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## A composite Pliocene record of sea surface temperature in the central Mediterranean (Capo Rossello composite section – South Sicily)

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1 Running head: Evolution of the Equatorial Pacific mean state over the last 10 Myr

2  
3 Title: Evolution of the zonal gradients across the Equatorial Pacific during the Miocene-  
4 Pleistocene

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21  
22 **ABSTRACT**

23 Combining  $U_{37}^{k'}$  - and  $TEX_{86}$ -derived temperatures, and oxygen isotopic values of mixed-  
24 layer and thermocline species from the IODP site U1338 (East Equatorial Pacific) and ODP  
25 Site 806 (West Equatorial Pacific) we assess the evolution of the zonal sea-surface  
26 temperature gradients and thermocline depth across the equatorial Pacific from the Late

27 Miocene through the Pleistocene. Data suggest a long-term shoaling of the thermocline along  
28 the equator throughout the Miocene-Pliocene that accelerated around 5.3 Ma. We identify a  
29 critical transition at about 3.8 Ma from a El-Niño-like-dominated mean state during the Late  
30 Miocene and Early Pliocene to a La-Niña-like-dominated state during the Late Pliocene-  
31 Pleistocene. This transition coincides with the restriction of the Indonesian seaway and the  
32 onset of ice growth in the northern hemisphere and in Antarctica that led to the long-term  
33 strengthening of the Walker circulation and affected low-latitude zonal gradient.

34

35 **KEY WORDS:** Equatorial Pacific, Miocene Pliocene climate, biomarker proxies, planktonic  
36 foraminifera, coccoliths.

37

## 38 INTRODUCTION

39 The equatorial Pacific Ocean plays an important role in the global climate by influencing the  
40 heat transport from low to mid- and high latitudes. The modern structure of the equatorial  
41 Pacific is characterized by a pronounced east-west asymmetry of the thermocline depth and of  
42 sea-surface temperature (SSTs) with the warm pool (~ 29°C) in the west and the cold tongue  
43 (~ 23°C) in the east (Fig. 1). The thermocline in the West Equatorial Pacific is deep and  
44 controls the flux of atmospheric latent heat (Tian et al., 2001) whereas in the east it is shallow  
45 and heat is absorbed (Boccaletti et al., 2004). The SSTs across the equatorial Pacific are  
46 affected by the El Niño–Southern Oscillation (Cane, 1998), one of the most important  
47 component of the global climate system, which alternates at inter-annual time scale between  
48 warm “El Niño” and cold “La Niña” phases.

49 The El Niño–Southern Oscillation is sensitive to the SST distribution across the equatorial  
50 Pacific through the link between temperatures and the strength of the trade winds (Neelin et

51 al., 1998). Changes in the meridional SST gradient in the equatorial Pacific have important  
52 consequences on the local and global climate.

53 The modern setting of the equatorial Pacific was established during the mid- to late Miocene  
54 (LaRivière et al., 2012) through the progressive closure of the Central American Seaway  
55 (CAS) and the Indonesian Seaway (Keigwin, 1982; Haug and Tiedemann, 1998; Srinivasan  
56 and Sinha, 1998; Schneider and Schmittner, 2006) together with Northern Hemisphere  
57 Glaciation (Mudelsee and Raymo, 2005) (Fig. 2). During this period, the onset of the modern  
58 thermohaline circulation and equatorial upwelling led to the progressive cooling of deep -  
59 water (Woodruff and Savin, 1989; Ravelo et al., 2004), upper-ocean stratification, and  
60 thermocline rise. Nathan and Leckie (2009) dated the proto warm pool in the West Equatorial  
61 Pacific (WEP) between 11.6 and 10 Ma. The thermocline depth asymmetry set up around 4.8  
62 Ma and induced a well-documented cold-water tongue in the Eastern Equatorial Pacific (EEP)  
63 between 4.4 and 3.6 Ma (Chaisson and Ravelo, 2000; Wara et al., 2005; Steph et al., 2010;  
64 Rousselle et al., 2013).

65 Although numerous studies (e.g., Raymo et al., 1996; Haywood and Valdes, 2004; Ravelo et  
66 al., 2004, 2006; Rickaby and Halloran, 2005; Wara et al. 2005; Barreiro et al., 2005; Fedorov  
67 et al. 2006, 2013; Dekens et al. 2008; Brierley et al., 2009) established that the equatorial  
68 Pacific zonal gradients were weaker during the early Pliocene warm period (El Niño-like, El  
69 Padre) and stronger during the mid-Holocene (La Niña-like; Koutavas et al. 2006), there is no  
70 consensus on the long-term evolution of the equatorial Pacific mean state since the Miocene  
71 (Fig. 2). This is a major limitation to predict El Niño–Southern Oscillation, in that its  
72 behavior is closely related to long-term changes in tropical Pacific oceanography that are still  
73 not fully understood. Some authors suggest that the equatorial thermal state remained  
74 unchanged over the last 12 Myr (Zhang et al., 2014) whereas others identified alternating El  
75 Niño-like and La Niña-like conditions (Wara et al., 2005; Ravelo et al., 2006; Nathan and

76 Leckie, 2009; Kamikuri et al., 2009; Drury et al., 2018) or a progressive shoaling of the  
77 thermocline over the last 13 Myr (LaRiviere et al., 2012).

78 Because the past variability of the equatorial Pacific mean state is also considered to have  
79 played a role in major climatic transitions (Yin and Battisti, 2001), reconstructing the long-  
80 term evolution of the thermal structure in this region is crucial.

