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Corresponding Author: Dr. Katharine Mary Grant, PhD

Corresponding Author's Institution: The Australian National University

First Author: Katharine Mary Grant, PhD

Order of Authors: Katharine Mary Grant, PhD; Rosina Grimm; Uwe Mikolajewicz; Gianluca Marino; Martin Ziegler; Eelco J Rohling

Abstract: The Mediterranean basin is sensitive to global sea-level changes and African monsoon variability on orbital timescales. Both of these processes are thought to be important to the deposition of organicrich sediment layers or 'sapropels' throughout the eastern Mediterranean, yet their relative influences remain ambiguous. A related issue is that an assumed 3-kyr lag between boreal insolation maxima and sapropel midpoints remains to be tested. Here we present new geochemical and icevolume-corrected planktonic foraminiferal stable isotope records for sapropels S1 (Holocene), S3, S4, and S5 (Marine Isotope Stage 5) in core LC21 from the southern Aegean Sea. The records have a radiometrically constrained chronology that has already been synchronised with the Red Sea relative sea-level record, and this allows detailed examination of the timing of sapropel deposition relative to insolation, sea-level, and African monsoon changes. We find that sapropel onset was near-synchronous with monsoon run-off into the eastern Mediterranean, but that insolationsapropel/monsoon phasings were not systematic through the last glacial cycle. These latter phasings instead appear to relate to sea-level changes. We propose that persistent meltwater discharges into the North Atlantic (e.g., at glacial terminations) modified the timing of sapropel deposition by delaying the timing of peak African monsoon run-off. These observations may reconcile apparent model-data offsets with respect to the orbital pacing of the African monsoon. Our observations also imply that the previous assumption of a systematic 3-kyr lag between insolation maxima and sapropel midpoints may lead to overestimated insolationsapropel phasings. Finally, we surmise that both sea-level rise and monsoon run-off contributed to surface-water buoyancy changes at times of sapropel deposition, and their relative influences differed per sapropel case, depending on their magnitudes. Sea-level rise was clearly important for sapropel S1, whereas monsoon forcing was more important for sapropels S3, S4, and S5.



18<sup>th</sup> March 2016

**Dr Katharine Grant** Postdoctoral Researcher Research School of Earth Sciences The Australian National University

Tel: (+61) 02 6125 3241 Fax: (+61) 02 6125 0738

katharine.grant@anu.edu.au

Editor Quaternary Science Reviews

Dear Henning,

On behalf of all authors, I hereby submit our revised manuscript "The timing of Mediterranean sapropel deposition relative to insolation, sea-level and African monsoon changes" for publication as an article in *Quaternary Science Reviews*.

We have addressed all comments of both reviewers by incorporating their suggestions into the text, and we also include results from the Kandiano et al. (2014) study in our revised Figure 4 and in the discussion of meltwater effects in section 3.2. Figures 1 and 3b have also been slightly amended, and the reference list has been extended in order to adequately address the reviewers' suggestions. Line references to added/amended text are given in our response to reviewers. The changes have not altered our original conclusions, but they have enriched the manuscript by providing a more comprehensive investigation of the environmental factors that could have contributed to negative  $\delta^{18}$ O anomalies in the Mediterranean at (or near) times of sapropel deposition.

We thank you for your patience in the revision process.

Yours sincerely,

Katharine Grant

# The timing of Mediterranean sapropel deposition relative to insolation, sea-level and African monsoon changes

K.M. Grant<sup>1\*</sup>, R. Grimm<sup>2</sup>, U. Mikolajewicz<sup>2</sup>, G. Marino<sup>1</sup>, M. Ziegler<sup>3</sup>, E.J. Rohling<sup>1,4</sup>

 Research School of Earth Sciences, The Australian National University, Canberra, ACT 2601, Australia

2. Max-Planck-Institute for Meteorology, Bundesstrasse 53, 20146 Hamburg, Germany

3. Department of Earth Sciences, University of Utrecht, Budapestlaan 455, Utrecht 3584 CD, The Netherlands

4. Ocean and Earth Science, University of Southampton, National Oceanography Centre, European Way, Southampton SO14 3ZH, UK

\* Corresponding author. E-mail address: <u>katharine.grant@anu.edu.au</u> (K.M. Grant)

*Keywords:* Eastern Mediterranean, sapropels, African monsoon, sea level, ice sheets, insolation, precession, meltwater pulses.

#### 1 Abstract

2 The Mediterranean basin is sensitive to global sea-level changes and African monsoon 3 variability on orbital timescales. Both of these processes are thought to be important to the 4 deposition of organic-rich sediment layers or 'sapropels' throughout the eastern 5 Mediterranean, yet their relative influences remain ambiguous. A related issue is that an 6 assumed 3-kyr lag between boreal insolation maxima and sapropel mid-points remains to be 7 tested. Here we present new geochemical and ice-volume-corrected planktonic foraminiferal 8 stable isotope records for sapropels S1 (Holocene), S3, S4, and S5 (Marine Isotope Stage 5) 9 in core LC21 from the southern Aegean Sea. The records have a radiometrically constrained 10 chronology that has already been synchronised with the Red Sea relative sea-level record, and this allows detailed examination of the timing of sapropel deposition relative to insolation, 11 12 sea-level, and African monsoon changes. We find that sapropel onset was near-synchronous

13 with monsoon run-off into the eastern Mediterranean, but that insolation-sapropel/monsoon 14 phasings were not systematic through the last glacial cycle. These latter phasings instead 15 appear to relate to sea-level changes. We propose that persistent meltwater discharges into the North Atlantic (e.g., at glacial terminations) modified the timing of sapropel deposition by 16 17 delaying the timing of peak African monsoon run-off. These observations may reconcile 18 apparent model-data offsets with respect to the orbital pacing of the African monsoon. Our 19 observations also imply that the previous assumption of a systematic 3-kyr lag between 20 insolation maxima and sapropel midpoints may lead to overestimated insolation-sapropel 21 phasings. Finally, we surmise that both sea-level rise and monsoon run-off contributed to 22 surface-water buoyancy changes at times of sapropel deposition, and their relative influences 23 differed per sapropel case, depending on their magnitudes. Sea-level rise was clearly 24 important for sapropel S1, whereas monsoon forcing was more important for sapropels S3, 25 S4, and S5.

#### 26 1. Introduction

27

46

28 The Mediterranean basin is an ideal natural laboratory for understanding the dynamic 29 interplay between key climatic and hydrological processes on a range of timescales. Over 30 millennial and longer intervals, changes in global ice volume and African monsoon dynamics 31 are both registered in eastern Mediterranean marine sediments (Rohling et al., 2014, 2015). 32 Detailed understanding of how these processes interact and translate into sedimentary signals 33 is far from complete, and first requires deeper insights into a characteristic feature of these 34 sediments: the periodic deposition of organic-rich layers, or "sapropels", in the eastern basin 35 (for overviews, see Rossignol-Strick 1985; Rohling and Gieskes, 1989; Rohling, 1994; Emeis 36 et al., 1996; Cramp and O'Sullivan, 1999; Rohling et al., 2015, and references therein). 37 38 The formation of these sapropels has been attributed to combinations of surface-water 39 freshening, reduced deep-water ventilation, and increased export production (e.g., Rossignol-40 Strick et al., 1982; Rossignol-Strick, 1985; Rohling, 1994; Higgs et al., 1994; Emeis et al., 41 1996; Jorissen, 1999; Thomson et al., 1999; Mercone et al., 2001; Casford et al, 2003; Abu-

Zied et al., 2008; Schmiedl et al., 2010). The main driver behind these processes was a
relative increase in surface-layer buoyancy, and the mechanism behind this buoyancy forcing

44 has long been attributed to monsoon-related freshwater inputs into the eastern Mediterranean

45 in response to periodic northward shifts of the intertropical convergence zone (ITCZ). Such

47 insolation), which caused the summer monsoon rain belt to intensify and migrate north over

shifts were associated with minima in the precession cycle (hence maxima in boreal summer

48 river catchments and North African wadi systems that drain into the eastern Mediterranean,

49 including the Nile (e.g., Rossignol-Strick et al., 1982; Rossignol-Strick, 1985; Rohling, 1999;

Rohling and De Rijk, 1999; Rohling et al., 2002, 2004; Larrasoaña et al., 2003; Scrivner et
al., 2004; Bianchi et al., 2006; Marino et al., 2007, 2009; Osborne et al., 2008, 2010).

53 This accepted scenario of surface buoyancy forcing primarily due to African monsoon 54 changes has recently been challenged for the most recent (early to middle Holocene) sapropel 55 'S1' (Grimm et al., 2015). Based on comprehensive simulations with a 3-dimensional ocean-56 circulation model coupled to a biogeochemical model and including a sediment module, 57 Grimm et al. (2015) concluded that the evolution of deep-water anoxia prior to S1 formation 58 was initiated by sea-level rise and sea surface warming, and that monsoon-related (i.e., 59 freshwater and nutrient input) forcings were not the sole cause for S1 formation because 60 simulated deep-water oxygen depletion started before the onset of enhanced monsoon run-61 off. However, the study also acknowledged that monsoon forcing potentially played a 62 significant role in determining the vertical extent of anoxia during S1 deposition. These 63 findings lead to the question of whether other sapropels were also intrinsically linked - at 64 least in part – to episodes of global ice melt. If so, this would represent a paradigm shift to the 65 common narrative of how sapropels were formed.

66

A link between sapropel formation and the melting of northern hemisphere ice sheets was 67 68 proposed in early studies of Mediterranean sediments (e.g., Kullenberg, 1951; Olaussen, 69 1960; Ryan, 1972). More recently, Troelstra et al. (1991) suggested that glacial meltwater 70 contributed towards preconditioning the Mediterranean for sapropel formation by lowering 71 surface-water salinities. They also suggested that this preconditioning process was interrupted 72 by the cold Younger Dryas (YD) interval. Rohling (1994) substantiated these claims using a 73 simple hydraulic model (based on Rohling and Bryden, 1994), by demonstrating that both 74 monsoon-related freshwater forcing and deglaciation may have contributed to S1 formation.

Rohling (1994) also suggested that a lag in S1 deposition relative to the nearest insolation
maximum may relate to the YD, which would have interrupted the progressive deterioration
of deep-water ventilation (hence anoxia) necessary for sapropel formation.

