### Dear Editor;

We have carefully reviewed the comments of the referees from the last revision and we have addressed all of them. As suggested by the reviewers we have complemented the discussion with new references in order to put the data in a regional context. We have improved the age model, and re-organized and re-wrote several paragraphs.

We reduced the introduction and descriptions of the study area and we included a paragraph to discuss Cu concentrations in recent times, according to the instructions of Referee # 1. In addition, we have improved the introduction and discussion section to respond to the comments of both reviewers. Misspelling errors have been also thoroughly checked.

It is important to mention that our records describe very well the observed climatic trends for the region in the Holocene, and our interpretations help to understand the effect of climatic variability on the oxygenation and productivity in the last 8000 years.

We hope that the reviewers positively evaluate the several improvements incorporated in this revised version of the manuscript.

Sincerely yours,

Práxedes Muñoz

On behalf of all authors

#### Referee answer

#### Referee#1

The paper has been greatly improved. I recommend to publish it with the following minor revisions:

- The abstract should be corrected for english. Several inacurate choices of words.

<u>Answer</u>. The revised version of the manuscript has been checked by the Elsevier Language Editing service.

- the introduction is still too long. I suggest deleting all the unfocused generalities from line 73 to line 111.

<u>Answer</u>. The introduction was modified and reduced (was cut in 13 lines). We re-wrote several paragraphs avoiding generalities but considering all the necessary aspects to establish the main objective of the project that was to identify the past variability in the main climatic forcing and oceanographic conditions that affected the area in the last 8000 years through the study of sedimentary records.

- the description of the study area is also long and includes unnecessary generalities. Delete from line 182 to line 190, at least.

<u>Answer</u>. The study area was reduced, 10 lines were eliminated removing repeated ideas that were mentioned in previous paragraphs in the text.

- There is a confusion with the calculated reservoir age: it is presented as a local deviation (DR) in the method (line 241), and as a reservoir age (R) in the results (line 318-322). The values in Carré et al., 2016 and Merino-Campos et al., 2018 are DR values. Check what the age of 411 years really corresponds to (DR or R) and correct the text accordingly (and the age model if necessary).

<u>Answer</u>. The text was corrected, the explanations for calculations were revised and also we changed the modeling method considering the hiatus observed by referee 2.

- line 585-586: I am surprised that the author do not relate the large Cu increase (and to a lesser extent Fe and Mn) in the last century to mining activities. It is said that it indicates an increase of productivity but this is not consistent with other productivity indicators, nor, as they mention, with the increase in oxygenation.

Answer. As suggested the increase of Cu in the last century is now discussed (lines 632 -646).

- the discussion about climatic implications (section 5.3.) is mostly focused on ENSO, which is somewhat speculative since the record does not have an interannual resolution, while in the other hand changes in the mean state, which is indeed what sediments most clearly record on the first order, is not sufficiently discussed. The most robust result here is the long-term trend from dry, disoxigenated, productive conditions in the early Holocene, towards more humid, more oxigenated and less productive conditions at present. This is a strong support for the scenario

proposing that early-midHolocene was characterizaed by a La Nina like mean conditions in response to insolation.

Answer. The discussion is separated in three main sections, first we discussed the main sedimentary composition, and then the temporal variability of the main productivity and oxygenation records, following with the main climatic implications. The PDO and ENSO are the main climatic drivers at centennial and decadal scale at eastern Pacific, we base our discussion in the variability imposed by these forcings, comparing with other studies in the area. We separate the records in periods according to their distribution and we identify similarities with other reports in the region, which based their observations on the variability of these processes. It is recognized that ENSO related variability has played a main role in the climatic expressions, but phases of PDO have also to be taken into account. The movement of the ITCZ depend on both, from which the humid and dry conditions have been predicted, all of them have been reported as the key drivers for the climatic variability from the mid-Holocene until today. We discussed our findings based on this information and highlight the variability of the OMZ effect over the shelf, which is an important characteristic at the East Pacific margin and has a significant impact on main biogeochemical cycles.

The La Niña-like conditions is mentioned in the discussion, but strangely, the authors cite De Pol-Holz et al., 2006 and Kaiser et al., 2008, who found warmer and more oxigenated conditions between 10 and 5ka further offshore in that area, in contradiction with the results presented here. The authors should discuss this discrepancy and present their results in the context of the early to mid Holocene La Niña-like hypothesis which was shown by Koutavas et al., Science, 2002, and by Carré et al., Quat. Int., 2012. This latter paper already discussed some coastal vs offshore discrepancies observed in Chile in this period.

Answer. De-Pol Holz indicated a decreasing trend of the denitrification rate from the early to mid-Holocene, but is not comparable with estimations at previous periods (> 15 kyr BP). The reduced ventilation during the Holocene was attributed to a decreased wind forcing and latitudinal SST gradient, but the higher denitrification rates indicated a more intense OMZ, compared to the estimations in the LGM. However, our sedimentary cores were taken at shallower depths, close to the coast, recording the direct effect of the upwelled waters and the surface variability of the oxygen minimum zone. Besides, the works of the cited authors do not refer directly to increasing oxygen concentrations, but to the OMZ response to the glacier and Patagonian dynamics. In the case of Koutavas et al., they estimated the surface temperatures deduced from the oxygen isotopic composition of foraminifers; they showed that mid-Holocene was the beginning of higher climatic variability. Our records match well with several descriptions for this period (~6500-8000) indicated for several authors, included De-Pol Holz and Koutavas, indicating that this period is the beginning of higher climatic variability towards present.

Several authors indicate clearly the predominance of La Niña at mid-Holocene, and in the case of the SST, the necessary conditions to develop La Niña condition is a higher zonal and meridional SST gradient. At tropics the surface temperature is higher around 21-22°C, in comparison with temperatures at southern sites (18°) in this period. However is relevant to understand that our cores are under the effect of the main upwelling center and the in-shore upwelling waters are cooler (~13°C) than of shore, as mentioned by Carré et al., 2012. Therefore is not a contradiction

to establish northward displacement of the ITCZ at mid-Holocene, which has been discussed by Koutavas et al., 2006 and others.

- Line 616-617: the La Niña-like conditions may be associated to reduced ENSO and a shifted ITCZ, but we cannot say it is caused by them.

Answer. This line was corrected

#### Referee#2

The manuscript by Muñoz et al. presents a multiproxy approach analysis of two short sediment cores retrieved off central Chile to reconstruct paleoceanographic and paleoclimatic variability during the last 8000 years. While several paleoceanographic studies have been conducted in southern Chile and off Peru, paleoceanographic records from central Chile covering the Holocene are extremely scarce. Thus, this study presents valuable data to understand millennial-scale changes in the Eastern South Pacific that can potentially attract the paleoceanographic community. I agree with most of the comments and observations made by the two anonymous reviewers on the first manuscript. After reading the revised version of the manuscript I think that the manuscript still needs substantial work before it is considered for publication in Biogeosciences.

My main criticism, also mentioned by both anonymous reviewers, is that the manuscript lacks a proper paleoceanographic interpretation combining relevant paleorecords from the Peruvian Upwelling system and from southern Chile. This could help to disentangle high latitude versus tropical forcing driving the observed changes in the data. During the last decade multiple studies focused on the Peruvian upwelling system were published that could help to gain insights on the mechanisms driving millennial-scale changes off Central Chile. Surprisingly these studies were not included in the revised version of the manuscript.

Answer. We have now complemented the discussion with new references in order to give a major scope of the data in a regional context, considering some works developed in south-central Chile and Peru. However this is limited to the mid and late-Holocene, and some information for the most recent time (two last centuries). Our cores record only ~8000 years and the sedimentation rates were relatively low, avoiding to obtain a higher resolution in recent times. Notwithstanding, our records establish relevant episodes of environmental changes that have been also documented at southern and far northern areas of our study site. In addition, this record is also in agreement with climatic descriptions for the south-central Chile and northern regions up to Peru. We consider that this version now includes a thorough overview of climate variability in the region, as requested by the reviewer, for the time interval recorded in our cores.

The second main problem that I see with the manuscript is that the Discussion lacks focus, probably because of the lack of a clear scientific question and the multiple types of proxies presented by Muñoz et al. I suggest including a clear scientific question in the Introduction to better guide the reader throughout the manuscript.

Answer. The aim of this project was to establish past variability of in the main oceanographic and climatic features in the study area a semiarid zone in the north-central Chile. The climatic fluctuations produce humid and arid conditions along the SE Pacific where the intensity of the wind remains the key factor for the strength of the upwelling and, therefore, for the supply of nutrients to the photic zone, being required for the development of the primary productivity. In our area, a main relevant upwelling zone has an impact over a narrow shelf and coastal zone along the year. In the same context, the oxygen conditions at bottoms and within the sediments are highly dependent on the oxycline, that normally is related with the intensity of upwelling and the oxygen consumption during the organic matter diagenesis, and the position of the oxygen minimum zone (OMZ). These characteristics have been described in the Introduction and Study Area sections, indicating the work done to establish the relationship between proxies measured in the sedimentary records with the main climatic variability described for the region. Our data indicates the greater relevance of the OMZ in the shelf over the oxygen consumption derived from the degradation of primary productivity, and that the latter had a variability that was not consistent with the variation in oxygen. Therefore, the oxygenation of the bottoms has been related with the variability of the oxycline position, which is affected today during warm events (El Niño), leading to the biggest changes in the biggeochemical cycles by the deepening of the oxycline and lower productivity. This is consistent with the main climatic shifts reported for the region from the mid-Holocene. Our work has showed the relevance of climatic variability at larger scales, both temporal and spatial, in the main environmental conditions in our region.

