

1 **Eastern Mediterranean Sea circulation inferred from the conditions of S1 sapropel**
2 **deposition**

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10

11 **Abstract**

12 Holocene Eastern Mediterranean Sea sediments contain an organic-rich sapropel S1 layer that
13 was formed in oxygen-depleted waters. The spatial distribution of this layer revealed that
14 during S1 deposition deep waters were anoxic below 1,800 m in water depth. **However,**
15 **whether this boundary permanently existed from early to mid-Holocene has not been**
16 **examined yet.** **To answer this question,** a multi-proxy approach was applied to a core retrieved
17 close to the 1,800 m boundary (at 1,780 m). We measured the bulk sediment elemental
18 composition, the stable isotopic composition of the planktonic foraminifer *Globigerinoides*
19 *ruber*, and the abundance of benthic foraminifera since the last deglaciation. The result
20 indicates that authigenic U and Mo accumulation began around 13-12 cal ka BP, in concert
21 with surface water freshening estimated from the *G. ruber* $\delta^{18}\text{O}$ record. The onset of
22 bottom/pore water oxygen depletion occurred prior to S1 deposition inferred from barium
23 enrichment. In the middle of the S1 deposition period reduced authigenic V, Fe and As
24 contents and Br/Cl ratio indicated short-term bottom water re-oxygenation. A sharp Mn peak
25 and maximal abundance for benthic foraminifera marked a total recovery for circulation at

26 approximately 7 cal ka B.P. Based on our results and existing data, we suggest that S1
27 formation within the upper 1,780 m of the Eastern Mediterranean Sea was preconditioned by
28 reduced ventilation, resulting from excess fresh water inputs due to insolation changes under
29 deglacial conditions, that initiated between 15 and 12 ka within the upper 1,780 m. Short-term
30 re-oxygenation in the Levantine Basin is estimated to have affected bottom water **at least as**
31 **deep as 1,780 m in response to cooling and/or reduction of fresh water inputs.** We tentatively
32 propose that complete ventilation recovery at the S1 termination **was depth-dependent with**
33 **earlier oxygenation within the upper 1,780 m.** Our results **provide new constraints of vertical**
34 **water column structure in the eastern Mediterranean Sea since the last deglaciation.**

35

36 **1. Introduction**

37 The Mediterranean Sea is located in a transition zone between subpolar depression and
38 subtropical high pressure and is known to be sensitive to on-going and past climate change
39 (Bethoux and Gentili, 1999; Roether et al., 1996). Holocene sediments obtained from the
40 eastern Mediterranean Sea often contain the most recent organic-rich sapropel deposit, S1
41 (10.8 ± 0.4 to 6.1 ± 0.5 cal ka B.P., De Lange et al., 2008), that formed due to a drastic
42 decrease in labile organic matter decomposition (Moodley et al., 2005). Reduced oxygen
43 supply to bottom waters has been suggested to be a precondition for sapropel formation
44 although increased biological productivity further promoted S1 deposition (Bianchi et al.,
45 2006; Myers et al., 1998; Rohling, 1994; Stratford et al., 2000).

46 Surface water density decreases due to excess fresh water inputs have played a pivotal
47 role in reducing Mediterranean Sea thermohaline circulation during sapropel formation
48 (Myers et al., 1998; Rohling, 1994; Stratford et al., 2000). By reinforcing Nile River
49 discharge toward the Levantine Sea (Emeis et al., 2003; Kallel et al., 1997; Revel et al., 2010;
50 Rohling, 1994; Rossignol-Strick et al., 1982), high summer insolation at minimum precession

51 is known to have had a fundamental impact (Kutzbach et al., 2014; Rohling, 1994 and
52 references therein; Ziegler et al., 2010). In parallel, due to an active Mediterranean storm
53 track, reduced boreal winter insolation could have favoured winter precipitation over the
54 Mediterranean Sea region (Kutzbach et al., 2014; Magny et al., 2013; Meijer and Tuenter,
55 2007; Rohling, 1994). The influence of a third freshwater source, Black Sea outflow, has been
56 estimated to be minor, since the time of Black and Mediterranean Sea connection (9 to 8 cal
57 ka B.P., Soulet et al., 2011; Vidal et al., 2010) had occurred later than the onset of S1
58 deposition. Deglacial conditions were not compulsory for sapropel deposition (Rohling, 1994)
59 even if sea-level rise and consequent incursion of deglacial fresh Atlantic water towards the
60 eastern Mediterranean Sea have possibly contributed to surface water density decrease from
61 14.5 ka (Rohling et al., 2015).

62 In addition to orbitally driven insolation changes in a precession cycle, Mediterranean
63 Sea bottom water oxygenation records indicate centennial to millennial variability (Abu-Zied
64 et al., 2008; Casford et al., 2003; De Rijk et al., 1999; Hennekam et al., 2014; Kuhnt et al.,
65 2007; Rohling et al., 1997; Schmiedl et al., 2010). One of the most prominent changes
66 occurred around 8 cal ka B.P., mainly affecting shallow water masses (Rohling et al., 2015) at
67 water depths ≤ 1500 m in the South Aegean Sea (ex. sites SL123 and LC-21, Figure 1a and b)
68 and ≤ 1200 m in the Adriatic Sea (ex. MD90-917, not shown in figure; Siani et al., 2013).
69 Both internal processes such as winter cooling in the northern high latitudes (Schmiedl et al.,
70 2010) and solar activity (Hennekam et al., 2014; Rohling et al., 2002) have been proposed to
71 produce centennial to millennial variability in the eastern Mediterranean Sea.

72 The spatiotemporal distribution of S1 deposition in the eastern Mediterranean Sea
73 could provide further insight into the water column structure during Holocene. A compilation
74 study revealed a presence of anoxic boundary at about 1,800 m in water depth (De Lange et
75 al., 2008). However, the stability of this boundary from early to mid-Holocene is poorly

76 **known.** Regional-scale modelling studies allowed an examination of the physical and
77 biogeochemical processes behind S1 formation (Adloff, 2011; Adloff et al., 2011; Bianchi et
78 al., 2006; Grimm, 2012; Meijer and Tuentner, 2007) **but proxy reconstructions that can be**
79 **compared with simulations** are still scarce for the Holocene. This is because, under low
80 oxygen conditions, conventional approaches such as benthic foraminiferal $\delta^{13}\text{C}$ measurements
81 suffer from epibenthic foraminifera paucity (Jorissen, 1999). Bulk sediment geochemistry
82 provides complementary information when post-depositional elemental redistribution is
83 carefully considered (Calvert and Fontugne, 2001; Reitz et al., 2006; Thomson et al., 1995;
84 Thomson et al., 1999; van Santvoort et al., 1996).

85 In this study, we investigated bottom water oxygenation conditions **using** a core
86 MD04-2722 (33°06'N, 33°30'E, 1,780 m water depth, Figure 1) **by analyzing major**, minor
87 and trace elemental concentrations within bulk sediments obtained from XRF (X-Ray
88 Fluorescence) scanning and ICP-MS measurements. Stable isotopic analyses of the surface
89 dwelling planktonic foraminifer *Globigerinoides ruber* (white) and benthic foraminiferal
90 abundance were also conducted. Based on twelve ^{14}C dates of *G. ruber*, we established well-
91 dated high-resolution records to provide a bottom water ventilation history for the deep
92 eastern Mediterranean Sea associated with S1 deposition.

93

94 **2. Modern Mediterranean circulation and the study area**

95 Present-day Mediterranean Sea thermohaline circulation is characterized by an anti-
96 estuary pattern (Tomczak and Godfrey, 1994). Atlantic surface water enters the surface
97 western and eastern Mediterranean Sea by passing through the Gibraltar and Siculo-Tunisian
98 Straits, respectively. Through excess evaporation, the salinity of surface water continues to
99 increase, forming Modified Atlantic Water (MAW). In the Levantine Sea (Figure 1a), MAW
100 flows along the African coast off the Nile River then around Cyprus (Pinardi and Masetti,

101 2000). Between Cyprus and Rhodes, MAW is cooled by cold winter winds and can be
102 transformed into Levantine Intermediate Water (LIW, 14°C, salinity of 38.7, with a potential
103 density anomaly $\sigma_0 = 29.05 \text{ kg/m}^3$, Figure 1b) (Tomczak and Godfrey, 1994). The water mass
104 occupies 200-500 m in the Levantine Sea and consists of the major water mass flowing back
105 toward the western Mediterranean Sea through the Siculo-Tunisian Strait (Tomczak and
106 Godfrey, 1994). Deep water found in the eastern Mediterranean Sea is an admixture
107 consisting of LIW and Adriatic Sea surface water cooled by winter Bora winds (Tomczak and
108 Godfrey, 1994). This Eastern Mediterranean Deep Water (EMDW, 13°C, salinity of 38.6, $\sigma_0 =$
109 29.19 kg/m^3 , Figure 1b) is less saline but colder and denser than LIW, bathing below 600 m
110 in the eastern basin (Tomczak and Godfrey, 1994). In addition to the Adriatic Sea, deepwater
111 formation was observed in the Aegean Sea in the 90s due to increased salinity (Roether et al.,
112 1996). Activity in the zone as the site of deepwater formation is poorly known for the
113 Holocene.

