

## **The C<sub>32</sub> alkane-1,15-diol as a proxy of late Quaternary riverine input in coastal margins**

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1 ABSTRACT

2 The study of past sedimentary records from coastal margins allows us to reconstruct variations  
3 of terrestrial input into the marine realm and to gain insight into continental climatic variability.  
4 There are numerous organic proxies for tracing terrestrial input into marine environments but  
5 none that strictly reflect the input of river-produced organic matter. Here, we test the fractional  
6 abundance of the C<sub>32</sub> alkane 1,15-diol relative to all 1,13- and 1,15-long-chain diols (F<sub>C<sub>32</sub> 1,15</sub>) as a  
7 tracer of input of river-produced organic matter in the marine realm in surface and Quaternary  
8 (0-45 ka) sediments on the shelf off the Zambezi and nearby smaller rivers in the Mozambique  
9 Channel (western Indian Ocean). A Quaternary (0-22 ka) sediment record off the Nile River  
10 mouth in the Eastern Mediterranean was also studied for long-chain diols. For the Mozambique  
11 Channel, surface sediments of sites most proximal to Mozambique Rivers showed the highest  
12 F<sub>1,15-C<sub>32</sub></sub> (up to 10%). The sedimentary record shows high (15-35%) pre-Holocene F<sub>1,15-C<sub>32</sub></sub> and low  
13 (<10%) Holocene F<sub>1,15-C<sub>32</sub></sub> values, with a major decrease between 18 and 12 ka. F<sub>1,15-C<sub>32</sub></sub> is  
14 significantly correlated (r<sup>2</sup>=0.83, p<0.001) with the BIT index, a proxy for the input of soil and  
15 river-produced organic matter in the marine environment, which declines from 0.25-0.60 for  
16 the pre-Holocene to <0.10 for the Holocene. This decrease of both F<sub>C<sub>32</sub> 1,15</sub> and the BIT is  
17 interpreted to be mainly due to rising sea level, which caused the Zambezi River mouth to  
18 become more distal to our study site, thereby decreasing riverine input at the core location.  
19 Some small discrepancies are observed between the records of the BIT index and F<sub>C<sub>32</sub> 1,15</sub> for  
20 Heinrich Event 1 (H1) and Younger Dryas (YD), which may be explained by a change in soil  
21 sources in the catchment area rather than a change in river influx. Like for the Mozambique  
22 Channel, a significant correlation between F<sub>C<sub>32</sub> 1,15</sub> and the BIT index (r<sup>2</sup>=0.38, p<0.001) is

23 observed for the Eastern Mediterranean Nile record. Here also, the BIT index and  $F_{C32\ 1,15}$  are  
24 lower in the Holocene than in the pre-Holocene, which is likely due to the sea level rise. In  
25 general, the differences between BIT index and  $F_{C32\ 1,15}$  Eastern Mediterranean Nile records can  
26 be explained by the fact that the BIT index is not only affected by riverine runoff but also by  
27 vegetation cover with increasing cover leading to lower soil erosion. Our results confirm that  
28  $F_{C32\ 1,15}$  is a complementary proxy for tracing riverine input of organic matter into marine shelf  
29 settings and, in comparison with other proxies, it seems not to be affected by soil and  
30 vegetation changes in the catchment area.

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32

### 33 **1. Introduction**

34 Freshwater discharge from river basins into the ocean has an important influence on the  
35 dynamics of many coastal regions. Terrestrial organic matter (OM) input by fluvial and aeolian  
36 transport represents a large source of OM to the ocean (Schlesinger and Melack, 1981). Deltaic  
37 and marine sediments close to the outflow of large rivers form a sink of terrestrial OM and  
38 integrate a history of river, catchment, and oceanic variability (Hedges et al, 1997).

39 Terrestrial OM can be differentiated from marine OM using carbon to nitrogen (C/N) ratios and  
40 the bulk carbon isotopic composition ( $^{13}\text{C}$ ) of sedimentary OM (e.g. Meyers, 1994). The  
41 abundance of N-free macromolecules such as lignin or cellulose result in organic carbon-rich  
42 plant tissues that lead to an overall higher C/N ratio for terrestrial OM compared to aquatic  
43 organisms (Hedges et al., 1986). However, this ratio may be biased when plant-tissues gain  
44 nitrogen during bacterial degradation and when planktonic OM preferentially lose nitrogen over  
45 carbon during decay (Hedges and Oades, 1997). Differences in the stable carbon isotopic  
46 composition may also be used to examine terrestrial input as terrestrial OM is typically depleted  
47 in  $^{13}\text{C}$  ( $\delta^{13}\text{C}$  of -28 to -25‰) compared to marine OM (-22 to -19‰). However,  $\text{C}_4$  plants have  
48  $\delta^{13}\text{C}$  values of around -12‰ (Fry et Sherr, 1984; Collister et al. 1994; Rommerskirchen et al.,  
49 2006) and thus a substantial  $\text{C}_4$  plant contribution can make it difficult to estimate the  
50 proportion of terrestrial to marine OM in certain settings (Goñi et al., 1997).

51 Biomarkers of terrestrial higher plants are also used to trace terrestrial OM input into marine  
52 sediments. For example, plant leaf waxes such as long-chain *n*-alkanes are transported and  
53 preserved in sediments (Eglinton and Eglinton 2008, and references cited therein) and can

54 provide information on catchment-integrated vegetation or precipitation changes (e.g. Ponton  
55 et al., 2014), while soil specific bacteriohopanepolyols (BHP) are biomarkers of soil bacteria and  
56 indicate changes in soil OM transport (Cooke et al., 2008). Similarly, branched glycerol dialkyl  
57 glycerol tetraethers (brGDGTs) are widespread and abundant in soils (Weijers et al., 2007, 2009)  
58 and can be used to trace soil OM input into marine settings via the branched and isoprenoid  
59 tetraether (BIT) Index (Hopmans et al., 2004). However, brGDGTs can also be produced in-situ in  
60 rivers (e.g. De Jonge et al., 2015) and thus the BIT index does not exclusively reflect soil OM  
61 input. Moreover, because the BIT index is the ratio of brGDGTs to crenarchaeol (an isoprenoidal  
62 GDGT predominantly produced by marine Thaumarchaeota; Sinninghe Damsté et al., 2002), the  
63 BIT index can also reflect changes in marine OM productivity instead of changes in terrestrial  
64 OM input in areas where primary productivity is highly variable, i.e. where the quantity of  
65 crenarchaeol is variable (Smith et al., 2012).

66 Although these terrestrial organic proxies are useful to trace soil, river or vegetation input into  
67 marine sediments, previously there were no organic geochemical proxies to specifically trace  
68 river-produced OM input. However, recently, the C<sub>32</sub> 1,15-diol, relative to all 1,13- and 1,15-  
69 long-chain diols (F<sub>C32 1,15</sub>), was proposed as a tracer for river-produced OM input (De Bar et al.,  
70 2016; Lattaud et al., 2017). Long-chain diols (LCD), such as the C<sub>32</sub> 1,15-diol, are molecules  
71 composed of a long alkyl chain ranging from 26 to 34 carbon atoms, an alcohol group at position  
72 C<sub>1</sub> and at a mid-chain position, mainly at positions 13, 14 and 15. They occur ubiquitously in  
73 marine environments (de Leeuw et al., 1979; Versteegh et al., 1997, 2000; Gogou and  
74 Stephanou, 2004; Rampen et al., 2012, 2014; Romero-Viana et al., 2012; Plancq et al., 2015;  
75 Zhang et al., 2011 and references therein), where the major diols are generally the C<sub>30</sub> 1,15-diol,

76 C<sub>28</sub> and C<sub>30</sub> 1,13-diols, and the C<sub>28</sub> and C<sub>30</sub> 1,14-diols. In marine environments the 1,14-diols are  
77 produced mainly by *Proboscia* diatoms (Sinninghe Damsté et al., 2003; Rampen et al., 2007) and  
78 the 1,13 and 1,15-diol are thought to be produced by eustigmatophyte algae (Volkman et al.,  
79 1999; Rampen et al., 2007, 2014b; Villanueva et al., 2014). Versteegh et al. (2000) showed that  
80 F<sub>C<sub>32</sub> 1,15</sub> was relatively higher closer to the mouth of the Congo River. Likewise, Rampen et al.  
81 (2012) observed that sediments from the estuarine Hudson Bay have a much higher F<sub>C<sub>32</sub> 1,15</sub> than  
82 open-marine sediments. More recent studies noted elevated amounts of F<sub>C<sub>32</sub> 1,15</sub> in coastal  
83 sediments, and even higher amounts in rivers indicating a continental source for this diol (De  
84 Bar et al., 2016; Lattaud et al., 2017). Since the C<sub>32</sub> 1,15-diol was not detected in soils distributed  
85 worldwide, production of this diol in rivers by freshwater eustigmatophytes is the most likely  
86 source of this compound which, therefore, can potentially be used as a proxy of river-produced  
87 OM input to marine settings.