### Some other issues that need to be addresses are:

1) I am completely surprised that coastal sediment cores of just ~110 cm long can contain records of ~8000 years, especially in areas associated with high marine productivity. After examining the age models and Fig. 5, it is evident that the sediment cores present large discontinuities or sections with extremely low sedimentation rates like during the mid-Holocene in core BGGC5. This observation merits an in-depth discussion in the manuscript given that Holocene records from the Peruvian Upwelling systems are well known for discontinuities and erosion events (e.g. Erdem et al. 2016). Specifically, what is the role of winnowing on the proxies presented in this manuscript?

Answer. The study area is considered as moderated in productivity and in consequence, the rate of organic carbon is lower than for other upwelling areas (e.g., off Concepción 36°S), however the upwelling here is perennial and the sedimentation rate is comparable to other shelf areas in northern and southern Chile (Muñoz et al., 2004), thus with these low accumulation rates we can confidently say that our cores contain the material accumulated in the last ~8000 years. Our sediments did not showed evidence of slumps, but the very low sedimentation rate in two sections of the core should be interpreted as a discontinuity for erosion, as was suggested by the reviewer. We analyzed the cores taking into account these discontinuities and thus we used other age modeling.

2) The authors show records of Mo, U and Re for both cores to infer changes in paleooxygenation. The use of Mo/U systematics is a powerful tool to reconstruct sub-oxic and anoxic conditions (Algeo and Tribovillard, 2006; Scholz et al., 2011; Salvatteci et al. 2016;). I recommend the authors to use this strategy to infer paleo-oxygenation (Section 5.3).

**Answer.** Figure 9 shows the U/Mo ratios, and we also include the Re/Mo. In addition, enrichment factors of several elements were also showed, indicating the relevance of authigenic processes related with the oxygen conditions at bottoms and the presence of sulfides. The Cd and Mo enrichment account for it. All were showed in the context of more or less reduced conditions at bottoms, productivity and climatic variability. We also compared our findings with the data reported for south central Chile and Peru at shallower zones over the shelf.

3) The results of some paleoceanographic studies need to be compared and contrasted with the results presented by Muñoz et al. to gain a mechanistic understanding of the processes driving the temporal trends in the data. For example, there are very few records of Mo, U and Re in the Eastern South Pacific (e.g. Muratli et al., 2010; Scholz et al., 2014; Salvatteci et al., 2016). A comparison and discussion with these records seems mandatory to better understand the mechanisms driving oxygen changes off central Chile. Other relevant studies to be consulted are: Doering et al. (2016), Mollier-Vogel et al. (2018), Salvatteci et al. (2016); Salvatteci et al. (2019).

Answer. The major variability observed for atmospheric conditions from the mid-Holocene onwards, reflected the main oceanographic features described for the eastern Pacific. We used pollen records to help us obtaining better approximations of climatic variability in the region, and our records fix very well with continental records in Laguna Aculeo and other data from Los Vilos, identifying dry/humid conditions in the same periods than our marine records, and in agreement to the millennial variability observed for all the region, from the north to central Chile. We used marine information than describes productivity variations and changes in the reduced conditions in the seafloor, the later related with the effect of the OMZ over the shelf. According with new references, some works have established that the ventilation of the OMZ occurs during the deglaciation, and during climatic shifts that operated off Peru at mid-Holocene, however, the variability of its position over shallower zones has not been established yet. We think that our work contributes to establish the variability of the oxycline over the shelf related with the deepening of the OMZ.

4) The authors should carefully revise the manuscript and the references for mistakes. There are several typos in the text (e.g. the use of BC and BP; misspelling names in the references, etc).

**Answer.** We checked all typing errors

- 1 Reconstructing past variations in environmental conditions and paleoproductivity
- 2 over the last ~8000 years off north-central Chile  $(30^{\circ} \text{ S})$

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### Abstract

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37 The aim of this project was to establish past variations in the main oceanographic and climatic features of a transitional semi-arid ecosystem in the north-central Chilean coast. 38 We analyzed recent sedimentary records retrieved from two bays, Guanaqueros and 39 40 Tongoy (30° S), for geochemical and biological analyses, including the following: sensitive redox trace elements, biogenic opal, total organic carbon (TOC), diatoms, and 41 stable isotopes of organic carbon and nitrogen. Three remarkable periods were 42 established with different environmental conditions and productivities: (1) > cal BP 43 44 6600, (2) cal BP 4500–1800, and (3) cal BP 100 to the present (CE 2015). The first period was characterized by a remarkably higher productivity (higher diatom 45 abundances and opal) in which large fluxes of organic compounds were also inferred 46 from the accumulation of elements, such as Ba, Ca, Ni, Cd, and P in the sediments. 47 48 Meanwhile, significantly reduced conditions at the bottom of the bays were suggested based on the large accumulation of Mo, Re, and U, showing a peak at cal BP 6600 when 49 sulfidic conditions could have been present. According to the pollen moisture index, 50 51 this was also identified as the driest interval. These conditions should be associated with 52 an intensification of the Southern Pacific Subtropical Anticyclone and stronger southerly western winds, emulating the La Niña-like conditions, as has been described 53 54 for the SE Pacific during the early Holocene and part of the mid-Holocene. During most 55 of the second period, lower productivity was observed; however, a small increase was identified between Cal BP 3400 and 4000, although lower amounts of diatom (valves g 56 1) and nutrient-type metal accumulations were evident. Anoxic conditions at the bottom 57 of the bays changed to an almost stable sub-oxic condition during this time interval. The 58 59 third period was marked by intense oxygenation after cal BP 1800, as observed by a drastic change in the accumulation of U, Mo, and Re. This was followed by a return to 60 more reduced conditions over the past two centuries, characterized by a small 61 62 productivity rise after cal BP ~130, as suggested by the opal accumulations. Overall, 63 lower primary productivity, lower reduced conditions at the bottom, and higher humidity conditions were established after cal BP 6600 to the present. We suggest that 64 65 the oxygenation might be associated with a weak effect from the oxygen minimum zone over the shelf and intensified El Niño activity, introducing oxygenated waters to the 66 coastal zones through the propagation of equatorial waves and establishment of 67

- conditions that reduced the primary productivity from the mid-Holocene toward the
- beginning of the modern era.
- 70 Keywords: paleoproductivity, paleoredox, trace metals, diatoms, opal, organic carbon,
- 71 Coquimbo, SE Pacific

#### 1. Introduction

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- 75 The mean climatic conditions in the SE Pacific are modulated by the dynamics of the
- 76 Southern Pacific Subtropical Anticyclone (SPSA) and Humboldt Current System. The
- 77 coastal wind pattern produced alongshore varies along the SE Pacific, showing lower
- seasonality between 18°-30° S and producing semi-permanent upwelling (Pizarro et al.,
- 79 1994; Figueroa and Moffat, 2000). This system is highly affected by the inter-annual
- 80 variability imposed by the El Niño Southern Oscillation (ENSO), impacting the wind
- intensity and, therefore, the productivity (Ruttland and Fuenzalida, 1991; Blanco et al.,
- 82 2002). Other climate patterns demonstrate impacts at longer timescales (inter-annual,
- decadal, inter-decadal), such as the Pacific Decadal Oscillation (PDO) and the Southern
- 84 Annular Mode (SAM). These patterns modify the strength and position of the southerly
- 85 western winds (SWW), producing cold/warm periods that affect mainly winter
- precipitation during the positive/negative trends of the SAM and lead to intense/weak
- 87 upwelling (Quintana and Aceituno, 2012; Ancapichún and Garcés-Vargas, 2015). In
- 88 addition, the orbitally induced variations in the austral insolation influences the extent
- 89 of the Antarctic sea ice and the Hadley cell, which act as important forces in the
- 90 latitudinal displacement of the Inter-tropical Convergence Zone (ITCZ; Kaiser et al.,
- 91 2008, and references therein). These fluctuations produce humid and arid conditions
- along the SE Pacific where the intensity of the wind remains the key factor in the
- 93 upwelling strength and, therefore, the supply of nutrients to the photic zone, all of which
- are required for the development of the primary productivity.
- 95 Off Coquimbo (30° S), there is normally semi-permanent and intense upwelling forced
- by local winds, strongly influenced by topographic features (Figueroa and Moffat,
- 97 2000) and ENSO variability (Schaffer et al., 1997; Escribano et al., 2004). During El
- Niño, the intensities of the mean winds alongshore are reduced (conversely, during La
- Niña) (Rahn and Garreaud, 2013), impacting the upper circulation of the ocean and
- affecting the oxygenation of the water column and strength of the upwelling. The high
- 101 productivity that takes place close to the coast during normal periods (Escribano et al.,

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Comentario [A2]: Lines were modified

