 10–15 L by using a SEP-PAK Classic C18 cartridge loaded with a HDEHP/H<sub>2</sub>MEHP. REEs were then extracted using a cationic resin (AG50W-X8) and Nd was purified by using Eichrom Ln-spec<sup>®</sup> resin chemistry.

**Table 2.** Locations of Seawater Stations Investigated in This Study, With Water Depth of the Sample<sup>a</sup>

Station	Latitude	Longitude	Depth	$\theta$ (°C)	S (psu)	$\sigma$ (kg.m <sup>-3</sup> )	$\epsilon$ Nd
Beta-1	35°17.46' N	6°47.16' W	10	19.48	36.43	25.99	-9.5 ±0.2
Beta-1	35°17.46' N	6°47.16' W	60	16.29	36.38	26.74	-9.7 ±0.2
Beta-1	35°17.46' N	6°47.16' W	110	15.57	36.27	26.83	-9.9 ±0.2
Beta-1	35°17.46' N	6°47.16' W	200	14.29	36.02	26.92	-10.2 ±0.2
Beta-1	35°17.46' N	6°47.16' W	250	13.73	35.92	26.97	-10.7 ±0.2
Beta-1	35°17.46' N	6°47.16' W	350	12.67	35.76	27.05	-11.8 ±0.2
Beta-1	35°17.46' N	6°47.16' W	400	12.41	35.72	27.08	-11.5 ±0.2
Beta-1	35°17.46' N	6°47.16' W	480	11.70	35.64	27.16	-11.9 ±0.3
Beta-1	35°17.46' N	6°47.16' W	510	11.44	35.62	27.19	-11.2 ±0.2
MD08-CTD3	35°18.87' N	6°48.05' W	500	11.38	35.60	27.19	-11.7 ±0.3

<sup>a</sup>Potential temperature ( $\theta$ ), salinity (S), and potential density ( $\sigma$ ) and  $\epsilon$ Nd data results are reported.

An Nd-oxide (NdO<sup>+</sup>) technique for thermal ionization mass spectrometry (TIMS) was used to analyze the <sup>143</sup>Nd/<sup>144</sup>Nd ratios of CWC from the Gulf of Cádiz [Copard *et al.*, 2010; Montero-Serrano *et al.*, 2011]. Purified Nd was loaded onto a single degassed Re filament and run using the H<sub>3</sub>PO<sub>4</sub>/silica gel method. Samples were analyzed on a six-Faraday collector Finnigan MAT 262 TIMS (LSCE, Gif-sur-Yvette), at a <sup>144</sup>Nd<sup>16</sup>O<sup>+</sup> signal intensity of greater than 500 mV, which was maintained for the duration of the 200 scans. Monitoring for Ce and Sm was conducted continuously during all of the NdO<sup>+</sup> analyses but they were not found to be present. PrO<sup>+</sup> isobaric interferences were measured and corrected, line by line, during offline analysis. The Nd isotopic ratios were corrected for mass fractionation relative to <sup>146</sup>Nd/<sup>144</sup>Nd = 0.7219 using a power law. Oxygen isotope ratios used for the corrections are <sup>18</sup>O/<sup>16</sup>O = 0.002085 and <sup>17</sup>O/<sup>16</sup>O = 0.000391. Replicate analyses of the La Jolla standard gave a mean <sup>143</sup>Nd/<sup>144</sup>Nd of 0.511858 ± 0.000010 (2 $\sigma$ ). This mean value is close to the certified value of 0.511850 ± 0.000013, suggesting a negligible (0.00001) machine bias that was, however, taken into consideration by removing the blank value.

The <sup>143</sup>Nd/<sup>144</sup>Nd ratios of the CWC from the Alboran Sea, and of seawater samples from the Gulf of Cádiz, were analyzed using a ThermoScientific Neptune<sup>Plus</sup> MC-ICPMS installed at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). The mass-fractionation correction was made by normalizing <sup>146</sup>Nd/<sup>144</sup>Nd to 0.7219 and applying an exponential-fractionation correction. During the analytical sessions, every two samples were bracketed with analyses of appropriate Nd standard solutions JNdi-1 and La Jolla, which are characterized by certified values of 0.512115 ± 0.000006 [Tanaka *et al.*, 2000] and 0.511858 ± 0.000007 [Lugmair *et al.*, 1983], respectively. Standard JNdi-1 and La Jolla solutions were analyzed at concentrations similar to those of the samples (5–10 ppb) and all of the measurements affected by a machine bias were corrected, when necessary, by using the La Jolla standard. The external reproducibility (2 $\sigma$ ) for time resolved measurement, deduced from repeated measurements of the La Jolla standard and JNdi-1, ranged from 0.2 to 0.4 Epsilon units for the different analytical sessions. The analytical error associated with each sample analysis is taken as the external reproducibility of the La Jolla standard for each session.

U-series dates were obtained for the CWC fragments from the Gulf of Cádiz (cores MD08-3231, GeoB 9022) and the Alboran Sea ('Brittlestar Big One Ridge') using ICP-QMS and ThermoScientific Neptune<sup>Plus</sup> MC-ICPMS following the methods described in Douville *et al.* [2010] and Pons-Branchu *et al.* [2014]. Moreover, we made use of previously published U-series and radiocarbon dates.

## 4. Results

### 4.1. Chronological Framework

Most of the <sup>230</sup>Th/U and radiocarbon dates for the CWCs investigated in the Gulf of Cádiz and the Alboran Sea (Table 1) have been reported in previous studies [Wienberg *et al.*, 2009, 2010; Frank *et al.*, 2011; Fink *et al.*, 2013]. Here, we provide eight additional <sup>230</sup>Th/U dates from fossil corals *L. pertusa*, and *D. alternata* of cores MD08-3231, GeoB 9022, and GP21 (Table 3).