88 Here we test the downcore application of this new proxy by analyzing F<sub>C<sub>32</sub> 1,15</sub> in a continental  
89 shelf record (0-45 ka) from the Mozambique Channel and a record (0-24 ka) from the Eastern  
90 Mediterranean Sea to reconstruct Holocene/Late Pleistocene changes in freshwater input of the  
91 Zambezi and Nile rivers, respectively. Analysis of surface sediments and comparison with  
92 previously published BIT index records (Castañeda et al., 2010; Kasper et al., 2015) allow us to  
93 assess the potential of the C<sub>32</sub> 1,15-diol as a tracer for riverine runoff, or more precisely, river-  
94 produced organic matter, in these coastal margins.

95

## 96 **2. Material and Methods**

### 97 *2.1. Study sites*

98 *2.1.1. Mozambique margin and Zambezi River*

99 The Mozambique Channel is located between the coasts of Mozambique and Madagascar  
100 between 11°S and 24°S and plays an important role in the global oceanic circulation by  
101 transporting warm Indian Ocean surface waters into the Atlantic Ocean. The Zambezi River is  
102 the largest river that delivers freshwater and suspended particulate matter to the Mozambique  
103 Channel (Walford et al., 2005). The Zambezi River has a drainage area of  $1.4 \times 10^6$  km<sup>2</sup> and an  
104 annual runoff between 50 and 220 km<sup>3</sup> (Fekete et al., 1999). It originates in northern Zambia,  
105 flows through eastern Angola and Mozambique to reach the Indian Ocean. The Zambezi delta  
106 starts at Mopeia (Ronco et al., 2006) and the Zambezi plume enters the Mozambique Channel  
107 and flows northwards along the coast (Nehama and Reason, 2014). The rainy season in the  
108 catchment is in austral summer when the Intertropical convergence zone (ITCZ) is at its  
109 southernmost position (Beilfuss and Santos, 2001; Gimeno et al., 2010; Nicholson et al., 2009).  
110 The seasonal variation of the Zambezi runoff varies between 7000 m<sup>3</sup>/s during the wet season  
111 to 2000 m<sup>3</sup>/s during the dry season (Beilfuss and Santos, 2001). A few smaller Mozambique  
112 rivers other than the Zambezi River flow into the Mozambique Channel (Figure 1): the Ligonha,  
113 Licungo, Pungwe and Revue in Mozambique (together with the Zambezi River, they are  
114 collectively called “the Mozambique rivers” here).

115 Past studies have shown that the deposition pattern of the Zambezi riverine detritus is variable  
116 with sea level, i.e. most of the time material was deposited downstream of the river mouth but  
117 during high sea level it was deposited northeast of the river mouth due to a shore current  
118 (Schulz et al., 2011). During the last glacial period the Zambezi riverine detritus followed a more  
119 channelized path (Schulz et al., 2011). Van der Lubbe et al. (2016) found that the relative

120 influence of the Zambezi river compared to more northern rivers in the Mozambique Channel  
121 varied during Heinrich event 1 (H1) and the Younger Dryas (YD). Schefuß et al. (2011) studied  
122 the  $\delta^{13}\text{C}$  and  $\delta\text{D}$  of *n*-alkanes, and the elemental composition (Fe content) of core GeoB9307-3,  
123 located close to the present day river mouth (Fig. 1), and reported higher precipitation and  
124 riverine terrestrial input in the Mozambique Channel during the Younger Dryas and H1. This is in  
125 agreement with more recent results from Just et al. (2014) on core GeoB9307-3 and Wang et al.  
126 (2013a) on core GIK16160-3, further away from the actual river mouth; both studies also  
127 showed an increased riverine terrestrial input during H1 and YD. To summarize, during H1 and  
128 the YD, the Zambezi catchment is characterized by higher precipitation and enhanced riverine  
129 runoff due to a southward shift of the Intertropical Convergence Zone (ITZC) resulting from  
130 Northern Hemisphere cold events, whereas during the Holocene drier conditions prevailed  
131 (Schefuß et al., 2011; Wang et al., 2013; van der Lubbe et al., 2014; Weldeab et al., 2014). The  
132 Last Glacial Maximum (LGM) in the Zambezi catchment is also recognized as an extremely wet  
133 period (Wang et al., 2013).

134

### 135 *2.1.2 Eastern Mediterranean Sea and Nile River*

136 The Eastern Mediterranean Sea is influenced by the input of the Nile River, which is the main  
137 riverine sediment supply with annual runoff of  $91 \text{ km}^3$  and a sediment load of about  $60 \times 10^9$   
138  $\text{kg}\cdot\text{yr}^{-1}$  (Foucault and Stanley, 1989; Weldeab et al., 2002). Offshore Israel, the Saharan aeolian  
139 sediment supply is very low (Weldeab et al., 2002). A strong north-eastern current distributes  
140 the Nile River sediment along the Israeli coast toward our study site. The Nile River consists of  
141 two main branches: the Blue Nile (sourced at Lake Tana, Ethiopia) and the White Nile (sourced



142 at Lake Victoria, Tanzania, Uganda). Precipitation in the Nile catchment fluctuates widely with  
143 latitude with the area north of 18°N dry most of the year and the wettest areas at the source of  
144 the Blue Nile and White Nile (Camberlin, 2009). This general distribution reflects the latitudinal  
145 movement of the ITCZ.

146 Castañeda et al. (2010) have shown that sea surface temperature (SST) (reconstructed with  
147 alkenones and TEX<sub>86</sub>) at the study site was following Northern Hemisphere climate variations  
148 with a cooling during the LGM, H1 and the YD and warming during the early part of the  
149 deposition of sapropel 1 (S1). Associated H1 and LGM cooling, extreme aridity in the Nile  
150 catchment is observed as inferred from the  $\delta D$  of leaf waxes. In contrast, during the early  
151 Holocene S1 deposition, a more humid climate and enhanced Nile River runoff prevailed  
152 (Castañeda et al., 2016). Neodymium ( $\epsilon_{Nd}$ ) and strontium ( $^{87}Sr/^{88}Sr$ ) isotopes (Castañeda et al.,  
153 2016; Box et al., 2011, respectively) show a relative increase in the contribution of Blue Nile  
154 inputs when the climate is arid (H1, LGM) and an increased contribution of the White Nile inputs  
155 when the climate is humid (S1). This change also affects the soil input into the Nile River, as  
156 inferred from the distribution of branched GDGTs, with a more arid climate reducing the  
157 vegetation in the Ethiopian highlands (source of the Blue Nile) and favoring soil erosion while  
158 during a more humid climate, vegetation increasing and soil erosion is less (Krom et al., 2002).  
159 To summarize, the climate of the Nile catchment area was colder and drier (Castañeda et al.,  
160 2010, 2016) during the YD, H1 and the LGM. The LGM and H1 were extremely arid events with  
161 the likely desiccation of the Nile water sources, i.e. Lake Tana and Lake Victoria (Castañeda et  
162 al., 2016). To the contrary, the time period during S1 sapropel deposition was warmer and

163 wetter resulting in an enhanced riverine runoff. The late Holocene is characterized by a  
164 decrease in precipitation (Blanchet et al., 2014).