81 While most published studies focus on very specific time intervals (e.g., Medina-Elizalde and  
82 Lea, 2010; Ford et al., 2015), the purpose of this study is to examine the long-term evolution  
83 of equatorial Pacific surface conditions over the last 10 Myr. To do so, surface and subsurface  
84 temperature records from IODP Site U1338 (EEP) (Fig. 1) are generated from both alkenones  
85 ( $U_{37}^{k'}$ ) produced by coccolithophores (marine unicellular haptophyte algae, Brassell et al.,  
86 1986) and glycerol- dialkyl glycerol- tetraethers (GDGTs,  $TEX_{86}^H$ ) produced by  
87 *Thaumarchaeota* (Archaea, Schouten et al., 2002; Kim et al., 2010). Additional hydrological  
88 information is provided by the oxygen-isotope composition of calcareous nannofossils  
89 *Noelaerhabdaceae* spp. calcifying in the photic zone and the thermocline-dweller  
90 foraminifera *Globorotalia menardii* (Martin, 1999; Spero et al., 2003) recovered from the  
91 ODP Site 806 (WEP) and IODP Site U1338 (Fig. 1).

## 92 **OCEANOGRAPHIC SETTING**

93 The equatorial Pacific surface ocean circulation, controlled by the Walker cell and Trade  
94 winds, results in two westward surface currents, the North and South Equatorial Currents,  
95 respectively (SEC and NEC; Fig. 1). The easterly winds pile warm waters in the WEP and  
96 create a subsurface eastward circulation known as the Equatorial Under Current (EUC),  
97 which brings cold and nutrient-rich waters to the surface in the EEP. The subsequent thinning  
98 of the mixed-layer and formation of a cold tongue in the EEP produce a strong W-E  
99 asymmetry of the equatorial thermocline depth, deeper in the West Pacific than in the East  
100 Pacific (Fig. 1). The modern equatorial SST gradient across the Pacific varies in response to

101 the inter-annual El Niño-Southern Oscillation. During El Niño years, the trade winds weaken,  
102 the thermocline deepens in the EEP, and the equatorial upwelling attenuates. As a  
103 consequence, the equatorial SST gradient is reduced, mean temperatures are warmer, and the  
104 extratropical heat distribution is impacted (Molnar and Cane, 2002; McPhaden et al., 2006).  
105 Since the Miocene, the Pacific zonal temperature gradients and the thermocline tilt have  
106 varied. Extended periods of reduced equatorial SST gradient and deep thermocline that mimic  
107 the modern El Niño events are called permanent El Niño-like states. Similarly, periods of  
108 long-term strong equatorial Pacific zonal gradients are called La Niña-like states.

## 109 **MATERIALS AND METHODS**

### 110 *IODP Site U1338 and ODP Site 806*

111 The IODP Site U1338 (2° 30.469' N, 117° 58.178' W, 4200 meters water depth, leg 321) was  
112 drilled in the EEP, in the modern cold tongue (Pälike et al., 2010) (Fig. 1). Despite the  
113 northward drift of the Pacific plate, this site remained in the equatorial band for the past 10  
114 Myr (Pälike et al., 2010). Sediments at Site U1338 were deposited above the calcite  
115 compensation depth (Pälike et al., 2012) and are composed of well-preserved biogenic  
116 material, predominantly nannofossil oozes, with variable abundances of diatoms, radiolarians,  
117 and foraminifera (Pälike et al., 2010). The age model of the core was constructed from  
118 biostratigraphic (nannofossils, foraminifers, diatoms, and radiolarians) and paleomagnetic  
119 data (Pälike et al., 2010; Backman et al., 2016). Average sedimentation rates are relatively  
120 high for the studied time interval (~ 27 m/Myr).

121 ODP Site 806 (0° 19,1' N, 159° 21,7' E, 2520 meters water depth, leg 130) is located on the  
122 northeast margin of the Ontong Java plateau in the area of the WEP warm pool (Fig. 1).  
123 Sediments are composed mostly of well-preserved calcareous nannofossils and planktic  
124 foraminifera. The age model is based exclusively on biostratigraphic data of calcareous  
125 nannofossils, foraminifera, diatoms, and radiolarians established by Kroenke et al. (1991),

126 later refined by Takayama (1993) and Chaisson and Leckie (1993). Ages are corrected based  
127 on biostratigraphic data of Backman et al. (2016) (see Supplement). The average  
128 sedimentation rate at this site is higher than at U1338, ranging from ~ 45 m/Myr between 9  
129 and 5 Ma to ~ 29 m/Myr during the Pliocene.

130 Proxy records at both sites are generated for the period between 10 and 0.18 Ma using  
131 samples from sections U1338B-22H7W through U1338B-1H2W and sections 806B-37X3W  
132 through 806B-1H1W. The average sampling resolution of these records varies between 100  
133 kyr and 400 kyr.

#### 134 *Oxygen Stable-Isotope Measurements*

135 To determine changes in the thermocline depth in the EEP and WEP through time, we  
136 measured the oxygen-isotope ratios ( $\delta^{18}\text{O}$ ) in the calcite of calcareous-nannofossil-enriched  
137 fractions and of the upper-thermocline-dweller planktonic foraminifera *Globorotalia*  
138 *menardii*. Calcareous nannofossils calcify mainly in the photic zone and thus are used to  
139 derive surface-water properties (Roth, 1986) whereas *G. menardii* is selected to assess  
140 subsurface water changes.

141  $\delta^{18}\text{O}$  values are measured on calcareous nannofossils *Noelaerhabdaceae* spp. isolated from  
142 the bulk sediment following the procedure of Minoletti et al. (2009).  $\delta^{18}\text{O}$  measurements are  
143 performed on the 2-5  $\mu\text{m}$  fractions at both sites. For site U1338, we use the  $\delta^{18}\text{O}$  data previously  
144 published by Rousselle et al. (2013) based on the same protocol as in the present study.  
145  $\delta^{18}\text{O}$  values are measured using a Finnigan Delta E mass spectrometer at 50°C, at a temporal  
146 resolution of 400 kyr. The quality of the granulometric separation based on 200 particles  
147 counting in smear slides is estimated to be at least 75% except between 8 and 9 Ma at Site  
148 806, where *Sphenolitus abies* dominated. These measurements are thus not considered here.