All of the corals analyzed have initial  $\delta^{234}\text{U}$  values within 7 ‰ of the modern day ocean (146.8‰) [Andersen *et al.*, 2010], suggesting a closed system for the U series [Robinson *et al.*, 2006]. The <sup>232</sup>Th concentrations of the samples range from 0.3 to 2 ppb, indicating that the cleaning protocol was largely successful in

**Table 3.** U Series Measurements and Ages of Fossil Corals Dated in This Study

Sample ID	Depth in Core (cm)	Species	Age (ka)	$^{238}\text{U}$ (ppm)	$^{232}\text{Th}$ (ppb)	$\delta^{234}\text{U}_{\text{measured}}$ (‰)	$\delta^{234}\text{U}_{\text{initial}}$ (‰)
MD08-3231	15	<i>L. pertusa</i>	20.72 ± 0.79	7.090 ± 0.015	0.594 ± 0.005	133.8 ± 4.3	141.8 ± 4.4
MD08-3231	22.5	<i>L. pertusa</i>	19.53 ± 0.99	5.494 ± 0.005	2.055 ± 0.023	139.7 ± 4.8	147.6 ± 4.8
MD08-3231	165	<i>L. pertusa</i>	28.33 ± 1.11	6.670 ± 0.025	0.384 ± 0.005	128.3 ± 3.9	139.0 ± 3.9
MD08-3231	265	<i>L. pertusa</i>	34.17 ± 1.10	5.779 ± 0.018	0.341 ± 0.004	127.8 ± 3.8	140.7 ± 3.8
GeoB 9022	0	<i>D. alternata</i>	0.582 ± 0.04	4.642 ± 0.004	0.396 ± 0.001	141.9 ± 1.1	142.1 ± 1.1
GP21 CF15	15	<i>L. pertusa</i>	13.90 ± 0.06	3.310 ± 0.002	0.527 ± 0.004	145.5 ± 1.9	151.4 ± 1.9
GP21 CF11	211.5	<i>L. pertusa</i>	28.56 ± 0.09	3.823 ± 0.003	1.373 ± 0.006	135.2 ± 1.2	146.6 ± 1.4
GP21 CF10	278	<i>L. pertusa</i>	43.04 ± 0.12	3.047 ± 0.003	0.474 ± 0.004	131.3 ± 1.0	148.3 ± 1.1

removing potential coatings of ferromanganese (Fe-Mn) oxides and hydroxides. The U/Th dates reported in Table 3 are corrected dates that were calculated using a simple correction model that considers the precipitation of initial  $^{230}\text{Th}$  from seawater, taking into account a seawater  $^{232}\text{Th}/^{230}\text{Th}$  activity ratio ranging from 6 to 14 [Frank *et al.*, 2004].

Four of the previously investigated CWCs from core MD08-3231 have  $^{232}\text{Th}$  concentrations of between 5 and 15 ppb, which required a significant correction for nonradiogenic  $^{230}\text{Th}$  [see Frank *et al.*, 2011, supplementary data]. We have considered that small amounts of  $^{232}\text{Th}$  must be due to contamination with  $^{232}\text{Th}$  from seawater, as the corals have been carefully cleaned so as to be free of visible remaining noncarbonate material. Thus, we used a seawater  $^{230}\text{Th}/^{232}\text{Th}$  activity ratio of  $10 \pm 4$  [Frank *et al.*, 2011] to account for non-carbonate initial  $^{230}\text{Th}$ , which led to an age correction of  $\sim 500$  years and an increase of relative age uncertainty due to error propagation of the correction model.

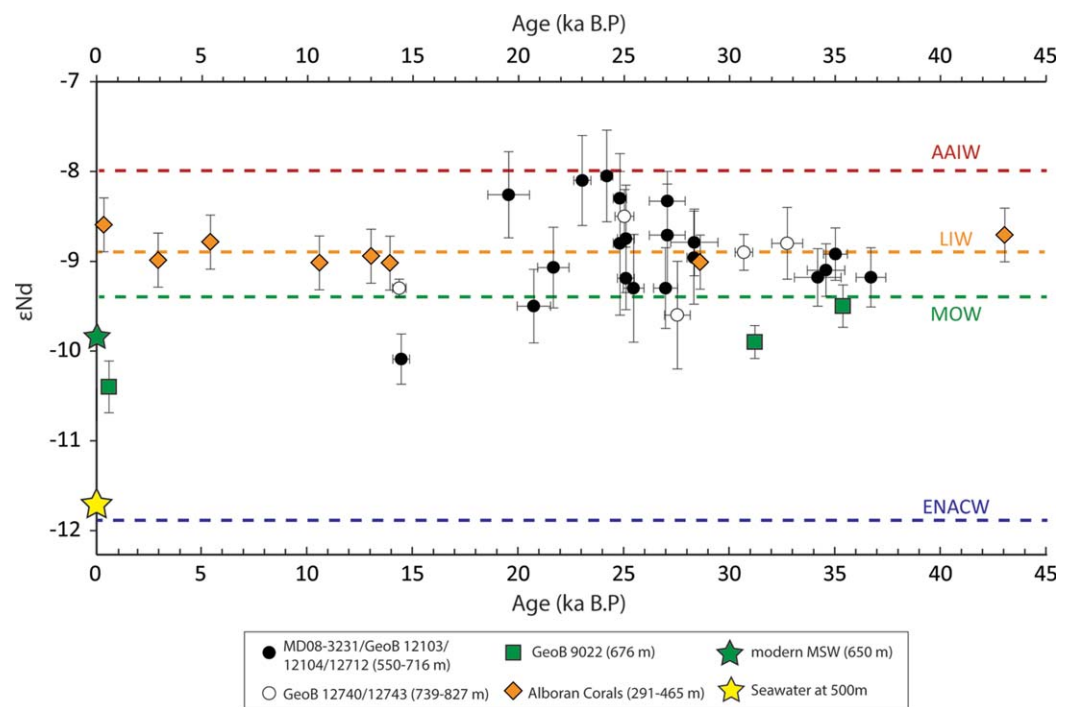
The  $^{230}\text{Th}/\text{U}$  ages of the MD08-3231 fossil corals obtained in this study range from  $19.53 \pm 0.99$  ka to  $34.17 \pm 1.10$  ka (Table 3). Table 1 illustrates this U/Th dating combined with available dates for CWC from the southern Gulf of Cádiz published by Frank *et al.* [2011] and Wienberg *et al.* [2010]. The lack of coral growth during the Holocene is in agreement with the observations of Wienberg *et al.* [2009, 2010] that scarcely any framework-forming cold-water coral existed in this region during interglacial periods, due to reduced upwelling and thus reduced productivity [Rogerson *et al.*, 2004].

In the Alboran Sea, five  $^{14}\text{C}$  dates have been published by Fink *et al.* [2013] and are augmented by three additional  $^{230}\text{Th}/\text{U}$  dates (Tables 1 and 3). The radiometric ages range from 0.34 to  $43.04 \pm 0.12$  ka (Table 1).

