

1 Middle-Late Pleistocene Eastern Mediterranean nutricline depth and coccolith
2 preservation linked to Monsoon activity and Atlantic Meridional Overturning
3 Circulation
4

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22
23
24 Abstract

25 The eastern Mediterranean Sea lies under the influence of high- and low-latitude
26 climatic systems. The northern part of the basin is affected by Atlantic depressions
27 and continental and polar air masses that promote intermediate and deep-water
28 formation. The southern part is influenced by subtropical conditions and monsoon
29 activity. Monsoon intensification results in enhanced freshwater discharge from the

30 Nile River and other (now dry) systems along the North African margin. This
31 freshwater influx into the Mediterranean Sea reduces surface water buoyancy loss.
32 Disentangling the influences of these diverse climatic forcings is hindered by inherent
33 proxy data limitations and by interactions between the climatic forcings. Here we use
34 a wealth of published and new paleoclimate records across Termination II to
35 understand the impacts of the higher latitude and subtropical/monsoon climate
36 influences on coccolithophore ecology and holococcolith preservation in Aegean Sea
37 sediment core LC21. We then use these findings to interpret coccolith assemblage
38 variations at Ocean Drilling Program Site 967 (located nearby LC21, at the
39 Eratosthenes Seamount) during multiple glacial-interglacial cycles across the Middle
40 Pleistocene (marine isotopic stages 14-9). The LC21 analysis suggests that
41 holococcolith preservation was enhanced during Heinrich Stadial 11 (~ 133 ka) and
42 cold spell C26 (~ 119 ka). These two events have been previously linked to cold
43 conditions in the North Atlantic and Atlantic Meridional Overturning Circulation
44 weakening. We propose that associated atmospheric perturbations over the
45 Mediterranean Sea promoted deep-water formation, and thus holococcolith
46 preservation. Similarly, in the Middle Pleistocene (MIS 14-9) of Site 967, we observe
47 temporal coincidence between ten episodes of enhanced holococcolith preservation
48 and episodes of Atlantic Meridional Overturning Circulation slowdown. In Site 967,
49 we also identified repeated fluctuations in placoliths and in *Florisphaera profunda*,
50 which indicate nutricline depth variations. The development of a deep chlorophyll
51 maximum is associated with the North Africa and wet phases, as recently observed
52 using elemental proxy records at Site 967, during the deposition of sapropel layers. A
53 further deep chlorophyll maximum development is identified during MISs 12 and 10,
54 as a result of pycnocline and nutricline shoaling within the lower part of the photic
55 zone due to glacial sea-level lowering and water mass transport reduction at both the
56 Gibraltar and Sicily Straits. Finally, enhanced holococcolith preservation during
57 cold/dry events is clearly correlated to weakened monsoon activity in both Africa and
58 Asia.

59

60 1 – Introduction

61 Paleoclimate reconstructions document the competing influence of southern *versus*
62 northern climate systems on the hydrography and hydrology of the eastern
63 Mediterranean Sea and its borderlands over a range of timescales (Emeis et al.,
64 2000b; Grant et al., 2017, 2016; Lourens, 2004; Rohling et al., 2002b). During
65 precession minima (Northern Hemisphere insolation maxima), the African monsoon
66 intensified and shifted northward, with attendant enhancement of the freshwater
67 release into the Mediterranean basin via large North African river systems and/or
68 currently inactive wadis (Amies et al., 2019; Marino et al., 2009; Osborne et al.,
69 2008; Rohling et al., 2002a; Rohling et al., 2015; van der Meer et al., 2007). This
70 impacted the basin's hydrography and weakened or even shut down dense water
71 formation, leading to oxygen starvation at depth and deposition of layers (sapropels)
72 with elevated organic carbon concentrations (De Lange et al., 2008; Rohling et al.,
73 2015; Rossignol-Strick et al., 1982). Millennial-scale climatic variations have been
74 less well documented and appear to be associated with variations in the strength of
75 the Atlantic Meridional Overturning Circulation (AMOC) (Grant et al., 2017, 2016;
76 Stockhecke et al., 2016).

77

78 Coccolithophores are marine unicellular phytoplankton organisms living in the upper
79 part of the water column. The ecology of coccolithophore species shows a strong
80 sensitivity to modern gradients within the Mediterranean Sea and different species
81 thrive in different areas, mainly in response to West-East temperature and nutrient
82 gradients, water column dynamics, and meso-scale oceanographic features (Bonomo
83 et al., 2012; D'Amario et al., 2017; Knappertsbusch, 1993; Oviedo et al., 2015). In
84 the sedimentary archive, calcite coccolithophore remains (coccoliths) have been used
85 successfully to infer orbital and suborbital variations in climate, productivity, and
86 nutricline depth in oceans and marginal seas (Beaufort et al., 1997; Flores et al.,
87 1997; Incarbona et al., 2013, 2010a; Marino et al., 2008; Molfino and McIntyre,

88 1990a; Rogalla and Andrulleit, 2005). In the eastern Mediterranean Sea, coccolith-
89 based paleoenvironmental reconstructions have been mostly aimed at assessing the
90 shallow *versus* deep position of the nutricline within the photic layer and its
91 relationship with the basin's freshwater budget, water mass circulation, and deep-sea
92 ventilation during sapropel deposition (e.g., Grelaud et al., 2012). These studies attest
93 to the development of a deep chlorophyll maximum (DCM) while organic carbon-
94 rich layers were accumulating on the oxygen-starved eastern Mediterranean seafloor
95 (Castradori, 1993; Giunta et al., 2003; Grelaud et al., 2012; Incarbona et al., 2019,
96 2011; Incarbona and Di Stefano, 2019; Maiorano et al., 2013; Negri et al., 1999;
97 Principato et al., 2006; Triantaphyllou et al., 2009b, 2009a), corroborating findings
98 based on other marine planktonic groups (Kemp et al., 1999; Meier et al., 2004;
99 Rohling and Gieskes, 1989).

