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

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

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ABSTRACT

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

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

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

INTRODUCTION

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

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

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

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

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

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

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

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

OCEANOGRAPHIC SETTING

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

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

MATERIALS AND METHODS

IODP Site U1338 and ODP Site 806

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

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

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

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

Oxygen Stable-Isotope Measurements

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

$\delta^{18}\text{O}$ values are measured on calcareous nannofossils *Noelaerhabdaceae* spp. isolated from the bulk sediment following the procedure of Minoletti et al. (2009). $\delta^{18}\text{O}$ measurements are performed on the 2-5 μm fractions at both sites. For site U1338, we use the $\delta^{18}\text{O}$ data previously published by Rousselle et al. (2013) based on the same protocol as in the present study. $\delta^{18}\text{O}$ values are measured using a Finnigan Delta E mass spectrometer at 50°C, at a temporal resolution of 400 kyr. The quality of the granulometric separation based on 200 particles counting in smear slides is estimated to be at least 75% except between 8 and 9 Ma at Site 806, where *Sphenolitus abies* dominated. These measurements are thus not considered here. In addition, five to seven specimens of *G. menardii* were hand-picked from the > 160 μm fractions for isotopic measurements. Analyses are performed on a VG ISOPRIME at 90°C

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

Alkenone Analyses

Alkenone-derived SSTs of Site U1338 previously reported by Rousselle et al., (2013) are supplemented in the present study with 23 additional samples selected from the Pliocene interval (U1338B-8H5W to U1338B-1H2W) to reach a temporal resolution of 100 kyr. Sedimentary alkenones are analyzed following the procedure described by Ternois et al. (2000). About 5 g of freeze-dried sediments are extracted in a mixture of CH₂Cl₂ / CH₃OH. Alkenones are isolated from the total lipid extract by silica-gel chromatography using solvents of increasing polarity. The fraction containing alkenones is concentrated, transferred into clean glass vials, and evaporated under a nitrogen stream. Gas-chromatography analyses are performed using a Varian 3400CX series equipped with a septum programmable injector (SPI) and a flame ionization detector (FID). We use a fused silica capillary column (Chrompack CP Sil5CB, 50 m long, 0.32 mm internal diameter, 0.25 µm film thickness) and helium as a carrier gas. The alkenone unsaturation ratio $U^{K'}_{37}$ ($C_{37:2}/(C_{37:2} + C_{37:3})$) is converted into SSTs using the Conte et al. (2006) calibration: $T = -0.957 + 54.293 (U^{K'}_{37}) - 52.894 (U^{K'}_{37})^2 + 28.321 (U^{K'}_{37})^3$, which provides more accurate estimates in warm-temperature range than Prahl et al. (1988). Average external precision of the SSTs using this calibration has been estimated at ~ 1.2°C (Conte et al., 2006). Replicate analyses indicate internal precision of ± 0.5°C. Because of the limited amount of sediment material at Site 806 (where alkenone concentrations are extremely low), previously published data from Pagani et al. (2010) provide data covering the past 5 Myr. Note that we recalculated SSTs from the $U^{K'}_{37}$ values of Pagani et al. (2010) using the Conte et al. (2006) calibration to avoid calibration bias between sites (see Supplement).

Forty samples from Site U1338 were analyzed for GDGTs at the Netherlands Institute for Sea Research (NIOZ, Texel, The Netherlands). About 5 g of sediment are freeze dried and extracted in a mixture of CH₂Cl₂ and CH₃OH. The total lipid extracts are separated following the protocol described in Schouten et al. (2002), using Al₂O₃ column chromatography to separate non-polar compounds from the polar GDGTs. Polar fractions are then filtered through a 0.4 µm pore-size filter, evaporated under nitrogen, and diluted to a concentration of 2 mg/ml. GDGTs are analyzed using an Agilent HP1100 (HPLC) equipped with a Prevail Cyano column (3 µm, 150 mm x 2.1 mm) following Schouten et al. (2007). The TEX₈₆ index is calculated using peak areas of the following GDGTs as defined by Schouten et al. (2002): $TEX_{86} = [GDGT2] + [GDGT3] + [GDGT5'] / [GDGT1] + [GDGT2] + [GDGT3] + [GDGT5']$. The TEX^H₈₆, defined as the log(TEX₈₆), is used to calculate temperatures over the past 9 Myr. Temperatures are calculated using the calibration established for temperatures above 15°C by Kim et al. (2010) ($T = 68.4 * TEX^H_{86} + 38.6$). The error on temperature estimates is ± 2.5°C. Other calibrations have been proposed for TEX₈₆ temperatures, in particular based on Bayesian statistics (Tierney and Tingley, 2015) but these yield results similar to those shown here, whereas the Kim et al. (2010) calibration is consistent with other studies from this area (e.g., Seki et al., 2010; Zhang et al., 2014). TEX^H₈₆ - derived temperatures are obtained at a mean temporal resolution of 230 kyr. At Site 806, because of the very low GDGT concentrations, we use previously published data from Zhang et al. (2014) covering the past 12 Myr for comparison with the data from Site U1338.