#### 4.2. $\epsilon\text{Nd}$ of Seawater Samples and Cold-Water Corals

The Nd isotopic compositions of the seawater samples from stations Beta-1 and MD08-CTD 3 are reported in Table 2 and Figure 2. At station Beta-1,  $\epsilon\text{Nd}$  values range from  $-9.5 \pm 0.2$  at the surface to  $-11.9 \pm 0.3$  at a water depth of 480 m (Figure 2d). Salinity and temperature decrease gradually from 36.4 to 35.6 and from 19 to 11°C, respectively (Figure 2d). The sea surface water is characterized by an  $\epsilon\text{Nd}$  value of  $-9.5 \pm 0.2$  that decreases significantly with depth to reach  $-10.7 \pm 0.2$  at a depth of 250 m. Below 250 m,  $\epsilon\text{Nd}$  values exhibit a narrow range from  $-11.2 \pm 0.3$  and  $-11.9 \pm 0.3$ . The  $\epsilon\text{Nd}$  value obtained at 500 m for station MD08-CTD 3 ( $-11.7 \pm 0.3$ ) is similar to that of the nearby station, Beta-1, and allows the present-day seawater  $\epsilon\text{Nd}$  signature to be constrained just above core MD08-3231.

CWC  $\epsilon\text{Nd}$  values from core MD08-3231 range from  $-8.1 \pm 0.5$  to  $-9.5 \pm 0.4$  between  $19.53 \pm 0.99$  and  $34.55 \pm 0.87$  ka, which differs strongly from the present-day seawater value on top of the Gamma mound ( $-11.7 \pm 0.3$ ) (Figures 2 and 3), located 2.5 km from the Beta mound. Note that replicate  $\epsilon\text{Nd}$  analysis of different coral portions collected on two single specimens (CWCs collected at 32.5 and 65 cm in core MD08-3231 and at 101 cm in core GeoB12103) yielded identical  $\epsilon\text{Nd}$  values, within analytical uncertainty (Table 1). Such a good reproducibility on  $\epsilon\text{Nd}$  analyses has also been obtained in previous studies for more than 12 CWC of the NE Atlantic by using the same analytical procedure [Colin *et al.*, 2010; Copard *et al.*, 2012]. This indicates an excellent level of reproducibility for the entire analytical procedure as well as homogeneous Nd isotopic composition within the coral skeleton of *L. pertusa* (Table 1). Such a good reproducibility of  $\epsilon\text{Nd}$  analyses in CWC indicates that variations of around 1 Epsilon unit could be considered as significant. In the southern Gulf of Cádiz, CWCs collected close to MD08-3231 display similar  $\epsilon\text{Nd}$  values for synchronous coral ages, suggesting the imprint of similar water masses. Therefore, it seems reasonable to compile all CWC



**Figure 3.**  $\epsilon_{\text{Nd}}$  values of CWC from the southern (black and open dots) and northern (green squares) Gulf of Cádiz and Alboran Sea (orange diamonds). The yellow star indicates the present-day  $\epsilon_{\text{Nd}}$  value at depth of the CWC investigated in the southern Gulf of Cádiz. Present-day  $\epsilon_{\text{Nd}}$  values of the intermediate water masses ENACW, MOW, LIW, and AAIW are also reported for comparison (dashed lines). CWC from the Gulf of Cádiz have been analyzed using TIMS with an Nd-oxide ( $\text{NdO}^+$ ) technique while CWC from the Alboran Sea have been analyzed using MC-ICPMS.

$\epsilon_{\text{Nd}}$  values ranging from  $-8.1 \pm 0.5$  to  $-9.6 \pm 0.6$  into one single record of temporal variability of seawater  $\epsilon_{\text{Nd}}$  representative of intermediate water masses (between 550 and 830 m) circulating in the southern Gulf of Cádiz from 15 to 40 ka (Figure 3).

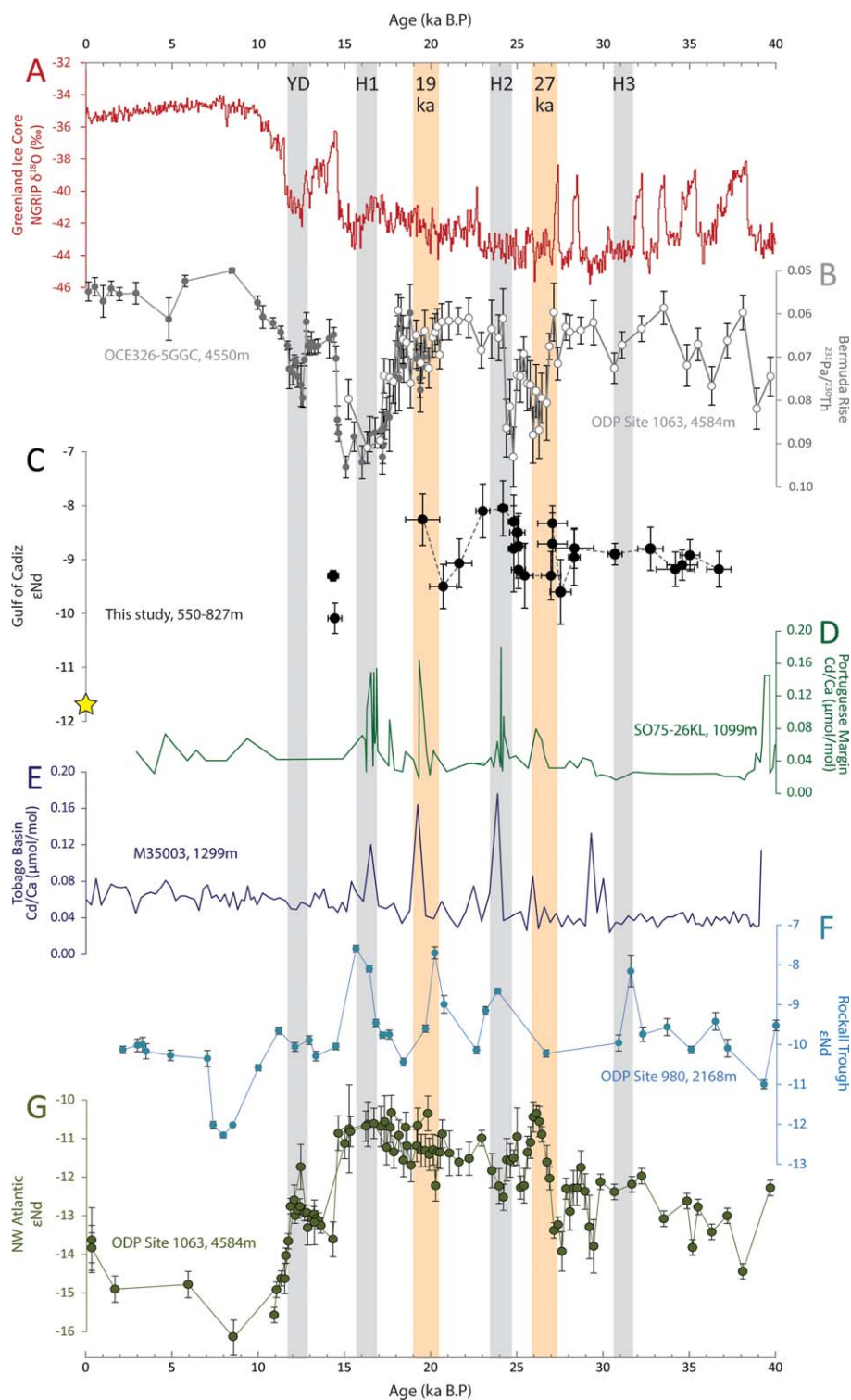
Only CWC  $\epsilon_{\text{Nd}}$  values from core GeoB 9022, located in the northern Gulf of Cádiz on the current MSW pathway [Zweng *et al.*, 2013, WOA data set], are shown separately because it is potentially bathed by different water masses (Figure 3). These CWCs, dated at 31 and 35 ka, display  $\epsilon_{\text{Nd}}$  values of  $-9.9 \pm 0.2$  and  $-9.5 \pm 0.1$ , respectively. In addition, one *D. alternata* specimen, dated to around 0.6 ka, shows an  $\epsilon_{\text{Nd}}$  value of  $-10.4 \pm 0.2$ . Note that the *D. alternata* species has not been the subject of any calibration for Nd isotopes. However, the  $\epsilon_{\text{Nd}}$  value seems consistent with the rest of the record.