149 In addition, five to seven specimens of *G. menardii* were hand-picked from the > 160  $\mu\text{m}$   
150 fractions for isotopic measurements. Analyses are performed on a VG ISOPRIME at 90°C

151 with a precision of 0.1‰ and expressed with reference to the VPDB international standard. 45  
152 samples at Site U1338 and 99 samples at Site 806 were analyzed. The average temporal  
153 resolution over the last 10 Myr is comprised between 120 and 150 ka.

#### 154 *Alkenone Analyses*

155 Alkenone-derived SSTs of Site U1338 previously reported by Rousselle et al., (2013) are  
156 supplemented in the present study with 23 additional samples selected from the Pliocene  
157 interval (U1338B-8H5W to U1338B-1H2W) to reach a temporal resolution of 100 kyr.  
158 Sedimentary alkenones are analyzed following the procedure described by Ternois et al.  
159 (2000). About 5 g of freeze-dried sediments are extracted in a mixture of CH<sub>2</sub>Cl<sub>2</sub> / CH<sub>3</sub>OH.  
160 Alkenones are isolated from the total lipid extract by silica-gel chromatography using  
161 solvents of increasing polarity. The fraction containing alkenones is concentrated, transferred  
162 into clean glass vials, and evaporated under a nitrogen stream. Gas-chromatography analyses  
163 are performed using a Varian 3400CX series equipped with a septum programmable injector  
164 (SPI) and a flame ionization detector (FID). We use a fused silica capillary column  
165 (Chrompack CP Sil5CB, 50 m long, 0.32 mm internal diameter, 0.25 μm film thickness) and  
166 helium as a carrier gas. The alkenone unsaturation ratio  $U^{K'}_{37}$  ( $C_{37:2}/(C_{37:2} + C_{37:3})$ ) is converted  
167 into SSTs using the Conte et al. (2006) calibration:  $T = -0.957 + 54.293 (U^{K'}_{37}) - 52.894$   
168  $(U^{K'}_{37})^2 + 28.321 (U^{K'}_{37})^3$ , which provides more accurate estimates in warm-temperature  
169 range than Prahl et al. (1988). Average external precision of the SSTs using this calibration  
170 has been estimated at ~ 1.2°C (Conte et al., 2006). Replicate analyses indicate internal  
171 precision of ± 0.5°C. Because of the limited amount of sediment material at Site 806 (where  
172 alkenone concentrations are extremely low), previously published data from Pagani et al.  
173 (2010) provide data covering the past 5 Myr. Note that we recalculated SSTs from the  $U^{K'}_{37}$   
174 values of Pagani et al. (2010) using the Conte et al. (2006) calibration to avoid calibration  
175 bias between sites (see Supplement).



176

## *GDGT Analyses*

177 Forty samples from Site U1338 were analyzed for GDGTs at the Netherlands Institute for Sea  
178 Research (NIOZ, Texel, The Netherlands). About 5 g of sediment are freeze dried and  
179 extracted in a mixture of CH<sub>2</sub>Cl<sub>2</sub> and CH<sub>3</sub>OH. The total lipid extracts are separated following  
180 the protocol described in Schouten et al. (2002), using Al<sub>2</sub>O<sub>3</sub> column chromatography to  
181 separate non-polar compounds from the polar GDGTs. Polar fractions are then filtered  
182 through a 0.4 μm pore-size filter, evaporated under nitrogen, and diluted to a concentration of  
183 2 mg/ml. GDGTs are analyzed using an Agilent HP1100 (HPLC) equipped with a Prevail  
184 Cyano column (3 μm, 150 mm x 2.1 mm) following Schouten et al. (2007). The TEX<sub>86</sub> index  
185 is calculated using peak areas of the following GDGTs as defined by Schouten et al. (2002):  
186  $TEX_{86} = [GDGT2] + [GDGT3] + [GDGT5'] / [GDGT1] + [GDGT2] + [GDGT3] + [GDGT5']$ . The  
187  $TEX_{86}^H$ , defined as the log(TEX<sub>86</sub>), is used to calculate temperatures over the past 9 Myr.  
188 Temperatures are calculated using the calibration established for temperatures above 15°C by  
189 Kim et al. (2010) ( $T = 68.4 * TEX_{86}^H + 38.6$ ). The error on temperature estimates is ± 2.5°C.  
190 Other calibrations have been proposed for TEX<sub>86</sub> temperatures, in particular based on  
191 Bayesian statistics (Tierney and Tingley, 2015) but these yield results similar to those shown  
192 here, whereas the Kim et al. (2010) calibration is consistent with other studies from this area  
193 (e.g., Seki et al., 2010; Zhang et al., 2014). TEX<sub>86</sub><sup>H</sup> - derived temperatures are obtained at a  
194 mean temporal resolution of 230 kyr. At Site 806, because of the very low GDGT  
195 concentrations, we use previously published data from Zhang et al. (2014) covering the past  
196 12 Myr for comparison with the data from Site U1338.

197

## **RESULTS**

198

### *Stable Isotopes of the Calcareous Nannofossil Fine Fractions and *G. menardii**

199

### **δ<sup>18</sup>O Record at Site U1338.---**

200 The oxygen stable-isotope signatures of the *Noelaerhabdaceae*-enriched fine fractions  
201 ( $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$ ) vary between -1.5‰ and 0.5‰ (Fig. 3A).  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  progressively  
202 increase between 8 Ma and 5 Ma, before declining to -1.3‰ until 2.7 Ma and rising again to  
203 0.4‰. The  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  record is similar to the higher resolution  $\delta^{18}\text{O}$  record of the  
204 surface dweller *G. sacculifer* from the nearby Site 847 covering only the last 5 Myr (Wara et  
205 al., 2005) (Figs. 1, 3A).