RESULTS

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

δ¹⁸O Record at Site U1338.---

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

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

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

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

EEP Temperature Reconstructions.

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

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

From the Late Miocene to the Pleistocene, TEX₈₆-derived temperatures (T_{TEX}) indicate a long-term cooling. Between 8 and 5 Ma, T_{TEX} and the SST_{U^{K'}37} vary in the same range of values (between 24°C and 27°C; Fig. 4A), although they are slightly cooler. From approx. 4.5 Ma, the two temperature proxies show marked temperature difference. Note that T_{TEX} increase around 7.5 Ma and 4.8 Ma, reaching values close to SST_{U^{K'}37}.

SST_{U^{K'}37} at Site U1338 are warmer than T_{TEX} by 2 to 3°C on average. Both progressively cool down between 9 Ma and 0.18 Ma but T_{TEX} decreases by 5°C while SST_{U^{K'}37} cools by only 3°C (Fig. 4A).

DISCUSSION

Surface and Subsurface Water Temperatures in the Equatorial Pacific

Warmer SST_{U^{K'}37} than T_{TEX}, as observed at Site U1338, is reported in several studies (e.g., Huguet et al., 2007; Lee et al., 2008; Leider et al., 2010; Lopes dos Santos et al., 2010; Rommerskirchen et al., 2011; Seki et al., 2012; Zhang et al., 2014) and explained by different depth habitat or season of production. However, because seasonality is weak at low latitudes, it is unlikely to explain a temperature differences of up to 4°C, at Site U1338. Unlike haptophytes thriving in the photic zone, *Thaumarchaeota*, the sources of the GDGTs, occur throughout the water column (e.g., Karner et al., 2001; Huguet et al., 2007), and the export of GDGTs can be integrated for a larger part of the upper part of the ocean, likely including a part of the thermocline. Wuchter et al. (2005) reported maximum concentrations of GDGTs between 100 and 200 m depth in the Pacific Ocean, coinciding with high ammonium concentrations below the photic zone (Murray et al., 1999; Karner et al., 2001; Wuchter et al., 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_{TEX} and $\text{SST}_{\text{U}^{K'}_{37}}$ 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_{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_{TEX} reflects the surface temperatures (Sinninghe Damsté
 260 et al., 2002; Wakeham et al., 2004) whereas under upwelling conditions, T_{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_{TEX} may reflect
 266 surface and subsurface conditions in response to different environmental factors (Hertzberg et
 267 al., 2016).

268 In the EEP, the $T_{\text{TEX}}\text{-SST}_{\text{U}^{K'}_{37}}$ temperature gradient (ΔT) is proposed to monitor changes of
 269 the thermocline depth (Seki et al., 2012) assuming that ΔT is driven by the depth migration of
 270 GDGT producers in response to upwelling activity. Therefore, at Site U1338, warming T_{TEX}
 271 and reduced ΔT reflect a deep thermocline and surface export of GDGTs, whereas a more
 272 pronounced cooling of T_{TEX} compared to $\text{SST}_{\text{U}^{K'}_{37}}$, (increased ΔT) reflects a shallow
 273 thermocline, upwelling activity, and subsurface export of GDGTs.