The lowest  $\epsilon_{\text{Nd}}$  values obtained from CWC in the southern Gulf of Cádiz range between  $\sim -9.5$  and  $-10$  during the glacial period ( $\sim 25$ – $27$  and 21 ka), whereas the highest values of  $\sim -8.0$  are observed at 19, 23–24, and 27 ka (Figure 3). The eight CWC samples collected in the Alboran Sea and dated to between 0.3 and 43 ka display a narrow range of  $\epsilon_{\text{Nd}}$  values between  $-8.6 \pm 0.3$  and  $-9.0 \pm 0.3$  (Table 1; Figure 3).

## 5. Discussion

Based on CWC from the Gulf of Cádiz and the Porcupine Seabight, the long-term evolution of the eastern North Atlantic seawater  $\epsilon_{\text{Nd}}$  for the last 250 ka is characterized by a significant decrease in  $\epsilon_{\text{Nd}}$  values along the north-eastern margin since the last interglacial period [Montero-Serrano *et al.*, 2011]. In this current study, we have significantly improved the resolution of the last glacial  $\epsilon_{\text{Nd}}$  record in the Gulf of Cádiz, by including new data that constrain the temporal evolution of the Nd isotopic composition of the Mediterranean end-member. Our data indicate that the seawater  $\epsilon_{\text{Nd}}$  values in the southern Gulf of Cádiz, at depths of between 550 and 827 m, decreased from a mean glacial value of  $-8.9 \pm 0.5$  to about  $-11.7 \pm 0.3$ , measured in modern seawater above the Gamma mound. The highest glacial  $\epsilon_{\text{Nd}}$  values (around  $-8$ ) observed at 19, 23–24, and 27 ka, occurred synchronously with abrupt changes in the deep limb of the AMOC as indicated by  $^{231}\text{Pa}/^{230}\text{Th}$  ratios from the Bermuda Rise (Figure 4b) [Gutjahr and Lippold, 2011; Böhm *et al.*, 2015].





**Figure 4.** Comparison of CWC  $\epsilon\text{Nd}$  from the southern Gulf of Cádiz with published climatic and paleohydrological records. (a) North Greenland Ice Core Project (NGRIP)  $\delta^{18}\text{O}$  record [NGRIP Project Members, 2004]. (b) Sedimentary  $^{231}\text{Pa}/^{230}\text{Th}$  activity ratios in sediments of core OCE326-5GGC (gray circles) [McManus *et al.*, 2004] and ODP Site 1063 (white circles) [Böhmer *et al.*, 2015]. (c)  $\epsilon\text{Nd}$  obtained from CWC from the southern Gulf of Cádiz (black dots). The yellow star indicates the present-day  $\epsilon\text{Nd}$  value at 500 m. (d) Benthic foraminiferal Cd/Ca ratio record of core SO75-26KL [Willamowski and Zahn, 2000]. (e) Benthic foraminiferal Cd/Ca ratio record of core M35003 [Zahn and Stüber, 2002]. (f)  $\epsilon\text{Nd}$  values extracted from the Fe-Mn oxyhydroxide fraction of bulk sediments of ODP Site 980 (55°29'N, 14°42'W, 2168 m) [Crocket *et al.*, 2011]. (g)  $\epsilon\text{Nd}$  values extracted from the Fe-Mn oxyhydroxide fraction of bulk sediments of ODP Site 1063 [Böhmer *et al.*, 2015]. Younger Dryas (YD) and Heinrich events (H1–H3) are indicated by gray bars.

In order to understand the variability in seawater  $\epsilon\text{Nd}$  in the Gulf of Cádiz, it is necessary to (i) assess the potential influences of lithogenic Nd inputs and regional “boundary exchange” on the  $\epsilon\text{Nd}$  of middepth water masses; (ii) determine the potential changes in the contribution of water masses and/or (iii) assess the potential change in Nd isotopic composition of the intermediate water masses entering in the Gulf of Cádiz since the last glacial period.