## 165 *2.2. Sampling and processing of the sediments*

### 166 *2.2.1. Mozambique Channel sediments*

167 We analyzed 36 core-top sediments (from multi cores) along a transect from the Mozambique  
168 coast to Madagascar coast (LOCO transect, Fallet et al. 2012). The LOCO core-tops have been  
169 previously studied by XRF and grain-size analysis (van der Lubbe et al., 2014, 2016) as well as for  
170 inorganic ( $\delta^{18}\text{O}$ , Mg/Ca) and organic (TEX<sub>86</sub>, Uk'<sub>37</sub>) temperature proxies (Fallet et al., 2012). 25  
171 core-top sediments (from grabs, gravity or trigger-weight corers) retrieved during the R/V  
172 Valdivia's Expeditions VA02 (1971) and VA06 (1973) (hereafter called VA; Schulz et al., 2011),  
173 comprising a north-south transect paralleling the East African coast, and spanning from 21°S to  
174 15°N (Fig. 1a) were also analyzed. These surface sediments have been studied previously for  
175 element content (TOC, TON), isotopic content ( $\delta^{18}\text{O}$ ,  $\delta^{13}\text{C}$ ) as well as for mineral and fossil  
176 (foraminifera) content (Schulz et al., 2011). Piston core 64PE304-80 was obtained from 1329 m  
177 water depth during the INATEX cruise by the RV Pelagia in 2009 from a site (18°14.44'S,  
178 37°52.14'E) located on the Mozambique coastal margin, approximately 200 km north of the  
179 Zambezi delta (Fig. 1a). The age model of core 64PE304-80 is based on <sup>14</sup>C dating of planktonic  
180 foraminifera (see supplementary information, van der Lubbe, 2014; Kasper et al., 2015) and by  
181 correlation of log (Ti/Ca) data from XRF core scanning with those of nearby core GIK16160-3,  
182 which also has an age model based on <sup>14</sup>C dating of planktonic foraminifera (see van der Lubbe  
183 et al., 2014 for details).

184 The LOCO sediment core-tops were sliced into 0 - 0.25 and 0.25 - 0.5 cm slices and extracted as  
185 described by Fallet et al. (2012). Briefly, ultrasonic extraction was performed (x 4) with a solvent  
186 mixture of dichloromethane (DCM)/methanol (MeOH) (2 : 1 v/v). The total lipid extract (TLE)  
187 was then run through a Na<sub>2</sub>SiO<sub>4</sub> column to remove water. The 25 VA core-tops from the  
188 Valdivia's expedition were freeze dried on board and stored at 4 °C. They were extracted via  
189 Accelerator Solvent Extractor (ASE) using DCM: MeOH mixture 9:1 (v/v) and a pressure of 1000  
190 psi at 100 °C using three extraction cycles.

191 We analyzed sediments of core 64PE304-80 for diols using solvent extracts that were previously  
192 obtained for determination of the BIT index and δD values of alkenones (Kasper et al., 2015).  
193 Briefly, the core was sliced into 2 cm thick slices and the sediments were ASE extracted using  
194 the method described above.

195 For all Mozambique Channel sediments, the total lipid extract (TLEs) were separated through an  
196 alumina pipette column into three fractions: apolar (Hexane : DCM, 9:1 v/v), ketone (Hexane :  
197 DCM, 1:1 v/v) and polar (DCM : MeOH, 1:1 v/v). The polar fractions, containing the diols and  
198 GDGTs, were dissolved into a mixture of 99:1 (v/v) Hexane : Isopropanol and filtered through a  
199 0.45 µm PTFE filters.

### 200 *2.2.2. Eastern Mediterranean sediment core*

201 Gravity core GeoB 7702-3 was collected during the R/V Meteor cruise M52/2 in 2002 from the  
202 slope offshore Israel (31°91.1'N, 34°04.4'E) at 562 m water depth (Pätzold et al., 2003,  
203 Castañeda et al., 2010). The chronology of this sedimentary record is based on 15 planktonic  
204 foraminiferal <sup>14</sup>C AMS dates (for details see Supplementary Information, Castañeda et al., 2010).

205 The sediments have previously been analyzed for GDGTs, alkenones,  $\delta D$  and  $\delta^{13}C$  of leaf wax  
206 lipids, and bulk elemental composition (Castañeda et al., 2010, 2016). Sediments were sampled  
207 every 5 cm and are 1 cm thick, and previously extracted as described by Castañeda et al. (2010).  
208 Briefly, the freeze-dried sediment were ASE extracted and the TLEs were separated using an  
209 aluminum oxide column into 3 fractions as described above.

### 210 2.3. Analysis of long-chain diols

211 Diols were analyzed by silylation of the polar fraction with 10  $\mu L$  N,O-Bis(trimethylsilyl)-  
212 trifluoroacetamide (BSTFA) and 10  $\mu L$  pyridine, heated for 30 min at 60°C and adding 30  $\mu L$  of  
213 ethyl acetate. Diol analysis was performed using a gas chromatograph (Agilent 7990B GC)  
214 coupled to a mass spectrometer (Agilent 5977A MSD) (GC-MS) and equipped with a capillary  
215 silica column (25 m x 320  $\mu m$ ; 0.12  $\mu m$  film thickness). The oven temperature regime was as  
216 follows: held at 70 °C for 1 min, increased to 130 °C at 20 °C/min, increased to 320 °C at 4  
217 °C/min, held at 320 °C during 25 min. Flow was held constant at 2 mL/min. The MS source  
218 temperature was held at 250 °C and the MS quadrupole at 150 °C. The electron impact  
219 ionization energy of the source was 70 eV. The diols were quantified using selected ion  
220 monitoring (SIM) of ions m/z 299.4 (C<sub>28</sub> 1,14-diol), 313.4 (C<sub>28</sub> 1,13-diol, C<sub>30</sub> 1,15-diol), 327.4 (C<sub>30</sub>  
221 1,14-diol), and 341.4 (C<sub>32</sub> 1,15-diol) (Versteegh et al., 1997; Rampen et al., 2012).

222 The fractional abundance of the C<sub>32</sub> 1,15-diol is expressed as percentage of the total major diols  
223 as follows:

$$224 F_{C_{32} 1,15} = \frac{[C_{32} 1,15]}{[C_{28} 1,13] + [C_{30} 1,13] + [C_{30} 1,15] + [C_{32} 1,15]} \times 100 \quad (1)$$

225

## 226 2.4. Analysis of GDGTs

227 GDGTs in the polar fractions of the extracts of the VA and LOCO core-top sediments were  
228 analyzed on an Agilent 1100 series LC/MSD SL following the method described by Hopmans et  
229 al. (2016). The BIT index was calculated according to Hopmans et al. (2004). We calculated the  
230 #ring tetra as described by Sinninghe Damsté et al. (2016) and the CBT index and soil pH as  
231 described by Peterse et al. (2012):

$$232 \text{ \#ring tetra} = \frac{GDGT\ Ib + 2 \times GDGT\ Ic}{GDGT\ Ia + GDGT\ Ib + GDGT\ Ic} \quad (2)$$

$$233 CBT = \log\left(\frac{GDGT\ Ib + GDGT\ IIb}{GDGT\ Ia + GDGT\ IIa}\right) \quad (3)$$

$$234 pH = 7.9 - 1.97 \times CBT \quad (4)$$

235

## 236 3. Results

### 237 3.1. Surface sediments of the Mozambique Channel

238  $F_{C_{32}\ 1,15}$  in surface sediments across the Mozambique Channel varies from 2.3 to 12.5% (Fig. 1d,  
239 1f) with one of the highest value in front of the Zambezi River mouth (10%). The core-tops  
240 located in front of other minor northern rivers (Licungo, Ligonha Rivers) are also characterized  
241 by values of  $F_{1,15-C_{32}}$  (>7.5%) higher than those further away from the coast (< 5%). The major  
242 diol in all Mozambique surface sediments is the  $C_{30}\ 1,15$ -diol ( $57.5 \pm 9.9\%$ ) with lower amounts of  
243 the  $C_{30}\ 1,14$ -diol ( $21.1 \pm 6.0\%$ ) and  $C_{28}\ 1,14$ -diol ( $13.2 \pm 4.9\%$ ) (Fig. 1f).