206  $\delta^{18}\text{O}$  values measured on the thermocline-dwelling planktonic foraminifera *G. menardii*  
207 ( $\delta^{18}\text{O}_{\text{G.menardii}}$ ) over the last 8 Myr (Figure 3A) demonstrate that values vary between -1.4‰  
208 and 1.25‰ and display trends similar to those of  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  until 5 Ma. Then,  
209  $\delta^{18}\text{O}_{\text{G.menardii}}$  become more positive than  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  and still increase until 0.4 Ma, as  
210 also recorded in the deep thermocline dweller *G. tumida*  $\delta^{18}\text{O}$  values ( $\delta^{18}\text{O}_{\text{G.tumida}}$ ) at Site 847  
211 (Figs. 1, 3A; Wara et al., 2005).

#### 212 **$\delta^{18}\text{O}$ Record at Site 806.---**

213  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  at site 806 vary within a range similar (between -2.8 and 0.9‰) to site  
214 U1338 (Fig. 3B).  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  values are relatively stable (around 0‰) between 7 Ma  
215 and 4.4 Ma and rapidly decline to more negative values between 4.5 Ma and 4 Ma, after  
216 which values remain stable until 0.38 Ma. The  $\delta^{18}\text{O}_{\text{G.menardii}}$  record at Site 806 shows a long-  
217 term decrease between 9.7 Ma and 7 Ma and becomes stable until 1.9 Ma. At 3.7 Ma oxygen-  
218 isotope values decrease sharply by 1‰, and again between 2.5 Ma and 1.8 Ma. The  
219  $\delta^{18}\text{O}_{\text{G.menardii}}$  values then increase to higher values (-0.4 ‰) until 0.5 Ma and further to 0.5‰  
220 at 0.2 Ma. Overall,  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  and  $\delta^{18}\text{O}_{\text{G.menardii}}$  reveal more positive values in the EEP  
221 than in the WEP and include larger-amplitude variations.

#### 222 *EEP Temperature Reconstructions.*

223 Alkenone-derived SSTs ( $\text{SST}_{\text{U}^{\text{K}^2}_{37}}$ ) at Site U1338 are relatively stable and warm ( $\sim 27^\circ\text{C}$ )  
224 between 10 Ma and 6.8 Ma (Fig. 4A). They cool by  $1^\circ\text{C}$  between 6.8 Ma and 5.6 Ma, and

225 show a broad warming until 3.7 Ma. From 3.7 Ma onwards, SSTs depict another cooling of  
226 about 4°C and superimposed cold episodes of 1 to 2°C amplitude around 2.9 Ma, 1.7 Ma, and  
227 0.5 Ma.

228 From the Late Miocene to the Pleistocene, TEX<sub>86</sub>-derived temperatures (T<sub>TEX</sub>) indicate a  
229 long-term cooling. Between 8 and 5 Ma, T<sub>TEX</sub> and the SST<sub>U<sup>K'</sup><sub>37</sub></sub> vary in the same range of  
230 values (between 24°C and 27°C; Fig. 4A), although they are slightly cooler. From approx. 4.5  
231 Ma, the two temperature proxies show marked temperature difference. Note that T<sub>TEX</sub>

232 increase around 7.5 Ma and 4.8 Ma, reaching values close to SST<sub>U<sup>K'</sup><sub>37</sub></sub>.

233 SST<sub>U<sup>K'</sup><sub>37</sub></sub> at Site U1338 are warmer than T<sub>TEX</sub> by 2 to 3°C on average. Both progressively cool  
234 down between 9 Ma and 0.18 Ma but T<sub>TEX</sub> decreases by 5°C while SST<sub>U<sup>K'</sup><sub>37</sub></sub> cools by only  
235 3°C (Fig. 4A).

236

## DISCUSSION

237

### *Surface and Subsurface Water Temperatures in the Equatorial Pacific*

238 Warmer SST<sub>U<sup>K'</sup><sub>37</sub></sub> than T<sub>TEX</sub>, as observed at Site U1338, is reported in several studies (e.g.,  
239 Huguet et al., 2007; Lee et al., 2008; Leider et al., 2010; Lopes dos Santos et al., 2010;  
240 Rommerskirchen et al., 2011; Seki et al., 2012; Zhang et al., 2014) and explained by different  
241 depth habitat or season of production. However, because seasonality is weak at low latitudes,  
242 it is unlikely to explain a temperature differences of up to 4°C, at Site U1338. Unlike  
243 haptophytes thriving in the photic zone, *Thaumarchaeota*, the sources of the GDGTs, occur  
244 throughout the water column (e.g., Karner et al., 2001; Huguet et al., 2007), and the export of  
245 GDGTs can be integrated for a larger part of the upper part of the ocean, likely including a  
246 part of the thermocline. Wuchter et al. (2005) reported maximum concentrations of GDGTs  
247 between 100 and 200 m depth in the Pacific Ocean, coinciding with high ammonium  
248 concentrations below the photic zone (Murray et al., 1999; Karner et al., 2001; Wuchter et al.,  
249 2005; Huguet et al., 2007; Pearson et al., 2007). GDGT producers can thus preferentially

250 develop in subsurface or deeper waters (Huguet et al., 2007; Kim et al., 2008; Lee et al.,  
251 2008; Huguet et al., 2011; Kim et al., 2012). The difference between  $T_{\text{TEX}}$  and  $\text{SST}_{\text{U}^{\text{K}'37}}$  in the  
252 EEP may then account for different production or export depth (Lopes dos Santos et al., 2010;  
253 Rommerskirchen et al., 2011; Schouten et al., 2013).