### 5.1. Modern Seawater $\epsilon\text{Nd}$ in the Southern Gulf of Cádiz

Core MD08-3231 is located in the Pen Duick Escarpment at a water depth of 550 m, with a present-day temperature and salinity of around 11.4°C and 35.6, respectively (Figure 2d), which corresponds to ENACW [Criado-Aldeanueva *et al.*, 2006]. Below 600 m water depth, a decrease in salinity and temperature has been shown to reflect the presence of a small contribution of EAAIW [Van Rooij *et al.*, 2011]. Seawater  $\epsilon\text{Nd}$  values at the depth of core MD08-3231 ( $\epsilon\text{Nd}$  of  $-11.2 \pm 0.2$  for station Beta-1 and  $-11.7 \pm 0.3$  for station MD08-CTD 3) are identical, within analytical uncertainty, to the  $\epsilon\text{Nd}$  of the ENACW ( $-11.9 \pm 0.6$ ) measured west of the Gulf of Cádiz [Piepgras and Wasserburg, 1983]. Based on multiparameter analysis of the distribution of water masses in the Gulf of Cádiz, Louarn and Morin [2011] have estimated a composition of around 90% ENACW and 10% EAAIW in the southern Gulf of Cádiz at the depth of the Pen Duick Escarpment (550 m). It is worth noting that the presence of MSW, generally characterized by salinity values higher than 36.2, is not detected at the Gamma Mound [Louarn and Morin, 2011; Van Rooij *et al.*, 2011; Mienis *et al.*, 2012]. Indeed, most of the MSW turns north-westward along the continental margin shortly after flowing out from the Strait of Gibraltar [Louarn and Morin, 2011]. An  $\epsilon\text{Nd}$  of  $-9.9 \pm 0.4$  has been obtained for MSW in the northern Gulf of Cádiz [Piepgras and Wasserburg, 1983]. Only a small fraction is deviated into the southern part of the Gulf of Cádiz through regional eddies which detach from the main flow [Fusco *et al.*, 2008].

### 5.2. Potential Influences of Lithogenic Nd Inputs on the $\epsilon\text{Nd}$ of Middepth Water Masses of the Gulf of Cádiz Since the Last Late Glacial Period

The Nd isotopic signature of intermediate water masses in the Gulf of Cádiz could potentially be modified over time along continental margins through the mechanism of regional “boundary exchange” [Lacan and Jeandel, 2001, 2004, 2005; Wilson *et al.*, 2012] and/or through the input of Saharan dust [Tachikawa *et al.*, 2004] and enhanced volcanic IRD-water exchange during glacial periods [Roberts and Piotrowski, 2015].

However, a previous study has stated that present-day seawater  $\epsilon\text{Nd}$ , measured in the core-top leachate sediments of the Gulf of Cádiz, is inconsistent with a contribution of unradiogenic eolian dust or margin sediments [Stumpf *et al.*, 2010]. On the other hand, during the cold intervals, the velocity of MOW is thought to have been higher [Voelker *et al.*, 2006; Toucanne *et al.*, 2007; Bahr *et al.*, 2015] which could have favored Nd exchanges with margin sediments of the Gulf of Cádiz characterized by unradiogenic  $\epsilon\text{Nd}$  values (around a value of  $-12$ ) [Grousset *et al.*, 1988, 1998]. In addition, the last glacial period is associated with higher inputs of Saharan dust ( $\epsilon\text{Nd}$   $-11 \pm 0.4$  to  $-14.4 \pm 0.1$ ) [Grousset *et al.*, 1988, 1998; Skonieczny *et al.*, 2011], associated with an aridification of North Africa, to the Tropical Northeast Atlantic and, to a lesser extent, to the Gulf of Cádiz [Sarnthein *et al.*, 1981; Hooghiemstra *et al.*, 1987; Moreno *et al.*, 2002; Wienberg *et al.*, 2010]. The increase in Saharan dust inputs is particularly evident during Heinrich Stadial events [Itambi *et al.*, 2009]. As  $\epsilon\text{Nd}$  values from CWC in the southern Gulf of Cádiz are more radiogenic during the last glacial than the modern seawater value, and even more during HS2, lithogenic Nd inputs to middepth water masses of the Gulf of Cádiz from boundary exchange or exchange with Saharan dust are, thus, likely to have been negligible, at least for the last 40 ka.

Furthermore, based on the analysis of five cores in the North Atlantic IRD belt, Roberts and Piotrowski [2015] have shown a general increase in  $\epsilon\text{Nd}$  from deep (4000 m) to intermediate water (1000 m) by 6  $\epsilon\text{Nd}$ -units during the last glacial period compared to the Holocene. The authors suggested an enhanced volcanic IRD-water exchange and the potential southward transport of this signal. Nevertheless, samples from the western midlatitude Atlantic [Böhm *et al.*, 2015], as well as our own samples, display glacial  $\epsilon\text{Nd}$  that is over 2  $\epsilon\text{Nd}$ -units less radiogenic, indicating that the southward propagation of this radiogenic Nd isotope labelling water is unlikely, or at least limited. We also draw attention to the discrepancy of about 5  $\epsilon\text{Nd}$ -units between the  $\epsilon\text{Nd}$  record produced by Roberts and Piotrowski [2015] and that of Crocket *et al.* [2011], for the same geographical area and water depth, during the Last Glacial Maximum (LGM).

### 5.3. Middepth Hydrological Reorganization Under Glacial Conditions

The  $\epsilon\text{Nd}$  values of CWC in the southern Gulf of Cádiz, at intermediate depth, display a mean glacial value of  $-8.9 \pm 0.5$  that is significantly higher than  $-11.7 \pm 0.3$ , measured in modern seawater. This last glacial radiogenic  $\epsilon\text{Nd}$  mean value seems to result from a major reorganization of the hydrography of the North Atlantic, involving more radiogenic water masses invading the Gulf of Cádiz during the glacial period or, alternatively, a change in the  $\epsilon\text{Nd}$  signature of the water masses entering in the Gulf of Cádiz over time.

At present time, MSW is the most radiogenic ( $\epsilon\text{Nd} = -9.9 \pm 0.4$ ) water mass in the Gulf of Cádiz but it does not contribute significantly to the water mass balance at intermediate water depth in the southern Gulf of Cádiz. A variable spatial influence of the MSW during glacial and interglacial marine isotopic stages (MIS) [Llave *et al.*, 2006] may have changed the Nd isotopic signature in this area. Under glacial conditions, when the MSW was denser, deeper, and flowed faster [Zahn *et al.*, 1987; Thunell and Williams, 1989; Zahn, 1997; Schönfeld, 1997; Cacho *et al.*, 2000; Voelker *et al.*, 2006; Toucanne *et al.*, 2007] the lower MSW core was enhanced and flowed more widely across the southern Gulf of Cádiz through increased meddy activity [Richardson *et al.*, 2000; Ambar *et al.*, 2008]. Conversely, during interglacial periods, the upper layer was the principal core and was largely confined to the Iberian margin. Therefore, because of the proximity of the studied site to the MSW pathways, we cannot exclude the possibility that, during glacial periods, variations in the hydrological conditions forced MSW to reach CWC mounds in the southern Gulf of Cádiz.