244 The values for the BIT index in surface sediments across the Mozambique Channel vary from  
245 0.01 to 0.42 (Fig. 1c). BIT values are highest in the most northern region (0.4) and in front of  
246 river mouths (0.2-0.3) compared to values found close to the coast of Madagascar (<0.04).  
247 Following Sinninghe Damsté (2016), we calculated the #ring tetra (the relative abundance of  
248 cyclopentane rings in tetramethylated branched GDGTs) to determine if the brGDGTs are in-situ  
249 produced in the surface sediments or derived from the continent. The #ring tetra has an  
250 average of  $0.39 \pm 0.03$  with higher values in front of the river mouths (with the highest values  
251 close to the Madagascar Rivers) and shows a clear decrease towards the open ocean (Fig. 1d).  
252 The low #ring tetra indicate that there is likely limited in-situ sedimentary production of  
253 brGDGTs in the sediments of the Mozambique coastal shelf area except for the samples closest  
254 to the Madagascar coast where high #ring tetra values and low BIT values indicate in-situ  
255 production of brGDGTs. However, for the Mozambique shelf, the brGDGTs are mostly derived  
256 from the continent, confirming the use of the BIT index as a tracer for riverine input in this  
257 region.

### 258 *3.2. Holocene and Late Quaternary sediments of the Mozambique Channel and Nile River*

259 In the sediments of the Mozambique Channel core 64PE304-80,  $F_{C32\ 1,15}$  shows a wide range; it  
260 varies from 2.4 to 47.6% (Fig. 2). Between 44 and 39 ka the values are relatively stable (average  
261 of  $27.6 \pm 4.5\%$ ), then they rapidly decline between 39 and 36 ka to 11%. From this point on they  
262 gradually increase, reaching 37.4% at 17 ka.  $F_{C32\ 1,15}$  is then rapidly decreasing until it reaches the  
263 lowest values of the record after 12 ka (average of  $4.9 \pm 1.4\%$ ). Holocene sediments (0-11 ka)  
264 show relatively low and constant values of  $F_{C32\ 1,15}$  ( $5 \pm 1.5\%$ ), similar to the values found in the  
265 surface sediments of the area, i.e.  $3.5 \pm 1.6\%$  (Figs. 1 and 2d).

266 The BIT index record (data from Kasper et al., 2015) shows similar changes as that of  $F_{C32\ 1,15}$ .  
267 Between 44 and 39 ka the average BIT value is  $0.43\pm 0.06$ , then the BIT value decreases to 0.36  
268 at 36 ka, followed by an increase until 17 ka to reach a value of 0.6, while the Holocene values  
269 are constant and average at  $0.1\pm 0.02$ . The #ring tetra of branched GDGTs is constantly low  
270 (average  $0.15\pm 0.01$ ; Fig. S1a) between 44 to 15.5 ka, then increases to 0.4 at 8 ka and stays  
271 constant until the end of the Holocene (average  $0.34\pm 0.03$ ). Overall, these values are low and  
272 do not approach the values (0.8-1.0) associated with in-situ production of branched GDGTs in  
273 coastal marine sediments (Sinninghe Damsté, 2016). The #ring tetra also shows a negative  
274 correlation with the BIT index throughout the record ( $R^2=0.74$ ,  $p<0.05$ ), indicating that when BIT  
275 values are high, #ring tetra is low. Therefore, high BIT values can definitely be associated with  
276 terrestrial brGDGT input. If we assume that the in-situ production of brGDGTs in the river (e.g.  
277 De Jonge et al., 2015; Zell et al., 2015) is minimal, we can then infer sources of soils from the  
278 different catchment areas by reconstructing the soil pH via the CBT index (see equation 3 and 4,  
279 Peterse et al., 2012). This showed a constant soil pH (average  $6.2\pm 0.1$ ) from 43 to 15 ka followed  
280 by a slight increase to 7 at 8 ka and constant (average  $6.8\pm 0.08$ ) at the end of Holocene (Fig.  
281 S1b).

282 In Eastern Mediterranean sediment core GeoB 7702-3,  $F_{C32\ 1,15}$  ranges from 3.9 to 47.0%.  
283 Between 24 and 15 ka the values are slowly decreasing from 41% at 24 ka to 7% at 15 ka.  
284 Subsequently,  $F_{C32\ 1,15}$  raises sharply until 11.7 ka (44%) followed by a sharp decrease down to  
285 16% at 10 ka.  $F_{C32\ 1,15}$  increases again until 7.5 ka up to 30%, followed by a slow decrease in the  
286 Late Holocene towards values as low as 6% (Fig. 3a). The BIT index (data from Castañeda et al.,  
287 2016) varies in a similar way as  $F_{C32\ 1,15}$ . It is constant between 24 and 17 ka (average  $0.37\pm 0.05$ ),

288 then decreases to 0.13 at 14.5 ka. It subsequently increases between 15.6 and 9 ka, before it  
289 decreases after 9 ka and stays constant in the Holocene (average  $0.17 \pm 0.05$ ). The #ring tetra of  
290 the brGDGTs (Fig. S1c) is constant from 24 to 15 ka ( $0.37 \pm 0.05$ ) then shows lower values from 15  
291 to 7 ka ( $0.29 \pm 0.04$ ) and, finally, increases again during the late Holocene ( $0.40 \pm 0.05$ ). The BIT  
292 index and #ring tetra do not show a clear negative correlation as observed for the Mozambique  
293 core. However, the values of #ring tetra values are well below 0.8-1.0, suggesting that in-situ  
294 production of brGDGTs does not play an important role, in line with the depth from which the  
295 core was obtained which is well below the zone of 100-300 m where in-situ production is most  
296 pronounced (Sinninghe Damsté, 2016). During parts of the record, low #ring tetra are associated  
297 with high BIT values, indicating that between 24 and 7 ka the brGDGT are mainly terrigenous.  
298 For the oldest part of the core, the soil pH shows a stable period from 24 to 14.8 ka (average  
299  $6.94 \pm 0.07$ ) then increases to 7.3 at 15 ka, followed by a large decrease (pH reaching 6.5 at 8.5  
300 ka). As the in-situ production of brGDGT is likely to be minimal in the latest part of the  
301 Holocene, and assuming that riverine production of brGDGTs is minimal, the soil pH can be  
302 reconstructed via the CBT index and shows a stable pH (average of  $6.8 \pm 0.1$ ).

#### 303 **4. Discussion**

##### 304 *4.1. Application of $C_{32}$ 1,15-diol as a proxy for riverine input in the Mozambique shelf.*

305 In the surface sediments of the Mozambique Channel  $F_{C_{32} 1,15}$  is relatively low overall (<10%) in  
306 comparison with other coastal regions with substantial river input (Fig. 1f), where values can be  
307 as high as 65% (De Bar et al., 2016; Lattaud et al., 2017). Moreover, the BIT values are also  
308 relatively low at 0.01-0.42. Further confirmation of the low amount of terrestrial input in the



309 analyzed surface sediments comes from the low C/N values (between 4.2 and 8.9 for the VA  
310 surface sediments; Schulz et al., 2011), characteristic of low terrestrial OM input (Meyers 1994).  
311 Nevertheless, the slightly higher values of both the BIT index and the  $F_{C32\ 1,15}$  near the river  
312 mouths indicate that both proxies do seem to trace present day riverine input into the  
313 Mozambique Channel in line with earlier findings of other coastal margins influenced by river  
314 systems (De Bar et al, 2016; Lattaud et al., 2017).