254 The interpretation of  $T_{\text{TEX}}$  temperatures across the equatorial Pacific is complicated by both the  
255 upwelling in the EEP and the different trophic conditions between the EEP and the WEP that  
256 influence the export depth of GDGTs (Seki et al., 2012). Numerous studies indicate that  
257 upwelling impacts on the depth of production and export of GDGTs to the sediment (Wuchter  
258 et al., 2005; Huguet et al., 2007; Lee et al., 2008; Lopes dos Santos et al., 2010; Seki et al.,  
259 2012). In the absence of upwelling,  $T_{\text{TEX}}$  reflects the surface temperatures (Sinninghe Damsté  
260 et al., 2002; Wakeham et al., 2004) whereas under upwelling conditions,  $T_{\text{TEX}}$  represent  
261 thermocline temperatures (Wakeham et al., 2002; Wuchter et al., 2005; Huguet et al., 2007).  
262 Furthermore, the nitrate maximum across the modern equatorial Pacific exhibits a clear  
263 asymmetry in the depth. Könneke et al. (2005) found that the production of *Thaumarchaeota*  
264 occurs in the subsurface nitrate maximum, which thus is deeper in oligotrophic regions such  
265 as the WEP (Brzezinski, 1988). As a consequence, within the same basin  $T_{\text{TEX}}$  may reflect  
266 surface and subsurface conditions in response to different environmental factors (Hertzberg et  
267 al., 2016).

268 In the EEP, the  $T_{\text{TEX}}-\text{SST}_{\text{U}^{\text{K}'37}}$  temperature gradient ( $\Delta T$ ) is proposed to monitor changes of  
269 the thermocline depth (Seki et al., 2012) assuming that  $\Delta T$  is driven by the depth migration of  
270 GDGT producers in response to upwelling activity. Therefore, at Site U1338, warming  $T_{\text{TEX}}$   
271 and reduced  $\Delta T$  reflect a deep thermocline and surface export of GDGTs, whereas a more  
272 pronounced cooling of  $T_{\text{TEX}}$  compared to  $\text{SST}_{\text{U}^{\text{K}'37}$ , (increased  $\Delta T$ ) reflects a shallow  
273 thermocline, upwelling activity, and subsurface export of GDGTs.

274 In the WEP, the interpretation of  $T_{\text{TEX}}\text{-SST}_{\text{U}^{\text{K}'37}}$  is not straightforward either. At Site 806,  
275  $\text{SST}_{\text{U}^{\text{K}'37}$  calculated from Pagani et al., (2010) are warm and stable, averaging 28 to 28.5°C  
276 over the last 5 Myr (Fig. 4C) except for a 2°C sharp cooling at ~ 1.5 Ma. High-resolution  
277 Mg/Ca-derived SSTs ( $\text{SST}_{\text{Mg/Ca}}$ ) corrected from  $\text{Mg/Ca}_{\text{seawater}}$  (Medina-Elizalde et al., 2008)  
278 from the same site show markedly warmer conditions in the surface of the WEP especially  
279 between 5 and 3.5 Ma, reaching up to 32°C. This difference with the alkenone-derived  
280 temperatures can be attributed to (1) the alkenone calibration being less accurate in the  
281 warmer temperature range (Conte et al., 2006), thus leading to the underestimation of  
282  $\text{SST}_{\text{U}^{\text{K}'37}$  (Tierney and Tingley 2018) or (2) the uncertainties on the seawater Mg/Ca  
283 correction. However, from 3 Ma onwards,  $\text{SST}_{\text{U}^{\text{K}'37}$  and  $\text{SST}_{\text{Mg/Ca}}$  temperatures display  
284 similar trends and values, except between 1.8 Ma and 1.2 Ma, when they diverge. Since this  
285 discrepancy coincides with isotopic changes (Fig. 3) we can reasonably conclude that the  
286 difference between the two records during the late Pliocene reflects oceanographic changes.  
287 During the early Pliocene, cooler  $\text{SST}_{\text{U}^{\text{K}'37}$  appear to be linked to a calibration limitation of  
288 the proxy at warm temperatures.

289 Comparison between the  $T_{\text{TEX}}$  record (Zhang et al., 2014) and subsurface temperatures  
290 derived from  $\text{Mg/Ca}_{G.tumida}$  (Ford et al., 2015) at ODP Site 806, highlights that from 5 to 3.5  
291 Ma both reconstructions agree well within their uncertainties (Fig. 4C). From 3.5 to 2 Ma,  
292  $T_{\text{TEX}}$  match with  $\text{SST}_{\text{U}^{\text{K}'37}$  and Mg/Ca-derived SSTs from the surface dweller *G. ruber*  
293 ( $\text{Mg/Ca}_{G.ruber}$  Medina-Elizalde et al., 2008), whereas  $\text{Mg/Ca}_{G.tumida}$  temperatures are  
294 systematically cooler than  $T_{\text{TEX}}$  (Fig. 4C). After 1.3 Ma,  $T_{\text{TEX}}$  values are intermediate between  
295  $\text{Mg/Ca}_{G.ruber}$  and  $\text{Mg/Ca}_{G.tumida}$  temperatures, while  $\text{SST}_{\text{U}^{\text{K}'37}$  tend to agree with  $\text{Mg/Ca}_{G.ruber}$   
296 SSTs. It is likely that in the WEP, the  $T_{\text{TEX}}$  reflect production in different water depth in  
297 response to trophic changes and environmental conditions. We speculate that during the warm  
298 early Pliocene and after 1.3 Ma,  $T_{\text{TEX}}$  reflect subsurface conditions and that during the Late

299 Pliocene, the export of GDGTs derived mainly from surface waters. This interpretation  
300 nuances the interpretations of Zhang et al. (2014), who considered that  $T_{\text{TEX}}$  reflect surface  
301 temperatures and could be used as an alternative to  $U^{\text{K}'}_{37}$  under warm conditions.