Modern  $\epsilon\text{Nd}$  values for MSW were reported in Piepgras and Wasserburg [1983] for hydrocast stations A-II 109 St. 95 ( $-9.8 \pm 0.6$  at 1000 m) and A-II 109 St. 101 ( $-9.9 \pm 0.5$  at 650m). The two glacial CWC samples from core GeoB 9022 present similar  $\epsilon\text{Nd}$  values (around  $-9.7 \pm 0.2$ ) to present-day seawater, suggesting stability in the Nd isotopic composition of the MSW since the last 35 ka. Such results are in agreement with data reported in Stumpf *et al.* [2010] that indicate constant Nd isotopic composition values for the MSW in the northern Gulf of Cádiz over the last 25 ka (around  $-9.3$ ).  $\epsilon\text{Nd}$  variations were less than 0.5  $\epsilon$ -unit, even during HS1 and YD [Stumpf *et al.*, 2010].

The depth range where CWC were collected (between 280 and 465 m water depth) in the Alboran Sea is occupied by modified Levantine Intermediate Water (LIW), which flows westward towards the Strait of Gibraltar and constitutes  $\sim 80\%$  of the MOW [Parrilla *et al.*, 1986; Kinder and Parrilla, 1987; Pinardi and Masetti, 2000].  $\epsilon\text{Nd}$  values of the Alboran CWC show a narrow range between  $-8.6 \pm 0.3$  and  $-9.0 \pm 0.3$ , suggesting that the Nd isotopic signature of the modified LIW has not changed significantly since the last glacial period (Figure 3). This is also in agreement with a recent study that has documented at low-time resolution the past seawater  $\epsilon\text{Nd}$  values of deeper water mass (WMDW), which also contributes to the MOW, from planktonic foraminifera in a core located in the Alboran Sea at 1989 m water depth [Jiménez-Espejo *et al.*, 2015]. This study indicates also for the WMDW a narrow range of  $\epsilon\text{Nd}$  values ( $-9.0 \pm 0.3$  to  $-9.7 \pm 0.3$ ) for the last 20 ka.

Consequently,  $\epsilon\text{Nd}$  of MSW must be considered constant, at around  $-9.7$ , but we cannot exclude the possibility that a larger fraction of MSW could have mixed into the southern Gulf of Cádiz during the last glacial period. However, glacial radiogenic  $\epsilon\text{Nd}$  values, reaching up to  $-8$  in the southern Gulf of Cádiz (Figure 3), cannot result from an enhanced contribution of MSW.

Currently, the North Atlantic water is the most unradiogenic end-member flowing into the Gulf of Cádiz. If we exclude a strong modification of the  $\epsilon\text{Nd}$  values of the North Atlantic water masses induced by an enhanced IRD flux during glacial time [Roberts and Piotrowski 2015], a more radiogenic signature for the northern-sourced water could be attributed to a change in the middepth subpolar gyre (mSPG) circulation. Montero-Serrano *et al.* [2011] have proposed that the weakening of mSPG during glacial periods limited the export of unradiogenic Nd isotopic composition of the subpolar middepth water to the temperate Atlantic and thus impacted upon the  $\epsilon\text{Nd}$  value of the ENACW. This mechanism could have limited the dilution of Mediterranean and southern-sourced water.

A possible source of radiogenic  $\epsilon\text{Nd}$  is the glacial EAAIW. Indeed, Nd isotopic composition of the AAIW stems from the mixing between radiogenic Pacific water ( $\epsilon\text{Nd} = -4$ ) and unradiogenic Atlantic source water ( $\epsilon\text{Nd} = -13.5$ ) in the Southern Ocean, which results in present-day  $\epsilon\text{Nd}$  values for EAAIW of between  $-7$  and  $-9$  [Jeandel, 1993; Stichel *et al.*, 2012]. EAAIW is modified during its northward penetration into the North Atlantic by boundary exchange and/or exchange with unradiogenic sediment from western North Africa [Rickli *et al.*, 2009]. Hence, at present, EAAIW displays  $\epsilon\text{Nd}$  values of around  $-11.5$  along the African margin [Tachikawa *et al.*, 1999; Rickli *et al.*, 2009; Stichel *et al.*, 2015].

A shift toward more radiogenic values was identified during the HS1 for the AAIW in CWC ( $\epsilon\text{Nd}$  of  $-6.5$ ) from the Drake Passage [Robinson and van de Flierdt, 2009]. This is probably a consequence of reduced export of NADW due to reorganization of AMOC [Piotrowski et al., 2005, 2012]. Further north, the  $\epsilon\text{Nd}$  signature of the glacial EAAIW, as well as its past modification during its northward penetration, remains unknown. Boundary exchange with volcanic sediments of the Canary Islands ( $\epsilon\text{Nd}$  of about  $+5.3$ ; cf. GEOROC), which are in the flow direction of southern component waters moving northward, is observed in the surface water in the environs of the Canary Islands ( $\epsilon\text{Nd}$  around  $-8$ ) [Rickli et al., 2010]. At the present time, this effect is probably spatially restricted (as observed in other volcanic areas) but we have to consider the possibility that an increase in volcanic sediment input from the Canary Islands, or simply a strengthening of northward currents, could have modified the southern water component by adding radiogenic  $\epsilon\text{Nd}$ , which may have propagated northward into the Gulf of Cádiz. However, the ability of such labeling to spread beyond a regional scale remains uncertain [Roberts and Piotrowski, 2015]. Nevertheless, assuming that the glacial EAAIW was not fully modified during its route from the Southern Ocean to the North Atlantic, as suggested for the glacial western branch of AAIW [Pahnke et al., 2008; Huang et al., 2014], our  $\epsilon\text{Nd}$  record could reflect a modification of the relative fractions of EAAIW and ENACW in the southern Gulf of Cádiz.

In the eastern North Atlantic, cadmium/calcium (Cd/Ca) and carbon isotope ( $\delta^{13}\text{C}$ ) records established for benthic foraminifera along sediment cores from the Moroccan (core M16004;  $29.59^\circ\text{N}$ ,  $10.39^\circ\text{W}$ ; 1512 m) and Portuguese (core SO75-26KL;  $37.49^\circ\text{N}$ ,  $09.30^\circ\text{W}$ ; 1099 m) margins [Zahn et al., 1987; Zahn, 1997; Willamowski and Zahn, 2000] indicate a greater influence of a southern-sourced nutrient-enriched water mass at intermediate-depth in the eastern Atlantic during the last glacial as far north as  $30^\circ\text{N}$ . Using micropaleontological data, Penaud et al. [2011] have documented a hydrologic front separating the Gulf of Cádiz from the south-western Iberian margin. The invasion of southern water in the temperate middepth Atlantic could result from a strengthening of AAIW and a reduction in the contribution of middepth subpolar waters, as suggested previously on longer-time scales [Montero-Serrano et al., 2011], and thus could have led to the increased  $\epsilon\text{Nd}$  values in our record spanning the last glacial period. In addition, northward penetration of the deep southern water component (AABW) below 2000 m water depth during last glacial is widely accepted [Yu et al., 2008] and has been evidenced by an  $\epsilon\text{Nd}$  record in the subtropical Northwest Atlantic (ODP Site 1063,  $33^\circ41'\text{N}$ ,  $57^\circ37'\text{W}$ , 4584 m). This record exhibits a similar glacial/interglacial pattern to that observed in the Gulf of Cádiz, with high  $\epsilon\text{Nd}$  values during the last glacial period [Böhm et al., 2015, Figure 5d]. All of these results point to an enhanced contribution of EAAIW to the thermohaline circulation between 20 and 40 ka.