315

#### 316 *4.2. Past variations in riverine input in the Mozambique Channel*

317 We compared the record of  $F_{C32\ 1,15}$  with previously published proxy records, in particular the BIT  
318 index (Kasper et al., 2015) and  $\log(Ca/Ti)$  (van der Lubbe et al., 2016). These two proxies show  
319 the same pattern as  $F_{C32\ 1,15}$  (Fig. 2). Indeed, the BIT index and  $F_{C32\ 1,15}$  are strongly correlated ( $r^2$   
320 =0.83,  $p<0.001$ ). Since the #ring tetra of brGDGTs varies between 0.06 and 0.4 (Supplementary  
321 fig. S1a), and is significantly negatively correlated with the BIT values, the brGDGTs are  
322 predominantly derived from the continent (cf. Sinninghe Damsté, 2016) and thus the BIT is likely  
323 reflecting riverine input in the marine environment. Furthermore,  $F_{C32\ 1,15}$  also shows a  
324 significant negative correlation with  $\log(Ca/Ti)$  ( $r^2=0.43$ ,  $p<0.0001$ , van der Lubbe et al., 2016).  
325 This is another proxy for riverine input since Ti is mainly derived from erosion of continental  
326 rocks transported to the ocean through rivers, whereas Ca derives predominantly from the  
327 marine environment.

328 The records of  $F_{C32\ 1,15}$  and BIT index show three major variations: a steep drop from 19 to 10 ka,  
329 a slow increase from 38 to 21 ka during the Last Glacial and a steep decrease between 40 to 38

330 ka. The largest change in the BIT index and  $F_{C_{32} 1,15}$  is between 19 to 10 ka, i.e. a major drop  
331 which coincides with an interval of rapid sea level rise (Fig. 2b). Following Menot et al. (2006),  
332 we explain the drop in the BIT index, and consequently also the drop in  $F_{C_{32} 1,15}$ , by the  
333 significant sea level rise occurring during this period. Rising sea level flooded the Mozambique  
334 plateau, moving the river mouth further away from the core site and establishing more open-  
335 marine conditions. This most likely resulted in lower  $F_{C_{32} 1,15}$  and BIT values, conditions that  
336 remained throughout the Holocene. The decrease in the delivery of terrestrial matter is also  
337 seen in element ratios (Fe/Ca) and organic proxies (BIT) in nearby core Geob9307-3 (Schefuß et  
338 al, 2011), which is located closer to the present day river mouth in the Mozambique plateau  
339 (Fig. 1a). Likewise, the gradual increase in the BIT index and  $F_{C_{32} 1,15}$  between 38 and 21 ka  
340 occurred at a time when sea-level was decreasing (Fig 2b., Grant et al., 2014; Rohling et al.,  
341 2014) and thus the river mouth came closer to our study site. The decrease of BIT values and  
342  $F_{C_{32} 1,15}$  during 40-38 ka coincides with Heinrich event 4 (H4), a cold and dry event in this part of  
343 Africa (Partridge et al., 1997; Tierney et al., 2008; Thomas et al., 2009), with dry conditions likely  
344 leading to a reduced riverine input into the ocean and thus a reduced input of brGDGTs and the  
345  $C_{32} 1,15$ -diol.

346 Interestingly, there are two periods where BIT and  $F_{C_{32} 1,15}$  records diverge (Fig. 2a): during the  
347 Younger Dryas (YD; 12.7-11.6 ka) and Heinrich event 1 (H1; 17-14.6 ka) with the BIT index  
348 decreasing ca. 1 ky later than  $F_{C_{32} 1,15}$ . Comparison with the Ca/Ti ratio shows that both during  
349 H1 and the YD, the Ca/Ti ratio increased at the same time as the  $C_{32} 1,15$ -diol but earlier than  
350 the BIT index, suggesting that the latter was influenced by other parameters. The BIT index is  
351 the ratio of brGDGTs (produced mostly in soil or in-situ in rivers in this area based on the low

352 values for #ring tetra; Sinninghe Damsté, 2016) over crenarchaeol (produced mainly in marine  
353 environment; Schouten et al., 2013 and references cited therein). As both the Ti/Ca ratio and  
354  $F_{C32\ 1,15}$  indicate a decrease in riverine input, a constant BIT index can be explained by two  
355 options: a simultaneous decrease in crenarchaeol (marine) production or a change in soil input  
356 with higher brGDGT concentrations eroding into the river. The concentration of crenarchaeol  
357 during H1 is relatively stable but there is a slight decrease of crenarchaeol during YD (Fig. S2b).  
358 Thus, the difference between BIT and  $F_{C32\ 1,15}$  during YD can be partly explained by decreased  
359 crenarchaeol production together with a decrease in branched GDGTs due to a reduced river  
360 flow leading to relatively stable BIT values. In contrast, crenarchaeol and brGDGT concentrations  
361 are relatively stable during H1 and thus the lower river input, as indicated by the Ca/Ti and  $F_{C32\ 1,15}$ ,  
362 apparently did not lead to a decrease in brGDGT input. This could be due to a shift of  
363 sources of soil which are eroded in the river, i.e. if in this period there is a shift towards soils  
364 with relatively higher brGDGT concentrations, the BIT index would remain high despite  
365 decreased river flow.

366 A shift in soil sources may be due to two major changes that happened during this period (and  
367 also during the YD), i.e. a shift in catchment area of the Zambezi River (Schefuß et al., 2011; Just  
368 et al., 2014) and a shift in the relative influence of the Zambezi River versus northern  
369 Mozambique rivers (van der Lubbe et al., 2016). The shift in catchment area is evident from the  
370 higher influx of kaolinite-poor soil into the marine system during H1 and YD (Just et al., 2014)  
371 coming from the Cover Sands of the coastal Mozambique area (Fig. 3d, blue circle), relative to  
372 the kaolinite-rich soils of the hinterlands (Fig. 3d, red circles). If the brGDGT concentrations from  
373 the latter region are higher, then this change of soil input could lead to a stable brGDGT flux into

374 the marine environment, despite decreasing Zambezi River runoff. Support for a shift in soil  
375 sources comes from the soil pH record reconstructed from brGDGTs, which during the YD shows  
376 a shift towards more acidic soils. However, no change in soil pH are observed during H1.

377 The relative influence of other rivers (Lurio, Rovuma Rivers) relative to the Zambezi River (Fig.  
378 3d green circle) was inferred from neodymium isotopes by Van der Lubbe et al. (2016), i.e. more  
379 radiogenic rocks are found in the northern river catchments in comparison to the rocks in the  
380 Zambezi catchment (Fig. 2b). These authors found that during H1 and YD, the relative  
381 contribution of the northern rivers is lower than normal, likely due to drought conditions north  
382 of the Zambezi catchment area (Tierney et al., 2008, 2011; Just et al., 2014). These northern  
383 rivers run through a catchment containing mainly humid highstand soils, which are different soil  
384 types than observed in the catchment area of the Zambezi River (van der Lubbe et al., 2016).

385 We hypothesize that higher brGDGT concentrations in the soils of the catchment areas of the  
386 Zambezi River can potentially explain the discrepancy between BIT and  $F_{C32\ 1,15}$ , i.e. during H1  
387 and YD there is more input of brGDGT-rich soils from the Zambezi than brGDGT-poor soils from  
388 the northern rivers leading to constant BIT values despite a dropping riverine input. Further  
389 research examining the brGDGT contents of soils in the different river catchment areas as well  
390 as surface sediments from offshore these northern rivers is required to distinguish between the  
391 different hypotheses.

#### 392 *4.3. Past variations in riverine input in the Eastern Mediterranean Sea*

393 With the Eastern Mediterranean Sea core, we compared  $F_{C32\ 1,15}$  in core GeoB7702 with other  
394 proxies including the BIT index,  $\log(Ca/Ti)$  and strontium isotopes, the latter to infer the relative

395 importance of the Blue Nile and the White Nile as source regions (Fig. 4c-e). The BIT values (data  
396 from Castañeda et al., 2010) show a significant positive correlation with  $F_{C_{32} 1,15}$  ( $r^2=0.38$ ,  $p <$   
397  $0.05$ ), while  $\log (Ca/Ti)$  shows an negative correlation to  $F_{C_{32} 1,15}$ , again supporting a continental  
398 origin of the  $C_{32} 1,15$ -diol.  $F_{C_{32} 1,15}$  and BIT records show much lower Holocene values ( $12\pm 6\%$ )  
399 compared to pre-Holocene ( $27\pm 11\%$ ), which again can be attributed to the sea level rise  
400 occurring during the last deglaciation, i.e. our study site was further away from the river mouth  
401 and the amount of continental-derived OM reaching the site decreased. Both records show low  
402 values during H1 comparable to the Holocene. These low values can be attributed to extreme  
403 aridity in the Nile River catchment (Castañeda et al., 2016), which we hypothesize led to a lack  
404 of vegetation and enhanced soil erosion but also leading to a severely reduced river flow,  
405 thereby decreasing the net amount of river borne OM reaching our core site.