114 At the location of core MD04-2722, surface water corresponds to the MAW and the
115 bottom water mass is estimated to be EMDW (Figure 1b).

116

117 **3. Materials and methods**

118 **3.1. Materials**

119 Core MD04-2722 (33°06'N, 33°30'E, 1,780 m water depth, total core length of 36.96
120 m) was collected in the south of Cyprus in the eastern Levantine Sea (Figure 1a) during the
121 VANIL cruise (R/V Marion Dufresne) conducted in 2004. In this study, we investigated the
122 first 2.5 m of sediments that covers the past 23.6 cal ka B.P. (Section 4). The sediment is
123 composed of a mixture of biogenic and fine terrigenous fractions (E. Ducassou, personal
124 communication, 2014). The first 48 cm of the sediments is homogeneous and presents
125 bioturbated hemipelagic mud facies. In the 48 to 62 cm interval, the core is brown with

126 typical oxidized sediments. The boundary surrounding 62 cm is not completely horizontal due
127 to sediment heterogeneity and/or coring processes. From 62 to 118 cm, sediments are dark
128 green with an oily aspect and do not contain visible laminations. The interval from 118 to 250
129 cm consists of bioturbated hemipelagic mud facies. The colour of sediments is represented in
130 grayscale values (Figure 2).

131 The core was sampled at every 2 cm interval for *G. ruber* (white) for stable isotope
132 analysis. Benthic foraminiferal abundance was studied at a 2 cm interval for **the first 134 cm**
133 **and with 2 to 10 cm resolution for the deeper part**. XRF scanning was performed every 5 mm
134 over the depths studied. The bulk sediment elemental composition was determined at a 2 cm
135 interval for 40 to 160 cm, and at a 4 to 20 cm interval for other depth ranges.

136

137 **3.2. Analytical methods**

138 All analyses were performed at CEREGE. High-resolution, non-destructive elemental
139 analyses were performed using an XRF core scanner (ITRAX, Cox Analytical Systems). The
140 relative abundance of S, Cl, Fe, Ti, V, Ca, Mn, and Br was measured under different
141 conditions. For Mn, V and Br measurements, a Mo tube was used as the X-ray source at 30
142 kV and 40 mA with 20 seconds of counting. For other elements, a Cr tube was used at 30 kV
143 and 30 mA with 20 seconds of counting. From the high-resolution optical image obtained by
144 ITRAX, a grayscale profile was extracted using ImageJ software (<http://imagej.nih.gov/ij/>).

145 To determine the concentrations of Al, Ca, Ti, Fe, Mn, Ba, Mo, U, V, As, Sb, Ni, Co,
146 Cu and Li in bulk sediments, well-homogenized, freeze-dried sediments (30 mg) were totally
147 dissolved in a mixture of ultrapure acids (1.7 ml of 15 M HNO₃, 1.3 ml of 22M HF and 0.1
148 ml of HClO₄). Obtained solutions were diluted and analyzed by an ICP-MS (Agilent 7500ce).
149 The accuracy of measurements was estimated using analyses of geostandards MAG-1 (marine
150 mud) and GSD12 (river sediment) that were subjected to the same digestion protocol as

151 samples. The analytical uncertainty was less than 5% and blank levels for the digestion
152 procedure were lower than 2% of the mean measured concentration for all of the analyzed
153 elements.

154 Calcite tests of *G. ruber* (white) were picked from the 250-355 μm size-fraction.
155 Foraminiferal $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ measurements were performed on a mass spectrometer (Finnigan
156 Delta Advantage) equipped with a carbonate device. The measured isotopic values were
157 normalized against NBS19. Mean external reproducibility was better than 0.05‰.

158 Benthic foraminifera abundance was determined by counting all existing calcareous
159 specimens in the $>150\ \mu\text{m}$ size fraction and dividing the number by the corresponding dry
160 bulk sediment weight. Since the benthic foraminiferal abundance became very high in
161 sediments below 134 cm, total benthic foraminiferal numbers were only counted for the first
162 132 cm. Dominant benthic foraminiferal species were identified for the studied interval.

163

164 **4. Chronology**

165 Chronology of core MD04-2722 is based on twelve AMS ^{14}C ages performed at the
166 ARTEMIS facility (Gif-sur-Yvette, France) on *G. ruber* (white) obtained from the $>150\ \mu\text{m}$
167 size fraction. Conventional radiocarbon ages were converted into calendar ages based on
168 MARINE13 (Reimer et al., 2013) using the ^{14}C calibration software CALIB 7.0.1 (Stuiver
169 and Reimer, 1986-2013) (Table 1). The calibration integrates a marine ^{14}C reservoir age of
170 400 years that agrees well with Mediterranean Sea surface water radiocarbon reservoir age of
171 390 ± 85 years (Siani et al., 2000; Siani et al., 2001).

172 The estimated mean sedimentation rate was 12 cm/ka. According to the age model, the
173 typical temporal resolution for XRF measurements (5 mm), stable isotopes (2 cm), and ICP-
174 MS measurements (2 to 10 cm) is approximately 40 years, 170 years, and 170 to 800 years,
175 respectively.

176

177 **5. Results**

178 For the studied interval, the Al and Ca concentrations in bulk sediments ranged
179 between 4.4 and 6.9% and between 10 and 17%, respectively (not shown in the figure). Based
180 on the assumption that the Al concentration in the detrital fraction was 8.0% (upper
181 continental crust; McLennan, 2001) and that all Ca was in the form of CaCO₃, we roughly
182 estimated that the two major components in the bulk sediments were detrital (55 to 86%) and
183 carbonate (25 to 42%) fractions. Considering the high proportion for the detrital component,
184 enrichment of the analyzed elements was evaluated using normalization against Al. By taking
185 into account the difficulty of precisely analyzing Al due to the attenuation of the XRF signal
186 by pore water (Tjallingii et al., 2007), a Ti normalization was used for the XRF results .

187

188 **5.1. Evaluation of the S1 layer**

189 In core MD04-2722, the preserved S1 layer was visually recognized as the dark color
190 between 60 and 120 cm with the slightly lighter colour interval from 80 to 100 cm shown in
191 the grayscale profile (Figure 2). High-resolution variability in organic matter content was
192 shown by a Br/Cl ratio profile obtained from a XRF scan (Figure 2). **Although Br XRF**
193 **intensity and Br/Cl ratio presented very similar variability (not shown in figure),** we used the
194 Br/Cl XRF intensity ratio to better illustrate changes in the organic matter content **(Cartapanis**
195 **et al., 2014)** since Br is incorporated in marine organic matter but also exists in pore water
196 (Cartapanis et al., 2011; Ziegler et al., 2008). High Br/Cl ratio values were found in the 55 to
197 120 cm interval and a clear decrease existed between 60 and 110 cm (Figure 2). Below 120
198 cm, the Br/Cl ratio was low and comparable to the core-top value (Figure 2).

199 Barium enrichment has often been used to localize initial sapropel layers, since it is
200 associated with biogenic barite (BaSO₄) that is an export production proxy resistant to post-

201 depositional oxidation (Calvert and Fontugne, 2001; De Lange et al., 2008; Thomson et al.,
202 1995; Thomson et al., 1999; van Santvoort et al., 1996). Both the Ba concentration and Ba/Al
203 ratio in core MD04-2722 displayed a similar convex-shaped peak from 55 to 120 cm,
204 consistent with the high Br/Cl ratio interval (Figure 2). The depth range of high Ba and
205 organic matter content at 55 to 120 cm corresponded to 6.8 to 10.4 cal ka B.P. (Table 1). This
206 time span is, in general, in agreement with the previously estimated S1 deposition period of
207 6.1 ± 0.5 to 10.8 ± 0.4 cal ka B.P. (De Lange et al., 2008).

208 Close to the upper end of the Ba/Al ratio peak, prominent Mn enrichment was found at
209 56 to 60 cm (6.8 to 7.0 cal ka B.P.) for both the Mn/Al and Mn/Ti ratio profiles (Figure 2).
210 The peak shape of Mn/Al and Mn/Ti ratios was slightly different around 62 cm, possibly due
211 to different sampling resolutions (1 cm for the Mn/Al ratio and 5 mm for Mn/Ti ratio) and the
212 irregular brown-coloured boundary at approximately 62 cm (see section 3.1). As the Mn peak
213 is mainly associated with the precipitation of Mn oxides under improved oxygenation, it often
214 marks the end of S1 (Reitz et al., 2006). Taken together, we defined the S1 layer of core
215 MD04-2722 at 55 to 120 cm (the dark gray band in Figure 2).

216 The number of benthic foraminifera gradually increased from the core-top to 56 cm
217 and reached a maximum value at 56 to 58 cm, with the Mn peak (Figure 2). Between 55 and
218 142 cm, benthic foraminiferal abundance displayed a concave shape (Figure 2). In the S1
219 layer, the sediment was void of benthic foraminifera at 104 cm (9.2 cal ka B.P.) and 108 cm
220 (9.4 cal ka B.P.). Below 142 cm (12.6 cal ka B.P.), benthic foraminiferal abundance was
221 higher than for the upper part. Major species identified within the studied interval included
222 *Gyroidina* spp., *Cibicidoides pachyderma*, *Hoeglundina elegans*, *Cibicidoides wuellerstorfi*,
223 *Uvigerina* spp., *Bolivina spatulata*, and *Globobulimina* spp. (not shown in the figure). The
224 first appearance of *Globobulimina* spp., indicators of low oxygen content (Jorissen et al.,

225 1995) is at 138 cm (12.2 cal ka B.P.). A detailed faunal assemblage will be provided
226 elsewhere.