78

79 Observational datasets to resolve the monsoon-versus-deglaciation question must include 80 high-resolution records of sapropel deposition, (east) African monsoon variability, and ice-81 volume/sea-level change on robust and chronologically consistent timescales. Such datasets 82 have not been available until recently (Grant et al., 2012, 2014). A key issue in this respect is 83 the fact that a widely accepted astronomically tuned timescale for Mediterranean sediments 84 assumes a 3-kyr lag between precession minima and sapropel mid-points (Hilgen et al., 1993; 85 Lourens et al., 1996, 2004). This assumption was derived from direct radiometric age 86 constraints for sapropel S1 (e.g., Lourens et al., 1996; Mercone et al., 2000; Casford et al., 87 2007; De Lange et al., 2008). Ziegler et al. (2010a) argued that radiometrically dated oxygen 88 isotope records derived from Chinese speleothems could be used as a tuning target for eastern 89 Mediterranean sapropels, in order to test if the specific S1 phase relationship can be 90 extrapolated to older sapropels. They found that the lag is variable for older sapropels but that 91 on average the assumption of a ~3 kyr lag is a good approximation for the Late Pleistocene. 92

Here we examine the detailed timing of sapropel deposition relative to insolation, sea-level,
and African monsoon changes, using highly resolved, co-registered signals (i.e., measured on
the same sample suite) of monsoon-related run-off and sapropels S1, S3, S4 and S5 in eastern
Mediterranean core LC21 (southeastern Aegean Sea; Fig. 1). These records have a
radiometrically constrained chronology which has already been synchronised (Grant et al.,
2012) with the Red Sea relative sea-level (RSL) record (Siddall et al., 2003, 2004; Rohling et
al., 2009), so inferred phase relationships between sapropels, monsoon forcing, and sea-level

100 changes are robust within quantified uncertainties. It has been demonstrated that the Red Sea 101 RSL record closely approximates global (eustatic) sea-level variations (see discussion and glacial isostatic modelling in Grant et al., 2014), which in turn reflect global ice-volume 102 103 variability on glacial-interglacial timescales. We can therefore use the tightly constrained relationships in our dataset to remove ice-volume effects from the LC21  $\delta^{18}$ O record of the 104 planktonic foraminifer *Globigerinoides ruber* (white) ( $\delta^{18}O_{ruber}$ ), to produce a robust, 105 106 deconvolved climate and hydrological record for the eastern Mediterranean, which extends 107 over the entire last glacial cycle. We also present new sediment geochemical records from 108 core LC21 that accurately delineate its sapropel intervals. By examining timing relationships 109 - on an internally consistent, U/Th-based chronology - between sapropel deposition and 110 changes in sea level, insolation and African monsoon run-off, we seek to clarify current 111 understanding of sapropel formation mechanisms under different glacial boundary conditions. 112 It is important to emphasise here that current knowledge of sapropel formation is largely 113 based on sapropel S1, followed by S5, because these are the most studied sapropels. Yet S1 114 was relatively weakly developed and was associated with lower insolation forcing, compared 115 to other sapropels. Also, both S1 and S5 were deposited after large deglaciations. These 116 sapropels are therefore not typical of all sapropels, hence the need to examine a range of 117 sapropels with different magnitudes of the postulated forcings.

118

119 An additional incentive for more detailed study of the dynamics behind sapropel formation is 120 the recent development of a method to reconstruct past sea-level changes using eastern 121 Mediterranean sediments (Rohling et al., 2014). This method relies on the sensitivity of 122 eastern Mediterranean seawater  $\delta^{18}$ O to glacial-interglacial sea-level changes in the Gibraltar 123 Strait. However, the conversion of Mediterranean  $\delta^{18}$ O to sea level is not systematically 124 straightforward, due to the periodic influx of <sup>18</sup>O-depleted monsoon run-off into the eastern

125	Mediterranean. As a result, the Mediterranean sea-level method currently involves identifying
126	sapropel intervals and removing them from the sea-level reconstruction. A better
127	understanding of eastern Mediterranean $\delta^{18}$ O changes across sapropel intervals, as presented
128	in this study, will therefore contribute towards improving the Mediterranean sea-level
129	method.
130	
131	2. Methods
132	
133	2.1 Geochemical analyses
134	
135	Scanning x-ray fluorescence (XRF) elemental analyses of the archive halves of sediment core
136	LC21 (southern Aegean Sea, $35^{\circ}$ 40' N, $26^{\circ}$ 35' E; Fig. 2) were performed at the British
137	Ocean Sediment Core Research Facility (BOSCORF) at the National Oceanography Centre,
138	Southampton, using an Itrax XRF core scanner (Cox Analytical Systems, Gothenburg,
139	Sweden). XRF data were collected every 0.5 mm down-core using a molybdenum tube set at
140	30 kV and 30 mA, and a sampling time of 40 seconds directly at the core surface. The
141	exposed core surface was covered with a 4 micron thin SPEX Certi Prep Ultralene1 foil to
142	avoid contamination of the XRF measurement unit and desiccation of the sediment.
143	Subsequent sub-sampling at continuous 1-cm intervals and stable isotope analyses of the
144	same core halves have been described in Grant et al. (2012). The foraminiferal stable oxygen
145	and carbon isotope records discussed below are for the surface-dwelling species
146	Globigerinoides ruber (white) ( $\delta^{18}O_{ruber}$ , $\delta^{13}C_{ruber}$ ) and the sub-surface dwelling species
147	<i>Neogloboquadrina pachyderma</i> (dextral) ( $\delta^{18}O_{pac}$ , $\delta^{13}C_{pac}$ ) (cf. Rohling et al., 2004). Tests of
148	these species were selected from >300 $\mu$ m and 150-300 $\mu$ m sieved sediment fractions,
149	respectively (see Methods in Grant et al., 2012).

### 151 2.2 Sapropel boundaries

153	Four sapropels (S1, S3, S4, S5) are visibly identified in core LC21 by distinct colour and
154	lithological changes, from olive-grey, fine-grained nannofossil ooze/clay admixtures to much
155	darker layers rich in organic matter (sapropels) (Fig. 2). An 'interruption' to sapropel
156	deposition is evident in S4 and to a lesser extent in S1 (Fig. 2). Because redox reactions at the
157	sediment-seawater interface, and within sediments, can cause downward reduction at the
158	sapropel base and downward oxidation (or 'burn down') at the sapropel top, sapropel
159	boundaries are best identified using elements that are enriched in sapropels and that
160	subsequently exhibit conservative behaviour within sediments (Thomson et al., 1995; De
161	Lange et al., 2008). Barium is ideal for this purpose because it is well-preserved in sediments
162	(Dymond et al., 1992), and enriched in sapropels (Thomson et al., 1995; De Lange et al.,
163	2008) due to its association with export productivity (Dymond et al., 1992). In core LC21,
164	pronounced increases in Ba at sapropel horizons are accompanied by elevated Vanadium (V).
165	Vanadium is a redox-sensitive element and precipitates under reducing conditions, so
166	although V can be mobile in sediments, the good agreement between increases in V and Ba in
167	core LC21 implies that – in this case – elevated V reliably indicates sapropel boundaries (see
168	Thomson et al., 1995; Nijenhuis et al., 1999; Table 1).
169	
170	2.3 Chronology

The age-depth model for core LC21 is described in detail in Grant et al. (2012). Briefly, it is
constrained by 14 direct radiocarbon datings and two geochemically 'fingerprinted' tephra
layers (the Minoan and Campanian Ignimbrite; Satow et al., 2015) for the interval 0-40 ka

175	BP, and by process-based correlation to the U/Th-dated Soreq Cave (Israel) speleothem $\delta^{18}O$
176	record for the interval >40-150 ka BP. All tie-points, radiometric ages (including all Soreq
177	speleothem datings) and their uncertainties were probabilistically assessed in a Bayesian
178	deposition model (using 'OxCal' software, Bronk Ramsey, 2008, 2009). Full error
179	propagation yielded age uncertainties (at $2\sigma$ ) of 0.4±0.2 kyr for the interval 0-40 ka BP, and
180	2.0±0.9 kyr for the interval >40-150 ka BP. The Red Sea RSL record was synchronised with
181	the LC21 chronology using the LC21 $\delta^{18}O_{pac}$ record (Grant et al., 2012); this record primarily
182	reflects glacial-interglacial sea-level changes due to a 'glacial concentration' effect on
183	Mediterranean seawater $\delta^{18}$ O similar to that in the Red Sea (see Rohling, 1999; Grant et al.,
184	2012; and Rohling et al., 2014 for more technical details).
185	
186	3. Deconvolving the LC21 $\delta^{18}O_{ruber}$ record
187	
188	3.1 Global ice-volume effects
189	
190	As mentioned before, global ice-volume changes strongly affect Mediterranean seawater $\delta^{18}$ O
191	due to the restriction of exchange flow through the Strait of Gibraltar with lowering of sea
192	level (Rohling, 1999; Rohling et al., 2014). The LC21 $\delta^{18}$ O records therefore contain an
193	important sea-level component (equations 1 and 2); removal of this component from the
194	$\delta^{18}O_{ruber}$ record – a proxy for eastern Mediterranean surface-water $\delta^{18}O$ – will therefore
195	isolate local climatic influences on Mediterranean surface-water $\delta^{18}O$ (see below).

196 Calculation of this 'residuals' signal first requires conversion of the Red Sea relative sea-

197 level (RSL) record (y) into equivalent eastern Mediterranean  $\delta^{18}$ O values (x), based on the

198 upper (equation 1) and lower (equation 2) probability limits for a quadratic relationship

199 determined by Rohling et al. (2014):

200

$$y = 18.23253367 - 54.32756406x + 2.68013962x^2 \tag{1}$$

$$y = -19.83859107 - 54.97329064x + 1.027303677x^2$$
(2)

201

202 Because the Red Sea RSL record is unreliable ca 14-23 ka BP (Fig. 3a; see Rohling et al., 2009), we have additionally converted into equivalent Mediterranean  $\delta^{18}$ O values the 203 204 probabilistic sea-level record of Stanford et al. (2011) for the last deglaciation (this record is based on numerous radiometrically dated sea-level indicators). The converted RSL records 205 (Fig. 3a) were normalised to the Late Holocene values for  $\delta^{18}O_{\text{ruber}}$  and then subtracted from 206  $\delta^{18}O_{ruber}$ . The resultant 'LC21 residuals' are defined by  $\delta^{18}O_{ruber}$  values lighter/heavier than 207 the upper/lower 95% probability intervals of the converted RSL records (Fig. 3b). While 208 209 these probability intervals should account for most of the sea-level/ice-volume component in the  $\delta^{18}O_{\text{ruber}}$  record, we note that there may be two short-term intervals (Section 3.2 below) 210 where glacial influences on Mediterranean  $\delta^{18}$ O have not been captured in the  $\delta^{18}$ O-to-sea-211 212 level conversion. 213

214 *3.2 Meltwater effects* 

215

Observational data and numerical modelling both suggest that the flow of Atlantic water into the Mediterranean was significantly increased during glacial and deglacial North Atlantic freshening events, or 'Heinrich Stadials' (HS), compared to background conditions (e.g., Cacho et al., 1999; Sierro et al., 2005; Rogerson et al., 2010). Sierro et al. (2005) demonstrated that the past five HS were associated with significant <sup>18</sup>O depletions in western Mediterranean surface waters, after correcting the foraminifera  $\delta^{18}$ O for temperature and icevolume effects. These depletions were observed in the Alboran Sea (core MD95-2043) as 223 well as north of the Balearic Islands (core MD99-2343) (Fig. 1). Subsequent studies have corroborated these findings. A western Mediterranean surface water <sup>18</sup>O depletion during HS 224 225 has been documented for: the Gulf of Cadiz during HS1 (Voelker et al., 2006), the Strait of 226 Sicily during HS1 (Essallami et al., 2007), the Gulf of Lion during HS1, HS2 and the YD 227 (Melki et al., 2009; Lombo-Tombo et al., 2015), the Menorca Rise (ODP site 975) during 228 HS11 (Kandiano et al., 2014), and the Alboran Sea (ODP site 976) during HS1 and HS11 (Jiménez-Amat and Zahn, 2015) (Fig. 1). Moreover, a synthesis of surface water  $\delta^{18}O$ 229 profiles from sediment cores from the Gulf of Cadiz and Alboran Sea covering the last ~30 230 ky revealed a reduced Atlantic-Mediterranean  $\delta^{18}$ O gradient during HS (Rogerson et al., 231 232 2010). In the same study, a one-dimensional hydraulic control model for the Strait of 233 Gibraltar confirmed that enhanced Atlantic inflow to the Mediterranean would result from a 234 freshening of the North Atlantic associated with iceberg meltwater. Such a scenario has 235 recently been corroborated by hosing experiments with a suite of CMIP5 models 236 (Swingedouw et al., 2013), which show that (isotopically light) meltwater from the subpolar 237 North Atlantic is directed towards the Gulf of Cadiz by the Canary current. 238 239 Comparison of the LC21 residuals with a record of sea-level change rates ('dRSL') on the 240 same LC21 chronology, and with the (radiometrically dated) timing of meltwater pulse 1a 241 (Deschamps et al., 2012) and HS1 (Álvares-Solas et al., 2011), and with the radiometrically 242 constrained timing of HS11 (Marino et al., 2015) (Fig. 3c), reveals that the timing of large 243 melting events during terminations I and especially II coincides with distinct intervals of 244 negative residuals. The timing of HS3 and HS5 can also potentially be correlated with pronounced negative LC21 residuals ca 28-30 and 46-48 ka, respectively (Fig. 3b), in line 245

with Sierro et al's (2005) observations of surface-water  $^{18}$ O depletions in the western

Mediterranean during the last five HS. This inference is possibly supported by dRSL for HS5,
but not for HS3 (Fig. 3c).