100

101 Here we present new data that complement a previous dataset of coccolith
102 assemblages from south-eastern Aegean Sea core LC21 (Grelaud et al., 2012), across
103 the penultimate glacial termination (termination II, T-II) and the last interglacial
104 period, with a precise, radiometrically constrained chronology (Grant et al., 2012).
105 This allows comparison of LC21 “coccolith proxies” with time series of
106 palaeoclimate variability in the monsoon and the North Atlantic region (Cheng et al.,
107 2009; Hodell et al., 2013), as well as atmospheric methane (CH₄) concentrations. Our
108 combined dataset is probabilistically evaluated to decipher the amplitude and timing
109 of change by quantitatively assessing the impact of chronological, analytical, and
110 proxy uncertainties. We use this analysis as a proof of concept for new, highly
111 resolved coccolith data from Ocean Drilling Program (ODP) Site 967 from the
112 Eratosthenes Seamount, South of Cyprus, within the Nile Delta Basin province
113 (Emeis et al., 1996). The new ODP 967 time series spans, at centennial-scale
114 resolution, three glacial/interglacial cycles of the Middle Pleistocene, from glacial
115 Marine Isotope Stage (MIS) 14 to interglacial MIS 9. Collectively, our new data and
116 analyses provide insights into climate variability at orbital and sub-orbital timescales

117 during both glacial and interglacial periods, complementing a wealth of existing
118 knowledge of the intervals of sapropel deposition. Specifically, we explore
119 modifications in nutrient dynamics and holococcolith preservation during the Middle
120 Pleistocene. These changes are compared with recently acquired variations in
121 elemental abundances, elemental ratios, and climate indices for ODP Site 967
122 (Section 6.3) that portray the alternation of wet and dry North Africa periods at both
123 orbital and sub-orbital timescales (Grant et al., 2017). Finally, we centre on the
124 correlation between holococcolith preservation, AMOC, and boreal monsoon activity
125 (both in Africa and in a wider Asian context) to assess: (i) the atmospheric impact of
126 continental/polar air outbreaks on the eastern Mediterranean deep-sea ventilation and
127 seafloor calcite preservation during cold stadials; and (ii) impact of millennial-scale
128 atmospheric perturbations on the eastern Mediterranean Sea.

129

130 2 – Environmental Setting

131 A negative hydrological balance maintains a robust antiestuarine thermohaline
132 circulation pattern in the Mediterranean Sea (Robinson and Golnaraghi, 1994).
133 Surface Atlantic water (Modified Atlantic Water – MAW) enters the Mediterranean
134 Sea and occupies the uppermost 100-200 m depth (Millot, 1999; POEM group,
135 1992). MAW spread out into the eastern Mediterranean Sea *via* the Mid-
136 Mediterranean Jet and reaches the Eratosthenes Seamount where a quasi-permanent
137 anticyclonic summer circulation exists, that is known as the Shikmona Gyre
138 (Malanotte-Rizzoli et al., 2014; Pinardi and Masetti, 2000; POEM group, 1992).
139 Levantine Intermediate Water (LIW) formation takes place close to the Eratosthenes
140 Seamount (Ovchinnikov I.M., 1984; POEM group, 1992). Eastern Mediterranean
141 Deep Water (EMDW) forms in the Adriatic and Aegean Sea (Fig. 1) due to winter
142 heat loss under the influence of intense Bora and Vardar winds (Malanotte-Rizzoli et
143 al., 2014; POEM group, 1992).

144

145 Today, the eastern Mediterranean Sea is one of the most oligotrophic areas globally.
146 Primary productivity is more than three times lower than in the western basin, in
147 accordance with a similar nutrient depletion trend (Krom et al., 2010, 1991). Primary
148 production is also seasonally controlled: higher productivity occurs in winter, after
149 winter convection, while severe oligotrophy occurs in summer due to deepening of
150 the thermocline and nutricline (Allen et al., 2002; Klein and Coste, 1984). The
151 Eratosthenes region is classified as a no-bloom area by satellite-based chlorophyll
152 analyses. The severe late spring-summer oligotrophy is followed by relatively higher
153 chlorophyll values in winter (D'Ortenzio and Ribera d'Alcalà, 2009).

154
155 High- and low-latitude climate systems impact on the eastern Mediterranean Sea. In
156 summer, subtropical high-pressure conditions cause stable dry and warm conditions
157 throughout the Mediterranean area (Lionello, 2012). In winter, the North African
158 subtropical high pressure is shifted southward, and cold and dry polar/continental air
159 outbreaks occur into the eastern Mediterranean from the north (Lionello, 2012;
160 Rohling et al., 2019, 2015). Expansion of the Siberian High is an important driver for
161 advection of cold air toward the eastern Mediterranean. Intensification of the Siberian
162 High during Holocene rapid climatic changes is thought to be an important driver of
163 surface water cooling and atmospheric perturbations in the central-eastern
164 Mediterranean Sea (Incarbona et al., 2008; Rohling et al., 2002b; Rohling et al.,
165 2019). Prolonged and strengthened polar/continental air outbreaks promote sea
166 surface heat loss and deep-water formation (Josey et al., 2011; Rohling et al., 2019;
167 Velaoras et al., 2017).

168

169 3 – Material and Methods

170 3.1 - Sediment cores

171 ODP Site 967 (34°04.098'N, 32°43.523'E, 2,553 m water depth) is located at the
172 base of the northern slope of the Eratosthenes Seamount, a structure that emerges
173 from the Nile Delta Cone (Fig. 1). Sediments are dominated by horizontal and sub-

174 horizontal brown and light gray, bioturbated nannofossil ooze and nannofossil clay,
175 intercalated with sapropels and turbidites (Emeis et al., 1996). Specifically, there are
176 five sapropel layers that show signs of moderate bioturbation (S13, S12, S11, b and
177 S10) in the studied interval (Emeis et al., 1996), while no turbidites and/or other
178 sedimentary disturbances were identified (Konijnendijk et al., 2014).

179
180 Sediment core LC21 (35°40'N, 26°35'E; 1,522 m water depth) was recovered in
181 1995 by *RV Marion Dufresne* in the southeastern Aegean Sea (Fig. 1). Lithology
182 consists of hemipelagic sediments, with visible sapropels (S1, S3, S4, and S5) and
183 tephra layers (Grant et al., 2016; Satow et al., 2015).

184 185 3.2 - Coccolith data

186 We carried out coccolith analyses at ODP Site 967 at 1 cm resolution between 14.80
187 and 21.49 m composite depth (mcd) (Emeis et al., 1996), for a total of 668 samples,
188 which were analysed with a polarized microscope at ~ 1000× magnification. Rippled
189 smear slides were prepared following standard procedures (Bown and Young, 1998).
190 On average 350 specimens were identified following the taxonomic concepts for
191 living coccolithophores of Young et al. (2003) and Jordan et al. (2004). Taxa were
192 grouped as ‘placoliths’, ‘miscellaneous group’, ‘upper photic zone (UPZ) group’,
193 ‘lower photic zone (LPZ) group’ and ‘holococcoliths’ (Di Stefano and Incarbona,
194 2004; Incarbona et al., 2010). Placoliths include small placoliths, small
195 *Gephyrocapsa*, *Gephyrocapsa muellerae*, and *Gephyrocapsa oceanica*. The
196 miscellaneous group includes *Helicosphaera* spp., *Coccolithus pelagicus*,
197 *Syracosphaera histrica*, *Pontosphaera* spp., *Calcidiscus leptoporus*, *Coronosphaera*
198 spp., *Braarudosphaera* spp., *Oolithotus fragilis*, *Calciosolenia* spp., and specimens of
199 all the other species. UPZ group includes *Syracosphaera pulchra*, *Umbellosphaera*
200 spp., *Discosphaera tubifera*, *Rhabdosphaera* spp., *Umbilicosphaera* spp., and
201 *Ceratolithus* spp.. LPZ group comprises *F. profunda*, which dominates the group,
202 with negligible amounts of *Gladiolithus flabellatus* in a few samples. Holococcoliths

203 include all the coccoliths produced during the haploid life-cycle stage (Incarbona et
204 al., 2019).