302 *Changes in the Equatorial Pacific Mean State over the Last 10 Myr*

303 To investigate the evolution of the surface stratification across the Equatorial Pacific, we  
304 calculate the oxygen-isotope gradients between *Noelaerhabdaceae* and *G. menardii* ( $\Delta\delta^{18}\text{O}$ )  
305 (Fig. 4B) (Nathan and Leckie, 2009; Beltran et al., 2014). Note that in doing so, the sea-water  
306  $\delta^{18}\text{O}$  component is factored out. Higher (lower)  $\Delta\delta^{18}\text{O}$  values reflect a shallower (deeper)  
307 thermocline (Ravelo and Fairbanks, 1992; Ravelo and Shackleton, 1995; Farrell et al., 1995;  
308 Nathan and Leckie, 2009, Beltran et al., 2014).

309 Variations in the gradient between  $T_{\text{TEX}}$  and  $\text{SST}_{U^{\text{K}'}_{37}}$  also can be used to evaluate changes in  
310 the thermocline depth.  $T_{\text{TEX}}$  and  $\text{SST}_{U^{\text{K}'}_{37}}$  values similar to those observed between 7.3 Ma  
311 and 6.8 Ma, and around 4.8 Ma at IODP Site U1338, indicate a deep thermocline (Fig. 4A).  
312 Some intervals are characterized by an increase in  $\Delta T$  (such as, for example, between 6.8 Ma  
313 and 5.3 Ma at IODP Site U1338) associated with a cooling trend of both proxies. These  
314 features indicate that GDGTs are produced deeper and that surface waters are concomitantly  
315 cooling, which is consistent with upwelling activity.

316 Based on temperatures and isotopic gradients between surface and subsurface waters from the  
317 EEP and WEP, the data reveal a pronounced oceanographic change at *ca.* 5 Ma with the  
318 thermocline progressively rising (Fig. 4A, 4B).

319 **Late Miocene [9 – 5.3 Ma] Interval.---**

320 During the late Miocene period, east and west  $\Delta\delta^{18}\text{O}$  are reduced and stable, indicating a deep  
321 thermocline across the equatorial Pacific (Fig. 4B). From 10 Ma to 6.3 Ma, data indicate  
322 warm SSTs ( $\sim 27\text{ }^\circ\text{C}$ ) in the EEP (Fig. 4A). This result coincides with the low abundances of  
323 upwelling radiolarian species reported between 9 and 7 Ma by Kamikuri et al. (2009). Similar

324 SST<sub>U<sup>K</sup>37</sub> and T<sub>TEX</sub> values at U1338 between 7.3 and 6.8 Ma also is consistent with a thick  
325 mixed layer in the EEP (Fig. 4A; Seki et al., 2012). In the WEP, Nathan and Leckie (2009)  
326 reported a similar decrease in the abundance of surface dwellers in favor of thermocline  
327 species, which suggests a deep thermocline. This reduced zonal gradient in SSTs and  
328 thermocline tilt across the equatorial Pacific corresponds to El Niño-like conditions, in  
329 agreement with earlier findings of Herbert et al. (2016), who indicate an expanded and weak  
330 Hadley circulation before 8 Ma.

331 The long-term Late Miocene El Niño-like state was interrupted by a brief interval (6.5 - 5.3  
332 Ma) during which the thermocline shoaled in the EEP, as shown by the  $\delta^{18}\text{O}$  gradient (Fig.  
333 4B). The SST cooling and decrease of surface salinity recorded at Site U1338 (Rousselle et  
334 al., 2013), supported by higher abundances of upwelling siliceous species during this interval  
335 (Kamikuri et al., 2009), suggest the first establishment of the cold tongue and of upwelling in  
336 the EEP. Increasing abundances of surface-dweller species at Site 806 (Nathan and Leckie,  
337 2009) (Fig. 3 and 4), this interval further supports the idea of a weak and transient La Niña  
338 like period. Drury et al. (2018) suggested a cooling of the thermocline waters related to the  
339 global late Miocene cooling (Federov et al., 2006; Drury et al., 2017). Although the data do  
340 not rule out this hypothesis, the T<sub>TEX</sub> cooling is supportive of a change in depth of GDGT  
341 production or export in response to the onset of the EEP upwelling.

#### 342 **Plio-Pleistocene [from 5.3 Ma] Interval.---**

343 The oxygen-isotope and temperature data indicate a long-term gradual thermocline shoaling  
344 and sea-surface cooling in the EEP (Fig. 4A, 4B) throughout this interval. In the WEP, the  
345 thermocline shoaled between 5.3 and 4 Ma, returning to a deep position between 4 and 1.8  
346 Ma and shoaling again after 1.8 Ma. This interval is characterized by the initiation of a  
347 progressive asymmetry in the thermocline depth across the equatorial Pacific.

348 After 5.3 Ma, SSTs at IODP Site U1338 were warm ( $\sim 27$  °C), in agreement with a deep  
349 thermocline until 4.5 Ma (Fig. 4A). This result is supported by a rapid increase of  $T_{\text{TEX}}$  at  
350 U1338. Moreover, abundances of upwelling radiolarian species decreased again in the EEP  
351 (Kamikuri et al., 2009). In the WEP, the proto-warm pool period ended (Nathan and Leckie,  
352 2009). This interval coincides with the Messinian salinity crisis (Hilgen et al., 2007), during  
353 which the isolation of the Mediterranean Sea would have triggered a reduction of the North  
354 Atlantic Deep Water formation (NADW) at  $\sim 6$  Ma because of changing water density of the  
355 North Atlantic waters (Pérez-Asensio et al., 2012). According to model simulations, this  
356 process would result in a deepening of the thermocline in the equator and El Niño-like  
357 conditions (Rühlemann et al., 2004; Timmermann et al., 2005; Barreiro et al., 2008), thus  
358 providing a possible explanation to the results herein.