102	2004 and references therein) maintains a zone of low dissolved oxygen content,	
103	reinforcing the oxygen minimum zone (OMZ; Helly and Levin, 2004, Ulloa et al.,	
104	2012); however, the opposite occurs during El Niño, in which oxygenated waters enter	
105	the coastal zone provided by the narrow continental shelf (Helly and Levin, 2004). This	
106	changes the normal suboxic conditions at the bottom, normal composition of	
107	macrofauna, and related geochemical characteristics of the sediments, with implications	
108	that persist for several years after the event (Gutiérrez et al., 2006; Sellanes et al., 2007).	Comentario [A3]: Lines were modified
109	These changes in primary productivity and oxygenation at the bottom can be observed	
110	in the sedimentary records that respond to the amount of organic carbon that has settled	
111	on the surface sediments under different oceanographic and climatic conditions. The	
112	diagenetic reactions during organic matter remineralization produce the enrichment or	
113	depletion of trace elements, which reflects the amount of settled organic matter but also	
114	reinforces the low oxygen conditions imposed by the OMZ, all of which promotes the	
115	enrichment or depletion of trace elements (Tribovillard, 2006). Their variability in	
116	sedimentary records has been extensively used to establish temporary changes in	
117	primary productivity and changes in the oxygenation at the bottom (Nameroff et al.,	
118	2002; Zheng et al., 2002; McManus et al., 2006; Siebert et al., 2003).	Comentario [A4]: Paragraph was modified
119	North-central Chile is a semi-arid zone that does not receive large fluvial contributions,	modified
120	except during abnormal periods such as in El Niño years, during which higher runoff	
121	has been recorded in austral winter (Valle-204; Levinson et al., 2000; Montecinos and	
122	Aceituno, 2003; Garreaud et al., 2009). Under this scenario, marine sediments are often	
123	highly influenced by primary production in the water column and terrestrial runoff;	
124	therefore, sedimentary records can reveal the past variability in primary production and	
125	oceanographic conditions over the shelf, which ultimately respond to the major	
126	atmospheric patterns in the region. We considered that redox trace elements off	Comentario [A5]: Paragraph modified
127	Coquimbo ( $30^{\circ}$ S) respond to changes in the local hypoxia (U, Mo, and Re); in addition,	
128	the nutrient-type elements are assumed to have followed the organic flux variability of	
129	the sediments (Ba, Ni Cu), according to the interannual and interdecadal variability	
130	described for the climatic and oceanographic settings in the region. Similarly, we	
131	measured Ca, K, and Pb to assess the terrigenous inputs from runoff and aeolian	
132	transportation, which is also impacted by Fe and Mn (Calvert and Pedersen, 2007). Ca	
133	accumulation depends, in turn, on carbonate productivity and dissolution, and has also	
134	been used as a paleoproductivity proxy (Paytan, 2008; Govin et al., 2012). We	Comentario [A6]: Paragraph modified
135	determined the enrichment/depletion of elements to establish the primary prevailing	

environmental conditions during the sedimentation of particulate matter (Böning et al., 2009). In addition, we considered the diatom assemblages with biogenic opal as a measurement of siliceous export production, total organic carbon (TOC), and stable isotopes to identify variations in the organic fluxes to the bottom. Moreover, pollen grains were used to identify environmental conditions based on the climate relationship of the main vegetation formations in north-central Chile. Based on our records we were able to identify wet/dry intervals, periods with high/low organic fluxes to the sediments, which are related to changes in primary production, and changes in the redox conditions at the bottom, which in turn, have been associated with the main climatic conditions described for the Holocene in this region.

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# 2. Study area

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The Coquimbo area (29–30 °S), in the southern limit of the north-central Chilean 148 149 continental margin, constitutes a border area between the most arid zones of northern 150 Chile (Atacama Desert) and the more mesic Mediterranean climate in central Chile 151 (Montecinos et al., 2016). Here, the shelf is narrow, and several small bays trace the 152 coast line. The Tongoy and Guanaqueros bays are located in the southern edge of a broad 153 154 embayment between small islands to the north (29 °S; Choros, Damas, and Chañaral) 155 and Lengua de Vaca Point to the south (30 °S) (Fig. 1), protected from southerly winds that are predominant in the region. Tongoy Bay is a narrow marine basin (10 km at its 156 maximum width) with a maximum depth of approximately 100 m. To the northeast lies 157 158 Guanaqueros Bay, a smaller and shallower basin. High wind events are evenly 159 distributed throughout the year and promote an important upwelling center at Lengua de 160 Vaca Point, resulting in the accumulation of high biomass along a narrow coastal area (Moraga-Opazo et al., 2011; Rahn and Garreaud, 2013) that reach concentrations of 161 approximately 20 mg m<sup>-3</sup> (Torres and Ampuero, 2009). In the shallow waters of Tongoy 162 163 Bay, the high primary productivity results in high TOC in the water column, allowing for the deposition of fine material to the bottom; TOC rises concurrently with periods of 164 165 low oxygen (Fig. 2; Muñoz et al., unpublished data). Recent oceanographic studies 166 indicate that low dissolved oxygen water intrusions from the shelf (Fig. 3) seem to be related to lower sea levels, resulting from annual local wind cycles at a regional meso-167 scale (Gallardo et al., 2017). Oceanographic time series indicate that transition times 168

develop in short periods due to changes in the direction and intensity of the winds along

170 the coast, with strong seasonality (http://www.cdom.cl/boyas-oceanograficas/boyatongoy). The spatial and temporal variability of these processes is still under study. In 171 addition, oceanic variability along the western coast of South America is influenced by 172 173 equatorial Kelvin waves on a variety of timescales, from intra-seasonal (Shaffer et al., 174 1997) and seasonal (Pizarro et al., 2002; Ramos et al., 2006), to inter-annual (Pizarro et al., 2002; Ramos et al., 2008). 175 Sedimentological studies are scarce with regard to the north-central shelf of Chile. A 176 177 few technical reports indicate that sediments between 27° S and 30° S are composed of very fine sand and silt with relatively low organic carbon content (< 3 and ~5%), except 178 179 in very limited coastal areas where organic material accounts for approximately 16% of 180 the total material (Muñoz, unpublished data; FIP2005-61 Report, www.fip.cl). Coastal 181 weathering is the main source of continental input owing to scarce river flows and little rainfall in the zone (0.5–80 mm y<sup>-1</sup>; Montecinos et al., 2016, Fig. 1). Freshwater 182 discharges are represented by creeks, which receive the drainage of the coastal range 183 forming wetland areas in the coast and even small estuaries, such as Pachingo, located 184 south of Tongoy (Fig. 1). These basins cover ~300 and 487 km<sup>2</sup>, respectively. The water 185 volume in the estuaries is maintained by the influx of seawater mixed with the 186 groundwater supply. Normally, a surface flux to the sea is observed. Freshwater 187

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# 3. Materials and methods

### 192 **3.1. Sampling**

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193 Sediment cores were retrieved from two bays in the Coquimbo region: Bahía

the coastal zone (Direccion General de Aguas, 2011).

Guanaqueros (core BGGC5; 30° 09' S, 71° 26' W; 89 m water depth) and Bahía Tongoy

discharges only occur through dry creeks that drain water during high rainfall periods in

- 195 (core BTGC8; 30° 14' S, 71° 36' W; 85 m water depth) (Fig. 1), using a gravity corer
- 196 (KC-Denmark) during May 2015, onboard the L/C Stella Maris II owned by the
- 197 Universidad Católica del Norte. The length of the cores was 126 cm for BGGC5 and 98
- 198 cm for BTGC8.
- 199 Subsequently, the cores were sliced into 1 cm sections, and subsamples were separated
- 200 for grain size measurements and determination of magnetic susceptibility, trace element
- and biogenic opal concentrations, C and N stable isotope signatures ( $\delta^{13}$ C,  $\delta^{15}$ N), and
- TOC content. The samples first were kept frozen (-20° C) and then freeze-dried before
- 203 laboratory analyses.

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205	3.2. Geochronology ( <sup>210</sup> Pb and <sup>14</sup> C)	
206	A geochronology was established combining ages estimated from <sup>210</sup> Pb <sub>xs</sub> activities	
207	suitable for the last 200 years and radiocarbon measurements at selected depths for	
208	older ages. The quantification of <sup>210</sup> Pb activities was performed through the alpha	
209	spectrometry of its daughter <sup>210</sup> Po following the procedure of Flynn (1968). The	
210	(unsupported) activities of $^{210}\text{Pb}_{xs}$ were determined as the difference between the $^{210}\text{Pb}$	
211	and <sup>226</sup> Ra activities measured in some intervals of the sediment column. Meanwhile,	
212	<sup>226</sup> Ra was measured by gamma spectrometry at the Laboratoire Géosciences of the	
213	Université de Montpellier (France). Standard deviations (SD) of the <sup>210</sup> Pb inventories	
214	were estimated by propagation of the counting uncertainties (Bevington and Robinson,	
215	1992) (Table S1, supplementary data). The ages were based on the Constant Rate of	
216	Supply Model (CRS, Appleby and Oldfield, 1978).	
217	Radiocarbon measurements were performed on a mix of planktonic foraminifer species	
218	in core BGGC5, whereas the benthic foraminifer species Bolivina plicata was selected	
219	for core BTGC8 (Table 1). The samples were submitted to the National Ocean Sciences	
220	AMS Facility (NOSAMS) of the Woods Hole Oceanographic Institution (WHOI). The	
221	timescale was obtained from $^{210}\text{Pb}_{xs}$ and $^{14}\text{C}$ measurements and from Bacon age-depth	
222	modeling open source software (Blaauw and Christen, 2011), considering the Marine	
223	curve <sup>13</sup> C (Reimer et al., 2013) (Fig. 4) and a reservoir deviation from the global mean	
224	reservoir age of 441 $\pm$ 35 y. This was estimated subtracting the $^{14}$ C age value	
225	corresponding at the historical dates 1828 AD and 1908 AD (499 $\pm$ 24 and 448 $\pm$ 23 $^{14}$ C	
226	y, respectively, Reimer et al., 2013) from the apparent <sup>14</sup> C age of the foraminifers	
227	measured at depths of 5 and 10 cm for cores BTGC8 and BGGC5, respectively	
228	(Sabatier et al., 2010; Table 2).	
229		
230	3.3. Geophysical characterization	
231	The magnetic susceptibility (SI $\times$ 10 <sup>-8</sup> ) was measured with a Bartington Susceptibility	
232	Meter MS2E surface scanning sensor at the Sedimentology Laboratory at Centro Eula,	
233	Universidad de Concepción. Mean values from three measurements were calculated for	

The grain size was determined using a Mastersizer 2000 laser particle analyzer (Hydro

2000-G, Malvern) in the Sedimentology Laboratory at Universidad de Chile. Skewness,

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each sample.

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- 237 sorting, and kurtosis were evaluated using the GRADISTAT statistical software (Blott
- and Pye, 2001), which includes all particle size spectra.