205

206 The holococcolith analysis at Aegean Sea core LC21 was carried out by observation
207 with a polarized microscope at about 1000× magnification, following the standard
208 procedure for rippled smear slides (Bown and Young, 1998). Holococcolith
209 percentage values were evaluated on 102 samples *versus* heterococcoliths specimens,
210 examining about 500 coccoliths. *Florisphaera profunda* percentage values at LC21
211 Aegean Sea core were presented before (Grelaud et al., 2012), following the same
212 procedure adopted in this study, and that earlier dataset is available at
213 <https://doi.pangaea.de/10.1594/PANGAEA.805357>.

214

215 3.3 - Statistical analysis of the time series

216 We use a Monte Carlo approach based on MATLAB coding (Marino et al., 2015;
217 Thirumalai et al., 2016) to: (i) stack the $\delta^{18}\text{O}$ time series for different stalagmites
218 (SB11, SB23, and SB25) from Sanbao Cave, China, that cover T-II and the last
219 interglacial period (Cheng et al., 2009); (ii) calculate rates of $\delta^{18}\text{O}$ change in the
220 Sanbao Cave stalagmites; (iii) probabilistically evaluate the chronological (Bazin et
221 al., 2013; Veres et al., 2013) and measurement uncertainties associated with the time
222 series of atmospheric methane (CH_4) concentrations from EPICA Dome C (EDC)
223 (Loulergue et al., 2008); and (iv) probabilistically evaluate chronological and
224 counting uncertainties associated with the *F. profunda* (Grelaud et al., 2012) and new
225 holococcolith records for core LC21.

226

227 Speleothem $\delta^{18}\text{O}$ time series from Sanbao Cave have been probabilistically evaluated
228 and stacked across the 140-110 ka interval. Input data for the Monte Carlo routine are
229 sample ages with 1σ uncertainties, and speleothem $\delta^{18}\text{O}$ with 1σ uncertainties (Cheng
230 et al., 2009). For each stalagmite (SB11, SB23, and SB25), individual data points are
231 then separately and randomly sampled 10,000 times within their chronological and

232 $\delta^{18}\text{O}$ uncertainties. The chronological uncertainties are evaluated using a random
233 walk Monte Carlo routine that employs a Metropolis–Hastings approach to reject
234 steps in the random walk that will result in age reversals (Rodríguez-Sanz et al.,
235 2017). That is, we imposed a stratigraphic constraint (monotonic increase of age with
236 depth, analogous to Rohling et al., 2014) to the data that are measured in a
237 stratigraphically coherent manner along individual stalagmites. All realizations are
238 then linearly interpolated on an equally spaced time scale and stacked to produce
239 10,000 speleothem $\delta^{18}\text{O}$ stacks with and without a correction that probabilistically
240 quantifies the impacts of the global ^{18}O enrichment/depletion (Schrag et al., 2002)
241 associated with ice-volume changes (Grant et al., 2012). Next, we calculated the 1st
242 time derivative, to obtain rates of speleothem $\delta^{18}\text{O}$ change for each of the 10,000 ice-
243 volume corrected ‘stacks’. This is done by smoothing each realization using 0.75 kyr
244 Gaussian window to remove sample-to-sample noise, which would result in spurious
245 jumps in the estimated rates of change, and by then differentiating the smoothed
246 realizations. Monte Carlo analysis of the EDC methane record and of the eastern
247 Mediterranean coccolith time series are performed using the same approach. Finally,
248 the 10,000 iterations of each of these time series are linearly interpolated and the
249 probability distribution assessed at each time step, thereby determining the 68%
250 (16th–84th percentile) and 95% (2.5th–97.5th percentile) probability intervals and the
251 probability maximum (PMAX, modal value) of the data.

252

253 4 – Coccolith taxon ecology

254 Placoliths are so-called ‘r-strategist taxa’ that rapidly exploit nutrients in the photic
255 zone (Baumann et al., 2005; Young, 1994). In the eastern Mediterranean Sea,
256 placoliths bloom in winter, after nutrient fertilization (Di Stefano et al., 2011;
257 Knappertsbusch, 1993; Triantaphyllou et al., 2004; Ziveri et al., 2000).

258 *Florisphaera profunda* is a deep photic zone species that indicates the occurrence of a
259 deep nutricline (McIntyre and Molino, 1996; Molino and McIntyre, 1990a). In low-
260 and middle-latitude open ocean regions, the relative abundance of this species is

261 anticorrelated with primary productivity (Beaufort et al., 2001, 1997; Hernández-
262 Almeida et al., 2019).

263

264 In the Mediterranean Sea, except for a limited area in the central part of the basin,
265 there is no apparent relationship between *F. profunda* abundance and satellite-
266 observed (surface) primary productivity levels (Hernández-Almeida et al., 2019;
267 Incarbona et al., 2008). However, *F. profunda* has been generally used to decipher
268 water column stratification and development of a deep nutricline due to monsoon-
269 fuelled freshwater discharge in the eastern Mediterranean and entrainment of
270 nutrients into the lower photic zone from below (Castradori, 1993; Grelaud et al.,
271 2012; Incarbona et al., 2019; Negri et al., 1999; Triantaphyllou et al., 2009b). This
272 occurs through the development of a distinct DCM in the eastern Mediterranean during
273 sapropel deposition. DCM development resulted from nutrient entrainment into the
274 photic zone from relatively buoyant intermediate waters that likely originated from
275 the Adriatic Sea, while the volume of MAW inflow through the Sicily Strait was
276 reduced (Myers et al., 1998; Rohling, 1991b; Rohling and Gieskes, 1989).
277 Accordingly, relative abundances of placoliths and *F. profunda*, or their ratio, provide
278 a robust indication of the depth of the nutricline in the eastern Mediterranean Sea.
279 Specifically, presence (absence) of placoliths (*F. profunda*) in the coccolith
280 assemblage reflects a shallow (deep) nutricline (Di Stefano et al., 2015; Flores et al.,
281 2000; Molino and McIntyre, 1990b).