249

If we consider sub-surface  $\delta^{18}$ O records from core LC21 and ODP site 975 that have been 250 tuned to the same age model (Marino et al., 2015; Figs. 1, 4a), we observe an isochronous <sup>18</sup>O 251 252 depletion at these sites during HS11. Although on a different age model, a depletion of similar timing and magnitude at ODP site 975 is recorded by surface-water  $\delta^{18}$ O records 253 (Kandiano et al., 2014; Fig. 4b), so the LC21<sup>18</sup>O depletion appears to be consistent with 254 255 widespread observations from the western Mediterranean. In that case, it seems likely that 256 negative spikes in the LC21 residuals during major deglaciation events are due to North Atlantic (meltwater-related)  $\delta^{18}$ O anomalies propagating into the eastern Mediterranean, and 257 these short-term signals are not captured by the RSL-to- $\delta^{18}$ O conversion for the 258 259 Mediterranean (Rohling et al., 2014).

260

261 An additional consideration is the direct input of glacial meltwater into the Mediterranean 262 from alpine deglaciation and/or retreat of the European ice sheet (EIS). Regarding alpine 263 meltwater, a synthesis of Mediterranean mountain glacial activity suggests that the last 264 deglaciation was staggered, with several glacial advances/retreats occurring between the last glacial maximum and the Holocene, yet there are no reliably dated records from many areas 265 266 (see Hughes & Woodward, 2009). The relatively small glaciers in this region would have responded rapidly to climate change and decayed much faster than extensive ice sheets 267 (Hughes et al., 2006), which implies that the largest meltwater effects on Mediterranean  $\delta^{18}$ O 268 would have occurred early in the deglaciation. It therefore seems unlikely that the negative 269 270 LC21 residuals ca 15 ka are primarily due to alpine meltwater. Nonetheless, a negative shift in surface water  $\delta^{18}$ O coinciding with HS1 has been recorded in cores from the Rhone 271

Canyon, in the far north Gulf of Lion (Lombo Tombo et al., 2015), which may reflect
enhanced Rhone run-off at that time. Also, Rohling et al. (2015) argued that alpine meltwater

from the Rhone offered the most plausible explanation for the onset of the most recent

275 organic rich layer (ORL1) in the western Mediterranean, at ~14.5 ka.

276

277 Even less information about Mediterranean alpine meltwater is available for the penultimate deglaciation. The negative LC21 residuals spike ca 133 ka occurs ~1-2 ky after the onset of 278 279 termination 2 (i.e., closer to the onset of deglaciation than during termination 1), so alpine 280 meltwater may have contributed to this isotopic depletion in LC21. However, the relatively small volume and <sup>18</sup>O depletion of alpine meltwaters compared to global ice sheets means 281 282 that North Atlantic meltwaters propagating into the eastern Mediterranean would likely dominate any  $\delta^{18}$ O meltwater signals in LC21. Further data are needed to address this 283 284 question.

285

286 Regarding meltwater from European ice sheets, recent work suggests that drainage of the EIS 287 at the end of the last glacial cycle was predominantly into the North Atlantic (Toucanne et al., 288 2015a), but there is also evidence that meltwater from the Fennoscandian ice sheet (FIS) flowed into the Black Sea during HS1, and that this overflowed into the northern Aegean Sea 289 290 (Soulet et al., 2013). Other studies of Black Sea sediments, however, suggest generally stable 291 environmental and hydrologic conditions through the last deglaciation until 14.5 ka BP, 292 despite an interruption between 16.5 and 14.8 ka BP which was linked to input of sediments 293 from a northern source (possibly the FIS) (Major et al., 2006; Bahr et al., 2006, 2008). Those 294 and other studies (Sperling et al., 2003; Rohling et al., 2015) agree that reconnection of the 295 Black Sea with the Mediterranean appears to have been established much later, ca 9 ka BP. 296 There are far fewer robust observations for the penultimate deglaciation, so in view of

297 available evidence, we surmise that any input of meltwater from European ice sheets directly 298 into the Mediterranean was likely relatively small for the last deglaciation, and therefore 299 would not significantly affect the LC21  $\delta^{18}$ O residuals.

300

301 In short, the timing, volume and isotopic composition of alpine run-off into the 302 Mediterranean over glacial-interglacial timeframes is not well constrained. While this freshwater source cannot be ruled out, the close timing between negative spikes in the LC21 303 residuals, <sup>18</sup>O depletions in the western Mediterranean, and HS suggests that the propagation 304 305 of meltwater from the North Atlantic into the eastern Mediterranean explains most of the 306 negative LC21 residuals at the end of HS1 and within HS11 (Fig. 3b). This is the first time such an inference has been made for the eastern Mediterranean, and it has important new 307 implications for the interpretation of eastern Mediterranean  $\delta^{18}$ O records, including those 308 309 from speleothems whose source waters derive from eastern Mediterranean sea-surface water.

310

311 3.3 Local climatic effects

312

313 Next we consider local climatic influences on Mediterranean surface-water  $\delta^{18}$ O, and how 314 these may manifest in the LC21 residuals. Such influences include sea surface temperature 315 (SST), regional precipitation/evaporation, and African monsoon run-off.

316

317 <u>3.3.1 Temperature</u>

318 The RSL-to- $\delta^{18}$ O conversion accounts for glacial-interglacial Mediterranean SST contrasts of

319 +5 °C (summer) and +3.5 °C (winter), with probabilistically determined uncertainties (see

Rohling et al., 2014). These gradients are based on reconstructions for the LGM-to-Present

321 (Hayes et al., 2005) and agree with isotope- and alkenone-derived SST estimates (Cita et al.,

322 1977; Emeis et al., 2003). Such reconstructions also imply, however, that the penultimate 323 glacial-interglacial SST gradient in the eastern Mediterranean was up to double that of the 324 LGM-to-Present (Cita et al., 1977; Emeis et al., 2003). In that case, the sharp rise in negative 325 LC21 residuals at the onset of sapropel S5 (Fig. 3b) might partly reflect a SST increase that 326 has not been accounted for in the RSL-to- $\delta^{18}$ O conversion.

327

We have converted existing SST data for core LC21 (Marino et al., 2007, 2009) to our U/Th-328 329 related chronology (Fig. 5a,b). Although the data only cover the intervals of sapropels S1 and 330 S5, detailed SST reconstructions for the eastern Mediterranean (Emeis et al., 2000, 2002, 331 2003) suggest that glacial-interglacial SST increases were greater immediately prior to S1 332 and S5 than for S3 and S4. Hence, any temperature effects on the LC21 residuals at the onset of S3 and S4 should be less pronounced than those for S1 and S5. The RSL-to- $\delta^{18}O$ 333 334 conversion used to calculate the LC21 residuals (Section 3.1) is based on LGM-to-Present 335 temperature gradients; therefore, the residuals record implicitly takes into account any SST 336 increases prior to the onset of sapropels S1, S3 and S4. 'Small-scale' temperature variations are not accounted for in the RSL-to- $\delta^{18}$ O conversion, although their effect on the LC21 337 338 residuals at sapropel onset would likely be negligible compared to the larger warming trends. Regarding sapropel S5, the LC21 (Fig. 5b) and comparable SST records (e.g., for southern 339 340 Aegean core MD40/67 in Emeis et al., 2003) suggest a glacial-interglacial temperature 341 gradient that was 3-4 °C (equivalent to a maximum of 0.8‰) greater than that of the last 342 deglaciation. Allowing for this ~1‰ change due to SST changes means that at least 0.5‰ of 343 the negative shift in residuals at the onset of S5 must still be explained. 344

345 3.3.2 Precipitation–evaporation

Any temperature effects on foraminiferal calcite  $\delta^{18}$ O would be (partly) offset by 346 fractionation effects on seawater  $\delta^{18}$ O due to evaporation. Hence, our inferred depletions in 347 LC21 residuals due to increased SSTs should be viewed as maximum estimates, and were 348 349 likely lower. The next question is therefore: to what extent do the remaining negative 350 residuals (Fig. 3b) reflect a decrease in net evaporation from the Mediterranean region during 351 times of sapropel deposition? This has long been debated, reflecting the contrasting and/or so far ambiguous proxy evidence (mainly pollen data) for hydrological changes in the eastern 352 353 Mediterranean region at times of sapropel formation (see Tzedakis, 2007, 2009 and Rohling 354 et al., 2015 for overviews).

355

356 For example, Kallel et al. (1997, 2000) inferred that the salinity of eastern Mediterranean 357 surface waters (SSS) was homogenous during sapropel deposition, and suggested that this reflected an increase in local precipitation. However, a statistical assessment of  $\delta^{18}$ O 358 359 gradients through the Mediterranean basin for the time of S1 deposition indicated that 360 strongest depletion clearly occurred in the Levantine region, in the vicinity of the Nile outlet (Rohling and De Rijk, 1999). Those authors also contested the calibration of  $\delta^{18}$ O into SSS as 361 used by Kallel et al. (1997, 2000), because the modern sea-surface salinity: $\delta^{18}$ O ratio in the 362 363 Mediterranean does not hold for periods of enhanced freshwater input to the basin; that 364 objection was further quantified by Rohling (1999) and Rohling et al. (2004). The importance 365 of 'other freshwater sources' (than the Nile) to the eastern Mediterranean at the beginning 366 and end of sapropel S5 deposition was also suggested by Scrivner et al. (2004), based on an apparent disparity between trends in Nd isotopes and residual  $\delta^{18}O$  (i.e., corrected for 367 368 temperature and ice-volume effects) across S5 at ODP site 967. However, the Nd record is of lower resolution compared to the  $\delta^{18}$ O residuals, and, even if the offset is real, run-off from 369 370 the wider North African margin could account for 'other freshwater sources', as was shown

371 by Osborne et al. (2008). Palynological studies of the eastern Mediterranean have also 372 inferred an increase in summer precipitation during times of sapropel formation (e.g., Rossignol-Strick, 1987, 1999; Wijmstra et al., 1990; Rossignol-Strick and Paterne, 1999; 373 374 Langgut et al., 2011). However, Tzedakis (2009) questioned the reliability of these 375 interpretations, and suggested that drought-resistant species or winter precipitation equivalent 376 to the present-day regime could explain the observed trends, without the need to invoke 377 enhanced summer precipitation. This interpretation is supported by Toucanne et al. (2015b), 378 who inferred an increase in winter precipitation over the northern Mediterranean borderlands 379 (NMB) during warm intervals of interglacials over the past 547 kyr, based on a suite of 380 geochemical and lithological tracers of Golo River run-off into the northern Tyrrhenian Sea 381 (core GDEC-4-2, Fig. 1).

382

383 In contrast, a comprehensive review of evidence for humidity changes in the Levant over the 384 last two glacial-interglacial cycles concluded that, in general, it was the glacial (rather than 385 interglacial) periods that were wetter (Frumkin et al. 2011). This is quantitatively supported 386 by calculations of Dead Sea (=Lake Lisan) levels over the past 120 ka BP (Rohling, 2013). 387 Similarly, groundwater isotope data suggest that Saharan aquifers were recharged during 388 glacial rather than interglacial periods (Abouelmagd et al., 2012, 2014). However, 389 speleothem growth periods in the Negev Desert (Israel) – indicative of wetter intervals – 390 generally coincide with interglacials and monsoon maxima/precession minima (i.e., sapropel 391 intervals), but not always: the Northern Negev appears to have been arid at 105 ka (~sapropel 392 S4) and 11 ka (≈S1) (Vaks et al., 2006).