227

228 **5.2. Indicators of dry/wet conditions**

229 The major detrital fraction in the study area originated from the Nile River particles
230 and Saharan dust (Krom et al., 1999). Ti/Al values have been used as an indicator of the
231 relative proportion of Nile River particles to Saharan dust (Wehausen and Brumsack, 2000;
232 Ziegler et al., 2010). In general, due to greater inputs of Al-rich river particles as compared to
233 the contribution of Ti-rich dust inputs (Wehausen and Brumsack, 2000; Ziegler et al., 2010),
234 the Ti/Al ratios of sediments far from the coastal zones in the eastern Mediterranean Sea were
235 lower under wet conditions. Ti/Al values in core MD04-2722 varied from 0.07 to 0.09 g/g for
236 the upper 250 cm with lower values between 35 and 230 cm (5.4 to 22.1 cal ka B.P.) (Figure
237 2).

238 In addition to Ti/Al ratios, *G. ruber* $\delta^{18}\text{O}$ values reflect wet/dry conditions and also
239 cool/warm surface water temperatures and continental ice volume. The isotopic values of core
240 MD04-2722 ranged between -1.4‰ and 3.5‰ with minimum and maximum values at 112 cm
241 (9.6 cal ka B.P.) and 220 cm (21.3 cal ka B.P.), respectively (Figure 2). The interval for low
242 *G. ruber* $\delta^{18}\text{O}$ values roughly agreed with the low Ti/Al interval (Figure 2).

243

244 **5.2. Redox sensitive elements**

245 Due to pyrite formation under reducing conditions, the sulphur content of bulk
246 sediments is high in sapropel layers (Passier et al., 1996; Rohling, 1994). Considering that
247 sulphur also exists in pore water in the form of the sulphate ion, we used the S/Cl ratio in core
248 MD04-2722 to extract a signal closely related to pyrite even if S XRF intensity and S/Cl
249 presented virtually identical profiles (not shown in figure). S/Cl values in core MD04-2722

250 indicated variability similar to the Ba/Al ratio (Figure 3) but enrichment below the S1 layer at
251 120 to 150 cm (the darkest gray band in Figure 3). This enrichment can be explained by
252 downward sulphidisation (see section 6.1). The downward sulphidisation could account for
253 high values for the S/Cl, Ni/Al, As/Al and Fe/Al ratios below the S1 layer (Figure 3) since As
254 and Ni (and also Co and Cu) are often adsorbed and/or incorporated in pyrite (Large et al.,
255 2014).

256 In general, high sedimentary Ni, Co and Cu concentrations are produced by enhanced
257 productivity via the transfer of these elements due to the organic matter flux and fixation in
258 sediments under reducing environments (Cartapanis et al., 2011; Nameroff et al., 2002). Ni/Al
259 values in core MD04-2722 were higher inside and below the S1 layer (Figure 3). The Co/Al
260 and Cu/Al ratios indicated variability comparable to the Ni/Al ratio (not shown in the figure).

261 A remarkable feature of the As/Al and Fe/Al profiles was strong depletion centred at
262 82 to 84 cm (around 8 cal ka B.P. inside the light gray band in Figure 3). Depletion was also
263 observed for the V/Al profile with a wider depth range between 80 and 110 cm (7.9 to 9.5 cal
264 ka B.P.) (Figure 3).

265 The U, Mo and Sb content of core MD04-2722 indicated a clear peak centred at the
266 interval of the high Ba/Al ratio but a detailed structure was specific to each element (Figure
267 3). The U/Al profile indicated a smooth convex shape between 60 and 148 cm (7.0 to 13.2 cal
268 ka B.P.), with a small negative spike at 84 cm (8.2 cal ka B.P., inside the light gray band in
269 Figure 3). The main peak for the Mo/Al and Sb/Al ratios ranged between 70 and 138 cm (7.6
270 to 12.2 cal ka B.P.), and 50 and 138 cm (6.3 to 12.2 cal ka B.P.), respectively (Figure 3). The
271 Mo/Al and Sb/Al ratios indicated a sharp peak from 56 to 60 cm where the benthic
272 foraminiferal number (Figure 2) and the Mn content were at a maximum (the orange band in
273 Figure 3). In addition to the Mo/Al and Sb/Al ratios, the Li/Al ratios of core MD04-2722
274 presented a peak synchronous with the high Mn content (Figure 3).

275

276 **6. Discussion**

277 Redox-sensitive elements and benthic foraminiferal abundance in core MD04-2722
278 can be used as an indicator of bottom/pore water conditions which, in turn, are modulated by
279 bottom water ventilation and biological productivity, whereas the Ti/Al ratio and *G. ruber*
280 $\delta^{18}\text{O}$ records reflect wet/dry conditions. In the following discussion, we first describe the
281 geochemical meaning of the different elemental ratios that can be used to infer Mediterranean
282 Sea ventilation. We then propose deepwater conditions for the onset, during and at the
283 termination of S1 deposition by combining the whole obtained data.

284

285 **6.1. Geochemical meaning of elemental ratios**

286 During sapropel deposition, sulphate reduction occurred, and HS^- excess for pyrite
287 precipitation migrated downwards whereas pore water Fe^{2+} existing below sapropel layer
288 moved upwards, leading to pyrite precipitation (Passier et al., 1996). The process is called
289 downward sulphidisation and impacts the distribution of trace elements such as Ni, Co, As
290 and Cu associated with pyrite (Passier et al., 1996). Hence, the enrichment of Ni, Co and Cu
291 below the S1 interval may not signify an increase in biological productivity prior to S1
292 deposition. Considering the difficulty in interpreting peaks below the S1 layer, here, we do
293 not discuss them further.

294 Due to insoluble chemical speciation in reducing environments, authigenic U and Mo
295 accumulate under suboxic and anoxic conditions, respectively (Algeo and Maynard, 2004;
296 Klinkhammer and Palmer, 1991; Tribovillard et al., 2006). The accumulation of these
297 elements does not necessarily indicate that the bottom water was permanently highly depleted
298 in oxygen. Enrichment could reflect suboxic/anoxic pore water conditions related to slow
299 ventilation and/or high organic rain. Taking this fact into account, we carefully evaluated

300 potential circulation changes by combining the U/Al and Mo/Al profiles of core MD04-2722
301 (Figure 3) with results from previous studies (see section 6.2).

302 In the middle of the S1 unit, the sediment colour was lighter and the sedimentary
303 organic matter (Br/Cl ratio) and redox sensitive element (Fe/Al, As/Al and V/Al ratios) were
304 lower (light gray band in Figures 2 and 3). The observed change could be explained by the
305 injection of dissolved oxygen into the water column via re-ventilation. The oxidation of
306 organic matter within the water column and on the sea floor can reduce sedimentary Br/Cl
307 values. The pyrite in sediments can be oxidized to iron oxides and/or to hydroxides when it is
308 in contact with dissolved oxygen (Chandra and Gerson, 2011). If these oxides and/or
309 hydroxides are again reduced in sediments, Fe and As associated with pyrite would be
310 released to pore water. Vanadium accumulation in anoxic sediments commonly takes place
311 via diffusion across the sediment/water interface and the release of this element occurs when
312 Mn is reduced (Nameroff et al., 2002). We investigate this issue in section 6.3.

313 The origin of the Mn peak at the end of S1 has been extensively discussed in previous
314 studies (Reitz et al., 2006; Thomson et al., 1995; Thomson et al., 1999; van Santvoort et al.,
315 1996). Core MD04-2722 only contains one Mn peak at the end of the S1 unit (Figure 2) and
316 the Mo/Al, Sb/Al and Li/Al ratios synchronously increase with Mn (Figure 3). Since Mo
317 (Shimmiel and Price, 1986; Tribovillard et al., 2006), As (Cutter et al., 2001 and references
318 therein) and Li (Reitz et al., 2006) can be scavenged by Mn oxides, the peaks observed for
319 core MD04-2722 were likely produced from water column oxygenation at the S1 termination
320 (section 6.4).

321 The following discussion focuses on the state of bottom water circulation in the deep
322 eastern Mediterranean Sea during the following periods: (i) the onset of S1 deposition, (ii)
323 oxygenation event(s) in the middle of the S1 unit, and (iii) the termination of S1 deposition.
324 We are aware that, due to diagenetic processes and post-despositional diffusion that can

325 modify boundary positions, precise timing for ventilation changes is difficult to obtain from a
326 single proxy profile. Considering the fact that distribution of each element is determined by
327 element-specific processes, we interpret and discuss the timing of ventilation changes when
328 several proxy reconstructions indicate consistent changes.

