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# Alkenone-based temperature patterns along the eastern South Pacific Coastal Ocean: the effect of upwelling and advection on the sedimentary alkenone unsaturation-index ( $U_{37}^{K'}$ )

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## Abstract

We report that the sedimentary record of alkenone unsaturation-index ( $U_{37}^{K'}$ ) in the eastern South Pacific displays a wide range of temperatures, in response to sea surface temperature (SST), as well as nutrient concentrations. The  $U_{37}^{K'}$  inshore-offshore trend shows low-correlation with SSTs at many transitional sites (50–200 km offshore), and in one oligotrophic coastal site (~20° S). In contrast, in open-ocean sites SSTs play the expected dominant role. The latitudinal trend shows that nitrate, phosphate and/or temperature affect distinctively the  $U_{37}^{K'}$ , suggesting progressive high-impacts in areas with permanent nutrient-stress conditions (~20° S), which is consistent with previous experiments. In contrast, this impact is slightly and not observable in transitional and open-ocean areas, respectively, where the coupling between offshore propagation of mesoscale eddies and upwelling filaments supply additional chlorophyll/nutrient. This study is a first field approximation to the impacts of thermal/non-thermal factors over the unsaturation-index in eastern Boundary Current systems, which would help in future temperature reconstructions.

## 1 Introduction

The alkenone unsaturation-index ( $U_{37}^{K'}$ ) is now accepted as an appropriate proxy of sea surface temperature (SST) and alkenone-integrated production temperature (alkenone-IPT) (Conte et al., 2006) over a wide range of latitudes. The statistically-based core-top calibration for the global ocean (60° S–60° N) (Müller et al., 1998) closely matches the original laboratory calibration for a single-strain 55a of *Emiliania huxleyi* grown in batch culture (e.g., Prahl et al., 1988), even though significant scatter in the overall dataset exists (e.g., Conte et al., 2006). Conte et al. (2006) concluded that the deviation of alkenone-IPT from Levitus estimates was best explained by a combined influence of seasonality in alkenone production and differential degradation of di- and tri-unsaturated alkenones.

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On the other hand, it has been suggested that the degree of variability in  $U_{37}^{K'}$  for a given mean annual SST (maSST) could be caused by non-thermal physiological growth factors such as nutrient- and light-stress which may have an effect on the accuracy of paleotemperature estimates based on  $U_{37}^{K'}$  determinations (e.g., Prahl et al., 2006).

Given the potential for non-thermal physiological effects on alkenone biosynthesis (e.g., Prahl et al., 2006) and for biosynthetic differences within a given genotype of alkenone-producing *Haptophytes* (Rontani et al., 2006),  $U_{37}^{K'}$  recorded in sediments from ocean margins and, in particular, from coastal upwelling areas might show temperature values inconsistent with global ocean core-top calibrations. Furthermore, the  $U_{37}^{K'}$ -signal may also be associated with different nutrient supply pathways produced by horizontal advection and/or vertical upwelling of nutrients due to cyclonic/anticyclonic mesoscale eddies and/or mode-water eddies (e.g., McGillicuddy et al., 2007).

By extrapolating information from laboratory experiments and some field studies we can infer that the  $U_{37}^{K'}$  may vary due to nutrient variability along the Peru-Chile margin, and from coastal to oceanic environments. This aspect motivated our selection of the Peru-Chile margin as a natural setting to explore the potential deviations that the  $U_{37}^{K'}$  may undergo due to the drastically fluctuating environmental conditions of this ecosystem (i.e., upwelling, filaments and eddies) (e.g., Correa-Ramirez et al., 2007).

Data from Eastern Boundary Upwelling Ecosystems are still insufficient, and the potential effects of nutrient variability derived from coastal upwelling and/or advection processes on the  $U_{37}^{K'}$ -signal exported to the sedimentary record is largely unknown. Our results will help in future interpretations of temperature reconstructions in these regions. Moreover, our findings can provide at least partial explanation for scatters and inconsistencies observed between  $U_{37}^{K'}$  measurements and maSST in the Peru-Chile Current System.