393

394 Despite these apparently disparate findings, a consensus is emerging that they can be

395 explained by increased seasonality and winter precipitation in the eastern Mediterranean

396 during precession minima (Tzedakis, 2007; Frumkin et al., 2011; Milner et al., 2012; Rohling 397 et al., 2015; Toucanne et al., 2015b). This argument is supported by recent simulations using 398 a high-resolution, fully coupled ocean-atmosphere general circulation model (Bosmans et al., 2015). If we assume the 'winter precipitation' hypothesis to be correct, the key question then 399 400 is what would be the effect of increased winter rainfall on eastern Mediterranean sea-surface  $\delta^{18}$ O, and could that explain negative LC21  $\delta^{18}$ O residuals during precession minima? In 401 402 Bosmans et al.'s (2015) simulations, the predicted winter precipitation was not uniform over 403 the Mediterranean but focussed over the Ionian and Levantine Seas, and was associated with 404 local convection rather than eastward propagating storm tracks from the North Atlantic. 405 Interestingly, no increase in either summer or winter rainfall over the NMB during precession 406 minima was indicated, whereas both summer and winter rainfall increased over the Levant. These observations suggest that the effect on surface water  $\delta^{18}$ O of any precipitation increase 407 408 would be localised; furthermore, this effect would likely be either negligible (i.e., balanced by increased evaporation from the same basin) or an increase in  $\delta^{18}$ O values (if locally 409 410 evaporated moisture precipitated outside the catchments of the eastern Mediterranean basin) 411 (Rohling et al., 2015).

412

To further investigate these effects, we developed a record of 'Soreq excess  $\delta^{18}$ O' ( $\delta^{18}$ O<sub>SOREO</sub>) 413 xs) for the Soreq cave (Israel) speleothem  $\delta^{18}$ O record ( $\delta^{18}O_{speleo}$ ) by subtracting the LC21 414  $\delta^{18}O_{ruber}$  record from it (Fig. 6). The  $\delta^{18}O_{SOREO XS}$  record thus represents a deconvolved local 415 416 eastern Mediterranean climate signal, from which any bias related to monsoon/riverine run-417 off or global glacial cycles has been removed. This calculation is possible because *i*) there is a direct evaporation-precipitation link between the  $\delta^{18}$ O composition of eastern 418 419 Mediterranean surface waters and speleothems from the Levant (Matthews et al., 2000; Bar-420 Matthews et al., 2003; McGarry et al., 2004; Kolodny et al., 2005; Almogi-Labin et al., 2009; 421 Marino et al., 2009; Grant et al., 2012), *ii*) temperature variations in eastern Mediterranean 422 surface waters and Soreq cave are coupled (Bar-Matthews et al., 2003; Affek et al., 2008), and *iii*) the  $\delta^{18}O_{\text{speleo}}$  and  $\delta^{18}O_{\text{ruber}}$  records have been previously synchronised (Grant et al., 423 2012). We first interpolated the  $\delta^{18}O_{speleo}$  and  $\delta^{18}O_{ruber}$  records to the same age-steps, 424 425 subtracted the mean from each record so that they were centred about a mean of zero, and then subtracted the zero-centralised  $\delta^{18}O_{\text{ruber}}$  from the zero-centralised  $\delta^{18}O_{\text{speleo}}$ . The 426 uncertainty of  $\delta^{18}O_{SOREQ XS}$  about the mean value (±0.38 at  $2\sigma$ ) was calculated using a root 427 428 mean squares calculation, and combines uncertainties for the average Late Holocene value of  $\delta^{18}O_{speleo}$  (±0.29) and  $\delta^{18}O_{ruber}$  (±0.24). 429

430

The  $\delta^{18}O_{\text{SOREO XS}}$  values suggest that most of the S1, S3, and S4 intervals were not 431 characterised by systematically wetter/warmer conditions in the Levant (Fig. 6b), although an 432 433 inferred decrease in net evaporation during brief (<1 ky) periods within the latter halves of 434 sapropels S1 and S4 can account for some of the variability in the residuals at those times. In contrast,  $\delta^{18}O_{SOREO XS}$  implies intermittently wetter/warmer conditions at Soreq Cave during 435 436 sapropel S5 (Fig. 6b). These interpretations are broadly consistent with the findings of Vaks et al. (2006), who noted that not all monsoon maxima/precession minima were associated 437 with more humid intervals in the Levant. Specifically, both  $\delta^{18}O_{\text{SOREO XS}}$  and the Vaks et al. 438 439 (2006) study imply that the interval of S5 was generally more humid, while the S4 and S1 intervals were relatively arid. The  $\delta^{18}O_{\text{SOREO XS}}$  record suggests that the S3 interval was also 440 441 relatively arid, although the Vaks et al. (2006) study suggests that this interval was more 442 humid. We note that the Bosmans et al. (2015) 'precession minima' simulations were performed for maximum eccentricity only, i.e., equivalent to the orbital configuration for S5; 443 444 future modelling of precession minima of different intensities is therefore needed to fully validate the observational data. Finally, positive  $\delta^{18}O_{SOREO XS}$  values in the earliest part of S5 445

446 imply drier conditions in the eastern Mediterranean at that time. This is consistent with 447 evidence for a peak in Mediterranean sclerophylls during the initial period (first  $\sim 2 \text{ kyr}$ ) of 448 interglacials, which strongly suggests summer aridity (Tzedakis et al., 2002, 2003; Tzedakis, 449 2009, and references therein). Hence, the sharp decrease in LC21 residuals at the onset of S5 (Fig. 3b) is not likely to represent decreased net evaporation in the eastern Mediterranean 450 451 region.

452

453 To sum up, minimal climatic influences can be attributed to the LC21 residuals through S1, 454 S3 and S4, and a maximum of -1‰ (likely less) of the LC21 residuals through sapropel S5 455 can be explained by local climate changes. We therefore conclude that most of the abrupt 456 negative shifts in LC21 residuals at the onset of sapropels are unlikely to be due to local 457 climatic changes, and instead reflect external, monsoon-related run-off into the eastern 458 Mediterranean from the Nile and wider North African margin (e.g., Rossignol-Strick, 1985; 459 Rohling, 1999; Rohling et al., 2002; Scrivner et al., 2004; Osborne et al., 2008, 2010). 460

#### 461 4. A proxy for African monsoon run-off

462

A useful means for validating our conclusion that the LC21 residuals predominantly reflect 463 464 African monsoon run-off into the eastern Mediterranean, is to compare them with past 465 changes in atmospheric methane ( $CH_4$ ) concentrations. Such changes are closely related to 466 the extent of tropical wetlands, and therefore also to changes in monsoon precipitation (Chappellaz et al., 1990; Spahni et al., 2011). Alternatively, we could compare the residuals 467 468 to other African monsoon proxy records, yet this is not straightforward due to i) large spatial 469 and temporal heterogeneity in African monsoon precipitation, *ii*) variable threshold responses 470 among different precipitation/monsoon proxies, *iii*) a dearth of suitable records extending

471 from the Holocene to MIS 5e, and iv) chronological uncertainties within and between 472 different proxy records (see Gasse, 2000). While ice-core CH<sub>4</sub> records are not without 473 chronological uncertainties, and do not unambiguously reflect monsoon variability (for 474 instance, they are also coupled to northern hemisphere temperature fluctuations; see 475 Baumgartner et al., 2014), they are advantageous because they provide a continuous, high 476 resolution, well-mixed (spatially and temporally) signal over the entire last glacial cycle that 477 is strongly coupled to boreal tropical hydrological changes. We therefore compare our LC21 478 residuals with ice-core CH<sub>4</sub> records from Greenland (North Greenland Ice-core Project 479 (NGRIP); Baumgartner et al., 2014) and from Antarctica (EPICA Dronning Maud Land 480 (EDML); Schilt et al., 2010; Bazin et al., 2013) (Fig. 5). Figure 5 reveals good agreement in 481 the timing of abrupt CH<sub>4</sub> increases and negative LC21 residuals prior to sapropel onset, for 482 all four sapropel intervals considered here.

483

484 The agreement is excellent for S3 and S5 at sapropel onset (Fig. 5b,c). A smaller peak in the 485 LC21 residuals before S5 (ca 132 ka) can be (partly) explained by isotopically light 486 meltwater reaching the eastern Mediterranean (section 3.2), so we do not compare the LC21 487 residuals and CH4 over this interval. For S4, methane levels start to increase ~0.5-1 kyr 488 before the LC21 residuals decrease (Fig. 5d). While this offset is within the age uncertainties 489 of both records, the timing of the LC21 residuals decrease is very closely aligned with a marked increase in  $\delta^{18}$ O in a U/Th-dated speleothem ('NALPS'; Boch et al., 2011) from the 490 491 European Alps (Fig. 5d). The NALPS record primarily reflects high-latitude temperature 492 variations in the wider North Atlantic, and is remarkably similar to NGRIP temperature 493 reconstructions (Boch et al., 2011). Given that NGRIP methane and temperature records are 494 directly in phase over the last glacial cycle (Baumgartner et al., 2014) (Fig. 5d), the U/Th-495 dated NALPS record can be used to independently validate the NGRIP chronology in this

interval. In detail, this reveals a clear offset between shifts in the NALPS and NGRIP records prior to the onset of sapropel S4 and within the S4 interruption (Fig. 5d), which suggests that the NGRIP chronology may be ~1 ky too old at around 108 ka BP. Interestingly, our U/Thbased chronology for the LC21 residuals yields a much closer agreement to the U/Th-dated NALPs record than to the NGRIP methane and temperature records (Fig. 5d). Thus, the  $CH_4$ and LC21 residuals records may be more closely in phase than figure 5d suggests, once the apparent age correction is implemented to the NGRIP records.

503

504 For sapropel S1, the timing of two abrupt CH<sub>4</sub> rises appears to be slightly offset from 505 decreases in the LC21 residuals (Fig. 5a). This offset may simply reflect age model 506 uncertainties (in LC21 and/or the ice cores), yet this is arguably the most reliably dated 507 section of all cores. An alternative explanation is that negative peaks in LC21 residuals at 508 15.5–14.3 ka BP, 12.5 ka BP, and at 11.8–11.6 ka BP may (partly) relate to meltwater release 509 into the North Atlantic during the last deglaciation. As explained in Section 3.2, such meltwater pulses would bring isotopically light  $\delta^{18}$ O into the Mediterranean, and this has not 510 been accounted for in the RSL-to- $\delta^{18}$ O conversion which is used to calculate the LC21 511 512 residuals. A third explanation is that northern hemisphere temperature, rather than tropical 513 precipitation, accounts for most of the CH<sub>4</sub> signal at this time. Both the NGRIP temperature 514 and CH<sub>4</sub> records clearly show the Bølling-Allerød warm interval (~13–15 ka BP), which is 515 less apparent in the LC21 residuals (Fig. 5a). Given these various explanations, we do not 516 attempt to interpret the LC21 residuals outside of the peak monsoon run-off interval 517 associated with sapropel S1. In that case, there is generally good agreement between the 518 LC21 residuals and CH<sub>4</sub> records from ~11 ka BP onwards (Fig. 5a). Thus, considering all 519 four sapropels in LC21, the  $CH_4$  records support our inference that the LC21 residuals

reliably track African monsoon run-off into the eastern Mediterranean at times of sapropeldeposition.