274 In the WEP, the interpretation of $T_{\text{TEX}}\text{-SST}_{\text{U}^{K'}_{37}}$ is not straightforward either. At Site 806,
 275 $\text{SST}_{\text{U}^{K'}_{37}}$ 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 Mg/Ca_{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 $\text{SST}_{\text{U}^{K'}_{37}}$ (Tierney and Tingley 2018) or (2) the uncertainties on the seawater Mg/Ca
 283 correction. However, from 3 Ma onwards, $\text{SST}_{\text{U}^{K'}_{37}}$ 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 $\text{SST}_{\text{U}^{K'}_{37}}$ appear to be linked to a calibration limitation of
 288 the proxy at warm temperatures.

289 Comparison between the T_{TEX} record (Zhang et al., 2014) and subsurface temperatures
 290 derived from 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_{TEX} match with $\text{SST}_{\text{U}^{K'}_{37}}$ and Mg/Ca-derived SSTs from the surface dweller *G. ruber*
 293 (Mg/Ca_{*G.ruber*} Medina-Elizalde et al., 2008), whereas Mg/Ca_{*G.tumida*} temperatures are
 294 systematically cooler than T_{TEX} (Fig. 4C). After 1.3 Ma, T_{TEX} values are intermediate between
 295 Mg/Ca_{*G.ruber*} and Mg/Ca_{*G.tumida*} temperatures, while $\text{SST}_{\text{U}^{K'}_{37}}$ tend to agree with Mg/Ca_{*G.ruber*}
 296 SSTs. It is likely that in the WEP, the T_{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_{TEX} reflect subsurface conditions and that during the Late

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

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

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

Variations in the gradient between T_{TEX} and $\text{SST}_{U^{K'}_{37}}$ also can be used to evaluate changes in the thermocline depth. T_{TEX} and $\text{SST}_{U^{K'}_{37}}$ values similar to those observed between 7.3 Ma and 6.8 Ma, and around 4.8 Ma at IODP Site U1338, indicate a deep thermocline (Fig. 4A). Some intervals are characterized by an increase in ΔT (such as, for example, between 6.8 Ma and 5.3 Ma at IODP Site U1338) associated with a cooling trend of both proxies. These features indicate that GDGTs are produced deeper and that surface waters are concomitantly cooling, which is consistent with upwelling activity.

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

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

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

SST_{U^K37} and T_{TEX} values at U1338 between 7.3 and 6.8 Ma also is consistent with a thick mixed layer in the EEP (Fig. 4A; Seki et al., 2012). In the WEP, Nathan and Leckie (2009) reported a similar decrease in the abundance of surface dwellers in favor of thermocline species, which suggests a deep thermocline. This reduced zonal gradient in SSTs and thermocline tilt across the equatorial Pacific corresponds to El Niño-like conditions, in agreement with earlier findings of Herbert et al. (2016), who indicate an expanded and weak Hadley circulation before 8 Ma.

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

Plio-Pleistocene [from 5.3 Ma] Interval.---

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

348 After 5.3 Ma, SSTs at IODP Site U1338 were warm ($\sim 27^{\circ}\text{C}$), in agreement with a deep
349 thermocline until 4.5 Ma (Fig. 4A). This result is supported by a rapid increase of T_{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 ~ 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, $\text{Mg}/\text{Ca}_{G.\text{ruber}}$ 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

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

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

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

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

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

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

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

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

CONCLUSION

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

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

447

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FIGURE CAPTIONS

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

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

Figure 3. A) $\delta^{18}\text{O}$ records in the EEP. $\delta^{18}\text{O}_{\text{Noelaerhabdaceae}}$ at IODP Site U1338 from Rousselle 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), $\delta^{18}\text{O}_{G. \text{menardii}}$ 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 806.

Figure 4. A) Temperature records at IODP Site U1338 (EEP): $\text{U}^{\text{K}'}_{37}$ -derived SST and TEX_{86} -derived temperatures. The shaded areas represent the error bars of the temperature estimates (light blue: TEX_{86} -derived temperatures and dark blue: $\text{U}^{\text{K}'}_{37}$ -derived SST). The arrows (black and blue) highlight the temperature trends. **B)** Isotopic gradient ($\Delta\delta^{18}\text{O}$) between surface and thermocline waters at IODP site U1338 (EEP) and ODP Site 806 (WEP). The evolution of the 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.