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# 3.4. Chemical analysis

- 241 Trace element analyses were performed via inductively coupled plasma-mass
- 242 spectrometry (ICP-MS) using an Agilent 7700x at Université de Montpellier (OSU
- OREME/AETE regional facilities). The analysis considered reference materials (UBN,
- BEN, and MAG1) that had an accuracy higher than ±5%; the analytical precisions were
- between 1% and 3%. Internal standardizations with In and Bi were used to deconvolve
- 246 the mass-dependent sensitivity variations of both matrix and instrumental origin
- occurring during the course of an analytical session. The analytical precisions attained
- were between 1% and 3%.
- TOC and stable isotope ( $\delta^{15}$ N and  $\delta^{13}$ C) analyses were performed at the Institut für
- 250 Geographie, Friedrich Alexander Universität (FAU) Erlangen-Nürnberg, Germany
- using a Carlo Erba elemental analyzer NC2500 and an isotope-ratio-mass spectrometer
- 252 (Delta Plus, Thermo-Finnigan) for isotopic analysis. Stable isotope ratios were reported
- 253 in the  $\delta$  notation as the deviation relative to international standards (Vienna Pee Dee
- Belemnite for  $\delta^{13}$ C and atmospheric N<sub>2</sub> for  $\delta^{15}$ N); thus,  $\delta^{13}$ C or  $\delta^{15}$ N = [(R sample/R
- standard) 1]  $\times$  10<sup>3</sup>, where R is  $^{13}$ C/ $^{12}$ C or  $^{15}$ N/ $^{14}$ N, respectively. The typical precision
- of the analyses was  $\pm 0.1\%$  for  $\delta^{15}$ N and  $\delta^{13}$ C.
- 257 Biogenic opal was estimated following the procedure described by Mortlock and
- 258 Froelich (1989). The analysis was performed by molybdate-blue spectrophotometry
- 259 (Hansen and Koroleff, 1999), conducted at the laboratories of Marine Organic
- 260 Geochemistry and Paleoceanography, University of Concepción, Chile. Values for
- biogenic opal were expressed by multiplying the Si (%) by 2.4 (Mortlock and Froelich,
- 1989). The analytical precision was  $\pm$  0.5%. Accumulation rates were determined based
- on the sediment mass accumulation rates and amount of opal for each core section in %.

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### 3.5. Microfossils analyses

- 266 Qualitative abundances of siliceous microfossils were determined for every 1 cm
- 267 following the Ocean Drilling Program (ODP) protocol, described by Mazzullo and
- 268 Graham (1988). This information was used to select sections every 4, 8, and 12 cm for
- 269 BGGC5 and every 6 cm for BTGC8, to determine quantitative abundances of

- 270 microfossils (diatoms, silicoflagellates, sponge spicules, crysophyts, and phytoliths).
- 271 Roughly 0.5 g of freeze-dried sediment was treated according to Schrader and Gersonde
- 272 (1978) for siliceous microfossils. They were identified and counted under an Olympus
- 273 CX31 microscope with phase contrast, in which 1/5 of the slides were counted at 400X
- 274 for siliceous microfossils and one transect at 1000X was counted for Chaetoceros
- 275 resting spores (Ch. RS). Two slides per sample were counted with an estimated
- 276 counting error of 15%. Total diatom abundances are given in valves g<sup>-1</sup> of dry
- 277 sediments.
- 278 Pollen analysis was conducted following the standard pollen extraction methodology
- 279 (Faegri and Iversen, 1989). The identification was conducted under a stereomicroscope,
- with the assistance of the Heusser (1973) pollen catalog. A total of 100–250 terrestrial
- pollen grains were counted in each sample. The pollen percentage for each taxon was
- 282 calculated from the total sum of terrestrial pollen (excluding aquatic taxa and fern
- spores). Pollen percentage diagrams and zonation were generated using the Tilia
- 284 software (Grimm, 1987).
- We further summarize pollen-based precipitation trends by calculating a pollen moisture
- 286 index (PMI), which is defined as the normalized ratio between Euphorbiaceae (wet
- 287 coastal scrubland) and Chenopodiaceae (arid scrubland). Thus, a positive (negative)
- value for this index point corresponds to relatively wetter (drier) conditions.

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# 4. Results

### 4.1. Geochronology

- The activity of <sup>210</sup>Pb<sub>xs</sub> (unsupported) was obtained from the surface to a depth of 8 cm
- in the two cores, with an age of ~AD 1860 at 8 cm in both (Table S1). Greater surface
- activities were obtained for core BGGC5 (13.48  $\pm$  0.41 dpm g<sup>-1</sup>) than core BTGC8 (5.80
- $\pm$  0.19 dpm g<sup>-1</sup>), showing an exponential decay with depth (Fig. 4). A recent
- sedimentation rate of  $0.11 \pm 0.01$  cm y<sup>-1</sup> was estimated.
- 297 The age—depth model provided a maximum age of cal BP 7990 for core BGGC5 and cal
- BP 8012 for core BTGC8 (Fig. 4). A mean sedimentation rate of  $0.026 \pm 0.012$  cm y<sup>-1</sup>
- was estimated for core BGGC5, with a period of relatively low values (< 0.01 cm y<sup>-1</sup>)
- 300 between cal BP 240 and 1500 and between cal BP ~5000 and 6400. This variation in the
- accumulation rates occurred over a few centimeters (5 and 7 cm, respectively); thus, this
- 302 rapid decrease was considered as a hiatus in the age-depth modeling. The model
- estimates the accumulation rates before and after the hiatus not auto-correlated,

obtaining variable sedimentation rates which are more accurate to the sedimentation process. We could not resolve the length and time of hiatuses; we assumed an elapsedtime of 1400 years based on the difference between the radiocarbon ages before and after the hiatus and a mid-depth corresponding to those gaps. Although we did not have stratigraphic evidence of these discontinuities in the sediment core, we believe that the assumptions considered allowed the development of reasonable age-depth models. Nevertheless, the interpretations of the proxy records were taken with caution in these age ranges. For BTGC8, mean sedimentation rates were less variable in the entire core at  $0.013 \pm 0.006$  cm y<sup>-1</sup>. The local reservoir deviation values were close to the global marine reservoir (Table 2) and higher than other estimations along the Chilean margin at shallower depths (146 ± 25 years at < 30 m water depth; Carré et al., 2016; Merino-Campos et al., 2018). Our coring sites are deeper (~90 m water depth) and influenced by upwelling water from Lengua de Vaca Point, which could explain such differences. However, moderate differences were observed between the models using both reservoir values. Thus, our estimations were based on two pre-bomb values established with <sup>210</sup>Pb measured in sediments and <sup>14</sup>C in foraminifers, used for the age modeling.

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### 4.2. Geophysical characterization

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Sediments retrieved from the bays showed fine grains within the range of very fine sand to silt in the southern areas. There, grain size distribution was mainly unimodal, very leptokurtic, more sorted, and skewed to fine grain when compared with sediments from the northern areas. Sediment cores obtained from the northern areas were sandy (coarse sand and gravel) with abundant calcareous debris. Longer cores of soft sediment were retrieved at the southernmost areas (BGGC5 and BTGC8, Fig. 1), where the silty component varied between 40% and 60% (Figs. 5a, 5b). The clay component was very low at both cores (< 2%). The sediment's color ranged from very dark grayish brown to dark olive brown (2.5Y 3/3-3/2) in Guanaqueros Bay (BGGC5) and from dark olive gray to olive gray (5Y 3/2-4/2) in Tongoy Bay (BTGC8). Visible macro-remains (snails and fish vertebrae) were found, as well as weak laminations at both cores. The magnetic susceptibility showed higher values close to the surface, up to  $127 \times 10^{-8}$  SI at BGGC5, and lower values ( $85 \times 10^{-8}$  SI) at BTGC8. At greater depths, however, the values were very constant, at  $5-8 \times 10^{-8}$  SI at BGGC5 core and  $12-20 \times 10^{-8}$  SI at BTGC8 core. In both cores, susceptibility rose substantially in the last century (Figs. 5a, 5b). Lower bulk densities were estimated at core BGGC5 (0.7-0.9 g cm<sup>-3</sup>), compared with core BTGC8

- $(> 1 \text{ g cm}^{-3})$  (Figs. 5a, 5b). Consistent with this, the mean grain size amounted to 60-80
- 339 μm in Guanaqueros Bay (BTGC8), compared with 50-60 μm in Tongoy Bay
- 340 (BGGC5). Both cores were negatively skewed, with values of −1 to −1.2 at BGGC5,
- and -1 to -2.5 at BTGC8. Minor increases toward coarser grain size were observed over
- 342 the past ~1000 years, especially in Tongoy Bay (BTGC8). In both cases, grain size
- 343 distributions were strongly leptokurtic. The Ca/Fe ratio also reduced with time, except
- at core BTGC8 where it was only observed during the last ~2000 years.

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### 4.3. Biogenic components

### 4.3.1. Siliceous microfossils and biogenic opal

- 348 The total diatom abundance fluctuated between  $5.52 \times 10^5$  and  $4.48 \times 10^7$  valves g<sup>-1</sup> at
- 349 core BGGC5. This abundance showed good correlation with biogenic opal content at
- BGGC5 ( $R^2 = 0.52$ , P < 0.5), with values increasing from 72 cm to the bottom of the
- 351 core, corresponding to cal BP 4900, and reaching their highest values before cal BP
- 352 6600. The opal percentage exhibited a maximum before cal BP 4900 (mode = 15.8%).
- 353 In contrast, the diatom abundance and biogenic opal were much lower at core BTGC8
- $(< 2 \times 10^5 \text{ valves g}^{-1} \text{ and } < 3\%, \text{ respectively})$ . Here, the siliceous assemblage was almost
- 355 completely conformed by *Ch.* RS (Fig. 6).
- 356 A total of 135 and 8 diatom taxa were identified in cores BGGC5 and BTGC8,
- 357 respectively, whereby core BTGC8 registered very low diatom abundances. In general,
- diatoms were the most important assemblage of siliceous microfossils (96%), followed
- by sponge spicules (3%). The contributions of phytoliths and chrysophyte cysts was less
- than 2% at core BGGC5. Ch RS was dominant in the diatom assemblage (~90%; Fig. 6)
- and included the species C. radicans, C. cinctus, C. constrictus, C. vanheurckii, C.
- 362 coronatus, C. diadema, and C. debilis. Other recorded upwelling group species (mainly
- at core BGGC5) were Skeletonema japonicum and Thalassionema nitzschioides var.
- 364 *nitzschioides* (Table S2). Other species range from 0.3% to 6% of the total assemblage.