406 In this core, there are 3 major discrepancies observed between the BIT index and  $F_{C_{32} 1,15}$ : (1)  
407 during the LGM, between 22-19 ka, where the  $C_{32} 1,15$ -diol shows a decrease while the BIT  
408 index remains constant, (2) during the onset of the deposition of S1 (6.1-10.5 ka, Grant et al.,  
409 2016) where the BIT index decreases later than the  $C_{32} 1,15$ -diol, and (3) after 2 ka when the BIT  
410 index increases while the  $C_{32} 1,15$ -diol decreases. For the LGM  $F_{C_{32} 1,15}$  is decreasing,  $\log (Ca/Ti)$   
411 is as well, but the BIT index remains constant (and the brGDGT concentration is also low; see  
412 supplementary figure S3) indicating that there is no significant decrease in terrigenous OM  
413 reaching the core site at that time. During the LGM, there is no significant change in continental  
414 climate, based on the findings of Castañeda et al. (2016), suggesting no change in vegetation  
415 cover or river flux. This suggests that the change in  $F_{C_{32} 1,15}$  is not due to a change in the input of  
416  $C_{32} 1,15$ -diol but in other, mainly marine derived, diols, in particular the  $C_{30} 1,15$ -diol. If this

417 hypothesis is true then an increase in this marine diol will lower the  $F_{C_{32} 1,15}$  but if the amount of  
418 crenarchaeol is not changing at the same time, the BIT values will remain unaffected.

419 The deposition of S1 is described as a period of increased freshwater input leading to  
420 stratification and anoxia (Rossignol-Strick et al., 1982). However, an increased river input is  
421 neither reflected in  $F_{C_{32} 1,15}$  nor in the BIT index, in fact both of them are asynchronously  
422 decreasing. Castañeda et al. (2010) showed that the decrease in the BIT index is due to a large  
423 increase in crenarchaeol (Supplementary figure S3), much larger than the increase in brGDGTs,  
424 due to increased productivity and preservation. A similar scenario may apply for the diols, i.e.  
425 the marine diols (in particular the  $C_{30} 1,15$ -diol, data not shown) are also increasing at that time  
426 more substantially than the  $C_{32} 1,15$ -diol, thus lowering  $F_{C_{32} 1,15}$ . However, there is a difference  
427 in timing, i.e. the BIT index decreases slightly later than the  $C_{32} 1,15$ -diol (9.1 and 10.5 ka,  
428 respectively). The decrease in  $F_{C_{32} 1,15}$  coincides with a substantial increase in sea level (Fig. 4b),  
429 which would cause the distance between the core site and the river mouth to increase, thereby  
430 decreasing the amount of terrigenous material reaching the site. This terrigenous decrease is  
431 also visible to some extent in the  $\log(Ca/Ti)$  but not in the BIT index. Possibly, like with the  
432 Mozambique Channel, the brGDGT concentrations in the river were much higher at that time.  
433 Indeed, the Sr isotopic record suggest a major shift from a Blue Nile to a White Nile source at  
434 10.5 ka, with the latter possibly containing more eroded soils with high brGDGT concentrations.  
435 This shift in soil sources is also shown in the change towards more acidic soil pH during that  
436 period based on the CBT index (Supplementary figure S1d).

437 For the most recent part of the record (0-5 ka), the BIT index increases, while  $F_{C_{32} 1,15}$  is slightly  
438 decreasing. The  $\delta D_{leaf waxes}$  (Fig 4.d) shows it was period of mild aridity which likely led to a

439 decreased riverine runoff and thus decreased river input. The reason the BIT index is increasing  
440 rather than decreasing is due to an increase in brGDGT concentrations (Fig. 3b), despite  
441 evidence for a decrease in river runoff. This can possibly be linked to the amount of vegetation  
442 in the Nile catchment, i.e. at that time there was a decrease in vegetation cover (Blanchet et al.,  
443 2014; Castañeda et al., 2016), which led to more soil erosion and thus potentially a higher  
444 brGDGT concentration in rivers and a higher BIT index. This hypothesis is supported by the log  
445 (Ca/Ti) (Fig. 4c), which is decreasing at this time, suggesting that soil runoff was increasing.

446 Our results from both the Nile and Mozambique Channel cores illustrate that  $F_{C_{32} 1,15}$  provides a  
447 suitable proxy for reconstructing past riverine input into coastal seas. Although some  
448 discrepancies are noted with other terrigenous proxies for both cores,  $F_{C_{32} 1,15}$  generally agrees  
449 well with these proxies. However, our interpretation of the  $C_{32} 1,15$ -diol record relies on the  
450 assumption that production of this diol in rivers is not changing with different hydroclimate  
451 fluctuations on land, something that needs to be tested. However, De Bar et al. (2016) showed  
452 that  $F_{C_{32} 1,15}$  in the Tagus River in Portugal did not significantly change over the course of a year,  
453 suggesting that this assumption might be valid. Since the  $C_{32} 1,15$ -diol is mainly produced in  
454 rivers itself, it is not impacted by vegetation abundance and soil composition, in contrast to  
455 other proxies like the BIT index and lignin concentrations. This may make it a potentially more  
456 reliable proxy to trace past river input into marine environments.

## 457 **6. Conclusion**

458 We studied core-tops in the Mozambique Channel and two sediment cores, in the Mozambique  
459 Channel, off the Zambezi River mouth and in the Eastern Mediterranean Sea, offshore the Nile

460 delta, to test  $F_{C_{32} 1,15}$  as a proxy for riverine input into the marine realm. The surface sediments  
461 show that the  $C_{32} 1,15$ -diol traces present day riverine input into the Mozambique Channel,  
462 supported by the BIT index. In both sediment records,  $F_{C_{32} 1,15}$  is significantly correlated with the  
463 BIT index showing the applicability of this proxy to trace riverine input, but also showed some  
464 discrepancies. This can be explained by the different sources of these proxies, i.e. the BIT index  
465 is reflecting soil and river-produced OM input and the  $C_{32} 1,15$ -diol is mainly reflecting river-  
466 produced OM input. Our multiproxy approach suggests that the timing of changes in the  
467 different terrestrial proxies records can differ due to changes in catchment area or to shifting  
468 importance of the different source rivers.

#### 469 **Author contribution**

470 S. S. and J. L. designed the study. J. Lattaud analyzed the surface sediments for diols and GDGTs and core  
471 GeoB 7702-3 for diols, I. C. sampled and extracted the surface sediments and the sediment cores  
472 64PE304-80 and GeoB 7702-3, D. D. analyzed the sediment core 64PE304-80 for diols. H. S. collected the  
473 VA core-tops, E.S. collected core GeoB7702-3. J. L., S. S., I. C. and J.S.S.D. interpreted the data. J. L. wrote  
474 the manuscript with input of all authors.

475 The authors declare that they have no conflict of interest.

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486

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706 **Figure Legend**

707 **Figure 1.** Map presenting (a) the location of the core-tops (LOCO transect in orange, VA core-  
708 tops in blue) and cores (stars), (b) the mean annual salinity, from NOAA 1x1° grid  
709 (<http://iridl.ldeo.columbia.edu>), (c) the BIT index (LOCO transect values, VA core-tops from this  
710 study), (d)  $F_{C_{32} 1,15}$  in the core-tops, (e) #ring tetra of the surface sediments (#ring tetra as  
711 defined by Sinninghe Damsté, 2016), (f) Ternary diagram of  $C_{28}$  (sum of  $C_{28} 1,13$  and  $C_{28} 1,14$ ),  
712  $C_{30}$  (sum of  $C_{30} 1,13$ ,  $C_{30} 1,14$  and  $C_{30} 1,15$ ) and  $C_{32}$  ( $C_{32} 1,15$ ) diols (LOCO transect in orange, VA  
713 core-tops in blue, data from Lattaud et al., 2017 in purple). The maps were drawn using Ocean  
714 Data View.