Therefore, the radiogenic  $\epsilon\text{Nd}$  mean values ( $-8.9 \pm 0.5$ ) recorded in the Gulf of Cádiz during the last glacial period could be explained by (i) a stronger influence of glacial MSW, (ii) a weakening of mSPG limiting the export of unradiogenic subpolar water, and (iii) an increase in the glacial EAAIW flowing northward, or a combination of the three. In addition, we cannot completely exclude the possibility that radiogenic labeling of a southern water component through an increase of local exchange with volcanic sediments from the Canary Islands [Rickli et al., 2010] could have propagated to the Gulf of Cádiz.

#### 5.4. Millennial-Scale Rapid Variability of Middepth Nd Isotopic Composition

The radiogenic peaks of  $\epsilon\text{Nd}$  at 23–24 and 27 ka, and to a lesser extent at 19 ka, coincide with a reduction of the AMOC as observed in the  $^{231}\text{Pa}/^{230}\text{Th}$  activity ratios in sediments from the subtropical western Atlantic (ODP Site 1063,  $33^\circ41'\text{N}$ ,  $57^\circ37'\text{W}$ , 4584 m) (Figure 4b) [Böhm et al., 2015]. Therefore, a northern source for radiogenic Nd is unlikely. As glacial MSW is characterized by nearly constant  $\epsilon\text{Nd}$  values (around  $-9.8$ ), the highest  $\epsilon\text{Nd}$  values (around  $-8$ ) observed in CWC, dated to around 19, 23–24, and 27 ka, most likely reflect stronger EAAIW export northward at these times. This would have been associated with a relatively reduced contribution of North Atlantic middepth subpolar waters [e.g., Montero-Serrano et al., 2011]. We can note that such hydrological changes could be rapid as three CWC characterized by an age around 27 ka (within the error bars of the U/Th dating) display  $\epsilon\text{Nd}$  between  $-9.6 \pm 0.6$  and  $-8.3 \pm 0.3$ . This suggests a rapid northward penetration of the EAAIW in agreement with the hydrological front that the Gulf of Cadiz constitutes. However, further investigations on well dated CWC would be necessary to improve seawater  $\epsilon\text{Nd}$  record of the southern Gulf of Cadiz to assess the rapidity of such hydrological changes.

At these times, the  $\delta^{13}\text{C}$  values in core SO75-26KL (Portuguese margin) and in core M35003 (12°5.4'N, 61°14.6'W, 1299 m) [Zahn and Stüber, 2002], located in the Tobago basin (western Atlantic), are depleted below mean glacial levels and Cd/Ca maxima observed (Figures 4d and 4e) are similar to values typically found in the Southern Ocean [Nolting *et al.*, 1991; Boyle, 1992; Frew and Hunter, 1992]. In the western tropical Atlantic,  $\epsilon\text{Nd}$  recorded by Fe-Mn oxides (core MD99-2198; 12.09°N, 61.23°W; 1330 m) has revealed higher radiogenic values coeval with cold glacial stages (HS1 and YD) which were interpreted as reflecting increased northward advection of AAIW during periods of reduced AMOC [e.g., Pahnke *et al.*, 2008]. Taken together, these results, and those obtained in this study, point to a basin-wide AAIW invasion of the North Atlantic at times of reduced AMOC.

The excursions of radiogenic  $\epsilon\text{Nd}$  values at 23–24 and 27 ka are coeval with increased iceberg discharge during the HS2 and the GS-3 (D-O stadial 3) [Elliot *et al.*, 2002]. These cold events occurred close to the LGM and were associated with massive expansions of marine and land-based ice sheets that caused the collapse of the AMOC, at least for the HS2 as observed in the  $^{231}\text{Pa}/^{230}\text{Th}$  reported in Figure 4b [Böhm *et al.*, 2015].

At greater water depths, ODP Site 1063 (4584 m water depth) also reveals slightly higher  $\epsilon\text{Nd}$  values between 27 and 15 ka, with radiogenic  $\epsilon\text{Nd}$  values peaking around 24–25 and 26–27 ka, suggesting a possible northward incursion of the deep southern-sourced water (AABW) as far as 33.7°N (Figure 4g) [Böhm *et al.*, 2015]. Indeed, AABW invasion during reduced AMOC is observed further north in the Rockall Trough: ODP Site 980 (55°29'N, 14°42'W, 2168 m) has revealed shifts towards more radiogenic  $\epsilon\text{Nd}$  values ( $\sim -8$ ) during the North Atlantic HS1 to HS3 cold events pointing to the same conclusion (Figure 4f) [Crockett *et al.*, 2011]. If so, this suggests that southern-sourced waters invaded the North Atlantic basin at bottom and mid-depth levels, during times of reduced AMOC, resulting in a smaller Nd isotopic composition gradient between water masses of the Southern Ocean and the North Atlantic ( $<2$   $\epsilon\text{Nd}$  units) compared to the present day configuration (3–6  $\epsilon\text{Nd}$  units).

Such results contradict previous studies that point to a smaller fraction of AAIW in the North Atlantic during cold events [Came *et al.*, 2008; Xie *et al.*, 2012; Huang *et al.*, 2014] but are in agreement with other studies that indicate an enhanced northward export of AAIW in the Pacific Ocean during cold periods in the northern hemisphere (HS events and the Younger Dryas) [Basak *et al.*, 2010; Pena *et al.*, 2013]. These cold events are also associated with warming in the Southern Hemisphere [Blunier and Brook, 2001; Pahnke *et al.*, 2003] and an increase in AAIW formation [Pahnke and Zahn, 2005; Bostock *et al.*, 2004; Saenko *et al.*, 2003; Weaver *et al.*, 2003].