282

283 The UPZ group consists of so-called ‘K-strategist taxa’, specialized to exploit a
284 minimum uptake of nutrients in surface water (Bazzicalupo et al., 2020; Di Stefano
285 and Incarbona, 2004; Young, 1994). Miscellaneous taxa reflect the lack of either an
286 apparent distinctive ecological preference or of an understanding of their ecological
287 preferences, with a potential weak K-strategy (Incarbona et al., 2010; Young, 1994).
288 Holococcoliths prefer dwelling in warm, oligotrophic surface water and are abundant
289 in the eastern Mediterranean Sea (D’Amario et al., 2017; Kleijne, 1991;

290 Knappertsbusch, 1993; Dimiza et al., 2015; Oviedo et al., 2015; Skampa et al., 2019).
291 Poor preservation of holococcoliths in sapropel S1 sediments was firstly recognised
292 by Crudeli et al. (2006) and was later confirmed across the whole eastern
293 Mediterranean, included the Eratosthenes Seamount (Incarbona et al., 2019;
294 Incarbona and Di Stefano, 2019) and Pliocene sapropel layers in sedimentary
295 sequences on Cyprus (Athanasidou et al., 2015). Importantly, potential preservation of
296 tiny holococcolith crystals improves when dense water renewal ensures vigorous
297 ventilation/oxygenation of the seafloor, even during short reventilation episodes that
298 “interrupt” sapropel deposition (Incarbona et al., 2019).

299

300 5 –Chronology

301 The original shipboard age model by Sakamoto et al. (1998) at ODP Site 967 has
302 since been revised, because of some inconsistent tuning to orbital insolation
303 (Konijnendijk et al., 2014; Lourens et al., 2001). More recently, Grant et al. (2017)
304 developed a monsoon runoff (sapropel) proxy from the principal component analysis
305 of sedimentary elemental data in ODP Site 967 that they tuned to precession minima.
306 They use a zero phase lag, which relies upon the assumption that little or no lag exists
307 when monsoon maxima did not immediately follow a high-amplitude glacial
308 termination (Grant et al., 2016; Lourens et al., 2001). In this study, we adopt the
309 chronology by Grant et al. (2017) for the coccolith data. Sedimentation rates between
310 MIS 14 and MIS 9 range from 1.4 cm kyr⁻¹ to 3.9 cm kyr⁻¹, implying that the mean
311 sampling resolution of our coccolith time series is about 340 years.

312 The LC21 chronology follows Grant et al. (2012) (see section 6.1). The mean
313 sedimentation rate is about 4.4 cm kyr⁻¹, with a mean sampling resolution of about
314 225 years.

315

316 6 – Results and Discussion

317 6.1 – Coccolith assemblages in Aegean Sea core LC21

318 *Florisphaera profunda* (Grelaud et al., 2012) and holococcoliths across T-II and the
319 last interglacial in south-eastern Aegean core LC21 are used to evaluate their
320 relationship with water column stratification and deep-sea ventilation, respectively.
321 Several features make this core and the timespan that we target ideal to provide a
322 ‘proof of concept’ for the interpretation of the new records from ODP Site 967 that
323 spans multiple glacial-interglacial cycles. First, core LC21 has a radiometrically
324 constrained chronology across T-II and the last interglacial period (Grant et al.,
325 2012). This has been corroborated through comparison with western Mediterranean
326 sediment cores and speleothem records, and it is consistent with the latest ice core
327 chronology across the study interval (Marino et al., 2015). Second, prominent
328 episodes of climate change punctuated T-II, including a multi-millennial Heinrich
329 stadial associated with major freshwater discharge into the North Atlantic and AMOC
330 slowdown (Deaney et al., 2017; Marino et al., 2015). Third, during the last
331 interglacial period, an organic rich layer (sapropel S5) was deposited in the eastern
332 Mediterranean under persistently anoxic or even euxinic conditions (Marino et al.,
333 2007; Rohling et al., 2015, 2006). Sapropel S5 conditions developed in response to
334 extensive monsoon-fuelled freshwater discharge along the North African Margin
335 (Amies et al., 2019; Osborne et al., 2008; Rohling et al., 2002a; Rohling et al., 2004)
336 that reduced surface salinity (van der Meer et al., 2007) and produced strong water
337 column stratification (Amies et al., 2019; Grelaud et al., 2012; Marino et al., 2007;
338 Rohling et al., 2006).

339

340 In Figure 2a-c we show *F. profunda* relative abundances from core LC21 (Grelaud et
341 al., 2012) with upper mixed layer depth fluctuations reconstructed for the same core
342 (Amies et al., 2019), and with the contemporaneous EDC time series of atmospheric
343 CH₄ concentrations and the Sanbao Cave $\delta^{18}\text{O}$ stack, which are thought to reflect
344 fluctuations in boreal monsoon intensity and attendant changes in the spatial coverage
345 of tropical wetlands (Cheng et al., 2016, 2009, 2006; Möller et al., 2013; Petrenko et
346 al., 2009). We present the *F. profunda* time series (Grelaud et al., 2012) on the

347 radiometrically constrained chronology of Grant et al. (2012), as natural logarithm of
348 the original data. Within uncertainties of the various records, we note a strong
349 similarity between the LC21 *F. profunda* record and variations of both Sanbao Cave
350 speleothem $\delta^{18}\text{O}$ (Fig. 2a) and EDC CH_4 (Fig. 2b). Notably, a distinct *F. profunda*
351 peak at ~ 129 ka appears contemporaneous with upper mixed layer thinning, with a
352 maximum in the rates of Sanbao Cave $\delta^{18}\text{O}$ change and with the CH_4 overshoot in
353 EDC. This suggests: (i) synchronous intensification of the African and Asian
354 monsoons at the onset of the last interglacial period, in line with what has been
355 documented for the early Holocene (Fleitmann et al., 2003; Marino et al., 2009;
356 Nicholson et al., 2020; Tierney et al., 2008); and (ii) a rapid increase in freshwater
357 discharge into the eastern Mediterranean at the onset of the last interglacial monsoon
358 maximum, quantified as up to ~ 8.8 times the modern pre- Aswan Nile discharge and
359 responsible for the most intense thinning of the upper summer mixed layer (Amies et
360 al., 2019). Observation (ii) is particularly relevant because it alludes to the influence
361 of the rates of monsoon intensification on stratification in the eastern Mediterranean.
362 When large amounts of monsoon-fuelled freshwater are rapidly added to the basin,
363 the strong evaporative climate of the Levant cannot keep up with sea-surface dilution
364 (Rohling et al., 1991b) and the water column becomes strongly stratified (Rohling
365 and Gieskes, 1989; Rohling et al., 2006; Marino et al., 2007; Athanasiou et al., 2015,
366 2017; Amies et al., 2019), causing shoaling of the pycnocline, the nutricline to be
367 positioned at the base of the photic layer and the attendant development of a
368 pronounced DCM.