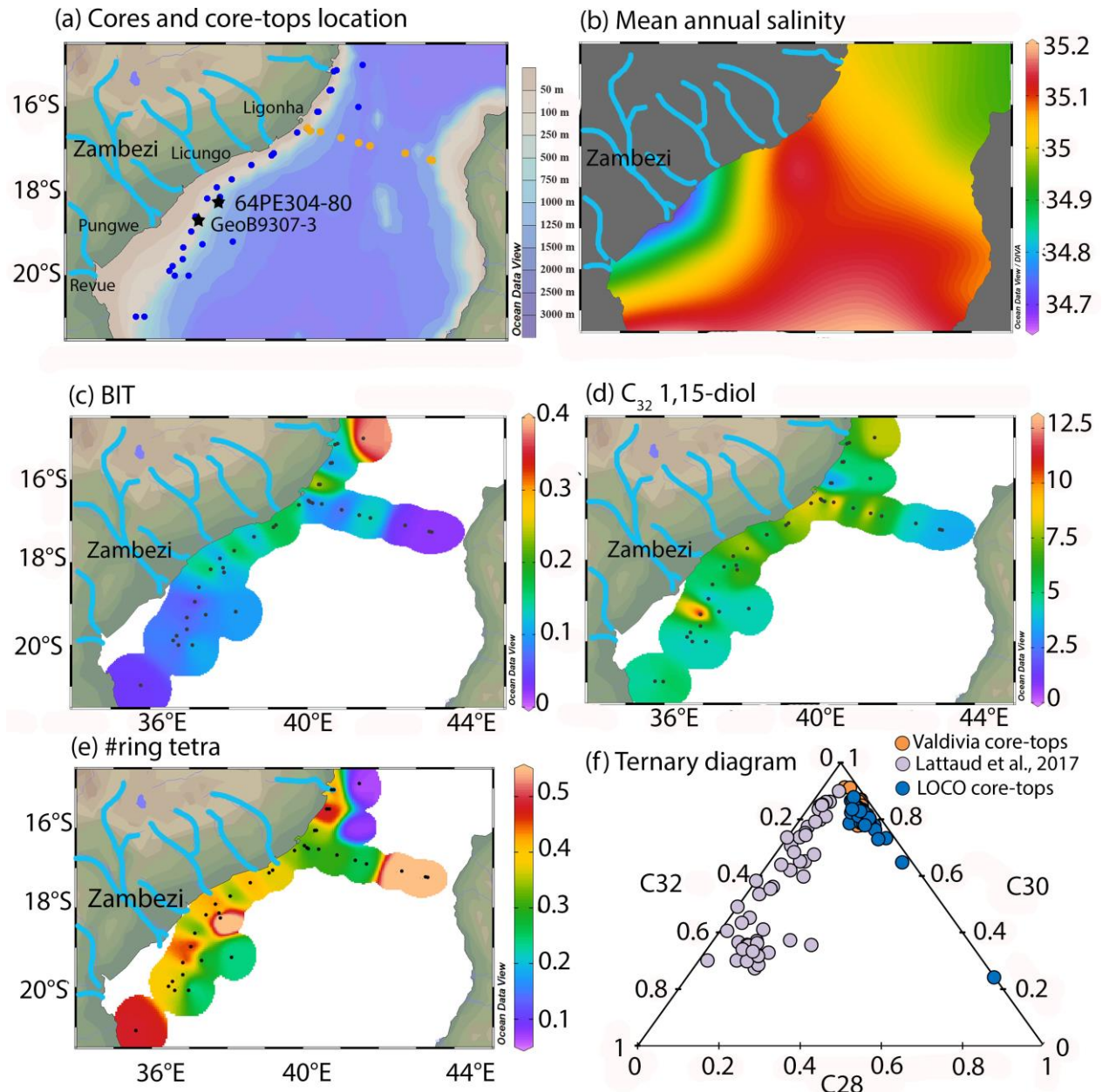
715 **Figure 2.** Organic and lithologic proxy records for core 64PE304-80 and parallel core GIK16160-  
716 3. (a) BIT index indicating soil and riverine input (Kasper et al., 2015) and  $F_{C_{32} 1,15}$  tracing riverine  
717 input (b) Red Sea Level changes (Grant et al., 2013) (c)  $\log(Ca/Ti)$  indicating terrestrial input (van  
718 der Lubbe et al., 2013), (d) reconstruction of  $\delta D$  precipitation based on leaf wax  $n$ - $C_{29}$  alkane of  
719 core GIK16160-3 (Wang et al., 2013),  $\epsilon_{Nd}$  signatures of the clay fraction document changes in  
720 riverine influence (van der Lubbe et al., 2016). The grey bars show the Younger Dryas (YD) and  
721 Heinrich event 1 (H1) and 4 (H4). Black triangles indicate positions where  $^{14}C$  AMS dates were  
722 obtained (Kasper et al., 2015)

723 **Figure 3.** Sources of riverine input in both area, (a) Location of core GeoB7702-3 (b) Close up  
724 location of core GeoB7702-3 (adapted from Castañeda et al., 2016) (c) source of the Nile river  
725 sediments (from Castañeda et al., 2016) and (d) Location of core 64PE304-80 and the  
726 Mozambique Channel (red circles shows source areas of the Zambezi river during dry conditions,

727 blue circle shows source area of the Zambezi river during wet conditions (Just et al., 2014), and  
728 green circle show northern rivers source area (van der Lubbe et al., 2016).

729 **Figure 4.** Organic and lithologic proxy records for core GeoB7702-3 and core 9509. (a) BIT index  
730 indicating soil and riverine input (Castañeda et al., 2010) and  $F_{C_{32} 1,15}$  tracing riverine input (b)  
731 Red Sea Level changes (Grant et al., 2013) (c)  $\log(Ca/Ti)$  indicating terrestrial input (Castañeda et  
732 al., 2016), (d) reconstruction of  $\delta D$  precipitation based on leaf wax n-C<sub>31</sub> alkane (Castañeda et  
733 al., 2016), (e)  $^{87}Sr/^{86}Sr$  signatures of the sediment core 9509 (offshore the Israeli coast)  
734 document changes in riverine influence (Box et al., 2011). The grey bars show the sapropel layer  
735 (S1), Younger Dryas (YD), Heinrich event 1 (H1) and the Last Glacial Maximum (LGM). Black  
736 triangles indicate  $^{14}C$  AMS dates (from Castañeda et al., 2010).