329

330 **6.2. Conditions of bottom water circulation prior to S1 deposition**

331 In core MD04-2722, U/Al and Mo/Al ratios began to increase from 13 cal ka B.P.
332 (148 cm) to 12 cal ka B.P. (138 cm, Figures 3 and 4). This time span corresponds also to the
333 beginning of decrease in benthic foraminiferal number (12.2 cal ka B.P., 138 cm; Figures 2
334 and 4) that was high in well-oxygenated glacial bottom waters. Two major factors affecting
335 benthic foraminiferal assemblage are food supply and water oxygenation (Jorissen et al.,
336 1995). Since the Ba/Al change indicates increasing trend of export production just before the
337 S1 deposition (Figure 2), it is logical to assume that the observed reduction of benthic
338 foraminiferal number was related to oxygen depletion. Therefore, the result can be interpreted
339 as the onset of oxygen depletion for bottom and/or pore waters at 1,780 m in water depth
340 started prior to S1 deposition. Since surface water freshening has been considered to be the
341 main factor affecting Mediterranean Sea thermohaline circulation (De Lange et al., 2008;
342 Emeis et al., 2000; Rohling, 1994), we examine timing for the onset of wet conditions,
343 changes in surface hydrology, and deepwater circulation using records obtained from core
344 MD04-2722.

345 Decreases for the Ti/Al ratio and *G. ruber* $\delta^{18}\text{O}$ in core MD04-2722 began prior to S1
346 deposition (Figure 4). To better illustrate surface water salinity changes in the Levantine Sea,
347 the *G. ruber* $\delta^{18}\text{O}$ record is combined with sea surface temperature (SST) reconstruction that
348 is based on planktonic foraminiferal assemblages obtained from a vicinity site (core MD84-
349 632, 32°28'N, 34°13'E; Essallami et al., 2007; Figure 1a). The present-day bottom water $\delta^{18}\text{O}$

350 value (Pierre, 1999) is then subtracted so that the local $\delta^{18}\text{O}_w$ anomaly can be calculated
351 (Figure 4). A marked decrease in the $\delta^{18}\text{O}_w$ anomaly began at approximately 12 cal ka B.P.
352 (Figure 4). Variability mainly stems from the core MD04-2722 *G. ruber* $\delta^{18}\text{O}$ record, not on
353 temperature because the major feature is maintained when another SST record from a nearby
354 site is applied (Castañeda et al., 2010). We note here that the large amplitude of seawater $\delta^{18}\text{O}$
355 anomaly change of 2.7‰ from 20.1 to 9.6 cal ka B.P. cannot be explained by global $\delta^{18}\text{O}$
356 change related to continental ice volume (Waelbroeck et al., 2002). Another record of *G.*
357 *ruber* $\delta^{18}\text{O}$ from a close site (core MD84-641; 33°02'N; 32°28'E, 1,375m water depth)
358 (Fontugne and Calvert, 1992) indicates a very similar variability to the core MD04-2722
359 record, attesting that the *G. ruber* $\delta^{18}\text{O}$ variability represents regional hydrological changes.

360 The $\delta^{18}\text{O}_w$ anomaly and Ti/Al records indicate the beginning of a fresher and wetter
361 period at approximately 12 and 15 cal ka B.P., respectively, comparable with the inception of
362 the African humid period at 12.5 cal ka B.P. (Adkins et al., 2006; deMenocal et al., 2000) and
363 increased Nile River discharge (Revel et al., 2014) in response to insolation changes (Laskar
364 et al., 2004) (Figure 4). Our result is consistent with the hypothesis that Nile River discharge
365 was one of the main fresh water sources for the Levantine Sea (Rossignol-Strick et al., 1982).
366 Indeed, the influence of Nile River discharge was estimated to have spread as far as Cyprus
367 (Almogi-Labin et al., 2009) (Figure 1a).

368 Spatial extension of weakly ventilated waters can be evaluated by comparing with
369 previously reported records from the eastern Mediterranean Sea. Based on the U-Th dating of
370 authigenic carbonates formed in reducing environments, the start of suboxic bottom water
371 conditions was estimated to be 12 cal ka B.P. on the Nile River deep-fan at 1,160 m in water
372 depth (“buildups”; Bayon et al., 2013) (Figures 1ab and 4). Based on decreasing benthic
373 foraminiferal $\delta^{13}\text{C}$ values that began at 15 cal ka B.P (Figure 4), weaker ventilation has also
374 been estimated for a water depth of approximately 700 m in the south Aegean Sea (site

375 SL123) and for a water depth of approximately 900 m in the southeast Levantine Sea (site
376 SL112) (Kuhnt et al., 2007; Schmiedl et al., 2010) (Figure 1ab). It is worth noting that the
377 decreasing trend of foraminiferal benthic $\delta^{13}\text{C}$ values does not necessarily mean complete
378 shutdown of ventilation. The $\delta^{13}\text{C}$ values (Figure 4) were still comparable with the late
379 Holocene values, and both benthic foraminiferal assemblage and trace elements (Figure 5) did
380 not indicate strong anoxic conditions before S1 interval. We thus propose reduced
381 intermediate/deepwater formation and consequently restricted extension of well-oxygenated
382 water mass(es) for this period. Productivity change is estimated to have a second role because
383 it would be more local/regional, reflecting coastlines, local nutrient supplies such as riverine
384 inputs, and the topography of lands that impact wind regimes.

385 The net increase in fresh water inputs reinforced vertical salinity gradients in the
386 Eastern Mediterranean Sea, which in turn shoaled the pycnocline at a depth shallower than the
387 euphotic layer, allowing the development of a deep chlorophyll maximum relative to present
388 day in the easternmost Mediterranean Sea (Castradori, 1993; Grelaud et al., 2012; Rohling,
389 1994). A shoaling pycnocline and a greater nutrient supply due to Nile River discharge (Herut
390 et al., 2000) could have contributed to enhanced biological productivity. Considering the time
391 required for circulation reorganization, the gradual consumption of existing dissolved oxygen
392 in the water column by organic decomposition, and the improvement of sedimentary organic
393 matter preservation under oxygen-depleted conditions (Hartnett et al., 1998), it is not
394 surprising that a net increase in the Br/Cl ratio in core MD04-2722 appeared later than the
395 onset of slow bottom water circulation (Figures 2 and 3).

396 Recent regional-scale modelling studies have proposed that initial glacial conditions
397 were a key parameter for forming S1 deposition (Adloff, 2011; Grimm, 2012). Sensitivity
398 experiments indicate that fresh water contributions and increased productivity are not
399 sufficient for attaining the observed bottom water oxygen depletion (Adloff, 2011; Grimm,

400 2012). Only a simulation that considered initial glacial conditions succeeded in maintaining
401 oxygen-depletion for several thousand years that were comparable to S1 duration (Grimm,
402 2012). The inception of weaker ventilation during the glacial-interglacial transition, as
403 proposed in this study, is in line with these recent simulations.

404

405 **6.3. Re-ventilation in the middle of the S1 period**

406 The lighter colour of the sediment and the clear decrease in the Br/Cl (Figure 2),
407 As/Al, Fe/Al and V/Al ratios (Figure 3) in core MD04-2722 indicate temporally improved
408 oxygenation in the water column and in pore water in the middle of the S1 period. Since the
409 Ba/Al ratio did not show clear diminution at this interval (Figure 2), biological productivity
410 was not responsible for the change. Improved oxygenation seems to be related to an active re-
411 ventilation that promoted organic matter degradation in the water column and post-
412 depositional oxidation (Thomson et al., 1999). To examine the variability in detail, we
413 combine highly resolved Br/Cl, Fe/Ti and V/Ti records with Fe/Al, As/Al and V/Al ratios
414 (Figure 5). The major decline in the Fe/Al, Fe/Ti and As/Al ratios is centred at 8.4 to 8.2 cal
415 ka B.P., whereas minimal Br/Cl, V/Al and V/Ti ratios extended between 9.5 to 7.9 ca ka B.P.
416 (Figure 5). A longer depletion period for the Br/Cl, V/Al and V/Ti ratios can be explained by
417 continuous organic matter oxidation with oxygen penetrated into sediments as well as the
418 subsequent reduction of Mn oxides (see section 6.1).

419 Our results indicate that re-ventilation event(s) affected bottom water located at the
420 upper limit of the anoxic layer at 1,800 m (De Lange et al., 2008). By combining the results
421 of core MD04-2722 with previously reported high-resolution records, we examine the spatial
422 coverage of re-oxygenation. In addition to shallow Adriatic Sea (MD90-917, 41°17'N, 17°37'
423 E, 1,010 m, not shown in the figure; Siani et al., 2013) and shallow south Aegean Sea (sites
424 LC-21 and SL123 ≤ 1500 m; Fig. 1), benthic foraminiferal assemblage indicates re-ventilation

425 in the Levantine Sea at water depths ranging from 900 m (site SL112, Fig. 1; Schmiedl et al.,
426 2010) to 2,300 m (site LC-31, Fig. 1; Abu-Zied et al., 2008; Schmiedl et al., 2010). We
427 propose two hypotheses about re-ventilation depths. Firstly, the re-ventilation affected water
428 column down to 2,300 m. The second possibility is the re-ventilation was limited to water
429 depths shallower than 1,800 m taking into account the fact that core LC-31 contains a small
430 slump confirmed by AMS ^{14}C dates inside of the S1 layer (Abu-Zied et al., 2008). In any
431 case, the large spatial distribution of the S1 interruption at least as deep as 1,780 m suggests a
432 basin-wide reorganization for ventilation.