369

370 In Figure 2d-f, LC21 holococcolith data are compared with North Atlantic and
371 western Mediterranean Sea records. It is evident that there is no or poor preservation
372 of holococcoliths during sapropel S5 (Fig. 2e). Peaks of holococcoliths below S5
373 correlate with low alkenone-derived SSTs in the Alboran Sea (Martrat et al., 2014)
374 (Fig. 2d) and with variations in AMOC strength proxies (Figs. 2e,f) from the Iberian
375 Margin. Specifically, $\log(\text{Ca}/\text{Ti})$, benthic foraminiferal $\delta^{13}\text{C}$, and C_{26}OH ratios

376 (Hodell et al., 2015; Martrat et al., 2007) unequivocally indicate that AMOC had
377 collapsed during Heinrich event HS11 (Böhm et al., 2015); we find that, at the same
378 time, holococcolith preservation was enhanced. A similar relationship is evident
379 above sapropel S5, especially with C₂₆OH ratio data (Fig. 2f), and may be correlated
380 with North Atlantic cold event C26 (Oppo et al., 2006, 2001; Tzedakis et al., 2018).
381 Coupled ocean-atmosphere hindcasts suggest that the AMOC slowdown/shutdown
382 may have propagated through the Mediterranean Sea in the form of major cooling
383 and intense atmospheric perturbations (Manabe and Stouffer, 1997; Vellinga and
384 Wood, 2002). Both cooling and atmospheric perturbations are major prerequisites for
385 surface water buoyancy loss and deep-water formation, explaining enhanced
386 Mediterranean bottom water ventilation during Heinrich and Stadial events in the last
387 glacial, based on benthic foraminifera $\delta^{13}\text{C}$ and alcohol index records (Cacho et al.,
388 2000; Sprovieri et al., 2012; Toucanne et al., 2012). Our new results from the Aegean
389 Sea explicitly link holococcolith preservation to Heinrich and Stadial events in
390 response to deep-water formation and seafloor ventilation increases in the
391 Mediterranean Sea during those events.

392

393 6.2 – Coccolith assemblages at ODP Site 967

394 At ODP Site 967, coccolith distribution patterns are compared (Figure 3) with: (i)
395 June 21st insolation at 65°N (Laskar et al., 2004); (ii) the benthic $\delta^{18}\text{O}$ composite
396 record from ODP Sites 967 and 968 (Konijnendijk et al., 2015); and (iii) the LR04
397 benthic $\delta^{18}\text{O}$ stack (Lisiecki and Raymo, 2005). Overall, coccolith assemblages are
398 dominated by placoliths, with percentages between 25 and 98% (average = 70%, Fig.
399 3D). The deep photic zone species *F. profunda* features high-amplitude fluctuations
400 between intervals in which it is barely occurring (1%) and intervals in which it
401 becomes the dominant (up to 74%), ~~and an average of 26%~~ (Fig. 3E). Placoliths'
402 peak (low) abundances occur when *F. profunda* percentages are at a minimum
403 (maximum), as highlighted by the pronounced anticorrelation ($R^2= 0.95$, $n = 668$)
404 displayed in Figure 4.

405

406 The observed alternating dominance of the placoliths and *F. profunda* (see above) at
407 ODP Site 967 suggests that between MIS 14 and MIS 9 the upper water column in
408 the easternmost Mediterranean Sea repeatedly switched between dominant winter-
409 induced fertilization (placoliths' peaks, shallow nutricline), similar to today's winter
410 conditions (Knappertsbusch, 1993), and a predominantly DCM-focused productivity
411 (*F. profunda*'s peaks, deep nutricline) during both glacial and interglacial periods.
412 Shoaling of the pycnocline, which promoted a deep nutricline (DCM), in association
413 with intensifications of the North African monsoon and sea-level lowering (Rohling
414 and Gieskes, 1989; Rohling, 1991a, 1991b) is the most plausible explanation for the
415 peak abundance of *F. profunda* at ODP Site 967 during sapropels and glacial periods,
416 respectively. Accordingly, *F. profunda* would be particularly sensitive to nutrient
417 (re)distribution within the photic zone, with intervals of positive shifts in the basins
418 freshwater budget (monsoon maxima, Rohling, 1991b) and reduced water exchange
419 at straits (glacial lowstands, Rohling, 1991a) both leading to enhanced stratification
420 in the upper water column, shoaling of the pycnocline, and development of a
421 nutricline at the based of the photic layer. This conceptual framework indicated by
422 box-model calculations (Rohling, 1991a,b) provides an explanation for the *F.*
423 *profunda* peaks both in sapropel layers (e.g., S12, S11, b, S10) and during MIS 12
424 and MIS 10 glacial periods (see also Section 6.3).

425 The remaining three coccolith groups at Site 967 are largely subordinate. Low
426 abundance of UPZ taxa (0.0-5.4%, 1.4% on average, Fig. 3F) and Miscellaneous taxa
427 (0.0-11.1%, 1.2% on average, Fig. 3G) is unexpected in the severe oligotrophy of the
428 eastern Mediterranean Sea, especially for UPZ taxa that include dominant
429 coccolithophore species in summer/autumn (Knappertsbusch, 1993; Malinverno et
430 al., 2009; Oviedo et al., 2015). However, living coccolithophore surveys show that
431 winter production (placolith blooming) is about an order of magnitude higher than
432 summer production, thereby explaining the apparent ecological contradiction in taxa
433 proportions (Knappertsbusch, 1993).

434 Holococcolith percentage values range between 0.0 and 20.8%, 1.9% on average
435 (Fig. 3H). As was the case in late Quaternary sapropels, tiny holococcoliths are again
436 not preserved in sapropel layers, but we also note that that poor preservation extends
437 far beyond sapropel layers.

438

439 6.3 – Comparison between coccoliths, element ratios and climatic indices at ODP
440 Site 967

441 *Florisphaera profunda* and holococcolith distribution patterns at ODP Site 967 (Figs.
442 5A-B) are compared with sedimentary elemental ratios and climatic indices from the
443 same sedimentary sequence (Grant et al., 2017). A principal component analysis
444 carried out on elemental proxies by these authors highlighted that principal
445 components PC1 (not shown) and PC2 (Fig. 5D) account for 79% of variance and
446 reflect terrigenous input and sapropel deposition (enhanced monsoon runoff),
447 respectively. Moreover, aeolian dust fluxes (Fig. 5E) and a North Africa
448 humidity/aridity index (Fig. 5F) were calculated and are also compared with
449 coccolith abundance fluctuations.