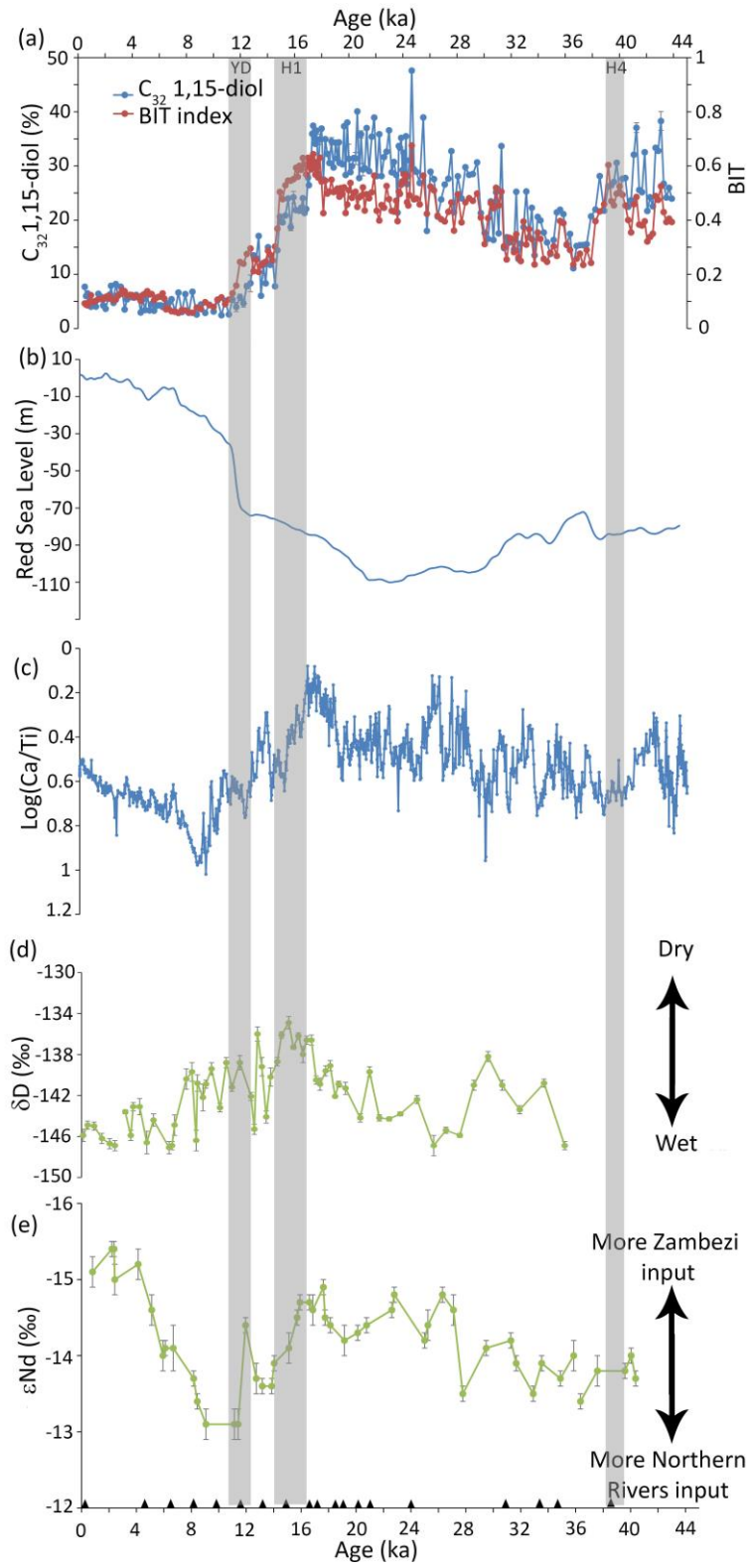
737



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739 Figure 1

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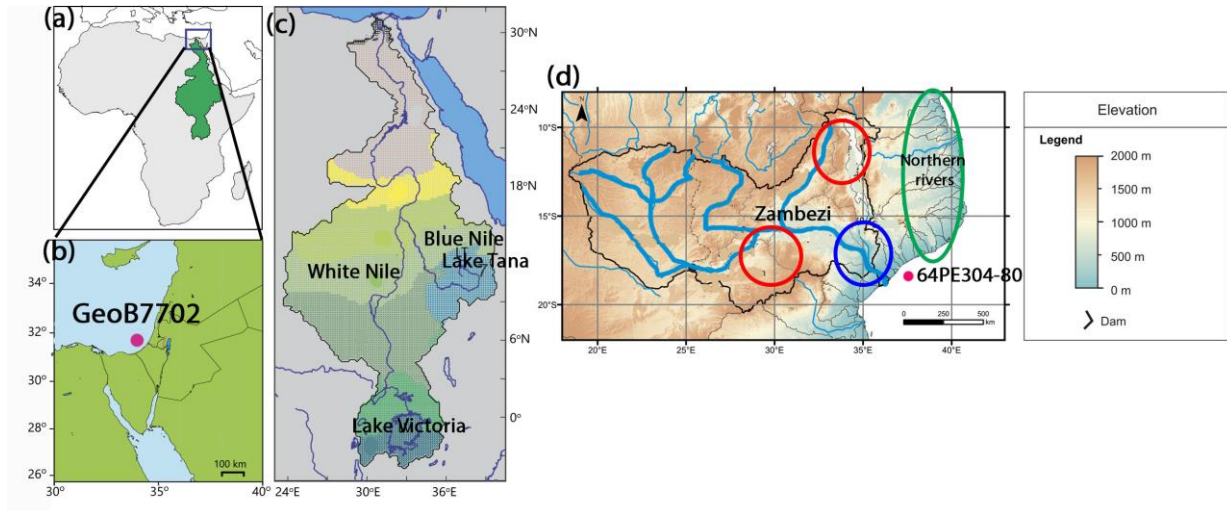


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742 Figure 2

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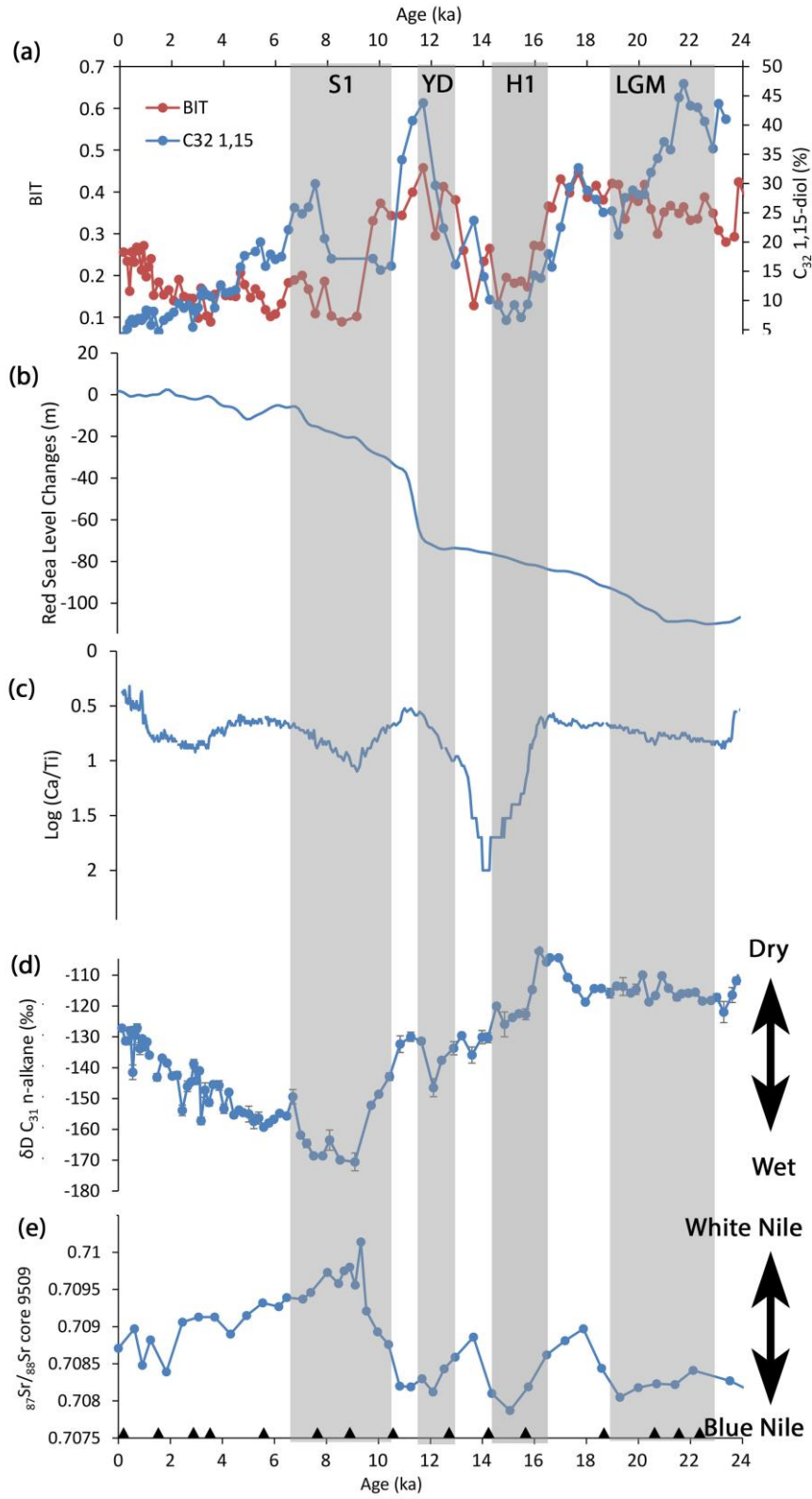


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746 Figure 3

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749 Figure 4