433 Cores presenting the S1 interruption are currently bathed in well-oxygenated EMDW
434 (Figures 1b and 5). At present, the formation of EMDW is controlled by the winter cooling of
435 surface water in the Adriatic Sea and occasionally in the Aegean Sea (Roether et al., 1996), as
436 well as by the salinity of LIW (section 2). Holocene surface water winter cooling in the
437 Aegean Sea has been demonstrated to occur with atmospheric circulation changes in relation
438 to the winter and spring Siberian High at 9.5 to 9.1 and 8.8 to 7.8 ka (Kotthoff et al., 2008;
439 Marino et al., 2009; Mayewski et al., 1997), and for the 8.2 ka event (Pross et al., 2009)
440 recorded in Greenland ice cores (Alley and Ágústsdóttir, 2005). Due to the cold air flux from
441 polar regions, surface water in convection zones could be cooled during winter, leading to the
442 activation of deep water formation in the Aegean (Schmiedl et al., 2010) and/or the Adriatic
443 (Siani et al., 2013) Seas. A numerical simulation has indicated that a surface eastern
444 Mediterranean Sea cooling of 2-3 °C could trigger deep convection in the Adriatic Sea and
445 intermediate water formation in the Aegean Seas, leading to oxygenation for the upper 1,250
446 m water masses of the eastern Mediterranean Sea (Myers and Rohling, 2000)

447 On the other hand, reconstructed Nile River discharge (Figure 4) presented a high-
448 frequency variability and a decline around 8 cal ka B.P. (Blanchet et al., 2014; Revel et al.,
449 2014). Planktonic foraminiferal Ba/Ca ratios indicate the fluctuation in Nile River discharge

450 at 8.4-8.2 cal kyr B.P. (Weldeab et al., 2014). Reduced Nile River discharge could have led to
451 a salinity rise for LIW that mixed with surface Adriatic Sea water, contributing to the
452 activation of EMDW formation. Thus, both temperature and salinity effects could be factors
453 for re-ventilation in the eastern Mediterranean Sea (Figure 6) in relation to northern high
454 latitude and tropical/subtropical climate conditions.

455 During the interruption period, vertical density gradient was attenuated, leading to
456 partial mixing between old dense glacial waters and overlaying lighter waters. Once cooling
457 and/or reduced fresh water input finished, the stagnant ventilation mode came back, which
458 suggests that the density barrier was still too strong to shift to the present-day circulation
459 mode. If the duration of cooling/less fresh water inputs, the size of this forcing and the
460 spatiotemporal S1 interruption are quantified, interruption can provide information regarding
461 the sensitivity of eastern Mediterranean thermohaline circulation.

462

463 **6.4. Total ventilation recovery at the S1 termination**

464 Mn/Al, Mn/Ti, Mo/Al, Li/Al and Sb/Al ratios (Figure 3), as well as the benthic
465 foraminiferal abundance of core MD04-2722 (Figure 2), indicated a synchronous increase at
466 7.0 cal ka B.P. (60 cm) to 6.8 cal ka B.P. (56 cm), suggesting oxygenation in the water
467 column at the core location. The age range for the increase was slightly earlier than the basin-
468 wide S1 termination of 6.1 ± 0.5 cal ka B.P. that was estimated using the cores from water
469 depths down to 3400 m (De Lange et al., 2008) although, considering the dating uncertainty,
470 the difference was subtle. If the age difference is real, the S1 termination may be
471 characterized by depth-dependant ventilation recovery with earlier oxygenation at depths
472 shallower than 1,800 m. Based on previous observations, we suggest this possibility. At first,
473 benthic ecosystem recovery at the S1 termination was depth-dependent, with a prior recovery
474 at shallower depths in the eastern Mediterranean Sea (Schmiedl et al., 2010). The second

475 point is the possible existence of dense deep water. Modelling studies have indicated that the
476 deep water mass was much denser than that of shallower waters because they contained saline
477 glacial (Grimm, 2012; Myers et al., 1998) (Figure 6). If the increase in surface water density
478 did not exceed the density of this dense water below, ventilation would affect only lighter
479 water masses at shallower depths.

480 Ventilation recovery can be explained both by the reduction of riverine fresh water
481 inputs and surface water cooling. A 9,000-yr-long transient model with insolation and
482 atmospheric greenhouse gas forcing simulated a gradual precipitation decline over eastern
483 Africa from the early to late Holocene (Renssen et al., 2006). Reconstructed Nile River
484 discharge (Blanchet et al., 2014; Revel et al., 2014; Weldeab et al., 2014) displayed a
485 decreasing trend from the early to mid-Holocene (Figure 4). Approximately 3°C of the SST
486 decrease during April-May was recorded in the south Adriatic between 7.1 and 6.9 cal ka BP
487 (Siani et al., 2013). In the south Aegean Sea during winter, an approximate 2 to 3 °C in
488 surface water cooling was observed from 7.5 to 7.0 cal ka B.P. (Marino et al., 2009). Once
489 surface water density within the deep/intermediate water formation zones exceeded the
490 threshold value, water convection could restart. The onset of these cooling records is
491 consistent with our estimation for total ventilation recovery at water depths shallower than
492 1,800 m from 7.0 to 6.8 cal ka B.P.

493 Considering the subtle density difference between the present LIW and EMDW
494 (section 2) and distinct climate background between the present and the moment of S1
495 termination, it is unattainable to precisely identify physical mechanisms of total ventilation
496 recovery. Nonetheless, if ventilation recovery was water depth dependent, the S1 termination
497 contrasted with re-ventilation event(s) during the S1 period, affecting waters below the
498 critical depth of 1,800 m in the Levantine Sea (Figures 5 and 6). The finding provides new
499 constraints for vertical structure within the eastern Mediterranean Sea during S1 deposition.

500

501 **Conclusions**

502 We analyzed the bulk sediment elemental composition, the $\delta^{18}\text{O}$ of *Globigerinoides*
503 *ruber* and the abundance of benthic foraminifera since the last deglaciation within core
504 MD04-2722 obtained from the Levantine Sea. Water depth at the core location was close to
505 the estimated anoxic layer upper limit for the most recent sapropel S1 deposition, 1,800 m, and
506 rendered the site highly sensitive to past circulation changes. By combining our new results
507 with previous studies, some fundamental features for ventilation in the eastern Mediterranean
508 Sea were identified.

509 Bulk sediment Ti/Al ratios and the surface water $\delta^{18}\text{O}$ anomaly calculated from the *G.*
510 *ruber* $\delta^{18}\text{O}$ obtained from core MD04-2722 indicated that a wet period and fresher surface
511 water appeared at the core site around 15 to 12 cal ka B.P. The enrichment of Mo and U, as
512 well as benthic foraminiferal density indicated that surface hydrological changes were
513 transferred to bottom water, leading to the reduction of intermediate/deep water formation and
514 consequent restricted expansion of oxygenated that began prior to S1 deposition. Our results
515 are consistent with previous reconstructions and regional-scale simulations, and support the
516 idea that reduced oxygen supply due to slow ventilation was a precondition of S1 formation.

517 Decreased Br/Cl, Fe/Al, Fe/Ti, V/Al, V/Ti and As/Al ratios indicated that temporal re-
518 oxygenation event(s) occurred during the middle of the S1 period. Improved oxygenation was
519 produced by active re-ventilation rather than reduced biological productivity and affected
520 water depths at least as deep as 1,800 m in the Levantine Basin. Winter cooling in the Aegean
521 and Adriatic Seas in relation to northern high latitude conditions and salinity increases related
522 to reduced Nile River discharge contributed to the temporal reactivation of thermohaline
523 circulation in the eastern Mediterranean Sea.

524 From the concomitant peak of Mn, Mo, Sb, and Li, and the **increased** abundance of
525 benthic foraminifera, a total recovery in ventilation at the core MD04-2722 site was estimated
526 to have occurred at 7.0 to 6.8 cal ka B.P. We tentatively propose a depth-dependent S1
527 termination with an earlier ventilation at water depths shallower than 1,800 m. This study
528 provides new constraints for the eastern Mediterranean Sea bottom water circulation since the
529 last deglaciation that could be examined by future modelling studies.

530

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540

541 **Figure captions**

542 Figure 1. The core location map. (a) The core MD04-2722 (33°06'N, 33°30'E, 1,780 m water
543 depth) location on the bathymetry map. Site locations of the records discussed in the text are
544 as follows: MD84-632 (32°28.2'N, 34°13.2'E) (Essallami et al., 2007); **MD84-641 (33°02'N;**
545 **32°28'E) (Fontugne and Calvert, 1992);** SL123 (35°45.3'N, 27°33.3'E, 728 m water depth);
546 SL112 (32°44.5'N, 34°39.0'E, 892 m water depth) (Kuhnt et al., 2007); LC-21 (35°39.7'N,
547 26°35.0'E, 1,520 m water depth); LC-31 (34°59.8'N, 31°09.8'E, 2,300 m water depth)
548 (Schmiedl et al., 2010); “buildups” (32°22'N, 31°42'E, 1,160 m water depth) (Bayon et al.,

549 2013). (b) The present-day density anomaly transect along the dashed blue line at 34.5-35.5N
550 in Figure 1a. The seawater temperature and salinity are from WOA09 (Antonov et al., 2010;
551 Locarnini et al., 2010). The dashed white line indicates the upper limit of the permanent
552 anoxic layer during S1 deposition at 1,800 m (see the text for detail). Figures were generated
553 using the Ocean Data View software (Schlitzer, 2009).