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# 4.3.2. TOC and stable isotope distribution

- 367 Consistent with opal and diatoms, core BGGC5 showed higher values of TOC
- 368 (between 2% and 5%) compared with less than ~1.5% at core BTGC8 (Figs. 5a, 5b).
- Furthermore,  $\delta^{13}$ C was slightly higher at core BTGC8 (-20% to -21%) compared with
- 370 core BGGC5 (-21% to -22%). The former also shows slightly higher values of  $\delta^{15}N$
- 371 from the deeper sections to the surface of the core (<7% to >10%). This increase

was less evident at core BGGC5, with values of ~9% at depth to > 10% at the surface 372 (Figs. 5a, 5b). The reduced TOC content was related to the slightly higher  $\delta^{13}$ C values 373 (approximately -20%) in both cores. 374 375 376 4.3.3. Pollen record Initial surveys at core BTGC8 (Tongoy Bay) revealed extremely low pollen 377 378 abundances, which hampered further palynology work. A comprehensive pollen analysis was conducted only for core BGGC5 (Guanaqueros Bay). The pollen record 379 380 of core BGGC5 consisted of 29 samples shown in Fig. 7. The record was divided into five general zones following visual observations of changes in the main pollen types 381 and was also assisted by CONISS cluster analysis. 382 Zone BG-1 (cal BP 7990–7600): This zone is dominated by the herbaceous taxa 383 384 Chenopodiaceae, Leucheria-type, Asteraceae subfamily (subf.) Asteroideae, and Apiaceae with overall high values for the wetland genus *Typha* spp. 385 Zone BG-2 (cal BP 7600–6700): This zone is also dominated by Chenopodiaceae, 386 Leucheria-type, and Asteraceae subf. Asteroideae. In addition, other non-arboreal 387 388 elements, such as Ambrosia-type, Poaceae, Brassicaceae, and Chorizanthe spp., increase considerably. 389 Zone BG-3 (cal BP 6700–3500): This zone is marked by a steady decline in 390 391 Chenopodiaceae and Leucheria-type and by the increase in several other herbaceous 392 elements, such as Euphorbiaceae, Baccharis-type, and Brassicaceae. 393 Zone BG-4 (cal BP 3500–50): This zone is mostly dominated by Ast. subf. 394 Asteroideae and is marked by a decline in Chenopodiaceae and Leucheria-type. Other 395 coastal taxa, such as Euphorbiaceae, Baccharis-type, Asteraceae subf. Chichorioideae, 396 Quillaja saponaria, Brassicaceae, and Salix spp., also increase in this zone. Zone BG-5 (cal BP 50–Present): The upper portion of the record is dominated by 397 Asteraceae subf. Asteroideae and Poaceae and is marked by higher amounts of 398 399 Geraniaceae, Asteraceae subf. Mutisieae, Myrtaceae, and Q. saponaria. Additionally, this zone includes introduced pollen types such as Rumex spp. and Pinus spp. The 400 latter is not shown in Fig. 7 because its abundance was minimal. 401 Overall, the most distinctive trend revealed by core BGGC-5 is a long-term decline in 402 Chenopodiaceae and higher amounts of Euphorbiaceae and Asteraceae subf. 403

Asteroideae. Along with these changes, a further increase of several other types of

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Comentario [A12]: The ages were corrected according to the new age model used, which was done for the entire manuscript.

pollen, representative of the coastal shrub land vegetation, began at approximately cal BP 6700.

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### 4.4. Trace element distributions

408 Trace element distributions are shown in Figs. 8a and 8b for Guanaqueros (BGGC5) 409 and Tongoy Bays (BTGC8), respectively. We use Al as a normalizing parameter for 410 the enrichment/depletion of elements due to its conservative behavior. The elements 411 412 are presented as metal/Al ratios. Trace metals are sensitive to the presence of oxygen (U, Re, Mo), showing an increasing metal/Al ratio from the base of core BGGC5 (cal 413 414 BP ~7990) up to cal BP 6600. After this peak, ratios showed a slight increase toward 415 cal BP 1800, close to the beginning of the recent era, followed by a sharp reduction 416 until present. The exception to this trend was Mo, which reached a maximum value up 417 to cal BP 6600 and then reduced steadily to the present. Similarly, metal ratios at core BTGC8 increase over time; however, the peak was observed at cal BP~1000 for U 418 and Re and at cal BP 6000 for Mo, with a second minor peak at cal BP 3400. Iron 419 420 revealed a clear upward trend at cal BP 3500-3800 for core BGGC5 and a second 421 peak between cal BP 4500 and 6500, which was not clearly observed at the Tongoy core (BTGC8). Instead, core BTGC8 showed higher values before cal BP 6400. In 422 423 both cores, Fe increased over the past ~80 years, whereas no clear trend could be established for Mn. In general, metal/Al values were higher at core BGGC5. 424 A second group of elements (metal/Al ratio), including Ca, Ni, Cd, and P (related to 425 primary productivity and organic fluxes), showed a pattern similar to that of Mo/Al of 426 427 core BGGC5, i.e., increasing values from cal BP ~7990, reaching the highest values near cal BP 6600-7000; afterwards, the values followed a constant reducing trend 428 429 toward the present. Otherwise, Cu/Al (a nutrient-type element) showed a different pattern, similar to the Fe/Al distribution, with a maximum value at cal BP 3500-3800 430 and a conspicuous upward trend over the past ~80 years. A third group, consisting of 431 432 Ba and Sr, exhibited a similar pattern but smoother, showing the maximum values before cal BP 6600. At BTGC8 core, a less clear pattern was demonstrated. Ca, Ni, 433 434 Cd, and P ratios at core BTGC8 showed only slightly decreasing values and very low

peak values compared with core BGGC5; however, Ni/Al showed increasing concentrations over the past 80 years, which was not observed at core BGGC5.

Metal/Al ratios of Ba and Sr showed no substantial variation in time. In general, all the

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elemental concentrations were lower than at core BGGC5 and presented similar long-438 term reduction patterns toward the present, except for Cu, Ni, and Fe. 439 The authigenic enrichment factor (EF) of elements was estimated as:  $EF = (Me/Al)_{sample}$ 440 441 / (Me/Al)<sub>detrital</sub>, where (Me/Al)<sub>sample</sub> is the bulk sample metal (Me) concentration normalized to the Al content, and the denomination "detrital" indicates a lithogenic 442 443 background (Böning et al., 2009). Detrital ([Me]<sub>detrital</sub> and [Al]<sub>detrital</sub>) concentrations were established considering the local metal abundance, which is more accurate than using 444 445 mean Earth crust values (Van der Weijden, 2002). We used average element concentrations on surface sediments (0-3 cm) of the Pachingo wetland (Table 3). The 446 447 values suggest a large enrichment of nutrient-type elements in a period prior to cal BP 6600, following the trend of the Me/Al ratios, except for Ba and Fe, which did 448

**Comentario [A15]:** This paragraph was re-organized

**Comentario [A16]:** We modified the Fig. 9 including the information of the Table 4 from the old version

### 5. Discussion

recent times after cal BP 80 (Fig. 9).

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### 5.1. Sedimentary composition of the cores: terrestrial versus biogenic inputs

not show authigenic enrichment. The EFs exhibited a sharp decrease in enrichment in

The sediments in the southern zones of the bays are a sink of fine particles transported from the north and the shelf (Figs. 5a, 5b), and respond to water circulation in the Guanagueros and Coquimbo Bays (Fig. 1) with two counter-rotating gyres moving counterclockwise to the north and clockwise to the south (Valle-Levinson and Moraga, 2006) (Fig. 1). The differences established by the sediment composition of the bays show that the sediments of Guanaqueros Bay better represent the organic carbon flux to the bottom, with higher accumulation rates (mean value: 16 g m<sup>-2</sup> y<sup>-1</sup>) and higher amounts of siliceous microfossils. Furthermore, is it a better zone than Tongoy for pollen identification (Figs. 5a, 6 and 7). Both areas have sediments composed by winnowed particles and relatively refractory material (C/N: 9-11), which has a slightly lower isotopic composition than the TOC composition in the column water (-18‰, Fig. 2) and is transported by water circulating over the shelf. The isotopic variations in  $\delta^{13}C$  and  $\delta^{15}N$  did not clearly establish differences between the sediments of the two bays; however, minor differences in  $\delta^{15}N$  would indicate a greater influence of the upwelling nutrient supply and OMZ on the shelf, resulting in a  $\delta^{15}N$  of 9–10% in the Guanaqueros Bay, values which are slightly higher than that in the Tongoy Bay sediments (Figs. 5a, 5b). This isotopic composition corresponds with 471 that of NO<sub>3</sub> in the upwelling waters (De Pol-Holz et al., 2007) in the range of those measured at north-central Chile (~11%; Hebbeln et al., 2000, De Pol-Holz et al., 472 2007, 2009). This is due to the isotopic fractionation of NO<sub>3</sub> during nitrate reduction 473 474 within the OMZ, which leaves remnant NO<sub>3</sub><sup>-</sup> enriched in <sup>15</sup>N (Sigman et al., 2009; Ganeshram et al., 2000 and references therein). This is particularly relevant because it 475 demonstrates the relevance of OMZ over the shelf sediments off Coquimbo at shallow 476 477 depths and the influence of the poleward undercurrent from the Perú OMZ (Mollier-Vogle et al., 2012). At sediment core BTGC8, lower values (< 8‰) measured at 478 greater depths within the core should account for a mix with isotopically lighter 479 480 terrestrial organic matter (Sweeney and Kaplan, 1980), owing to its proximity to a 481 small permanent wetland in the southern side of Tongoy Bay (Pachingo), the sediments of which have  $\delta^{15}N$  of 2–6% (Muñoz et al., data will be published 482 elsewhere). This suggests that Tongoy sediments contain a combination with 483 484 continental material (Fig. 5b). Thus, cores BGGC5 and BTGC8 in the Guanaqueros and Tongoy Bays record the 485 variability in oceanographic conditions; however, in the Tongoy core, the 486 487 concentration of oceanographic proxies is diluted owing to the input of terrigenous material. This helps to decipher the climatic variability, considering that the main 488 input of clastic material to the area takes place during major flooding events. 489 490 Additionally, the main circulation of the bay system leads to favorable conditions for 491 the sedimentation and preservation of organic marine proxies in Guanaqueros Bay,