522

523 **5. Discussion** 

524

525 5.1 Monsoon–sapropel phasing

526

527 Our reconstructions suggest that the increases in African monsoon run-off associated with 528 sapropels S3, S4 and S5 are sharply delineated, and coincide closely with the starts of 529 sapropel deposition. For sapropel S1, the onset of the monsoon signal seems somewhat 530 masked in the LC21 residuals, likely due to meltwater effects (Fig. 3; Section 3.2). 531 Nonetheless, an abrupt increase in monsoon run-off to peak values coincides with the onset 532 of S1. These coherent observations suggest that monsoon-related freshwater inputs to the 533 eastern Mediterranean were a primary driver in triggering the onset of sapropel formation, 534 consistent with conventional views (e.g., Rossignol-Strick, 1985; Emeis et al., 1996; Ziegler 535 et al., 2010a).

536

537 The timing of sapropels (and monsoonal flooding) relative to insolation/precession, however, 538 varies by up to ~3 kyr, irrespective of whether the sapropel onset or mid-point is used for 539 phase calculations (Table 2, Fig. 7). We focus on sapropel mid-point phasings in order to 540 compare our results with previous studies (Lourens, 1994; Lourens et al., 1996, 2004; Ziegler 541 et al., 2010a). The mid-points of sapropels S3 and S4 in core LC21 occurred within 0.1-0.8 542 kyr of precession minima and insolation maxima (Table 2; Fig. 7c,d), whereas the mid-points 543 of sapropels S1 and S5 lag the nearest precession minimum/insolation maximum by 2.1-3.3 544 kyr (Table 2; Fig. 7a,b). Previous studies inferred a lag of ~3 kyr between the mid-point of S1 and the nearest precession minimum/insolation maximum (Lourens, 1994; Lourens et al.,

546 1996, 2004; Ziegler et al., 2010a), and we find a comparable phase offset for S1. We find that

547 it also appears to be valid for S5, but not for S3 or S4. We note, however, that the mid-point

548 ages for S1, S3, S4 and S5 in Ziegler et al. (2010a) are all within chronological uncertainties

549 of the new mid-point ages presented here, when we consider what Ziegler et al. (2010a)

termed "precursory events" of S3 and S4 as part of the sapropel.

551

552 It is worth bearing in mind that the timing of a sapropel mid-point depends on the duration of 553 sapropel deposition, as noted by Emeis et al. (2003), and this clearly varies (Table 1). In core 554 LC21, the largest discrepancy between sapropel durations is between S1 and S5 (2.4 kyr, 555 Table 1), yet both sapropels exhibit a similar precession-lag. Furthermore, S4 and S5 have 556 similar durations, yet very different insolation phasings (Tables 1,2). Therefore, differences 557 among sapropel mid-point phasings with respect to precession are not simply an artefact of 558 variable sapropel deposition periods. An alternative approach is to consider 559 sapropel-insolation phasings based on the onset rather than mid-point of a sapropel. This 560 makes sense because African monsoon precipitation is influenced by positive vegetation-561 albedo feedbacks which vary with insolation intensity (Nicholson, 2009). These feedbacks are therefore unlikely to be the same for all sapropels. However, at the onset of elevated 562 563 monsoon run-off (= sapropel deposition), such feedbacks would not yet have had time to fully develop. Hence, the timing of sapropel onsets should be less biased by feedback 564 processes than the timing of sapropel mid-points. As noted above, in LC21 we observe 565 566 similar (relative) differences among sapropel-insolation phasings using the onset and midpoint criteria (Table 2), which suggests that our inferred phase relationships are robust. 567 568

Regardless of possible explanations for the above phase differences, our observations suggest that 1) the concept of a consistent 3-kyr lag between precession minima/insolation maxima and an interval of sapropel deposition is valid only within broad uncertainties of a few thousand years, and 2) insolation is not the sole driver of African monsoon dynamics, given that insolation–monsoon phasings do not seem to be consistent among the sapropels considered here.

- 575
- 576 5.2 Sea level–sapropel phasing
- 577

578 In core LC21, the deposition of sapropels S3, S4 and S5 began after sea level had risen to a

579 relative 'highstand', despite differences in the height of these highstands (Fig. 7b-d).

580 Conversely, sapropel S1 was deposited when sea level was still rising, and its onset occurred

581 ~4 kyr before a highstand (Fig. 7a). These observations suggest that the relative timing of the

582 Holocene sapropel (S1) was unique for the last glacial cycle. However, when considering

583 rates (dRSL) and amplitudes of sea-level change, rather than RSL itself, we see another

pattern, namely a clear discrepancy between sea level–sapropel phasings between early

585 interglacial and glacial-inception periods (Fig. 7). In the interglacial case (S1 and S5),

sapropel deposition began after sea-levels had risen ~90 m, and after a 50-75% reduction in

587 rates of sea-level rise from their peak rates (Fig. 7a,b). In the case of glacial inception times

588 (S3 and S4), the onset of sapropel deposition occurred after substantially smaller sea-level

rises (~15-35 m), and just after sea-level rise rates peaked (Fig. 7c,d).

590

591 These results imply that over the last glacial cycle, sapropel deposition was sensitive to sea-

level changes, in agreement with Grimm et al.'s (2015) model experiments for sapropel S1.

593 In those experiments, monsoon forcing alone could not explain S1 formation because the

594 development of deep-water anoxia took several millennia, i.e., longer than the timespan 595 between monsoon onset and S1 deposition. However, Grimm et al.'s (2015) simulations also 596 showed that monsoon forcing nonetheless contributed to S1 deposition because the additional 597 freshwater inputs led to strong changes in density gradients; these in turn affected the vertical 598 extent and duration of S1 anoxia. We observe close timing relationships for both sea-level 599 and monsoon changes among all sapropels considered here, which occurred under a wide 600 range of insolation and ice-volume forcings. We also observe that the relationships with sea-601 level change are not straightforward, and that different interpretations arise depending on 602 comparison with sea-level itself, or with rates of sea-level change (see below).

603

#### 604 5.3 Sapropel formation: sea-level versus monsoon control

605

606 Our data strongly suggest that the onset of sapropel deposition over the last glacial cycle was 607 sensitive to both sea-level change and monsoonal flooding of the eastern Mediterranean. 608 While increased freshwater input to the Mediterranean from the northern Mediterranean 609 Borderlands (NMB) has frequently been proposed to have also contributed to sapropel 610 formation (section 3.3.2), and cannot be entirely ruled out, the effect of this run-off on net 611 buoyancy forcing has not yet been adequately quantified. Given that most precipitation over 612 the wider eastern Mediterranean (including the NMB) is sourced from the basin itself, that 613 implies little or no net effect on surface buoyancy changes (see section 3.3.2). Hence, 614 monsoon- and deglaciation-related changes in basin hydrography remain the most plausible 615 mechanisms of surface buoyancy forcing at the time of sapropel deposition. 616 617 Given that our data show: *i*) synchronous onsets of sapropel deposition and monsoonal

618 flooding into the eastern Mediterranean, and *ii*) variable sea-level (ice-volume) histories

619 immediately prior to and at the onset of sapropel deposition among different sapropels,
620 despite broad similarities among these histories, the most straightforward interpretation
621 would be that the onset of sapropel deposition was closely related to monsoon forcing, and
622 not so systematically controlled by deglaciation effects.

623

624 Nonetheless, especially for sapropel S1, a strong sea-level (and warming) influence may still 625 exist (e.g., Grimm et al., 2015) because, out of the four sapropels considered here, S1 is the 626 only one that was deposited immediately after a substantial sea-level rise, and sea level 627 continued to rise throughout S1 deposition. A link between sapropel S1 formation and 628 deglaciation was first demonstrated quantitatively by Rohling (1994) and Béthoux and Pierre 629 (1999), who showed that the latter would lead to decreased surface- and intermediate-water 630 salinities, which in turn would inhibit deep-water ventilation. This is because rising sea levels 631 would have enhanced the exchange of Atlantic and Mediterranean waters at the Strait of 632 Gibraltar by increasing the cross-sectional area of the strait (e.g., Bryden and Kinder, 1991; 633 Rohling, 1999; Rogerson et al., 2012). The resultant decrease in residence time of 634 Mediterranean waters, combined with the reduced salinity of inflowing Atlantic water due to melting ice sheets, would cause progressive surface buoyancy gain in the Mediterranean 635 636 (Rohling et al., 2015). This relationship – between sea-level change and Mediterranean 637 salinity – is well-defined and non-linear on glacial-interglacial timeframes (Rohling and 638 Bryden, 1991) (as opposed to the modern stochastic relationship observed inter-annually by 639 Pinardi et al. (2014)).

640

A relatively weak insolation maximum during the Holocene provides further support for the
proposed role of sea-level rise in S1 deposition, because the monsoon maximum was
probably less intense (de Noblet et al., 1996) than for other sapropels (namely S5; Rohling et

644 al., 2004) and therefore may not have provided sufficient buoyancy forcing (via freshwater 645 run-off) for deep-water stagnation. This argument is somewhat supported by the LC21 646 residuals through S1, in that they are roughly half the magnitude of those for S5 (Fig. 3), 647 which was deposited during a relatively strong insolation maximum (Fig. 7b). Notwithstanding possible temperature effects on the LC21 residuals, our interpretation is 648 further supported by a detailed study of stable isotopes ( $\delta^{18}O$ ,  $\delta^{13}C$ ) in multiple for a miniferal 649 species through sapropels S1 and S5 in the eastern Mediterranean (Rohling et al., 2004). That 650 651 study concluded that freshwater dilution of surface waters was much reduced during S1 compared to S5. Similarly, the duration of S1 is distinctly shorter (4.4 kyr) than that of S5 652 653 (6.8 kyr) (Table 1), which may be consistent with weaker monsoon-forcing of S1 compared 654 to S5.

655

656 There is little differentiation among the LC21 residuals values for S1, S3 and S4 (Fig. 3b), or 657 among the corresponding insolation values (Fig. 7), implying that sapropels S3 and S4 may 658 have been comparable to S1 in terms of relatively weak monsoon forcing. However, the 659 durations of S3 and S4 exceed that of S1 (Table 1), and opposite sea-level trends are 660 observed for S3 to S5 relative to S1: sea level either declined (by ~20 m during S3 and S5) or hovered around a broad plateau (S4) (Fig. 7). Furthermore, sea-level histories are noticeably 661 662 different for the 5-6 kyr preceding S3 and S4 compared to S1 (this is the timeframe that 663 Grimm et al. (2015) suggest to be crucial for a deglaciation influence on sapropel deposition). Falling sea level would have reduced the cross-sectional area of the Strait of Gibraltar and 664 665 Strait of Sicily, leading to reduced Atlantic-Mediterranean exchange, longer Mediterranean 666 residence times, and increased Mediterranean surface-water salinity in the more restricted and 667 highly evaporative basin (e.g., Rohling 1999; Rohling et al., 2014). The resultant increase in 668 surface density would have promoted deep-water formation rather than stagnation. Although

sea level was still rising at the very onset of S5, similar to S1, S5 deposition began 3-4 kyr after the main, rapid sea-level rise associated with termination II. It is hard to reconcile such timings with timescales for deep-water stagnation based on deglaciation effects alone (see below). For S3 and S4, sapropel onset coincides with peak rates of sea-level rise (Fig. 7c,d), which may imply some degree of sea-level control on the timing of sapropel onset, in addition to monsoon forcing.