In the eastern basin, CWC from the Gulf of Cádiz as well as Cd/Ca data of core SO75-26KL [Willamowski and Zahn, 2000] reveal a stronger influence of EAAIW between 550 and 1099 m water depth during HS events. However, core M16004 located at 1512 m off the Moroccan margin [Willamowski and Zahn, 2000] does not show more EAAIW influence at these times. In the western basin, several studies investigating cores between 600 and 1100 m water depth have demonstrated a reduction of the western AAIW (WAAIW) influence during YD and HS1 [Came *et al.*, 2008; Xie *et al.*, 2012; Huang *et al.*, 2014], while at deeper depth, between 1299 and 1330 m, two other studies point to an increased WAAIW during North Atlantic cold events [Zahn and Stüber, 2002; Pahnke *et al.*, 2008]. Therefore, we suggest that a deepening of WAAIW (around 1300 m) occurred during the North Atlantic Cold events, while in the eastern basin, EAAIW has influenced the 550–1100 m interval of the water column. Further investigations need to be done to understand this complex and fragmentary observation of the geographical distribution of the EAAIW and WAAIW during glacial times.

According to the benthic  $\delta^{13}\text{C}$  record in the southwest Pacific [Pahnke and Zahn, 2005], the most recent excursion of EAAIW observed in the Gulf of Cádiz at  $\sim 19$  ka does not seem to be linked to any increase in the production of AAIW. At the same time, a high-amplitude anomaly occurred in the Cd/Ca record in core SO75-26KL along the Portuguese margin (Figure 4d) as well as in core M35003 in the middle depths of the tropical Atlantic Ocean (Figure 4e). A similar radiogenic  $\epsilon\text{Nd}$  excursion was also observed both in core MD99-2198 in the Tobago Basin and at ODP Site 980 in the Rockall Trough (Figure 4f). Pahnke *et al.* [2008] have correlated the northward penetration of AAIW at this time to reduced GNAIW formation [Zahn *et al.*, 1997; Hüls and Zahn, 2000; Hall *et al.*, 2006]. Indeed, this time interval is associated with a meltwater pulse from the Northern Hemisphere ice sheets and an abrupt rise in global sea level [Scourse *et al.*, 2000; Yokoyama *et al.*, 2000; Clark *et al.*, 2004]. Hanebuth *et al.* [2009] have documented a rapid rise in global sea

level starting around 19.6 ka with a duration of  $\sim 800$  years and a magnitude of  $\sim 10$  m. *Toucanne et al.* [2010] have demonstrated that the southward flow of meltwater derived from the Fennoscandian ice sheet, and freshwater discharge from the Manche River, peaked around 18–20 ka, which coincides with increased iceberg discharge shown in previous studies [*Knutz et al.*, 2006, 2007; *Peck et al.*, 2007]. The ice sheet retreat at 20–18 ka may have contributed to a destabilization of the AMOC, as suggested by a sharp increase in the  $^{231}\text{Pa}/^{230}\text{Th}$  ratio in the North Atlantic region [*McManus et al.*, 2004], thereby allowing an increased northward penetration of AAIW at the expense of northern Atlantic water. Such results are in line with a reversed phasing at millennial timescales between northward export of Southern Ocean water and southward export of North Atlantic water, a pattern which is supported by modeling studies [*Keeling and Stephens*, 2001; *Saenko et al.*, 2003; *Weaver et al.*, 2003; *Stouffer et al.*, 2007].

## 6. Conclusions

$\epsilon\text{Nd}$  of seawater and CWC samples, collected at depths of 280–827 m in the Gulf of Cádiz and the Alboran Sea, have been investigated in order to constrain hydrological variations in the intermediate water of the southern Gulf of Cádiz over the past 40 ka.

The Nd isotopic signatures of the modified LIW in the Alboran Sea and the MSW in the Gulf of Cádiz display a narrow range from  $-8.6 \pm 0.3$  to  $-9.0 \pm 0.3$  and  $-10.4 \pm 0.2$  to  $-9.5 \pm 0.1$  over the last 45 ka, respectively. In contrast, the  $\epsilon\text{Nd}$  record obtained from seawater and CWC in the southern Gulf of Cádiz presents a strong contrast between present-day values ( $-11.6 \pm 0.3$  to  $-10.4 \pm 0.3$ ) and those of the last glacial period ( $-9.6 \pm 0.6$  to  $-8.1 \pm 0.5$ ). The more radiogenic glacial values might be the result of a higher relative proportion of MSW, an enhanced northward inflow of glacial EAAIW and/or an accompanied retraction of mSPG. The retraction of mSPG may have limited the export of less radiogenic subpolar middepth water. Moreover, we cannot exclude the possibility that enhanced boundary exchange along the Canary Islands may have contributed radiogenic Nd to the glacial Gulf of Cádiz via the EAAIW.

Higher radiogenic  $\epsilon\text{Nd}$  values ( $\sim -8$ ) have been observed at 23–24 and 27 ka, coeval with increased iceberg discharge during the HS2 and the GS-3 (stadial DO3) and a significant reduction in the AMOC. Since the  $\epsilon\text{Nd}$  values of the MSW have remained constant over the last 35 ka ( $-10.4 \pm 0.2$  to  $-9.5 \pm 0.1$ ), such radiogenic values could reflect a major northward spread of EAAIW into the southern part of the Gulf of Cádiz, which is in agreement with previous nutrient-proxies and  $\epsilon\text{Nd}$  records from the North Atlantic basin [*Willmowski and Zahn*, 2000; *Zahn and Stüber*, 2002; *Pahnke et al.*, 2008]. Such results are consistent with reduced GNAIW formation and a larger export of AAIW towards the North Atlantic [*Mangini et al.*, 2010]. Furthermore, an EAAIW excursion around 19 ka has been revealed by CWC of the southern Gulf of Cádiz: this excursion was coeval with a meltwater pulse from Northern Hemisphere ice sheets [*Hanebuth et al.*, 2009]. The meltwater pulse could have resulted in a destabilization of the AMOC, as evidenced by  $^{231}\text{Pa}/^{230}\text{Th}$  activity ratios in sediments, [*McManus et al.*, 2004; *Böhm et al.*, 2015] leading to a substantial volume of the Atlantic Ocean being filled by southern-sourced water.

The CWC  $\epsilon\text{Nd}$  from the southern Gulf of Cádiz suggests a basin-wide invasion of AAIW at times of reduced AMOC, which is in agreement with previous studies. This supports modelling studies pointing to an interhemispheric seesaw [*Keeling and Stephens*, 2001; *Saenko et al.*, 2003; *Weaver et al.*, 2003; *Stouffer et al.*, 2007].

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