554

555 Figure 2. Characterization of the S1 interval recorded in core MD04-2722. Grayscale value,
556 bulk elemental composition, benthic foraminiferal abundance (number of tests per unit of
557 weight for dry bulk sediment), *G. ruber* $\delta^{18}\text{O}$ as a function of depth in core (cm). The S1 layer
558 (55 to 120 cm) is shown with a dark gray band, the interval of low grayscale values (80 to 100
559 cm) with a light gray band, and the interval of Mn enrichment (56 to 60 cm) with an orange
560 band. Triangles indicate the depth levels dated by AMS ^{14}C (see Table 1 for details).

561

562 Figure 3. The elemental concentrations and the element/Al ratios for the bulk sediment of
563 MD04-2722 as a function of depth in core, together with the S/Cl XRF intensity. Dark and
564 light gray bands indicate the S1 layer determined from Ba enrichment (55 cm to 120 cm) and
565 the interval of low grayscale values (80 to 100 cm), respectively. The darkest gray band and
566 the orange band indicate the interval affected by downward sulphidisation (Passier et al.,
567 1996) (120 to 150 cm) and Mn enrichment (56 to 60 cm), respectively.

568

569 Figure 4. The last 23 ka variability of proxies for ventilation (U/Al, Mo/Al and benthic
570 foraminiferal density) and wet/fresh conditions (Ti/Al and seawater $\delta^{18}\text{O}$ anomaly) based on
571 core MD04-2722 results as compared with previous studies. The surface water $\delta^{18}\text{O}$ anomaly
572 was calculated by combining the *G. ruber* $\delta^{18}\text{O}$ record from core MD04-2722 with the SST
573 reconstruction (see text for detail). Suboxic conditions on the Nile deep-fan were estimated to

574 have lasted for 12 to 7 ka B.P. at the “buildups” location (Figure 1 a and 1b) (Bayon et al.,
575 2013). Benthic foraminiferal $\delta^{13}\text{C}$ records were based on the epibenthic foraminifer species
576 *Planulina ariminensis* (Kuhnt et al., 2007; Schmiedl et al., 2010). Changes in Nile River
577 discharge are shown with the log-scale Fe/Ca ratio for core MS27PT located close to the
578 Rosetta mouth of the Nile River (Revel et al., 2014). The precession parameter is from Laskar
579 et al. (2004). The blue zone indicates the African Humid Period (AHP, 12.5 to 5.5 cal ka
580 B.P.) based on Adkins et al. (2006) and deMenocal et al. (2000). The gray band indicates the
581 S1 period from 10.4 to 6.8 cal ka B.P. estimated for core MD04-2722. The dashed line
582 presents the possible onset for weaker ventilation at 15 cal ka B.P. in the eastern
583 Mediterranean Sea. LGM = Last Glacial Maximum.

584

585 Figure 5. Re-oxygenation in the middle of the S1 interval as inferred from the bulk
586 geochemistry (Br/Cl, Fe/Al, Fe/Ti, As/Al, V/Al and V/Ti ratios) in core MD04-2722 and the
587 benthic foraminiferal oxygen index obtained at sites LC-31 and SL123 (Schmiedl et al., 2010)
588 (Figure 1ab). Dark and light gray bands indicate the S1 unit (10.4 to 6.8 cal ka B.P.) and the
589 interval of low grayscale values (9.0 to 7.9 cal ka B.P.) in core MD04-2722, respectively.

590

591 Figure 6. Schematic ventilation patterns in the eastern Mediterranean Sea at present, S1
592 interruption and 15-12 ka intervals.

593

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874

875

875 Table 1. Radiocarbon ages for core MD04-2722

Depth in core (cm)	AMS sample n°	¹⁴ C age ± 1σ (yr BP)	Cal. age* (yr BP)	95.4 % (2σ) cal age ranges (yr BP)
2	SacA26352	3765 ± 30	3700	3604 - 3812
20	SacA26353	4400 ± 30	4551	4433 - 4678
48	SacA31590	5760 ± 40	6188	6061 - 6277
64	SacA26354	6790 ± 30	7318	7249 - 7396
80	SacA31591	7485 ± 45	7945	7835 - 8042
112	SacA26355	8980 ± 35	9629	9525 - 9763
136	SacA26356	10680 ± 40	12048	11843 - 12292
150	SacA26357	11925 ± 40	13377	13275 - 13483
176	SacA26358	13675 ± 45	15955	15762 - 16130
192	SacA26359	15170 ± 50	17964	17793 - 18139
220	SacA26360	18030 ± 60	21313	21040 - 21555
250	SacA26361	20000 ± 70	23611	23354 - 23878