675

676 To help clarify the processes leading to sapropel deposition, we can also consider general trends in the LC21  $\delta^{13}$ C records, together with previously published records from LC21 677 678 (Marino et al., 2007; Abu-Zied et al., 2008; Grelaud et al., 2012) which have now been 679 converted to the radiometrically-based chronology used here (Fig. 8). We focus on sapropel 680 onset and the prior 5-6 kyr because, as suggested by Grimm et al. (2015) for sapropel S1, at least 5.5 or 6 kyr (based on modelling and benthic  $\delta^{13}$ C data, respectively) of progressive 681 682 deep-water stagnation and oxygen consumption was required to initiate S1 deposition and anoxia. There is no consistent trend among the LC21  $\delta^{13}$ C profiles in the 5-6 kyrs preceding 683 sapropel onset (Fig. 8), implying that the processes necessary for sapropel deposition 684 (reduced ventilation, increased biological production) developed differently for each 685 sapropel. The most notable disparity is between S1 and S3-S5. For S1, the  $\delta^{13}C_{rub-pac}$  gradient 686 is minimal for most of the pre-sapropel period, and both  $\delta^{13}C_{rub}$  and  $\delta^{13}C_{pac}$  start to decrease 687 ~2-3 kyr before the S1 onset. This decline is mirrored in  $\delta^{13}$ C values for G. inflata (Fig. 8), a 688 planktonic foraminifer with a similar apparent ecological niche to *N. pachyderma*. In 689 contrast, prior to S3-S5 there is a 1-2‰ gradient between  $\delta^{13}C_{pac}$  and  $\delta^{13}C_{rub}$ , and a negative 690 691 shift in both occurs much closer to sapropel onset, synchronous with a negative shift in the 692 LC21 residuals. For S3 and S4, these shifts also coincide with rising sea levels, so without 693 further proxy records it is difficult to conclusively distinguish between monsoon and

694 deglaciation effects. However, the fact that  $\delta^{13}$ C trends prior to S3 and S4 are arguably more 695 similar to those pre-S5 than pre-S1, hints at a common development mechanism for S3, S4, 696 and S5.

697

698 The most extensive datasets in LC21 are for S1 and S5 (Fig. 8). These two sapropels have 699 already been studied extensively elsewhere (see Rohling et al., 2015, and references therein). 700 However, the advantage of our study is that we can now unambiguously examine timing 701 relationships between sea-level rise, monsoon run-off into the eastern Mediterranean, and key 702 proxy records, on an independent and chronologically consistent age-scale (Fig. 8). 703 Regarding S1, a spike in benthic foraminiferal abundances ('benthics/g') coincides with the 704 YD (Fig. 8a); abundances then decline until the S1 onset. This decline coincides with rapidly rising sea levels, decreasing  $\delta^{13}$ C values, and a sharp increase in oxygen deficient species 705 706 (ODS) within the LC21 benthic foraminiferal assemblage (Abu-Zied et al., 2008; chronology 707 after Grant et al., 2012) (Fig. 8a). Importantly, the rise to peak monsoon run-off occurs after 708 the shift in benthic assemblages. This is compelling evidence for a key role of deglaciation in 709 instigating vertical stratification and deep-water anoxia prior to S1 formation. At the same 710 time, the synchronous onset of S1 and peak monsoon run-off into the eastern Mediterranean 711 is equally strong evidence that S1 deposition was tied to monsoon forcing.

712

In contrast, benthic fossils in core LC21 disappeared at the onset of (rather than prior to)
sapropel S5 (Marino et al., 2007). Similar observations for S5 have been made previously.
For example, Schmiedl et al. (2003) studied a southeast Aegean core from a similar water
depth to LC21, and observed relatively high benthic foraminiferal abundances right up until
S5 onset. They concluded that bottom waters were sufficiently ventilated until the start of S5
deposition, despite an inferred stepwise reduction in oxygen content over the preceding ~3

719	kyr. Capotondi et al (2006) also found a close agreement between the onset of benthic anoxia
720	and S5 deposition, and Jorissen (1999) noted differences in benthic foraminifera assemblages
721	between S1 and S5. In LC21, the onset of benthic azoic conditions during S5 coincides with
722	abrupt increases in organic carbon ( $C_{org}$ ) deposition, isorenieratene concentrations, and
723	abundances of Florisphaera profunda coccoliths (Grelaud et al., 2012), as well as with
724	negative peaks in the $\delta^{13}$ C and residuals records (Fig. 8d). The C <sub>org</sub> increase is unlikely to be
725	due solely to increased preservation, because the values are unusually high (up to 14%) for
726	Aegean sediments (cf typically 2-3%; Mercone et al., 2001; Thomson et al., 2004). Elevated
727	isorenieratene concentrations are indicative of anaerobic phototrophic Chlorobiaceae, while
728	increased abundances of F. profunda are associated with an increase in subsurface rather than
729	surface primary productivity. Together, these records point to the rapid development of a
730	deep chlorophyll maximum and euxinic conditions at the onset of S5 in LC21 (Marino et al.,
731	2007; Grelaud et al., 2012). Thus, for S5, there is a strong timing relationship between
732	substantial monsoon run-off into the eastern Mediterranean, benthic anoxia, increased
733	(subsurface) productivity, and increased $C_{org}$ deposition.
734	
735	5.4 African monsoon variability
736	
737	Our datasets, in line with previous studies based on less well constrained chronologies and/or
738	fewer sapropel events (Rohling et al., 2002, 2004; 2006; Emeis et al., 2003; Marino et al.,
739	2007; De Lange et al., 2008; Hennekam et al., 2014), strongly suggest that the timing of
740	monsoon run-off into the eastern Mediterranean was crucial in triggering surface buoyancy
741	changes necessary for sapropel deposition. Our results also show that the timing of this
742	monsoon run-off, relative to insolation changes, varied over the last glacial cycle (Fig. 7).
743	These timings were broadly similar for sapropels S1 and S5 (Fig. 7a,b) and for sapropels S3

and S4 (Fig. 7c,d). Comparable phasings were observed among sea level–sapropel
relationships, whereby longer lags between insolation maxima and sapropel mid-points were
observed for larger sea-level changes (Fig. 7; Table 2). These observations suggest that
global ice-volume changes exceeding a certain magnitude interfere with the precessionpacing of the African monsoon.

749

750 An empirical link between African monsoon precipitation and glaciation has been inferred 751 from model simulations. Freshwater hosing experiments for the North Atlantic suggest that the African monsoon weakened in response to (glacial) meltwater pulses (Chang et al., 2008; 752 753 Tjallingii et al., 2008; Kageyama et al., 2013), due to atmosphere-ocean feedbacks associated 754 with a reduced Atlantic meridional overturning circulation (AMOC) and a southward shift in 755 the intertropical convergence zone (ITCZ) (Chang et al., 2008). Transient simulations (Otto-756 Bliesner et al., 2014) and a model set-up that included remnant ice sheets in addition to a 757 North Atlantic freshwater influx (Lézine et al., 2011; Marzin et al., 2013) show similar results 758 to the previous hosing experiments. Notably, Lézine et al. (2011) and Marzin et al. (2013) 759 concluded that a freshwater flux into the North Atlantic would have had a larger impact on 760 African monsoon precipitation than the presence of remnant ice sheets alone. This is in line 761 with our observations: the magnitude of sea level rise (and therefore meltwater effects), rather 762 than sea level at sapropel onset, distinguish the more insolation-lagged sapropels S1 and S5 763 (Section 4.2; Fig. 7).

764

A link between past African monsoon variability and global ice-volume changes appears to
be supported by proxy data. For example, precession-scale variability in a West African
monsoon proxy record was interpreted in terms of large-scale changes in global ice volume
(Weldeab et al., 2007), while Heinrich Stadials (HS) have been invoked to explain millennial-

769 scale dry periods in North Africa (Tjallingi et al., 2008), a delayed response of the East 770 African monsoon to precession/insolation forcing (Ziegler et al., 2010a), as well as a 771 southward shift in the Sahara-Sahel boundary (Collins et al., 2013). In particular, Ziegler et 772 al. (2010a) proposed that African monsoon intensity (and by inference, sapropel formation) 773 varies in phase with northern hemisphere summer insolation but was suppressed during 774 Heinrich-type cold events, possibly associated with periods of meltwater release and sea-level 775 rise. This suppression led to an apparent lag of the African monsoon behind insolation 776 maxima. Ziegler et al. (2010a) proposed that this lag – while variable – averaged 2-3 kyr 777 between precession minima and African monsoon maxima/sapropel mid-points. Our results 778 for the Holocene and Last Interglacial support this hypothesis: S1and S5 (and coeval 779 increases in monsoon run-off) are preceded by North Atlantic cold events and meltwater 780 pulses (dRSL>0) (Fig. 9a,b), and the mid-points of these sapropels lag maximum insolation 781 by 2-3 kyr (Fig. 7a,b). However, the relationship between these phenomena is not consistent 782 for all sapropels/monsoon intervals. Meltwater pulses and North Atlantic cold events C21 and 783 C24 preceded intervals of monsoon run-off and sapropel deposition in the eastern 784 Mediterranean during MIS 5a (sapropel S3) and 5c (sapropel S4), and cold event C23 785 interrupted S4 deposition (Fig. 9c,d), yet here the mid-point of these sapropels (and of the 786 concurrent monsoon run-off intervals) is in direct phase with an insolation maximum. Hence, 787 cold events and meltwater pulses do not necessarily induce a precession-lag in African 788 monsoon /sapropel timing.

789

The maximum rates of sea-level rise prior to S3 are comparable to those for S5 (and for S1, if we consider the probabilistic dRSL records) (Fig. 9), implying that the rate of meltwater addition to the global ocean does not determine sapropel/monsoon lag. However, total sealevel change prior to S1 and S5 is tens of metres greater than for S3 and S4. If melting rates 794 are comparable, this suggests that it is the duration of the meltwater addition that disrupts the 795 precession-pacing of the African monsoon. Calculating from the start of the meltwater 796 addition ('maximum probability' dRSL>0) to the onset of monsoon run-off and sapropel 797 deposition, we find meltwater durations of 7 kyr (for S5), 2 kyr (for S4) and 1.6 kyr (for S3) 798 (Fig. 7). For S1, the Stanford et al. (2011) dRSL curve suggests that the main period of 799 meltwater addition to the global ocean began ca 16.5-17 ka BP (Fig. 7a). This is consistent 800 with a more recent estimate of 16.5 ka BP for the start of the main phase of deglaciation, 801 although the initial phase began earlier (~20 ka BP) (Lambeck et al., 2014). Hence, the 802 duration of meltwater addition prior to S1 was likely 6-9 kyr.

803

804 Modelling experiments suggest that the sensitivity of the AMOC and ITCZ (hence African 805 monsoon precipitation) to meltwater pulses is greatest under full-glacial conditions 806 (Ganopolski and Rahmstorf, 2001; Swingedouw et al., 2009; Zhang et al., 2015). Our African 807 monsoon run-off reconstructions cover both glacial-inception and early interglacial periods, 808 so it is unlikely that our observations primarily reflect climate-dependent sensitivities of the 809 African monsoon. This gives further weight to our preferred interpretation that the duration of 810 meltwater input to the global ocean was critical in determining the timing of the African 811 monsoon response to insolation forcing. Interestingly, the inclusion of variable northern 812 hemisphere ice sheets in transient model simulations of African monsoon variability (Ziegler 813 et al., 2010b; Weber and Tuenter, 2011) produced zero phase lag in the precession band, in 814 line with previous simulations based on stationary ice-sheets (Tuenter et al., 2005; Kutzbach 815 et al., 2008). While Weber and Tuenter (2011) acknowledged that the crude resolution of ice 816 sheets and ocean circulation, and lack of atmospheric mid-latitude dynamics in these transient 817 models may undermine their reliability, these simulations are nonetheless quasi-consistent 818 with our observations: we observe a tight phasing between sapropel mid-points (hence

819 African monsoon intervals) and insolation maxima (= precession minima) in MIS 5a and 5c 820 (Fig. 7c,d; Table 2). The fact that this observed phasing is not systematic among successive 821 insolation maxima, may be the reason for model-data offsets if only 'average' phasings are 822 considered. For example, lag correlation analyses (Caley et al., 2010) of a proxy record for 823 African monsoon run-off into the eastern Mediterranean (Revel et al., 2010) revealed an 824 average insolation-lag of 0.7 kyr for African monsoon maxima over the past 45 kyr, but a 825 longer lag when the Holocene monsoon maximum was considered alone. Thus, statistical 826 analyses may not capture the detail of monsoon-ice-volume phasings.