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### 5.2. Temporal variability of primary productivity and the oxygenation of bottoms

making the sedimentary records of these sites complementary.

Ca, Sr, Cd, and Ni profiles suggest a lower share of organic deposition over time (Figs. 8a, 8b), consistent with the slight reduction in TOC content observed in the sediments (Figs. 5a, 5b) and concomitant with the other elements related to organic fluxes to the bottom and primary productivity. Similarly, the maximum Ba concentrations indicate higher productivity before cal BP 6600. The same is true for Ca, Cd, and Ni, suggesting that the maximum productivity and organic fluxes to the bottom occurred during this period. After this age, the reduction in TOC and other nutrient-type elements (Ni, Sr, Ca, Cd) to the present is consistent with the increase in oxygen at the bay bottom. Hence, the slight rise in Ba in the last 100 years (Fig. 8a) is a response to this less anoxic environment, owing to better preservation within the

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sediments in less anoxic environments with moderate productivity (Torres et al., 1996; 505 Dymon et al., 1992) as is the case with our study site (Gross Primary Productivity 506 = 0.35 to 2.9 g C m<sup>-1</sup>d<sup>-1</sup>; Daneri et al., 2000). This leads to a negative correlation with 507 508 TOC (-0.59; Table 4), owing to the remobilization of Ba under anoxic conditions before cal BP 6600. Meanwhile, the P distribution showed a trend similar to that of 509 TOC and the other elements related to the organic fluxes to the bottom (Ni, Cd), 510 although with a lower correlation (~0.6). This is consistent with the distributions 511 512 observed for U, Re, and Mo at core BGGC5, which indicate that anoxic or suboxic conditions were developed from cal BP 7990 to 1800 but were stronger before cal BP 513 514 6600 (Figs. 8a, 8b). After this period and to the present, a remarkable reduction in 515 their concentration suggests a more oxygenated bottom environment, concurrent with 516 lower organic fluxes to the sediments. The Re profile shows the influence of suboxic 517 waters not necessarily associated with higher organic matter fluxes to the bottom. 518 Since this element is not scavenged by organic particles, its variability is directly related to oxygen changes (Calvert and Pedersen, 2007, and references therein). 519 Otherwise, the accumulation of P depends on the deposition rate of organic P (dead 520 521 plankton, bones, and fish scales) on the bottom and is actively remineralized during aerobic or anaerobic bacterial activity. P and TOC showed a declining trend toward 522 523 the present, suggesting a reduction in flux of organic matter over time, which was also 524 observed for Ni and Cd distributions. Alternatively, the reducing fluxes of organic proxies could be explained by the higher remineralization of organic material settled at 525 the bottom due to higher oxygen availability, as shown by U, Mo, and Re distributions 526 (Figs. 8a, 8b). However, the lower  $\delta^{15}N$ , depending on the denitrification process, is 527 similar to that at deeper environments in the zone (De-Pol Holz et al., 2009), 528 suggesting the influence of the reductive environment of OMZ over the shelf. The 529 influence of the primary productivity on oxygen consumption at the bottom over time 530 would be secondary in this system, which is considered to be moderated in 531 productivity compared with upwelling centers in north and south Chile. 532 Productivity reconstructions were based on the qualitative relative abundances of 533 diatom and sponge spicules, quantitative diatom counts (valves g<sup>-1</sup>), and biogenic opal 534 content only in core BGGC5, since core BTGC8 registered low valve counts (< 1% in 535 relative diatom abundance). However, in both cores, diatom assemblages were 536 represented mainly by Ch. RS, which are used as upwelling indicators (Abrantes 1988, 537

Vargas et al., 2004). The downcore siliceous productivity based on opal distribution

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(Figs. 6 and 9) distinguished three main time intervals of higher productivity, which coincided with the ages highlighted by the distribution of the sedimentary proxies noted previously: (1) > cal BP 6600, (2) cal BP 4500–1800, and (3) cal BP ~100 to recent times (CE 2015). Other periods between cal BP 6600 and cal BP 4500 and between cal BP 1800 and cal BP 100 did not experience higher productivities.

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At first period (> cal BP 6600), the opal accumulation rate was remarkably high, amounting to  $\sim 35 \pm 18$  g m<sup>2</sup> y<sup>-1</sup> (range: 16–119 g m<sup>-2</sup>y<sup>-1</sup>, Fig. 9) when *Chaetoceros* spores were predominant, indicating an intensification in upwelling. During this period, all metal proxies suggest that primary productivity increases before cal BP 6600, owing to the high concentrations and major enrichment of Ni, Ca, and P that occurred in this period, concomitant with higher opal accumulation within the sediments (Fig. 6 and 9). From these elements, Ni is the best indicator of organic sinking flux related with diatom productivity in organic-rich upwelling sediments (Böning et al., 2015), which helps to sustain our statement. In addition, the authigenic enrichments of Cd were very high (> 100, Fig. 9) resulting in high Cd/U ratios (> 2; Fig. 9), indicative for anoxic conditions as this ratio could vary between 0.2 and 2, from suboxic to anoxic environments (Nameroff et al., 2002). The Cd accumulation in this period was higher than that reported for a highly productive zone off Concepción in periods of high organic carbon accumulation in the sediments (~5, Muñoz et al., 2012). Additionally, the high enrichment of Mo (~20) indicates the prevalence of anoxic conditions at the bottom in this period due to the control by sulfide concentrations (Huerta-Diaz and Morse, 1992; Chaillou et al., 2002; Nameroff et al., 2002; Sundby et al., 2004, Tribovillard et al., 2004). Our low U/Mo ratio (~0.3, Fig. 9) corroborates this assumption, as similar to those values reported today at shallower anoxic zones off Perú interrupted by seasonal oxygenation (McManus et al., 2006; Sholz et al., 2011, Salvatteci et al., 2016; Vergara et al., 2016). This is similar to our shelf, notwithstanding the prevalence of very reduced conditions within the sediments. The enhanced reduced conditions in this period, probably sulfidic, favor the accumulation of Mo and Cd over that of U, occurring in anoxic environments where the chemocline is close to the water-sediment interface or above it, allowing the formation of authigenic Mo that exceed the U uptake within the sediments (Algeo and Tribovillard, 2009 and references therein). Re is enriched in less reduced conditions than Mo, resulting in the lowest Re/Mo in this period (Fig. 9). This is congruent with the environmental conditions at the bottom in zones of high productivity and intense

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upwelling, where sulfidic conditions are developed owing to oxygen consumption in 573 the shallower zones and linked to the OMZ, as occur at northern Chilean regions, 574 where the main productivity is developed over the narrow shelf. Thus, the high 575 576 productivity before cal BP 6600 could result from a more intense upwelling that generated permanent reduced conditions that became very anoxic at the bottom in this 577 period. Even so, the low oxygen conditions prevailed in the subsequent periods but 578 were less intense than before. 579 After cal BP 6400 until 4500 we obtained little information owing to a gap in the 580 sedimentary record, which made it difficult to visualize changes in the oxygenation 581 and productivity proxies in this interval. However, in the next period (cal BP 4500-582 1800), we observe that the opal accumulation was lower than in the previous recorded 583 period,  $12 \pm 4$  g m<sup>2</sup> y<sup>-1</sup> (range: 6–20 g m<sup>-2</sup>y<sup>-1</sup>, peaking at cal BP 3400–4000; Fig. 9), 584 which is partially consistent with nutrient-type element distributions and element 585 enrichment (Fig. 8a, Fig. 9). Fe clearly shows higher values at approximately cal BP 586 587 3500 (Fig. 8a), which helped to boost primary productivity at this time, with a small 588 increase in diatom, measured as valves per gram and abundance (%) (Fig. 6). Other 589 elements showed less prominent accumulations (Ni, Cd, Ba, Ca, and P), pointing to a lower organic matter deposition into the sediments during this period (Fig. 8a). Thus, a 590 591 decreasing trend in the primary productivity from cal BP 6600 is observed, which is 592 also consistent with observations off south-central Chile (36° S, Concepción shelf) where lower accumulations of nutrient-type elements were also observed at cal BP 593 3600-4000 and cal BP 2600 than at cal BP 6200 (Muñoz et al., 2012). However, low 594 595 oxygen conditions within the sediments were maintained, which could be more related to the manifestation of the OMZ close to the coast, favoring Mo and Re accumulation 596 until cal BP 1700-1800 (Fig. 8a). Lower Cd/U ratios (~1; Fig. 9) were estimated, 597 suggesting higher variations in the primary productivity but with moderate changes in 598 the oxygen conditions at the bottom. High Re/Mo and U/Mo ratios could indicate a 599 600 shift toward less reduced conditions but still anoxic, since U, Re, and Mo are highly enriched (6, 20, and 15, respectively; Fig. 9). U and Re accumulations occur in 601 602 conditions that exhibit less intense reduction but are not very favorable for Mo 603 accumulation (Morford et al., 2009). This could be caused by a lower C rain rate due

to lower productivity, producing low oxygen consumption and a less sulfidic

environment along the central-Chilean margin (30-36° S), which is in agreement with

the lower biogenic opal flux and diatom abundance after cal BP 6600 (Figs. 6, 9).