359 Numerous studies have focused on the magnitude of Pliocene equatorial Pacific warmth (4.5  
360 – 3.5 Ma) and the factors responsible for it (e.g., Raymo et al., 1996; Haywood and Valdes,  
361 2004; Ravelo et al., 2004; Barreiro et al., 2008; Rickaby and Halloran, 2005; Brierley et al.,  
362 2009; Scroxton et al., 2011; Ford et al., 2015). It is accepted that during the mid-Piacenzian  
363 Warm Period, persistent El-Niño like conditions prevailed. This interval, also known as El  
364 Padre mean state (Ford et al., 2015), is characterized by a deep and warm thermocline and  
365 warm surface conditions across the equatorial Pacific.

366 The low-resolution record presented here suggests a brief interval centered around 4 Ma when  
367 the thermocline deepened in the east and shoals in the west. SSTs in the EEP reach 30°C and  
368 around 32°C in the WEP (Fig. 4A, 4C,  $\text{Mg}/\text{Ca}_{G.ruber}$  SSTs; Medina-Elizalde et al., 2008).  
369 From 3.5 Ma, a pronounced zonal gradient of the thermocline depth and of the SSTs  
370 developed across the Pacific. The SST cooling trend and shoaling of the thermocline  
371 identified in the EEP coincides with the permanent establishment of the cold tongue  
372 (Groeneveld et al., 2006; Dekens et al., 2007; Steph et al., 2010; Rousselle et al., 2013). This



373 result is in agreement with Chaisson and Ravelo (2000) and Kamikuri et al., (2009), who  
374 report higher abundances of subsurface-dweller foraminifera and radiolarian upwelling  
375 species in the EEP at that time. Simultaneous with the changes in the EEP, the warm pool  
376 became established and the thermocline deepened in the WEP, as suggested by warmer  
377 surface waters and reduced  $\Delta T$  (Fig. 4C, Wara et al., 2005; Medina-Elizalde et al., 2008) and  
378 planktonic foraminifera abundances (Cannariato and Ravelo, 1997; Chaisson and Ravelo,  
379 2000; Li et al., 2006). This La Niña-like pattern is consistent with earlier studies (Rickaby and  
380 Halloran 2005; Kamikuri et al., 2009; Steph et al., 2010; Ford et al., 2012 and Rousselle et al.,  
381 2013) in favor of an active equatorial upwelling and a strong Walker cell.  
382 Between 1.8 and 1.3 Ma, data indicate that the thermocline tilt and the SST gradient across  
383 the Pacific were reinforced (Fig. 4). This result is in agreement with the setting of the modern  
384 cold tongue identified by Martinez-Garcia et al. (2011).

#### 385 *Evolution of the Pacific Mean State in a Regional and Global Context*

386 Prior research has attributed changes in the thermocline tilt to local tectonic events such as the  
387 opening and closing of the Panama and Indonesian oceanic gateways (e.g., LaRiviere et al.,  
388 2012), and changes in sea-surface-temperature zonal gradients to global climate perturbations  
389 or changes in regional wind patterns and strength (Hovan, 1995; Ford et al., 2012).  
390 However, there is still a strong debate about the timing of the closure of these oceanic  
391 gateways and their role in controlling the equatorial Pacific mean climate state. Montes et al.,  
392 (2012, 2015), Osborne et al. (2014), and Sepulchre et al. (2014) suggested that the Central  
393 American Seaway was restricted by 10-11 Ma, which contradicts the commonly accepted late  
394 Pliocene timing of its closure (Keigwin, 1982; Duque-Caro, 1990; Haug and Tiedemann,  
395 1998; Haug et al., 2001). On the other hand, Molnar and Cronin (2015) showed a marked  
396 restriction of the Indonesian Seaway from 5 Ma. This interpretation implies that the changes

397 of the equatorial Pacific mean state during the Miocene would have been controlled mainly by  
398 global climate changes.

399 The brief La Niña-like interval between 6.5 and 5.3 Ma may be attributable to the  
400 combination of the restricted flow through the Central American Seaway (Keigwin, 1982;  
401 Keller et al., 1989) and the 25 m sea-level drop at ~ 6.5 Ma (Haq et al., 1987; Hardenbol et  
402 al., 1998; Haq and Schutter, 2008) due to the expansion of ice sheets in Greenland (Larsen et  
403 al., 1994), Alaska (Bradshaw et al., 2012) and Antarctica (Billups, 2002; Williams et al. 2010)  
404 and the 2 Myr-long Northern Hemisphere glaciation (Larsen et al., 1994). This global cooling  
405 is thought to have contributed to the reinforcement of the equatorial upwelling and EUC.

406 Data from sediments younger than ~ 5 Ma indicate an important change in the equatorial  
407 Pacific mean state with the dominance of La Niña-like intervals. The onset of this long-term  
408 state coincides with a drastic restriction of the Indonesian Seaway (Srinivasan and Sinha,  
409 1998; Molnar and Cronin, 2015), and changes in the global climate. The Indonesian Seaway  
410 restriction is thought to have contributed to the cooling of the eastern tropical Pacific over the  
411 last 5 Myr through the strengthening of the Walker circulation, leading to the increase in the  
412 Pacific zonal temperature gradient (Fig. 3, 4).