827

828 **6.** Conclusions

829

830 We have established independent (radiometric-based) datings for sapropels S1, S3, S4 and S5 831 in eastern Mediterranean core LC21, and show that, in detail, insolation-sapropel phasings 832 were not systematic through the last glacial cycle. This observation potentially has 833 implications for the application of an assumed 3-kyr lag between insolation 834 maxima/precession minima and sapropel mid-points, and hence for the astronomical 835 chronology of eastern Mediterranean sedimentary sequences. For instance, previous sapropel 836 mid-point ages established by a lagged orbital tuning may be up to 3 kyr too young, in cases 837 where the mid-points should actually be in direct phase (zero lag) with insolation maxima. 838 However, it is unlikely that sapropel ages established by that method are too old. We suggest 839 that persistent meltwater discharge into the North Atlantic over several kyrs, such as during 840 glacial terminations, modified the timing of sapropel deposition via a delay in the timing of 841 African monsoon run-off into the eastern Mediterranean. We show that the onset of this 842 monsoon run-off and sapropel deposition was near-synchronous (within 0.5 kyr) for all 843 sapropels considered here, and conclude that monsoon forcing was important in instigating

844 the formation of these sapropels; furthermore, for sapropels S3–S5, monsoon forcing was 845 probably more important than sea-level rise. However, it is likely that sea-level rise, and 846 attendant hydrographic changes, strongly contributed to sapropel S1 deposition. Thus, 847 deglaciation affected sapropel deposition directly (via changes in Mediterranean 848 hydrography) and indirectly (via the response of the African monsoon to meltwater pulses). 849 The fact that sapropel S1 was probably affected by both of the above mechanisms may 850 explain why it exhibits the longest insolation lag compared to S3 to S5. That in turn would 851 suggest that the 3-kyr lag assumption for sapropel tuning (which is based on S1) results in 852 overestimated insolation lags for many sapropels. In this context, our observations may 853 reconcile apparent model-data offsets with respect to the orbital pacing of the African 854 monsoon, by demonstrating that insolation-monsoon phasings varied with the magnitude of 855 meltwater pulses.

856

857 Finally, we conclude that deciphering the degree to which deep-water anoxia/sapropel 858 deposition can be attributed to either sea-level rise or African monsoon run-off remains 859 challenging, because both processes respond to boreal summer insolation, and both lead to a 860 loss of surface buoyancy in the eastern Mediterranean and an attendant reduction in deep-861 water ventilation. This buoyancy sensitivity likely dates back to when the Mediterranean 862 basin first became semi-enclosed from the open ocean during the Middle Miocene, which 863 coincides with the first appearance of sapropels at ~15.4 Ma (Taylforth et al., 2014). Detailed 864 process modelling of sapropels older than S1, and including the western Mediterranean basin, 865 will help to elucidate how changes in sea level, SST, and monsoon run-off each contributed to Mediterranean palaeohydrography and sapropel formation. 866

867

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875	
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#### Table 1

timing of the respo	ective monsoo	n signal.		
Age:	Sapropel		Sapropel	Monsoon
(ka BP)			mid-point	onset
S1	6.1-10.5	[4.4]	8.3	10.8
S3	80.8-85.8	[5.0]	83.3	85.8
S4	101.8-107.8	[6.0]	104.8	108.0
S4 interruption	104.0-105.4	[1.4]	104.9	
S5	121.5-128.3	[6.8]	124.9	128.3

Age boundaries, duration [kyr, in parentheses] and mid-point of core LC21 sapropels, and timing of the respective monsoon signal.

#### Table 2

Lag between sapropel mid-points/onset and maxima in June-July insolation at 65°N (INSOL<sub>MAX</sub>), maxima in the summer inter-tropical insolation gradient (SITIG<sub>MAX</sub>), and precession minima (P<sub>MIN</sub>). Negative values mean that sapropel mid-points/onset lead the equivalent insolation maxima/precession minima. Red text denotes phase offsets >2 kyr.

Phase:	Insolation:Sapropel <sub>MID</sub>			Insolat	Insolation:Sapropel <sub>ONSET</sub>		
(kyr)	(INSOL <sub>MAX</sub> )	(SITIG <sub>MAX</sub> )	(P <sub>MIN</sub> )	$(INSOL_{MAX})$	(SITIG <sub>MAX</sub> )	(P <sub>MIN</sub> )	
<b>S</b> 1	2.5	2.8	3.3	0.3	0.6	1.1	
<b>S</b> 3	0.1	0.8	-0.3	-2.4	-1.7	-2.8	
S4	-0.5	-0.2	0.7	-3.6	-3.2	-2.3	
S5	2.1	2.6	2.1	-1.3	-0.8	-1.3	

#### **Figure captions**

**Figure 1** Location of marine cores discussed in the study and schematic of surface circulation in the eastern Mediterranean Sea.

**Figure 2** Photograph of core LC21 (in metres below sea floor, mbsf) with scanning XRF records of barium and vanadium (grey, with 41-point moving averages in black). Sapropels are indicated (grey rectangles).

**Figure 3 a)** Planktonic foraminiferal (*G. ruber*)  $\delta^{18}$ O record from core LC21 (red), superimposed on a maximum probability sea-level record (Grant et al., 2012, 2014, based on the Red Sea sea-level reconstruction method of Siddall et al., 2003, 2004 and Rohling et al., 2009) which has been converted into equivalent Mediterranean  $\delta^{18}$ O values following Rohling et al. (2014) (darker blue shading = 68% confidence limits; lighter blue shading = 95% confidence limits). The RSL record is unreliable ca 14-23 ka BP so the probabilistic sealevel reconstruction of Stanford et al. (2011), (which is based on radiometrically dated sealevel indicators), has also been converted into equivalent Mediterranean  $\delta^{18}$ O values (black dashed lines =  $2\sigma$  error). **b**) LC21  $\delta^{18}$ O 'residuals' beyond the 68% (pale red) and 95% (dark red) confidence limits of the Red Sea sea-level equivalent  $\delta^{18}$ O variations, and beyond the 95% (black line) confidence limits of the Stanford et al. (2011) sea-level equivalent  $\delta^{18}$ O variations. A potential correlation between negative residuals and Heinrich Stadials (HS) 3 and 5 is based on the timing of Dansgaard-Oeschger stadials in Greenland ice (Rasmussen et al. 2014). Boreal summer insolation at 65°N (orange; after Laskar, 1990) is also shown. c) Maximum probability record of rates of sea-level change (dRSL) with its 95% probability interval (blue envelope; equivalent to  $2\sigma$ ) (Grant et al., 2012, 2014), based on the Red Sea sea-level reconstruction method (Siddall et al., 2003, 2004; Rohling et al., 2009). The record is unreliable ca 14-23 ka BP so meltwater pulse 1A (Deschamps et al., 2012) is also indicated (blue square with  $2\sigma$  error bars), as well as Stanford et al.'s (2011) record of sea-level change rates (grey shading = 95% probability interval). The timing of sapropels S1-S5 (grey rectangles), the Younger Dryas (Rasmussen et al., 2014) and Heinrich Stadials 1 (Álvares-Solas et al., 2011) and 11 (Marino et al., 2015) (green rectangles), and an inferred sharp increase in monsoon run-off into the eastern Mediterranean (yellow lines) are also indicated.

**Figure 4** Stable oxygen isotope records through sapropel S5 of **a**) *N. pachyderma* from core LC21 (red) and ODP Site 975 (black; Marino et al., 2015), on the radiometrically constrained chronology of Grant et al. (2012), and **b**) *N. pachyderma* (black; Marino et al., 2015), *Globigerinoides bulloides* (orange), and *G. ruber* (blue) (Kandiano et al., 2014) from ODP Site 975. The timing of Heinrich Stadial 11 (Marino et al., 2015) is also indicated (green rectangle).

**Figure 5** Ice-core methane (CH<sub>4</sub>) variations from Greenland (NGRIP; Baumgartner et al., 2014) (**a**, **c**, **d**) and from Antarctica (EDML; Schilt et al., 2010) on the AICC2012 chronology (Bazin et al., 2013) (**b**), compared with LC21 residuals beyond the 68% (orange) and 95% (red) confidence limits of sea-level equivalent  $\delta^{18}$ O variations. In (**a**), LC21 residuals are also calculated using the Stanford et al. (2011) sea-level reconstruction (open red circles) (see Fig. 3). The NGRIP temperature record is also in **a** and **d** (black; Kindler et al., 2014) and a U/Th-dated speleothem  $\delta^{18}$ O record (brown) from the northern European Alps (NALPS; Boch et al., 2011) is shown in **d**. Intervals shown are for full interglacial conditions (**a**, **b**) and periods of glacial inceptions (**c**, **d**). Previously published sea-surface temperature (SST) records from LC21 (blue) were derived from alkenones (**b**; Marino et al., 2007) and from planktonic foraminiferal data (**a**; Marino et al., 2009), and have been converted to the Grant et al. (2012) chronology used here.

**Figure 6 a**) Soreq cave speleothem  $\delta^{18}$ O (red; Grant et al., 2012) and LC21 *G. ruber*  $\delta^{18}$ O (blue). **b**) Soreq cave 'excess'  $\delta^{18}$ O ( $\delta^{18}$ O<sub>SOREQ XS</sub>) after removal of source-water  $\delta^{18}$ O signal (green; see section 3.3.2) for sapropels S1, S3, S4 and S5 (grey rectangles).

**Figure 7** Ice-volume and insolation changes spanning sapropel intervals under full interglacial conditions (**a**, **b**) and periods of glacial inceptions (**c**, **d**). Maximum probability curves of relative sea-level (RSL, blue) and its rates of change (dRSL, black) with their 95% probability intervals (paler envelopes; equivalent to  $2\sigma$ ) (Grant et al., 2012, 2014) are based on the Red Sea sea-level reconstruction method (Siddall et al., 2003, 2004; Rohling et al., 2009). The 95% confidence intervals for all RSL data (blue dashed lines) are also shown. The Red Sea RSL and dRSL data are unreliable ca 14-23 ka BP so in (**a**) we also show Stanford et al.'s (2011) 95% probability intervals of sea level and sea-level change rates (grey shading), as well as meltwater pulse 1A (Deschamps et al., 2012). Insolation curves are for boreal

summer at 65°N (red) and the summer inter-tropical insolation gradient (SITIG; green) (based on Laskar, 1990), with filled circles indicating maxima. Sapropels (grey rectangles) and an inferred sharp increase in monsoon run-off into the eastern Mediterranean (yellow lines) are also indicated.

**Figure 8** Planktonic foraminiferal stable carbon isotope records from core LC21, for the surface-dwelling *G. ruber* ( $\delta^{13}C_{ruber}$ ; green) and sub-surface dwelling *N. pachyderma* ( $\delta^{13}C_{pac}$ ; purple) and *G. inflata* (panel **a** only; pink = normal test, blue = smooth test). Red Sea relative sea-level record (RSL; blue) and the Stanford et al. (2011) sea-level record (grey shading) as in Fig. 7. LC21 residuals (red with orange shading) as in Fig. 5). The timing of sapropels S1-S5 (grey rectangles) and of cold episodes in the North Atlantic (the Younder Dryas, YD (Rasmussen et al., 2014), and Heinrich Stadials 1 (Álvares-Solas et al., 2011) and 11(Marino et al., 2015) (green rectangles) are indicated. Previously published data from LC21 are also shown in panels **a** and **d**, after conversion to the Grant et al. (2012) chronology: Total number of benthic foraminifera (Benthics/g; grey line with dots) and percent oxygen deficient species (ODS, grey shading), from Abu-Zied et al., (2008) (panel **a**); weight percent organic carbon (C<sub>org</sub>; brown shading), isorenieratene concentration (black line with dots), and percent *F. profunda* (pink), after Marino et al (2007) and Grelaud et al. (2008) (panel **d**).