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## 2 Methods

Alkenone analysis was performed on the uppermost centimeter of sediment cores ( $n = 39$ ) collected with Multi corer and Box corer during several cruises along the Peru-Chile margin. 5–6 g of freeze-dried sediments samples were extracted by sonication with 25 mL of MeOH for 30 min at room temperature (x2) and centrifuged at 2000 rpm for 3 min. A recovery standard ( $C_{35}$  straight chain ketone) was added to the samples before the extraction. The resultant lipid residue was transferred to a separatory funnel and 25 mL of a mixture of DCM/MeOH was added (3:1) (x3). The extracts were combined and separated with hexane 60 mL (x2) and 40 mL (x1). The hexane fraction was washed first with 40 mL of Milli-Q water (washed w/DCM) and later with a solution of 5% NaCl. The organic phase was dried overnight with  $Na_2SO_4$  anhydrous, and concentrated on a rotavapor. The concentrate was subjected to column chromatography with 7 g of 5% deactivated silica-gel, to separate lipid functional groups of total extract (Prahl et al., 1989). Different fractions were concentrated on a rotavapor and dried with  $N_2$  within a GC vial (with DCM). Fractions were diluted with iso-octane and an internal standard ( $5\alpha$ -cholestane) was added.

The different fractions were analyzed in a GC SHIMADZU, equipped with a capillary column (RTX-SMS, 0.25  $\mu$ m d.f., 0.32 mm i.d.  $\times$  30 m of longitude, RESTEK) and a flame ionization detector (FID). The separation of compounds was development with hydrogen as carrier gas (~10 psig column head pressure, 6.37 mL/min column flow). The temperature regime for gas chromatography was as follows: initial column temperature at 50 °C for 1 min, increased to 120 °C at 30 °C/min, increased to 300 °C at 6 °C/min, isothermal at 300 °C/48 min, total time 81.3 min.

$U_{37}^{K'}$ -index calculations were conducted according to Prahl and Wakeham (1987), and  $U_{37}^{K'}$  values were converted to alkenone-based temperature ( $U_{37}^{K'}\text{-SST}$ ) using the equation of Prahl et al. (1988), i.e.,  $U_{37}^{K'} = 0.034T + 0.039$  (1), where  $T$  = temperature (°C). Analytical accuracy was  $\pm 0.2$  °C.

We also included 44 data points previously published for the area; the complete set of

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samples ( $n = 83$ ) covered the area between  $0^{\circ}$  and  $50^{\circ}$  S (see data supplement: <http://www.biogeosciences-discuss.net/7/545/2010/bgd-7-545-2010-supplement.pdf>). Although the locations studied here display differences in sedimentation rates and bioturbation depths (Muñoz et al., 2004) and may cover the last several hundred years of deposition (e.g., Prahl et al., 2006), they are all of Holocene age, and thus are considered suitable for the objectives of this study. Chlorophyll-a climatology data from the SeaWiFS sensor was derived by time-series files, from September 1997 to August 2007. Maximum/minimum values of Chlorophyll-a concentration were identified for each site along the Peru-Chile margin. The maSSTs and nutrients datasets were extracted from the NOAA World Ocean Atlas 2001 (Conkright et al., 2002). The SeaWiFS data used in this study was acquired using the GES-DISC Interactive Online Visualization AND aNalysis Infrastructure (Giovanni) as part of the NASA's Goddard Earth Sciences (GES) Data and Information Services Center (DISC) (Available at: <http://reason.gsfc.nasa.gov/OPS/Giovanni/ocean.seawifs.shtml/>, 2008).

### 15 3 Results

#### 3.1 Coastal upwelling to open ocean areas overview

The  $U_{37}^{K'}$ -masST relationship for the Peru-Chile margin displays a significant linear regression, but a relatively low-coefficient ( $r^2=0.66$ ). Moreover, the  $U_{37}^{K'}$ /SST does not change seasonally, and is not significantly different from the annual temperatures (summer:  $r^2=0.70$ ; winter:  $r^2=0.71$ ), and the coefficients are also low (Table 1). Thus,  $U_{37}^{K'}$  in our study area would record the masST. The lower regression coefficients found for the study area would be explained by implication of coastal sites that are affected by upwelling-associated nutrient injection/consumption. Simple-linear regressions were developed between the  $U_{37}^{K'}$  vs. masST, nitrate, and phosphate concentrations for the three areas defined above (Fig. 1). Also, the  $U_{37}^{K'}$  vs. masST relationship was compared