413 The La Niña-like event between 1.8 and 1.3 Ma coincides with pronounced ice growth in  
414 Antarctica (McKay et al., 2012) and the development of the northern-hemisphere ice sheets.  
415 The gradual increase of the pole-to-equator temperature gradient is thought to have reinforced  
416 the Walker cell, and affected the low-latitude zonal gradient (Molnar and Cane, 2002; McKay  
417 et al., 2012). Furthermore, modelling experiments show a positive feedback of the Pliocene  
418 low-latitude cooling on the global cooling (Fedorov et al., 2006). The shoaling of the  
419 thermocline increased the climate sensitivity of this area by strengthening the temperature  
420 gradient through the tropics and subsequently the Walker and Hadley cells (Brierley et al.,  
421 2009; Brierley and Fedorov, 2010). These processes led to the final setting of upwelling areas

422 (Philander and Fedorov, 2003; Dekens et al., 2007; Brierley et al., 2009; Brierley and  
423 Fedorov, 2010).

424

## CONCLUSION

425 This study demonstrates the complexity of reconstructing the evolution of the equatorial  
426 Pacific mean climate state and oceanographic features and the need of a multi-proxy approach  
427 to get a comprehensive understanding of the mechanism at play over the past 10 Myr.  
428 Combined isotopic zonal gradients between the surface (*Noelaerhabdaceae*) and subsurface  
429 (*G. menardii*) waters from the east and west equatorial Pacific reveals important steps towards  
430 the establishment of what is known today as El Niño–Southern Oscillation. The record  
431 suggests a general shoaling of the thermocline along the equator since the Late Miocene. The  
432 data show the appearance of the cold tongue at ~ 6.8 Ma before its proper establishment  
433 around 4.4 to 3.6 Ma (Chaisson 1995; Cannariato and Ravelo, 1997; Chaisson and Ravelo,  
434 2000; Molnar and Cane, 2002; Ravelo et al., 2004, 2006; Sato et al., 2008; Steph et al., 2010;  
435 Rousselle et al., 2013) and its modern configuration ~ 1.8 Ma. This result is in agreement with  
436 the findings of LaRivière et al. (2012) and Zhang et al., (2014), who suggested progressive  
437 shoaling of the thermocline over the last 13 Myr.

438 The data indicate two critical transitions at about 5.3 Ma, when the thermocline globally  
439 started shoaling across the equatorial Pacific and around 3.8 Ma from an El-Niño-like-  
440 dominated mean state during the Late Miocene to a La-Niña-like-dominated state during the  
441 Plio-Pleistocene. These transitions coincide with marked regional and global changes.  
442 Locally, the restriction of the Indonesian Seaway and subsequent intensification of the Walker  
443 circulation increased the Pacific zonal temperature gradient over the last 5 Myr. From a global  
444 point of view, the growth of ice sheets in the northern hemisphere and in Antarctica most  
445 likely affected the Walker cell and the low-latitude zonal gradient.

446

447

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833

## FIGURE CAPTIONS

834 **Figure 1. A)** Modern equatorial Pacific sea-surface temperatures (SSTs) from Ocean Data  
835 View. The white dots show the location of the study sites, ODP 806 and IODP U1338. The  
836 gray dots indicate the sites of previously published oxygen-isotope records (ODP 847 and  
837 1241). WPWP, Western Pacific Warm Pool; EPWP, Eastern Pacific Warm Pool; NEC, North  
838 Equatorial Current; SEC, South Equatorial Current; EUC, Equatorial Under Current; NECC,  
839 North Equatorial Counter Current, PC, Peru Current (modified after Pisias et al., 1995). **B)**  
840 Cross sections of temperature (colors) and currents (vectors) averaged over 2.5°S-2.5°N  
841 (modified after US CLIVAR).

842

843 **Figure 2.** Comparison of El Niño-like (in red) and La Niña-like (in blue) time periods  
844 inferred from previous studies using different proxies from the EEP and WEP over the past 12  
845 Myrs.

846

847 **Figure 3. A)**  $\delta^{18}\text{O}$  records in the EEP.  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  at IODP Site U1338 from Rousselle  
848 et al. (2013),  $\delta^{18}\text{O}_{G. \text{sacculifer}}$  and  $\delta^{18}\text{O}_{G. \text{tumida}}$  at site 847 from Wara et al. (2005),  $\delta^{18}\text{O}_{G. \text{menardii}}$   
849 at Site U1338. **B)**  $\delta^{18}\text{O}$  records in the WEP.  $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$  and  $\delta^{18}\text{O}_{G. \text{menardii}}$  at ODP Site  
850 806.

851

852 **Figure 4. A)** Temperature records at IODP Site U1338 (EEP):  $\text{U}^{\text{K}'}_{37}$ -derived SST and  $\text{TEX}_{86}$ -  
853 derived temperatures. The shaded areas represent the error bars of the temperature estimates  
854 (light blue:  $\text{TEX}_{86}$ -derived temperatures and dark blue:  $\text{U}^{\text{K}'}_{37}$ -derived SST). The arrows (black  
855 and blue) highlight the temperature trends. **B)** Isotopic gradient ( $\Delta\delta^{18}\text{O}$ ) between surface and  
856 thermocline waters at IODP site U1338 (EEP) and ODP Site 806 (WEP). The evolution of the  
857 thermocline depth in the EEP and the WEP over the last 10 Ma according to the  $\Delta\delta^{18}\text{O}$  is also

858 shown. C) Temperature records at ODP Site 806:  $U^{K'}_{37}$ -derived SST (Pagani et al., 2010) in  
859 green,  $TEX_{86}$ -derived temperatures (Zhang et al., 2014) in red,  $Mg/Ca_{G.ruber}$ -derived SST  
860 (Medina-Elizalde et al., 2008) in brown and  $Mg/Ca_{G.tumida}$ -derived temperature (Ford et al.,  
861 2015) in orange.