876 * Median probability

877

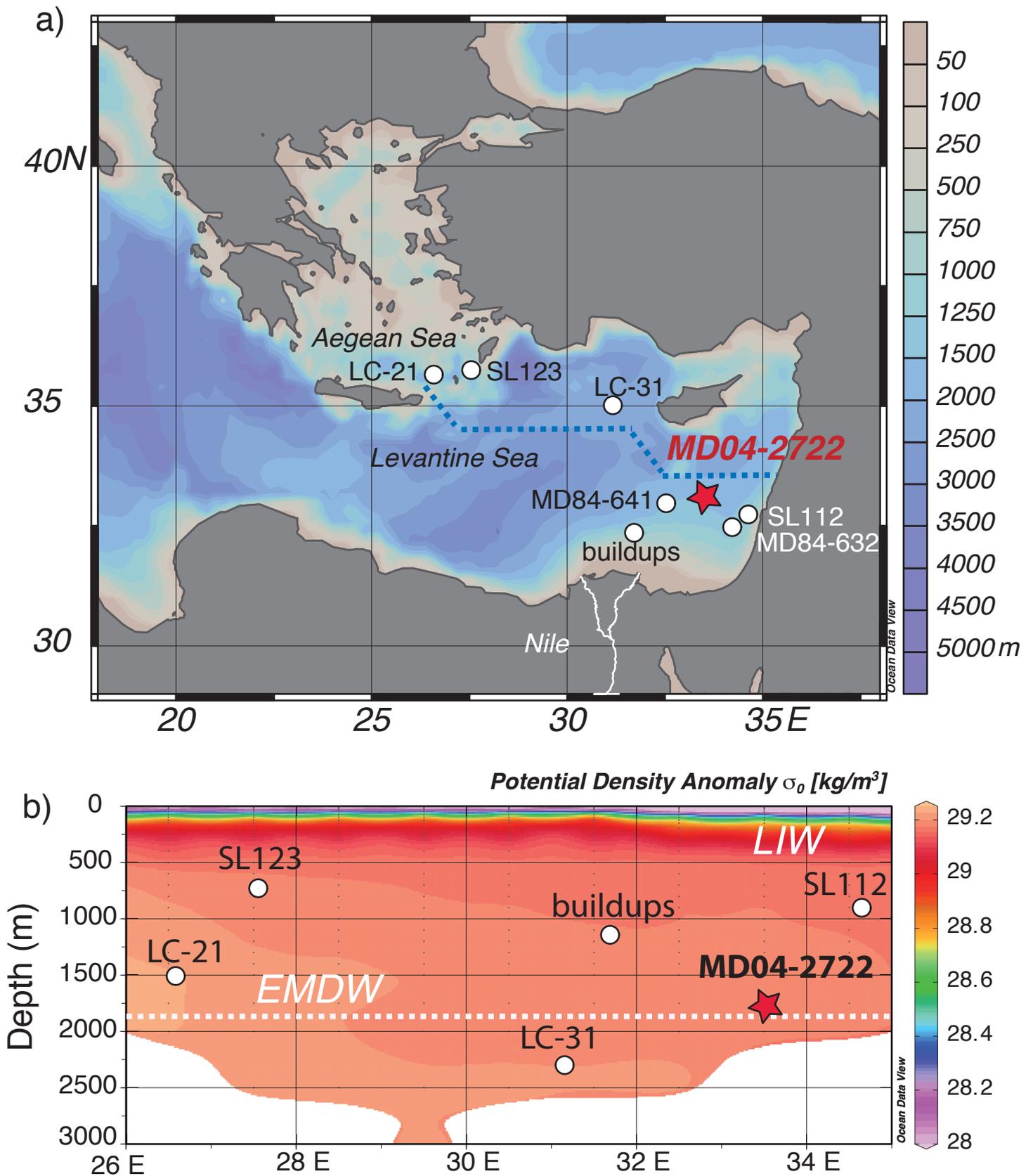


Figure 1

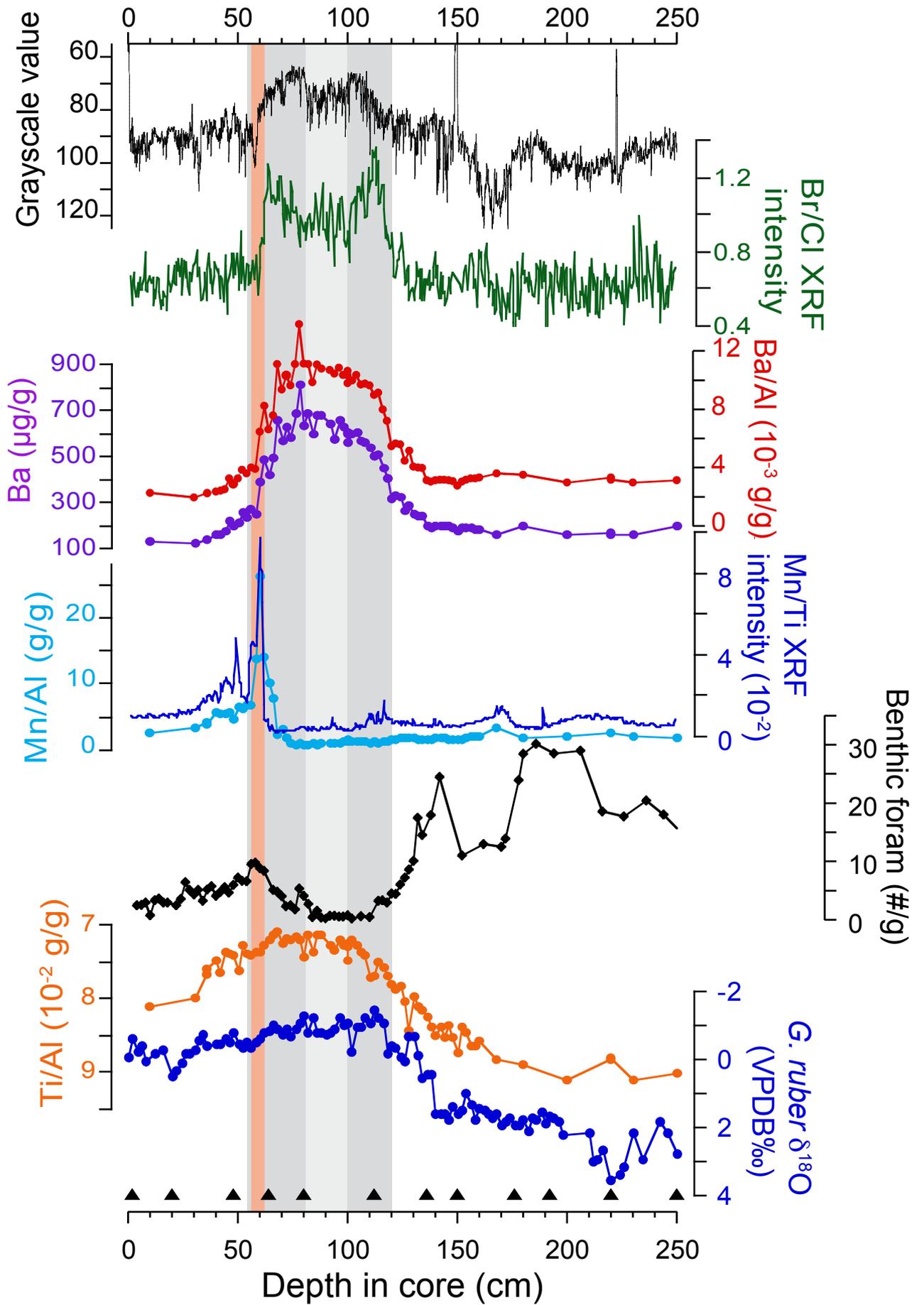


Figure 2

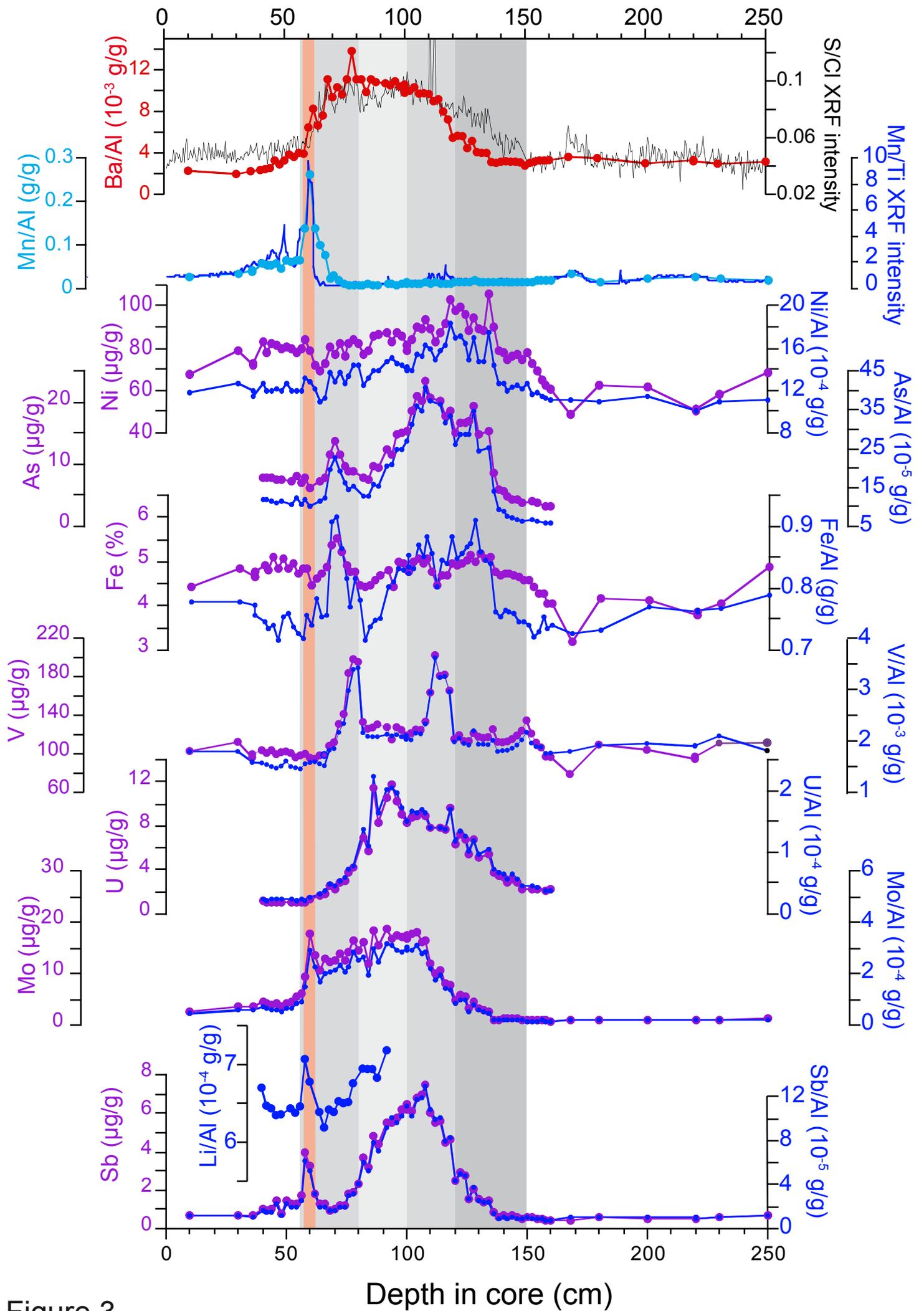