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with the original laboratory calibration (Fig. 1) (Prahl et al., 1988). A “poor” correspondence between  $U_{37}^{K'}$  and maSST within the coastal upwelling ( $r^2=0.18$ ) and transition areas ( $r^2=0.37$ ) was observed (Fig. 1). Moreover, regression lines from upwelling and transition areas do not exhibit a close relationship with the original laboratory calibration. On the other hand, only the sites within the coastal upwelling boundary displayed significant correlations between the  $U_{37}^{K'}$  and nitrate and phosphate (Fig. 1). In contrast, the high- $U_{37}^{K'}$ /maSST coefficient in open-ocean areas is consistent with the regional SST-trend and *E. huxleyi* strain 55a culture experiment (Fig. 1).

### 3.2 Potential impact of non-thermal factors on the $U_{37}^{K'}$

In order to evaluate the impact of non-thermal factors on the  $U_{37}^{K'}$ , we calculated the residuals of temperatures for each data point (Fig. 2a). They display a wide range of temperature differences for the study area (between 2 °C and –6 °C, with one data point at –7.9 °C), and a standard deviation of  $\pm 2.3$  °C (Fig. 2a). Our  $\pm 2.3$  °C standard deviation is 0.8 °C higher than the one estimated for the global ocean calibration from Müller et al. (1998) ( $\pm 1.5$  °C, 60° S–60° N, see Table 1), probably due to the several data points with cold  $U_{37}^{K'}$ -SST in our study area (Fig. 2a).

Figure 3 displays the latitudinal distribution of  $U_{37}^{K'}$ -SST from surface sediments along the Peru-Chile margin, and their relationship with the maSST, nitrate and phosphate, chlorophyll-*a* concentration, and the upwelling-index. The latter shows prominent maxima at 15° S and 30° S, and minimum values around 20° S; southward of 40° S is a non-upwelling system (Fig. 3b). Seasonal differences in the upwelling-index are only important off central/southern Chile (~34°–36° S).

Around 25% of the total samples collected in the coastal upwelling and transition areas off Peru (~12° S) and central/southern Chile (~36° S and ~40° S) lie within the standard deviation (black rectangles in Fig. 3a). Coldest  $U_{37}^{K'}$ -SSTs were detected off northern Chile mainly in the coastal upwelling zone at ~20° S, the transition area at

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~28° S, and off central Chile (~34° S). Together, these sites comprise 20% of the total (Fig. 3a). Non-linearity in the  $U_{37}^{K'}$ -SST/maSST relationship is clearly present here (Fig. 3a), and thus this scatter could reflect an influence of non-temperature related factors on the  $U_{37}^{K'}$ .

5 Within the area where the coldest  $U_{37}^{K'}$ -SST were found, the nitrate concentrations were lowest between 20° and 24° S (<1  $\mu$ M), and increased toward the south (~5  $\mu$ M at ~34° S). Phosphate concentrations ranged between ~0.5 and ~0.7  $\mu$ M in this area (Fig. 3a). The area between 20° S and 24° S could be considered as a potential low-nutrient/low-chlorophyll-*a* area (LNLC, Fig. 3a).

10 In the coastal upwelling area, samples with lowest  $U_{37}^{K'}$ /highest maSST (between 20 °C and 22 °C, dashed area in Fig. 1) exhibit a close relationship with low- $U_{37}^{K'}$ /low-nitrate (between 0 and 2  $\mu$ M, Fig. 1). Likewise, the same samples exhibit a similar relationship with low- $U_{37}^{K'}$ /low-phosphate (Fig. 1). This finding would suggest that when nitrate and phosphate are low in the seawater they potentially control the  $U_{37}^{K'}$  rather 15 than temperature, a scenario that would be possible in areas where upwelling events are permanent and nutrient-uptake by phytoplankton occurs year-round (e.g., northern Chile).