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Slight increasing values of Re/Mo ratios until ~cal BP 3500 suggest a decreasing trend in the reduced conditions, which became stronger after cal BP 1800. This time was also highlighted in the sedimentary records off Concepcion shelf (36° S, Muñoz et al., 2012) showing maximum enrichment of U and Cr near cal BP 1800, both indicating less reduced conditions toward the present compared with previous periods. After this age, no comparison could be made owing to a discontinuity in the sedimentary records off Concepción. Notwithstanding, the suboxic conditions have prevailed until today at Central Chile, where the oxygenation seems has been stronger off Coquimbo. It could be caused by eddies related to the instabilities of the Peru Undercurrent (Vergara et al., 2016), which seem to start operating more frequently from cal BP 1800 to the present. After this age to cal BP 100, higher productivities were not found, and a second discontinuity (cal BP 1500-240) impeded environmental reconstructions, with the very low estimated sedimentation rate hindering the realization of sufficient time resolution for the proxies in this interval. After cal BP ~100 to recent times (CE 2015) (third period mentioned before), the productivity increased substantially, deduced from the rise in opal accumulations toward the present (mean opal value of  $21 \pm 18$  g m<sup>2</sup> y<sup>-1</sup>, range: 8–34 g m<sup>2</sup> y<sup>-1</sup>; Fig. 9); however, this corresponded with lower diatom abundances, which were observed from cal BP 1800 to the present (range: 0.5-4.9  $\times$  10<sup>6</sup> valves g<sup>-1</sup>, Fig. 6). This is likely caused by the fact that only a few sections of the core in this interval were analyzed for diatoms, leading to a low resolution for this measurement in the most recent period. Another possibility is that the opal flux was overestimated owing to the fact that the flux calculations were based on recent sedimentation rates, an estimation that tends to be higher than at longer timescales (Sadler et al., 1999). However, the slight increase in the Cd/U ratio and P and Ni enrichment could suggest an increase in the primary productivity and organic fluxes to the bottom in more recent times. In addition, the main trend established before and after the hiatus indicates an increase in the marine productivity, which would not be as high as in the first period (before cal BP 6600). After cal BP 1800, there is an evident change to a less reduced environment toward the present, suggesting a more oxygenated bottom environment concurrent with a reduction in primary productivity, except for the last 100-130 years. Contrary to other metals, there is a conspicuous upward trend for Cu/Al, Fe/Al, and Mn/Al in recent times, which is consistent with the decreasing trend of EFs of Re, U, and Mo (Fig. 8a, 8b, Fig. 9); these estimations would not be influenced by the sedimentation rates but rather the presence of oxygen.

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Otherwise, the highest enrichment of Cu could suggest the presence of particulate forms and oxide formation (Peacock and Sherman, 2004; Vance et al., 2008; Little et al., 2014) occurring in the presence of an oxygenated environment that results in a high metal enrichment of Cu (EF<sub>cu</sub> =  $4.6 \pm 0.5$ , Fig. 9); however, suboxic conditions have prevailed, indicated by the U/Mo ratios in the range of the reduced sediments, which are less than in the sediments of the Peru shelf (Scholz et al., 2011; Salvatteci et al., 2016). In addition, the Cu enrichment coincides with the growing trend of industrialization in the area, mainly the mining activity, which has been the main economic source for Coquimbo region since 1890; therefore, the exposition of mineral ores and mine residues to the environment by natural processes as intemperization and wind transportation deserve attention.

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oxygenated environment, which is actually contradictory. We assume that episodic oxygenation changes the original extent of the accumulation of these sensitive redox

We suggest that higher productivity in the last 100 years has occurred in a more

trace element accumulations because of their remobilization to soluble forms (Morford

and Emerson, 1999; Morford et al., 2009). The main processes involved in the OMZ

ventilation at longer timescales are related to El Niño (Vergara et al., 2016 and references therein); thus the increased frequency and intensity of El Niño would result

in a mean effect, which is observed as a gradual change in metal enrichment over time.

Several observations made at the central Peruvian and south-central Chilean coasts

(12°-36° S) reveal that the present-day wet/dry variability associated with ENSO has

662 a strong impact on the bottom ocean oxygenation (Escribano et al., 2004; Gutiérrez et

al., 2006; 2008; Sellanes et al., 2007), suggesting a large increase in the oxygen levels 664 at the bottom during El Niño events, which change the sediment geochemistry, the

effects of which can be observed several months later. Other oxygenation mechanisms

can result from coastal-trapped Kelvin waves originating from the equator and 666

propagating along the coast, which modify the stability of the regional current system

and the pycnocline and trigger extra-tropical Rossby waves (Pizarro et al., 2002;

Ramos et al., 2006; 2008). This oceanographic feature may have operated more 669

670 frequently in the last century, changing the oxygen content in the bays with major

671 impact on redox-sensitive elements in the surface sediments. Comentario [A32]: New paragraph

### 5.3. Main climatic implications

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According to paleoenvironmental records, the past climate and oceanographic 674 variability have been interpreted mainly based on the past variability in the intensity of 675 the SWW and latitudinal position of the ITCZ (Veit et al., 1996; Hebbeln et al., 2002; 676 677 Lamy et al., 1999; Maldonado and Villagrán, 2002). The ITCZ movements from the northernmost or southernmost latitudinal position depend on the different phases of 678 ENSO and PDO variability (Yang and Oh, 2020), as the main regulators of the climate 679 at the centennial and decadal scales. This has an impact over relevant oceanographic 680 characteristics, such as sea surface temperature (SST), upwelling, and accordingly, 681 productivity at the SE Pacific. We established marked differences in paleo-productivity 682 proxies and paleo-oxygenation in the last ~8000 years (Figs. 6, 8), indicating that high 683 marine productivity prevailed during our first period (cal BP 8000-6600), according to 684 685 what was established for central Chile between 10 and 5 ky owing to sustained mean La Niña-like conditions associated with the cold phase of the PDO (positive phase) (De 686 Pol-Holz et al., 2006; Kaiser et al., 2008; Lamy et al., 2010), concomitantly with 687 688 reduced ENSO variability and a northward ITCZ displacement, which implies more 689 permanent southeast tradewinds and, hence, the upwelling of rich-nutrient cold waters 690 at eastern Pacific (Koutavas and Lynch-Stieglitz, 2004; Koutavas et al., 2006). Our high productivity records associated with low oxygen conditions at the bottom, both reaching 691 692 a maximum level at cal BP 6600, corresponds to the highest productive period and the 693 most reductive environment at the bottoms over the past 8000 y. At the Peruvian margin, this period has also been described as being drier with the dominance of La 694 Niña-like conditions according to the northerly position of both westerlies and the ITCZ 695 696 (Mollier-Vogel et al., 2019). Our pollen records also point to the driest conditions 697 during this period (PMI, Fig. 9), which matches with other reports in the region; this 698 indicates that an arid phase was developed at mid-Holocene affecting the eastern margin of Pacific from central Chile to the Galapagos (Carré et al., 2012). For central Chile, the 699 700 aridity conditions were extended until cal BP 5700 (Jenny et al 2002, Maldonado and 701 Villagrán, 2006) or cal BP 4200 (Maldonado and Rozas, 2008; Maldonado and Villagrán, 2002, 2006), characterized by reduced rainfall but intense coastal humidity, 702 703 which have been associated with coastal fogs that frequently occur during the spring 704 owing to a strengthening of the SPSA (Vargas et al., 2006; Garreaud et al 2008; Ortega et al., 2012) and La Niña-like conditions, which explains the main variability of the 705 SPSA (Ancapichún and Garcés-Vargas, 2015). Others have suggested a reduced ENSO 706 variance during the early and mid-Holocene (Rein et al., 2005), which indicates a less 707