450

451 Intensification of East African monsoon precipitation coincided with northward
452 displacement of the Intertropical Convergence Zone (ITCZ) and subsequent
453 intensification of precipitation over the catchment basins of the Nile and other rivers,
454 which fuelled enhanced freshwater discharge along the wider North African margin
455 into the eastern Mediterranean (Ehrmann et al., 2016; Rohling et al., 2002a; Rohling
456 et al., 2015). *Florisphaera profunda* proliferates because it benefits from the
457 nutricline positioned in deep photic zone and the establishment of a DCM
458 (Castradori, 1993; Gironé et al., 2013; Grelaud et al., 2012; Incarbona et al., 2011;
459 Negri et al., 1999), following the rationale outlined above (Myers et al., 1998;
460 Rohling, 1991a; Rohling and Gieskes, 1989). This scenario has been described for
461 eastern Mediterranean sapropels and is further supported by the coupling of the *F.*
462 *profunda* signal and enhanced humidity in North Africa that leads to enhanced

463 monsoon runoff into the eastern Mediterranean, especially during S12, S11, b and
464 S10 (Fig. 5). Also evident is decoupling between *F. profunda* and the
465 humidity/aridity index, for instance during glacial sapropel S13 and during sapropel
466 layer b. This suggests that the climatic indices at ODP Site 967 reflect increased
467 rainfall over North Africa and Sahara, without sufficient northward monsoon
468 penetration that determines the intensity of monsoon-related runoff into the eastern
469 Mediterranean Sea (Grant et al., 2017). Also, different nutrient dynamics may have
470 developed during the glacial sapropel S13, with surface fertilization that supports
471 placolith-bearing species competition.

472
473 However, the coccolith record at Site 967 suggests that *F. profunda* increases are a
474 recurring feature of the record that is not necessarily associated with the deposition of
475 organic-rich layers deposition on the eastern Mediterranean seafloor, especially in
476 glacial episodes (Fig. 5A). The Ba/Al signal indicates that many sapropel layers have
477 been partially oxidized (Fig. 5G), so that their currently visible extents do not
478 represent the original thickness of anoxic sediments. Yet, we argue that *F. profunda*
479 peaks are not exclusively linked to the environmental changes associated with
480 sapropel formation. Positive *Florisphaera profunda* peaks without a corresponding
481 sapropel layer (be it visible, or oxidized) are evident in all eastern Mediterranean
482 records that span sufficiently long time intervals (Castradori, 1993; Giunta et al.,
483 2003; Maiorano et al., 2013; Negri et al., 1999; Triantaphyllou et al., 2010). The
484 occurrence of *F. profunda* during glacial periods identifies a second mode of DCM
485 development in the eastern Mediterranean, as proposed on the basis of planktonic
486 foraminiferal assemblages (Rohling and Gieskes, 1989) and modeling (Myers et al.,
487 1998; Rohling, 1991a). Again, the DCM development would be driven by the
488 pycnocline and nutricline shoaling within the lower part of the photic zone. But, for
489 glacials, the eustatic sea-level drop (Fig. 5C) would have been the main trigger
490 mechanism, after reducing water mass transport at both Gibraltar and Sicily Straits
491 and ultimately shoaling the pycnocline and nutricline depth up to ~ 80 m (Myers et

492 al., 1998; Rohling, 1991a). However, the scattered *F. profunda* abundance signal in
493 MISs 12 and 10 suggests that other factors may operate in conjunction with sea-level
494 fall, for reducing water transport across the Gibraltar and Sicily Straits. Among these,
495 less frequent and intense northern outbreaks in deep water production sites and the
496 inflow of low-density meltwater from the Atlantic Ocean may have weakened the
497 Mediterranean thermohaline circulation and may have caused transient reductions in
498 the water transport at straits, like during last glacial cold spells (Sierro et al., 2005;
499 Sprovieri et al., 2012; Toucanne et al., 2012; Azibeiro et al., 2021).

500

501 Holococcoliths are made of small calcite rhombohedra, arranged in different patterns.
502 They are the most vulnerable coccoliths to selective dissolution (Roth and Coulbourn,
503 1982). Comparison between the new ODP 967 holococcolith record and the
504 humidity/aridity index (Figs. 5B, F) shows strong similarities throughout the time
505 span studied, which clearly point to the dissolution of haploid-life calcite remains at
506 the Erathostenes seamount during humid phases. This agrees with the notion that
507 surface buoyancy gain by freshwater river discharge negatively impacted deep-water
508 formation in the eastern Mediterranean Sea, which enhanced organic carbon
509 preservation (De Lange et al., 2008; Myers et al., 1998; Rohling et al., 2015) and,
510 thus, worsened holococcolith preservation, like during sapropel S1 (Crudeli et al.,
511 2006; Incarbona et al., 2019; Incarbona and Di Stefano, 2019) and sapropel S5 (Fig.
512 2, this study).

513

514 It is worth noting that the three major aeolian dust peaks during MIS 13-12 (Fig. 5E)
515 did not match with increasing holococcolith preservation but fall within ‘no
516 preservation’ intervals. Aeolian dust peaks are associated with weakened monsoon
517 activity and North Africa dry periods, as seen from the Ti/Al ratio (Konijnendijk et
518 al., 2015; Lourens et al., 2001; Wehausen and Brumsack, 2000; Ziegler et al., 2010),
519 and would imply a lower freshwater input into the eastern Mediterranean Sea.

520 Ideally, these conditions would enhance deep water formation, reduce organic matter

521 preservation on the seafloor and increase preservation of holococcolith calcite.
522 However, dust accumulation in marine cores depends on wind strength and direction
523 (Moulin et al., 1997; Zabel et al., 1999). Thus, aeolian dust peaks are not necessarily
524 tied to cooling and atmospheric perturbations that, for instance during the latest
525 Quaternary, led to enhanced Mediterranean bottom water ventilation in coincidence
526 of glacials and stadials (Cacho et al., 1999; 2000; Sprovieri et al., 2012; Toucanne et
527 al., 2012).

528

529 6.4 – AMOC slowdown, holococcolith preservation and monsoon weakening
530 In Figure 6, North Atlantic geochemical records (Fig. 6A-B), Mediterranean
531 holococcolith data (Fig. 6C), and oxygen isotopes of China speleothems (Fig. 6D) are
532 plotted using their own original age models (Cheng et al., 2016; Grant et al., 2017;
533 Hodell et al., 2015; Martrat et al., 2007). The grey boxes used for correlation are
534 drawn and link correlative events in these various Northern Hemisphere records.
535 Records shown in Figure 6 help with outlining a concept of atmosphere/ocean
536 interactions that resulted in the correlation between Northern Hemisphere climate
537 variability and surface water ecosystem modifications and seafloor diagenesis in the
538 eastern Mediterranean.