20 On the other hand, the samples located between ~30° and ~34° S (Fig. 3a) show a mixed signal potentially derived from significant changes of nutrient input between summer and winter. In this area, the main nutrient input by upwelling events occurs during summer, while winter is a non-upwelling period (Fig. 3b). No-additional contribution of nutrients from riverine sources is expected since large rivers are only found 25 at 36° S and southward (Fig. 3b). However, as mentioned above (see Introduction) additional contribution of nutrients/chlorophyll-*a*, as well as salt and heat, may have an influence in the region due to offshore transport by mesoscale eddies.

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## 4 Discussion

### 4.1 Possible factors controlling the alkenone unsaturation-index signal in surface sediments from coastal upwelling to open-ocean areas

The overall trend of increasing  $U_{37}^{K'}$ /nutrient regressions from the open-ocean to the 5 coast suggests a nutrient effect on the  $U_{37}^{K'}$  in coastal upwelling areas (Fig. 1). In contrast, the effect of temperature on the  $U_{37}^{K'}$  increases offshore (Fig. 1), probably masking the nutrient effect toward open-ocean, and showing that the index faithfully records water temperatures in oceanic areas, as has been shown previously (Müller et al., 1998).

10 From culture experiments Prahl et al. (2003) observed that a decrease in nitrate (from ~20 to 0  $\mu\text{M}$ ) and phosphate (from ~0.5 to 0.1  $\mu\text{M}$ ) concentrations induces a change in the  $U_{37}^{K'}$  from 0.5 to 0.41, and suggested that the effect of nutrient-stress in the seawater changes the  $U_{37}^{K'}$ -SST by ~3 °C. Thus, nutrient-stress condition alone could cause a decrease on  $U_{37}^{K'}$ , producing unusually cold-temperature estimates. Therefore, we may 15 assume that the atypical cold-residual temperatures found in the Peru-Chile upwelling system (Fig. 2a) could mirror the impacts derived from nutrient-stress conditions. Our data can be compared with those previously published for other coastal upwelling systems (e.g., Müller et al., 1998) where similar anomalously low- $U_{37}^{K'}$ -SST have been observed, with residual values that range from -1.5 ° to -5.5 °C for the California margin, 20 and from -1.0 ° to -3.0 °C for the Benguela-Angola margin (Fig. 2b and c). In these areas, the low-values also coincide with low-nitrate concentrations (<2  $\mu\text{M}$ ) (Conkright et al., 2002).

25 Lowest temperatures found in the near coastal area off northern Chile (~20° S) may well be related to nutrient-stress and could be explained by both the year-round low-volume of upwelled nutrient-rich Equatorial Subsurface Waters (ESSW, see upwelling-index in Fig. 3b) limited to the nearshore area (<60 km) (e.g., Strub et al., 1998); and

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between the coastal and the oligotrophic open-ocean areas (e.g., Alvarez-Salgado et al., 2007). The potential accumulation of organic material at the surface waters would favor the growth of bacteria, and hence nutrient regeneration, and, in turn, phytoplankton growth and chlorophyll-*a* increase in oceanic areas (Dandonneau et al., 2003). On the other hand, the vertical nutrient fluxes into the euphotic zone associated with cyclonic eddies, could be an alternative source of nutrients to the oligotrophic open-ocean waters, which should be sufficient to support the new primary production in these areas (e.g., McGillicuddy et al., 2007). The oceanic core-sites around 20° S (Fig. 3a) could be recording a  $U_{37}^{K'}$ -signal derived from potentially healthy cell conditions, due to enough nutrient supply by cyclonic eddies transported offshore. In fact, eddies display the highest kinetic energy values here ( $>100 \text{ cm}^2 \text{ s}^{-2}$ , Fig. 3a) (Correa-Ramirez et al., 2007).