Figure 3

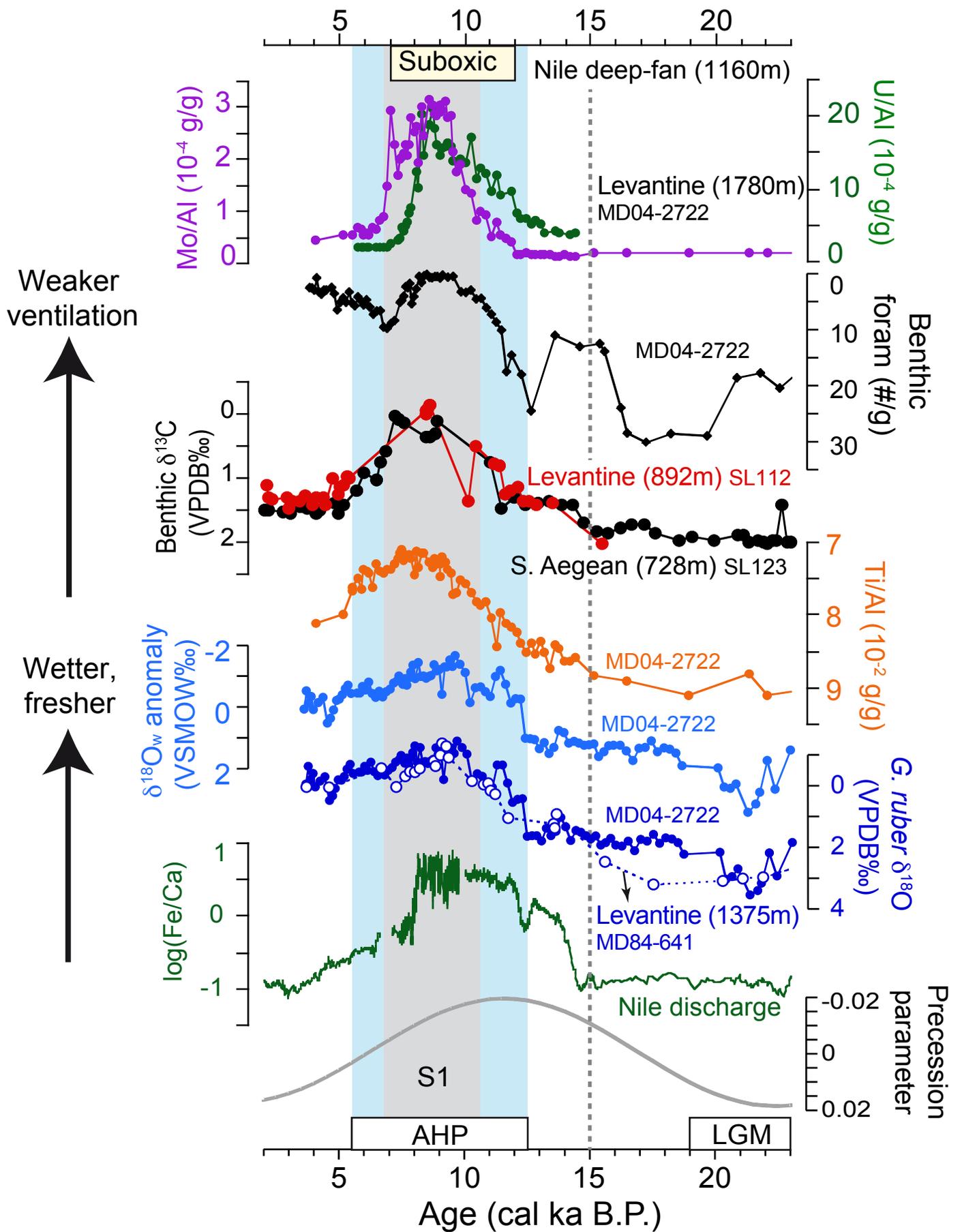


Figure 4

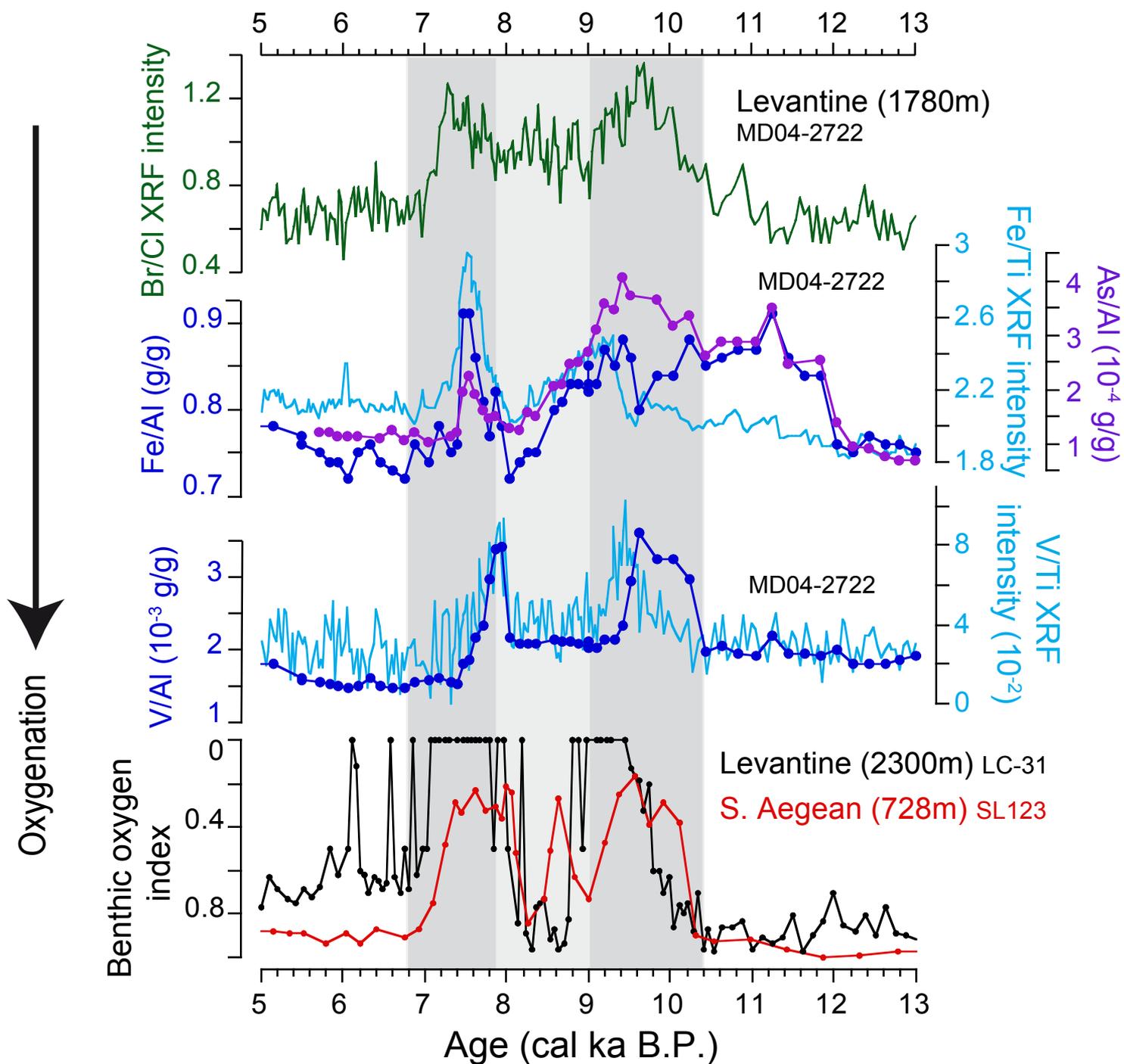


Figure 5

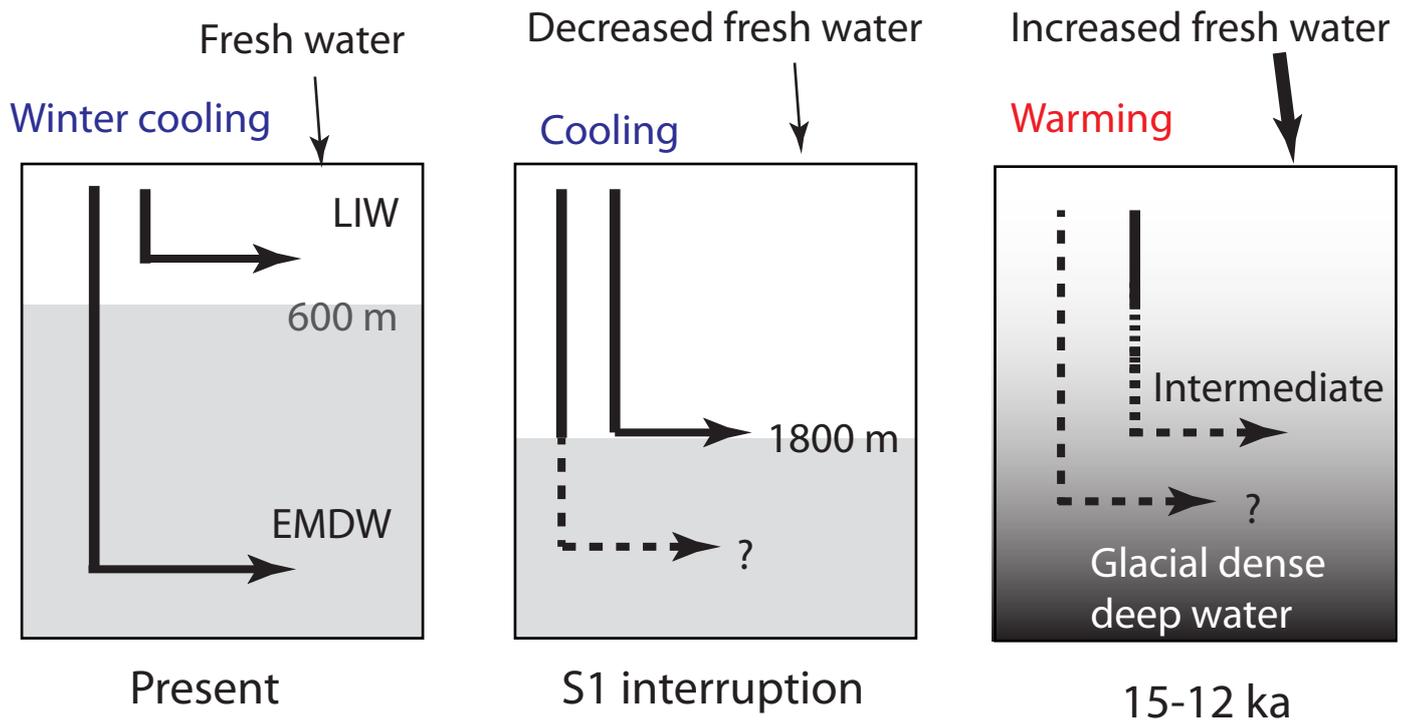


Figure 6