**Figure 9** Timing of North Atlantic cold events, meltwater events and East African monsoon maxima under full interglacial conditions (**a**, **b**) and periods of glacial inceptions (**c**, **d**). Cooling in the North Atlantic region is indicated by the NGRIP ice-core temperature record (green; Kindler et al., 2014) (**a**,**c**,**d**) and by a maximum probability, alkenone-derived sea surface temperature (SST) record from ODP Site 976 (**b**) (grey; Martrat et al., 2014) on a radiometric-based chronology (Marino et al., 2015). A U/Th-dated speleothem  $\delta^{18}$ O record from the northern European Alps (NALPS; Boch et al., 2011), approximating Greenland temperature variations, is also shown (**c**,**d**). Cold events include the Younger Dryas (YD) (Rasmussen et al., 2014) and Heinrich Stadials 1 (Álvares-Solas et al., 2011) and 11 (Marino et al., 2015) (green rectangles), as well as events C21, C23, C24 (McManus et al., 1994). Meltwater events are inferred from rates of sea-level change (dRSL; blue) from Grant et al. (2012, 2014) (blue shading = 95% probability intervals of the maximum probability dRSL; line = maximum probability dRSL), and from Stanford et al. (2011) (grey shading in **a**; see Fig. 7). Also shown in (**a**) is MWP-1a at 46 m/kyr (Deschamps et al., 2012). LC21 residuals (red, orange) and sapropel intervals (grey rectangles) are the same as in Fig. 5.











Figure 4





Figure 6





Figure 7













## The timing of Mediterranean sapropel deposition relative to insolation, sea-level and African monsoon changes

K.M. Grant, R. Grimm, U. Mikolajewicz, G. Marino, M. Ziegler, E.J. Rohling

#### Highlights

- 1. Chronological framework for sapropels S1–S5, African monsoon, and sea-level change
- 2. Insolation-monsoon/sapropel phasings not systematic over the last glacial cycle
- 3. Meltwater release at glacial terminations delays monsoon and sapropel onset
- 4. Sea-level rise and monsoon run-off both important for sapropel formation

#### **Editorial comments**

You would need to consider more clearly the fact of deglacial freshwater input into the western Mediterranean from other sources, e.g., the Alpine region specifically.

#### Done – see our responses to reviewer #2.

On this note, Fig. 4 deals with your own data (ODP 975) only. However, MIS5e has been worked out in detail by Kandiano et al. 2014 (GPC).

We have amended Fig. 4 to include the results of Kandiano et al. (2014), and we now also discuss their results in comparison to ours over Heinrich Stadial 11 and termination 2 (lines 252-255).

#### **Reviewers' comments**

#### **Reviewer #2 (Remarks to the Author):**

# Figures. Figures presented by Grant and collaborators are very relevant. Nevertheless, Figure 1 have to be improved in order to facilitate the reading / understanding of the manuscript (and of the discussion in particular). I strongly invite the authors to add / draw on Figure 1: (i) the rivers (Nile, wadi-systems of North Africa, or Rhône, see below) that funnel freshwater into the eastern Mediterranean; (ii) the location of both marine (i.e., ODP 967, ODP 975, MD40/67) and continental (i.e., Soreq Cave) records mentioned in the main text; and (iii) the counter-clockwise surface flow pattern in the eastern Mediterranean discussed in part 3.2 of the manuscript.

#### Done.

# Meltwater effects. In part 3.2 of their manuscript, Grant et al. "consider whether glacial meltwater or monsoon runoff best explain the negative LC21  $\delta$ 180 residuals coincident with HS1 and HS11". This is an interesting / balanced discussion. Nevertheless, I'm puzzled with the idea that monsoon runoff could explain this signal during HS1/11 since it is well-known that monsoon runoff occurred at time of / or around precession minima...that means, for its last occurrence (ca. 12 kyr BP), about 6 kyr after the onset of HS1 (ca. 18 kyr BP). As a result, is it really useful to discuss this possibility (i.e, monsoon runoff during HS1) in detail?... to finally conclude that "all these observations are in line with the argument that negative spikes in the LC21 residuals during major deglaciation events are likely due to North Atlantic (meltwater-related) salinity anomalies propagating into the eastern Mediterranean". To my view, this part of the discussion should be shortened (without consequences on the main conclusion of the manuscript).

#### We agree with the reviewer that this discussion is unnecessary and have cut it from the text.

I would invite the authors (i) to consider (recent) results from the Laurentide and European ice-sheets, rather than results from numerical modeling (Alvarez-Solas et al., 2011; see line 230), for the timing / source of the "large melting events during terminations" (and during HS especially) discussed in lines 228-233;

This is an interesting discussion but beyond the scope of the present study. The timing and sources of meltwater pulses (MWPs) are still much debated, especially regarding the Laurentide Ice Sheet, and most observations are for the last deglaciation only. For instance, Stanford et al. (2011QSR) provide a comprehensive synthesis of the timing and sources of MWPs during the last deglaciation, and show that there was significant waxing and waning among all global ice sheets ca 17.5–15.5 ka BP. Furthermore, in our case it is not necessary to consider specific meltwater sources into the North Atlantic when we are only concerned with whether the associated light  $\delta^{18}$ O anomalies were propagated into the Mediterranean. (Note that we do now cite evidence from the European Ice Sheet, but in reference to point (iii) below).

(ii) to cite Sierro et al. (2005, Paleoceanography) (in addition to Rogerson et al., 2010, G3) when they discuss evidences for the flow of North Atlantic water into the Mediterranean during Heinrich Stadials (line 215-218);

#### Done. We've also added other citations to support this discussion (lines 216-229).

(iii) to consider a possible mediterranean origin for freshwater pulses at time of deglacial HS. Indeed, growing evidences suggest that the Rhône River (France) funnelled large volume of meltwater from the Alpine Ice-Sheet into the western Mediterranean during HS1 (e.g., Lombo-tombo et al., 2015, Sedimentary Geology). This source could contribute, in addition to the North Atlantic influence, to the decrease in the planktonic 🗉 180 observed in the western Mediterranean (ODP 975; Figure 4).

Done (lines 262 to 306). We have significantly expanded section 3.2 in order to consider these other potential sources (although we find, on balance, that the North Atlantic influence likely dominates the  $\delta^{18}$ O depletions in question).

#### # Others.

The authors discuss the evidences for humidity increases in winter during intervals of precession minima. Recent results published by Toucanne et al. (2015, Quaternary Science Reviews) and Bosmans et al. (2015, Quaternary Science Reviews) should be cited / added here. On this point, it is interesting to note that the  $\delta$ 18Osoreq xs support this hypothesis.

We agree with the reviewer that the Toucanne and Bosmans papers should be cited here (these studies had not been published at the time of our manuscript submission). In addition, in light of the continued debate regarding humidity increases during sapropel deposition/precession minima, we have expanded section 3.3.2 (see lines 356-411 and 436-449) so that it now includes a more lengthy discussion of the key evidence contributing to this debate, and how our  $\delta^{18}O_{SOREQ XS}$  record fits with this evidence.

Considering that this humidity is likely related to enhanced westerlies over the northern Mediterranean borderlands (see references above), it should be relevant to consider this forcing as well in the process leading to sapropel deposition.

At face value this appears to be a logical comment. However, it assumes *i*) that humidity did indeed increase during the sapropel intervals considered here, *ii*) that a humidity increase was related to enhanced westerlies, and – most importantly – *iii*) that an increase in humidity would cause sufficient surface buoyancy forcing to inhibit deep-water ventilation (and thus promote sapropel formation). These points are now covered in our expanded section 3.3.2. We find that *i*) on balance the evidence suggests that only for sapropel S5 is there strong, unambiguous evidence for increased humidity; *ii*) recent modelling disputes the 'enhanced westerlies' hypothesis, and *iii*) the effect of increased humidity on surface buoyancy forcing was likely secondary to monsoon- and sea-level driven surface-water freshening. Nonetheless, the effect of increased humidity on surface buoyancy forcing remains poorly constrained and so we cannot rule it out; we have amended the first paragraph of section 5.3 (lines 608-615) to reflect this.

- Lines 396-397: the authors write: "An alternative explanation is that negative peaks in LC21 residuals prior to S1 (ca 11.5-15.5 ka BP) may party relate to...". Considering that S1 is dated at 10.2-6.4 ka BP (e.g., Mercone et al., 2000, Paleoceanography), and that the negative peaks in LC21 prior to S1 occurred between 16-14 ka BP and from ca. 13 ka BP (Figure 5a), I do not understand the reference to "(ca 11.5-15.5 ka BP)" in line 397. Please correct (or explain).

We have re-written this sentence to avoid confusion (the reference "ca 11.5-15.5 ka BP' was not to S1 but to the pre-S1 interval of negative residuals). We now clarify the timing of negative residuals in line 508.

- The reference "(Taylforth et al., 2014)" (line 728) cannot be found in the 'Reference list'.

Done.

#### **Reviewer #3 (Remarks to the Author):**

My only concern is the following: authors make a linear relationship between meltwater and sea level rise in the Mediterranean Sea and then sea level rise and lower salinity waters. Recently (Pinardi et al., Journal of Climate, 2014, explained the precise relationship between Gibraltar inflow, salinity and mean sea level in the Mediterranean Sea. It is shown that this relationship is not simple but has also stochastic components due to the balance between the Mediterranean surface water budget and the Gibraltar inflow. Atlantic ice melting in the Mediterranean Sea would in fact enhance the stochasticity of the Gibraltar inflow contribution to mean sea level tendency in the Mediterranean Sea and this could account at least in part for the differences found between S3, S4 and sea level changes with respect to S1-sea level relationship. In addition changes in salinity induced by a larger Gibraltar inflow from the Atlantic could be different in the different cases. I am suggesting that the authors reflect about the possibility of also considering a complex relationship between meltwater events, Gibraltar inflow changes, sea level rise and freshening of the Mediterranean Sea.

The paper that the reviewer refers to considers present-day mean sea-level changes over a decade, where the maximum amplitude of sea-level variations is ~1 m. Hence, while he/she is correct that such variability has a strong steric component and is stochastic, on the timescales that we consider sea-level variability is tens of m and is mostly driven by glacioeustasy. Nonetheless, we now provide more detail about the relationship between changes in sea level and Mediterranean salinity, and explicitly state that this relationship in non-linear (lines 627-639).

Furthermore, the statement at line 545-546: "Falling sea level would have led to increasing Mediterranean surface-water salinity, due to inflow of more saline global ocean waters" is probably too simplistic. I recommend to weaken the statement.

We have re-written this statement more comprehensively (lines 664-667), so that the well-understood link between falling (global) sea level and an increase in Mediterranean salinity on glacial-interglacial timescales is explained in more detail and referenced (see also previous comment).

Line 40: please add reference to paper about all the sapropel deposition factors: D. Bianchi, M. Zavatarelli, N. Pinardi, R. Capozzi, L. Capotondi, C. Corselli and S. Masina, 2006. "Simulations of ecosystem response during the sapropel S1 deposition event." Palaeogeography, Palaeoclimatology, Palaeoecology, 235, pp. 265-287, doi:10.1016/j.palaeo.2005.09.032

Done.