Cold-coastal filaments and mesoscale eddies have been described off northern and central Chile (e.g., Correa-Ramirez et al., 2007). The surface transport/accumulation of organic material to the oceanic oligotrophic areas during summer/winter suggests that the alkenone signature found in surface sediments from oceanic areas reflects both a coupling between nutrient regeneration process by accumulation of allochthonous organic material during winter (Correa-Ramirez et al., 2007), vertical nutrient-pumping from cyclonic eddies (e.g., McGillicuddy et al., 2007), and seaward nutrient-rich upwelling filaments during summer (e.g., Correa-Ramirez et al., 2007). All these mechanisms provide enough nutrients to alkenone-producing algae into oceanic areas, and as a result, the  $U_{37}^{K'}$  is dependent on the temperature regimes solely (Fig. 1). Although the nutrient vertical and horizontal advection could inflict physiological-stress on the alkenone-producing algae (i.e., alkenone/alkenoate ratio) (Prahl et al., 2006), this effect was not observed in the  $U_{37}^{K'}$ -SSTs from open-ocean (Fig. 3a). Alternatively, different alkenone-producing *Haptophytes* species may have different physiological responses, and consequently different effect on the  $U_{37}^{K'}$ -signal (Rontani et al., 2006).

For the transitional area (50–200 km offshore), the pattern of the  $U_{37}^{K'}$ /nutrient re-

gressions (Fig. 1) may represent a signal of temporary advective processes offshore from coastal sites, as follows. Cyclonic/anticyclonic eddies generated along the continental margin drift in a general westward direction through the transitional area off central Chile (30°–36° S), transporting high-chlorophyll-nutrient levels to ~500 km offshore, especially during winter (e.g., Correa-Ramirez et al., 2007). These features are stable for several months (~6 months), and could be a potential transport mechanism of cold-water anomalies from the coastal upwelling areas.

On the other hand, for the transition samples located between 27° S and 32° S, with high-nutrient/low-chlorophyll-*a* conditions (HNLC, Fig. 3a), micronutrients such as iron is the most important limiting factor for the carbon pump (Torres and Ampuero, 2009). Here, riverine discharges are negligible (Fig. 3b). Thus, low-temperatures estimated in this area (Fig. 3a) may be a direct effect of iron-limitation on nutrient uptake processes.

This study is a first field approximation of the possible impacts of thermal/non-thermal factors on the alkenone unsaturation-index in one Eastern Boundary Current System (Peru-Chile), and raises the importance of onshore-offshore advective processes which could be helpful for the interpretation of paleorecords.

## 5 Concluding remarks

The sedimentary  $U_{37}^{K'}$  record along the Peru-Chile margin is a signal derived from ecological and/or physiological changes, as a result of the systematic changes in seawater temperature, nutrient availability conditions, or a combination of both. These changes can be mainly explained by upwelling events and/or offshore transport of heat and high-chlorophyll-*a*/nutrient concentration by mesoscale eddies. Lowest  $U_{37}^{K'}$ -temperatures in the near coastal area off northern Chile (e.g., ~20° S), are shaped by an alkenone-signal from a water column permanently depleted in nutrients. Alternatively, these temperatures also reflect strongest upwelling events, and/or a cold-signal derived from the alkenone-production at the thermocline depth. In contrast, the  $U_{37}^{K'}$ -temperatures at

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~20° S in the open-ocean area describe additional nutrient supply by eddies mechanisms. Moreover, the coupling between the offshore propagation of mesoscale eddies and upwelling filaments explain the  $U_{37}^{K'}$  in agreement with maSSST found in the open-ocean sites, suggesting that the temperature is the controlling factor for  $U_{37}^{K'}$  in these areas. In fact, through the transitional area, the effect of temperature on the  $U_{37}^{K'}$  increases offshore, while the nutrient effect disappears, as result of offshore advection of mesoscale eddies.

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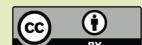
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**Table 1.** Simple linear regression parameters for the relationship  $U_{37}^{K'}$  vs. mean annual sea surface temperature (maSST), nitrate (maNitrate), and phosphate (maPhosphate), respectively.  $U_{37}^{K'} = a + b_1 X$ , for the study area ( $n$ =number of samples;  $r^2$ =regression coefficient).