539

540 The correlation boxes in Figure 6 are drawn assuming that AMOC
541 slowdown/shutdown caused an atmospheric perturbation that impacted a vast area of
542 the northern Hemisphere, including the eastern Mediterranean Sea, East African and
543 Asian monsoon sites (Vellinga and Wood, 2002). The log (Ca/Ti) record from the
544 Iberian Margin (Fig. 6B) is a proxy for millennial-scale variability (Hodell et al.,
545 2015): minima indicate stadial/Heinrich phases during which the AMOC was
546 severely weakened or collapsed (McManus et al., 2004, 1999). Previous studies based
547 on $\delta^{13}\text{C}$, alcohol index and sortable silt records support the hypothesis that
548 Mediterranean bottom water ventilation at least in the western sub-basin was more
549 intense during glacial periods and stadial events (Cacho et al., 2000, 1999; Frigola et

550 al., 2008; Sprovieri et al., 2012; Toucanne et al., 2012) and strengthened
551 Mediterranean outflow into the Gulf of Cadiz and in the Iberian Margin (Sierro et al.,
552 2020). Simultaneous enhancement of EMDW ventilation would have enhanced
553 holococcolith preservation, as observed below and above sapropel S1 (Incarbona et
554 al., 2019; Incarbona and Di Stefano, 2019) and during HS11 and C26 in the Aegean
555 Sea (this study, Fig. 2). The physico-chemical processes in the seafloor microsystem
556 by which tiny holococcolith calcite rhombohedra are preferentially preserved under
557 oxic conditions is not clear yet. Incarbona et al. (2019b) hypothesized a possible
558 detrimental action of organic acid produced by bacteria under a dysoxic/anoxic state
559 on the water/sediment interface, in analogy with results from modern surveys in the
560 Gulf of California (Ziveri and Thunell, 2000). However, specific studies are needed
561 to better understand the processes involved.

562

563 The link between AMOC slowdown/shutdown and East African, Indian, and Asian
564 monsoon weakening is well-established (Deplazes et al., 2014; Porter and Zhisheng,
565 1995; Rohling et al., 2002; Rohling et al., 2006; Schulz et al., 1998; Sirocko et al.,
566 1993; Tjallingii et al., 2008), and may involve severe drought development due to
567 southward ITCZ displacement (Krebs and Timmermann, 2007; Vellinga and Wood,
568 2002; Zhang and Delworth, 2006). Yet, an ITCZ shift and associated Hadley cell
569 changes likely explain only part of the impacts on monsoon circulation, which also
570 depends on regional and local processes (Donohoe et al., 2013; Geen et al., 2020).
571 For example, recent modeling shows that Northern Hemisphere glacial cooling,
572 increased ice-sheet albedo, and sea-level lowering produce an anomalous thermal
573 gradient between the Arabian Peninsula and the Arabian Sea that results in a
574 weakened Walker circulation in the Indian Ocean and drought in the monsoon
575 systems of the Indo-Pacific region (DiNezio et al., 2018). As mentioned before for
576 the HS 11 and C26 cold spells (section 6.1), AMOC slowdown/shutdown also
577 produces cooling and intense atmospheric perturbations in the Mediterranean region
578 (Manabe and Stouffer, 1997; Vellinga and Wood, 2002), facilitating deep-water

579 formation and bottom water ventilation (Cacho et al., 2000; Sprovieri et al., 2012;
580 Toucanne et al., 2012). Thus, AMOC slowdown/shutdown may have both
581 strengthened cold and dry polar/continental air outbreaks and reduced Indian-African
582 monsoon activity, with both processes conducive to enhanced EMDW production
583 (the prerequisite for holococcolith preservation).

584 In Fig. 6, grey correlation boxes indicate a link between AMOC slowdown/shutdown
585 phases (Fig. 6B) and both the heaviest oxygen isotopic values in Chinese Sanbao
586 Cave speleothems (Fig. 6D) and enhanced holococcolith preservation in the
587 Mediterranean (Fig. 6C).

588 The atmospheric CH₄ record from Antarctica ice cores (Fig. 6E) reflects emissions
589 from boreal and monsoonal wetlands (Guo et al., 2012; Landais et al., 2010). The
590 signal has been separated into three principal components, a glacial-interglacial
591 forced component, a bi-hemispheric insolation driven component, and a millennial-
592 scale oscillatory component, which respectively explain 80%, 15% and 5% of the
593 variance (Guo et al., 2012). Glacial-interglacial cycles force globally synchronous
594 monsoonal variations in methane emission. The other two forcings are hemispheric in
595 nature, because of the two hemispheres' anti-phasing for low-latitude summer
596 insolation changes in ITCZ oscillations and for millennial-scale bipolar see-saw
597 (strengthening/weakening) AMOC circulation (Guo et al., 2012). The limited number
598 of grey correlation boxes between the Antarctic CH₄ signal and of Asian monsoon
599 activity (Fig. 6D-E) suggests that the activity of Southern Hemisphere monsoon
600 systems (South America, South Africa, Australia) have been a major driver of
601 methane emission between 550 and 300 ka. However, we note the occurrence of three
602 different peaks of Antarctic CH₄ during MIS 13, a signal which is also visible in
603 Mediterranean holococcoliths and the China speleothem record (Fig. 6). This feature
604 supports reduced emission from southern hemisphere methane sources during MIS 13
605 and an increased and synchronous emission from northern monsoons and boreal
606 wetlands (Guo et al., 2012).

607

608 7 – Conclusions

609 The chronology and age uncertainties of SE Aegean Sea core LC21 coccolith data,
610 Iberian Margin geochemical records, $\delta^{18}\text{O}$ in the Sanbao Cave stalagmites, and
611 atmospheric methane concentrations from EDC have been probabilistically assessed
612 for the last interglacial and TII. The *F. profunda* peak at the base of sapropel S5 layer
613 is contemporaneous with a maximum in the rates of $\delta^{18}\text{O}$ change in Sanbao Cave and
614 with the CH_4 overshoot in EDC, which suggests an African and Asian monsoon
615 intensification at the onset of the last interglacial and the raising of the summer upper
616 mixed layer depth. Holococcolith preservation was enhanced during Heinrich event
617 HS11 and the C26 cold spell, when AMOC was severely weakened or collapsed.
618 Cooling and atmospheric perturbations promoted surface-water buoyancy loss and
619 deep-water formation, and thus increased Mediterranean bottom water ventilation
620 rates. These results for LC21 explicitly link holococcolith preservation to episodes of
621 enhanced deep-water formation and seafloor ventilation in the eastern Mediterranean
622 Sea during Heinrich and Stadial events.

623

624 We also analysed 668 samples from ODP Site 967 to reconstruct variations in
625 coccolithophore ecology in the region of the Eratosthenes Seamount (eastern
626 Mediterranean Sea) between about 550 and 300 ka (Middle Pleistocene, MIS 14-9).
627 Placoliths and *F. profunda* abundance fluctuations are strongly anticorrelated ($R^2 =$
628 0.95 , $n = 668$) and together dominate the coccolith assemblages at Site 967, which
629 suggests that the surface water environment repeatedly switched between
630 predominant winter-induced fertilization (shallow nutricline) and predominant
631 monsoon-induced deep nutricline conditions (and Deep Chlorophyll Maximum
632 development) associated with a shoaling of the pycnocline to a position within the
633 lower photic zone. In analogy with results for core LC21, Middle Pleistocene phases
634 of enhanced monsoon runoff into the eastern Mediterranean appear to have caused
635 shoaling of the pycnocline into the lower photic layer, which provides a deep
636 nutricline that fosters *F. profunda* proliferation. A second mode of *F. profunda*

637 proliferation and DCM development is seen in glacials, when the nutricline shoaled
638 into the lower photic zone due to sea-level lowering that reduced water-mass
639 transport through the main Mediterranean straits. Thus, our coccolithophore
640 assemblage analyses corroborate earlier suggestions of such a phenomenon based on
641 planktonic foraminiferal analyses (Rohling and Gieskes, 1989) and modeling (Myers
642 et al., 1998; Rohling, 1991a).