Relationship	Eq#	a	b <sub>1</sub>	n	r <sup>2</sup>	Reference
<b>Peru-Chile margin</b>						
maSST	1	0.045	0.031	83	0.66*	This study
Summer SST	2	0.033	0.029	83	0.70*	This study
Winter SST	3	0.110	0.030	83	0.71*	This study
<b><math>U_{37}^{K'}</math>-SST agree with annual SST variability</b>						
maSST	4	0.047	0.034	28	0.91*	This study
<b>Upwelling area (&lt;50km)</b>						
maSST	5	0.017	0.285	15	0.18*	This study
maNitrate	6	0.456	0.026	15	0.50*	This study
maPhosphate	7	0.316	0.316	15	0.63*	This study
<b>Transition area (50-200km)</b>						
maSST	8	0.073	0.028	32	0.37*	This study
maNitrate	9	0.512	-0.004	32	0.01	This study
maPhosphate	10	0.424	0.111	32	0.04	This study
<b>Open-ocean area (&gt;200km)</b>						
maSST	11	0.047	0.033	28	0.95*	This study
maNitrate	12	0.633	-0.005	28	0.01	This study
maPhosphate	13	0.559	0.065	28	0.01	This study
<b>Culture experiments</b>						
<i>E. huxleyi</i> (strain 55a)	14	0.039	0.034	22	0.99	Prahl et al. (1988)
<b>Core-Tops</b>						
Global Ocean Calibration (60°N-60°S)	15	0.044	0.033	370	0.98	Müller et al. (1998)

(\*) p<0.05; significant regression

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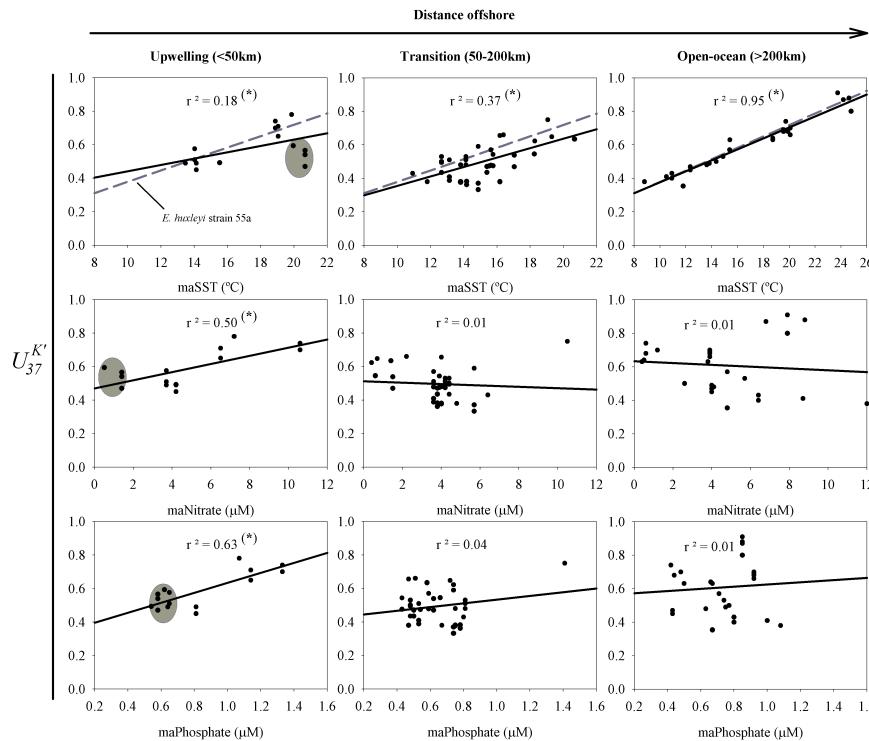
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**Fig. 1.** Linear regressions analyses between  $U_{37}^{K'}$  and mean annual sea surface temperature, nitrate, and phosphate from coastal to open-ocean areas. Grey dotted line represents the batch culture experiment of *Emiliania huxleyi* strain 55a (Prahl et al., 1988). (\*)  $p < 0.05$ ; significant regression. Grey circles denote the lowest  $U_{37}^{K'}$  from upwelling sites. Sediment samples were grouped according to latitude and offshore boundary of coastal upwelling influence. Three areas were defined: (a) coastal upwelling boundary (<50 km offshore); (b) transition area influenced by upwelling filaments and mesoscale eddies (from 50 to 200 km offshore); and (c) open-ocean (>200 km offshore).

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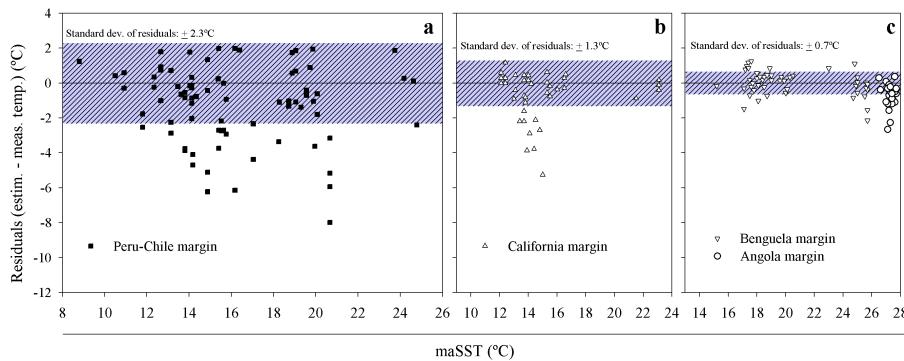
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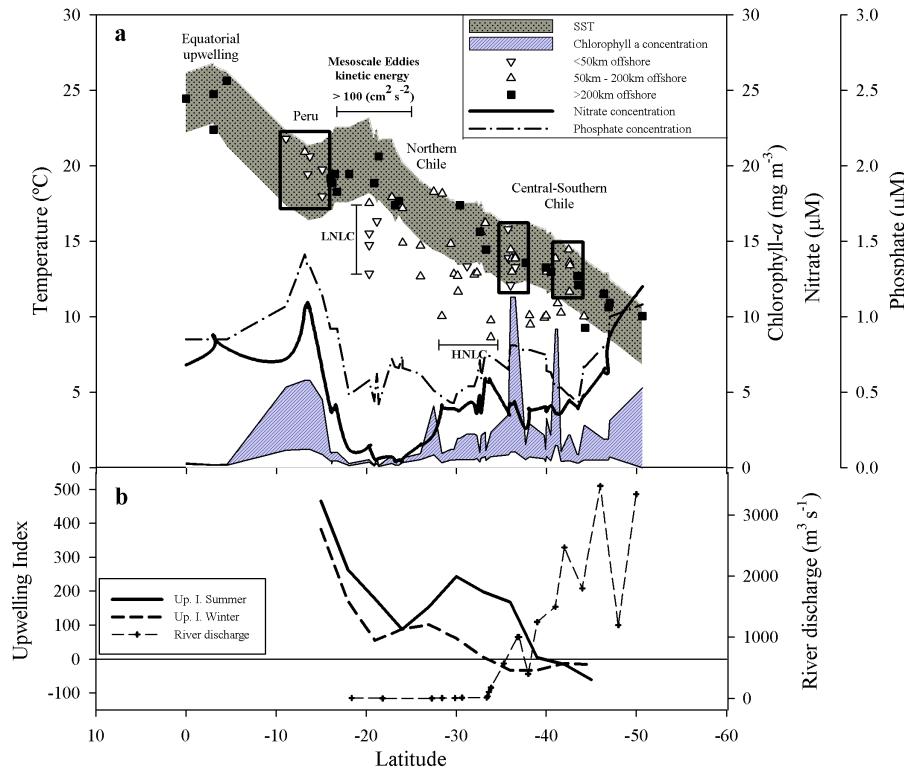


**Fig. 2.** Relationship between the maSST and the residuals of temperatures for three important upwelling areas: (a) Peru-Chile, (b) California and (c) Angola-Benguela margin. Residual temperatures were calculated from the difference between  $U_{37}^{K'}$ -SST and the maSST for each data point.

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**Fig. 3.** Latitudinal distribution of: (a)  $U_{37}^{K'}$ -SST estimated from surface sediments. The grey dotted area displays the standard deviation of the maSST ( $\pm 2.3^{\circ}\text{C}$ ). Rectangles include upwelling points with warm  $U_{37}^{K'}$ -SST. Chlorophyll- $\alpha$  represents the maximum/minimum values from the SeaWiFS sensor. (b) Summer/winter upwelling-index, and river discharges (Dirección General de Aguas, available at: <http://www.dga.cl/>, 2009; Davila et al., 2002). Upwelling-index:  $\text{m}^3$  of water upwelled/100-m coastline/s (available at: <http://www.pfel.noaa.gov/>, 2008).