643

644 Scattered but statistically significant peaks of holococcoliths (up to ~ 20%) mark
645 increased carbonate preservation on the seafloor due to enhanced production of
646 EMDW, which match changes in recently proposed North Africa aridity indices
647 (Grant et al., 2017). Finally, holococcolith enhanced preservation during cold spells is
648 linked to increased deep-water formation in the Mediterranean Sea, in response to
649 Atlantic Meridional Overturning Circulation slowdown and weakened monsoon
650 activity during Northern Hemisphere cold events.

651

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659

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1235 Captions

1236 Fig. 1: location of ODP Site 967 and of records used for correlation. A) Red arrows
1237 indicate the location of International Ocean Discovery program (IODP) Site U1385 in
1238 the Iberian Margin, ODP Site 967 in the eastern Mediterranean, Core LC21 in the
1239 Aegean Sea, Sanbao Cave in China and EPICA Dome C in Antarctica. B) Inset map
1240 of the red box in A). The blue and red circles respectively indicate the location of
1241 core LC21 and ODP Site 967. The dashed line shows the Nile cone province. Black
1242 arrows point out the path of Eastern Mediterranean Deep-water from Adriatic and
1243 Aegean Sea.

1244

1245 Fig. 2: plot of coccolith data in the Aegean Sea core LC21 and comparison with
1246 selected records. a) *Florisphaera profunda* percentage values (blue circles),
1247 expressed as natural logarithm (Grelaud et al., 2012). The 1st derivative of Sanbao
1248 Cave speleothem $\delta^{18}\text{O}$ (orange line) for each of the 10,000 ice-volume corrected
1249 ‘stacks’, following the chronology by Cheng et al. (2009) and after ice-volume
1250 correction. b) *Florisphaera profunda* percentage values (blue line), expressed as
1251 natural logarithm (Grelaud et al., 2012). Epica Dome C CH_4 (grey circles and grey
1252 line), following the Antarctic Ice Core chronology AICC2012 (Bazin et al., 2013) c)
1253 *Florisphaera profunda* percentage values (blue line), expressed as natural logarithm
1254 (Grelaud et al., 2012). Upper Summer Mixed Layer depth in the three different
1255 experiments by Amies et al. (2019) (respectively pink, orange and red lines for
1256 experiments A, B and C). d) Alkenone-derived SST (red circles) from the Alboran
1257 Sea (Martrat et al., 2014). e) Holococcoliths percentage values (blue circles, this
1258 study) and superimposed $\delta^{13}\text{C}$ values of the benthic foraminifera species *Cibicidoides*
1259 *wuellerstorfi* from the Iberian Margin (green circles) (Martrat et al., 2007). f) Ca/Ti
1260 ratio, expressed as logarithm (green line), in sediments from the Iberian Margin
1261 (Hodell et al., 2015), superimposed to the C_{26}OH ratio from the same area (red
1262 circles) (Martrat et al., 2007). Thick lines represent the 3-pt running average and
1263 coloured shadows indicate the 95% confidence level. The sapropel S5 and HS11
1264 extent is also shown.

1265

1266 Fig. 3: plot of coccolith data at ODP Site 967 and comparison with selected records.
1267 A) June 21st insolation curve at 65°N (Laskar et al., 2004). B) Benthic $\delta^{18}\text{O}$
1268 composite record from ODP Sites 967 and 968 (Konijnendijk et al., 2015).
1269 C) LR04 benthic $\delta^{18}\text{O}$ stack (Lisiecki and Raymo, 2005). D) Downcore percentage
1270 variations of Placoliths. E) Downcore percentage variations of *F. profunda*. F)
1271 Downcore percentage variations of UPZ taxa. G) Downcore percentage variations of
1272 Miscellaneous taxa. H) Downcore percentage variations of holococcoliths. Black
1273 horizontal bars show the error associated to countings for a 95% confidence level.

1274 Red filling indicates values higher than the total average percentage. Horizontal thick
1275 black lines indicate MIS boundaries from Lisiecki and Raymo (2005). Horizontal
1276 yellow boxes show visible sapropel layers in the ODP 967 composite section (Emeis
1277 et al., 2000a).

1278

1279 Fig. 4: scatter plot of placoliths and *F. profunda* percentage values at ODP Site 967.
1280 The black line shows the linear fit. The equation of the linear fit, R^2 correlation index
1281 and number of samples are also reported.

1282

1283 Fig. 5: coccolith, element ratios, indices and principal component scores at ODP Site
1284 967. A) *Florisphaera profunda* percentage values (black circles, the black line is the
1285 3-pt running average, this study). B) Holococcolith percentage values (blue circles,
1286 the blue line is the 3-pt running average, this study). C) Relative sea-level (Spratt and
1287 Lisiecki, 2016). D) PC2 of elemental proxies that reflects sapropel/monsoon runoff
1288 layers (Grant et al., 2017). E) The Aeolian residual by Larrasoña et al. (2003)
1289 plotted by the Grant et al. (2017) chronology. F) The North Africa humidity/aridity
1290 index (Grant et al., 2017). G) The Ba/Al ratio (Grant et al., 2017). Horizontal thick
1291 black lines indicate MIS boundaries from Lisiecki and Raymo (2005). Horizontal
1292 yellow boxes show visible sapropel layers in the ODP 967 composite section (Emeis
1293 et al., 2000a).

1294

1295 Fig. 6: correlation among Mediterranean, North Atlantic, Asian and Antarctica
1296 records. A) Alkenone-based SSTs (°C) at Site MD01-2444, Iberian Margin (Martrat
1297 et al., 2007). B) Log (Ca/Ti) at IODP Site U1385, Iberian Margin (Hodell et al.,
1298 2015). Note the reversed axis. C) 3-pt running average of holococcolith percentage
1299 values at ODP Site 967, eastern Mediterranean Sea (present study). Note the reversed
1300 axis. D) $\delta^{18}\text{O}$ speleothem data at Sanbao Cave, China (Cheng et al., 2016). Note the
1301 reversed axis. E) Epica Dome C CH_4 , Antarctica (Bazin et al., 2013).

1302 Grey boxes indicate correlations among different records: cooling and AMOC
1303 weakening/shutdown in the Iberian Margin (A, B), holococcolith preservation in the
1304 eastern Mediterranean seafloor (C), monsoon activity weakening in China (D) and
1305 EDC methane concentration decrease in Antarctica (see Section 6 for further
1306 explanation).
1307