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producing moderate El Niño events at the CP and strong La Niña off Peru (Carré et al., 709 2014, Mollier-Voguel et al., 2019). This was favorable for upwelling and primary 710 711 productivity development along the Chilean and Peruvian margin. In addition, Braconnot et al. (2012) indicated that this lower ENSO was linked to fresh water 712 melting that counteracted the insolation regime, pointing a more limited cold-dry period 713 between 6700-7500 years ago, which matches our records of maximum productivity 714 715 (Figs. 6, 9) concomitantly with the lowest bottom oxygen conditions and indicates a greater influence of the OMZ over the shelf at the central-Chilean margin. 716 After the maximum productivity recorded, a decreasing trend occurred under warm and 717 humid climatic conditions, which would be because of an enhancement in regional 718 719 precipitation in the northern margin of SWW (Jenny et al., 2003; Maldonado and Villagrán, 2006), consistent with the southern movement of the ITCZ, leading to wetter 720 721 climatic conditions in the southern tropics regions (Koutavas and Lynch-Stieglitz, 2004). A gradual rise in K/Ca, Fe, Al, and Pb distributions was observed in our cores 722 723 (Figs. 5, 9), usually considered to be an indicator of continental input by fluvial or aerial 724 transport (Calvert and Pedersen, 2007; Kaiser et al., 2008; Govin et al., 2012; Ohnemus and Lam, 2015; Saito et al., 1992; Xu et al., 2015). This indicated that the precipitation 725 726 has been increasing through the mid- and late Holocene, except for a period of reduced 727 (or weak) ENSO activity reported between cal BP 6000 and 4000 (Koutavas and Joanides, 2012; Carré et al., 2014). It is also consistent with the pollen records of central 728 Chile, which suggest an arid phase from cal BP 6200 until cal BP 4200 (Maldonado and 729 730 Villagrán, 2006). The lack of records between these ages in our cores (hiatus) prevented 731 the search for evidence to account for this period; consequently, no sharply contrasting dry/humid periods were identified after cal BP 6600. Mostly, a gradual increase in 732 humidity and a weakening in paleo-productivity proxies after cal BP 4500 (Figs. 8, 9) 733 734 were observed, which would be consistent with the beginning of higher ENSO 735 variability for central-Chile after cal BP 5700 (Jenny et al., 2002, Maldonado and Villagrán, 2002, 2006). In general, this is a period of increased ENSO variability (from 736 737 cal BP 5700) and stronger El Niño events after cal BP 4000-4500, concomitant with the 738 high variability of latitudinal displacements of the ITCZ related to the seasonality of insolation described for the region at the mid- and late Holecene (Haug et al., 2001; 739 Toth et al., 2012; Carré et al., 2014). This is consistent with the occurrence of alluvial 740

episodes in the area caused by more frequent or heavier rainfall events over time,

frequent or less intense warm anomaly related to a Central Pacific (CP)-mode ENSO,

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related to intensified westerlies and increased El Niño events observed from Peru to 742 central Chile (Jenny et al., 2002; 2003; Rein et al., 2005; Sandweiss et al., 2007; Ortega 743 et al., 2012; Ortega et al., 2019). A consequence is greater continental inputs, as 744 745 suggested by our sedimentary records in agreement with the pollen moisture index that indicated more humid conditions through the mid-Holocene to the present. This was 746 747 concomitant with greater oxygenation at the bottom and reduced primary productivity. Nonetheless, between cal BP 4500 and 3000, a slight increase in diatom abundance and 748 opal concentrations was observed, along with a slight accumulation in nutrient elements 749 (Ni, Cd, Fe, and Ca concentrations; Fig. 8). Small increases in the organic carbon flux 750 and Cd/U ratios (Fig. 5, 9) suggest that the increase in primary productivity could be 751 752 boosted by continental nutrients (Dezileau et al., 2004; Kaiser et al., 2008). This period 753 has been documented for the tropical east Pacific as a peak of La Niña activity (cal BP 754 3000–4000; Toth et al., 2012). This would also explain the increase in the productivity proxies. 755 756 Despite the dominance of warm events described for the mid- and late Holocene, they 757 were not strong enough to change the suboxic conditions at the bottom in the north-758 759 760 761

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central Chilean margin, which varied little until cal BP 1800 (Figs. 8, 9; see U, Mo, and Re). Actually, the periodicity of El Niño was similar between cal BP 5000 and cal BP 3000 and lower than modern times (Sandweiss et al., 2007), supporting the observation of relatively low variability of the oxygen proxies in the sediments dependent on the OMZ influence over the shelf. This implies that the upper limit location of the OMZ did not drastically change during most of the mid- and late Holocene. Contrary to our observations, the sediments at the Peruvian shelf were less reduced in the late-mid Holocene than at the present, which was due to a deepening in the OMZ by the increased advection of waters enriched in oxygen from the Equatorial Undercurrent and the shifting of the OMZ center toward the Chilean margin (Mollier et al., 2019). Therefore, the enhanced oxygenation of Peru and OMZ deepening translated into a decrease in the oxygen conditions off north-central Chile. This period is followed by an increased El Niño frequency that has been consistent with the intensification and frequency of flooding events recorded in Peru and central Chile in the last ~2000 years (Rein et al., 2005; Sandweiss et al., 2007; Jenny et al., 2002; Toht et al., 2012), which is concomitant with the drastic oxygenation at the bottom observed in our records after cal BP 1800. In this regard, the oxygen variation at the bottom would be related to a less intense effect of the OMZs over the shelf at the central Chilean margin during the warm

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El Niño-like phases, owing to a deepening of the oxycline (and vice versa during La 776 Niña). These tend to be associated with low productivity and, in turn, a reduction in the 777 organic fluxes and oxygen consumption during organic matter diagenesis. 778 After cal BP 1800, few records were obtained until cal BP 130, when we observed the 779 restoration of more reduced conditions, although lower than during previous periods. 780 This corresponds to the time of Peruvian upwelling shift due to the northward 781 displacement of the ITCZ to the modern position and the enhancement of the Walker 782 783 circulation (Gutiérrez et al., 2009), which establishes an intensification of the upwelling in the eastern Pacific; consequently, an increase in the primary productivity, producing 784 high demand for oxygen during organic matter remineralization, as observed today, 785

which leads to stronger oxygen consumption in the northern part of the eastern margin.

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#### 6. Conclusions

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808 809 Our results suggest that the geochemistry and sedimentary properties of the coastal shelf environments in north-central Chile have changed considerably during the Holocene period, suggesting two relevant changes in the redox conditions and productivity, which point to a more reducing environment and higher productivity around cal BP 6600. Afterwards, a less reducing environment along with decreasing trends in primary productivity and increased humid conditions occurs with time. The oxygenation of the surface limit of the OMZ has been proposed as the main mechanism that controls the reduced conditions over the shelf and slope sediments during the mid-Holocene, which mainly affected the Peruvian margin closed to the OMZ edge. This led to contrasting conditions in the central-Chilean margin where the most reduced conditions were observed, which was maintained with low variability until cal BP 1800. After this age, the OMZ expression over the shelf was weak, returning to more reduced conditions in recent times (two last centuries), similar to the Peruvian margin but weaker at north-central Chile. The northward shifts of the SWW belt, in addition to an increased frequency in El Niño events, have been proposed as the main drivers for climatic conditions during this period. These elements have introduced high variability in the primary productivity during this time interval. This also impacted the accumulation of organic matter due to an intensification of its remineralization, showing a decreasing trend in the buildup of nutrient-type elements and organic carbon burial rates toward the present. Otherwise, decreasing oxygen content at the bottom is highly influenced during El Niño events,

810	something that seems to have been operating at higher frequencies after cal BP 1800		
811	and, especially after cal BP 130, when the most extreme events become more frequent.		
812	Thus, the El Niño phenomenon and ITCZ latitudinal displacement have greatly		
813	contributed to the climatic and oceanographic features in the eastern Pacific, linked to		
814	the positive or negative phases of the PDO, which all has a relevant effect on the OMZ		
815	position in the Chilean margin.		
816	Finally, these changes highlight the sensitivity of these environments to climate		
817	variability at different timescales, which is consistent with the description of past		
818	regional climatic trends. Based on the dramatic changes observed in the past centuries,		
819	future changes are expected in the context of global warming at unprecedented rates.		
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Table 1. Radiocarbon dates for BGGC5 and BTGC8 sediment cores collected from mixed planktonic foraminifera and monospecific benthic foraminifera (*Bolivina plicata*), respectively. The <sup>14</sup>C-AMS was performed at NOSAM-WHOI. The lab code and conventional ages collected from each core section are indicated. For error calculations see <a href="http://www.whoi.edu/nosams/radiocarbon-data-calculations">http://www.whoi.edu/nosams/radiocarbon-data-calculations</a>.

				Modern			
Core		Mass	Lab Code	fraction		Conventional	1σ
identification	Material	(mg)	NOSAM	pMC	1σ error	Age BP	error
	Planktonic						
BGGC5	foraminifera						
10-11	Mix	1.8	OS-122160	0.8895	0.0027	940	25
18-19	Mix	1.1	OS-122141	0.7217	0.0024	2,620	25
31-32	Mix	2.7	OS-122161	0.6590	0.0021	3,350	25
45-46	Mix	2.0	OS-122162	0.6102	0.0017	3,970	25
55-56	mix	1.6	OS-122138	0.5864	0.0025	4,290	35
66-67	mix	2.8	OS-122304	0.5597	0.0018	4,660	25
76-77	mix	2.6	OS-122163	0.4520	0.0016	6,380	30
96-97	mix	1.1	OS-122139	0.4333	0.0033	6,720	60
115-116	mix	4.7	OS-122164	0.3843	0.0016	7,680	35
	Benthic						
BTGC8	foraminifera						
5-6	Bolivina plicata	4.2	OS-130657	0.8953	0.0017	890	15
20-21	Bolivina plicata	7.7	OS-123670	0.7337	0.0021	2,490	25
30-31	Bolivina plicata	13.0	OS-123671	0.6771	0.0016	3,130	20
40-41	Bolivina plicata	11.0	OS-123672	0.6507	0.0019	3,450	25
50-51	Bolivina plicata	8.7	OS-123673	0.5877	0.0014	4,270	20
60-61	Bolivina plicata	13.0	OS-123674	0.5560	0.0018	4,720	25
71-72	Bolivina plicata	10.0	OS-123675	0.4930	0.0013	5,680	20
80-81	Bolivina plicata	7.3	OS-123676	0.4542	0.0012	6,340	20
90-91	Bolivina plicata	6.8	OS-123677	0.4259	0.0015	6,860	30
96-97	Bolivina plicata	6.8	OS-123678	0.3903	0.0013	7,560	25

Table 2. Reservoir age estimation considering the <sup>210</sup>Pb age determined with the CRS model (McCaffrey and Thomson, 1980) at selected depth sections of the core, as compared with <sup>14</sup>C ages (y BP) from the marine13.14 curve (Reimer et al., 2013), according to Sabatier et al. (2010).

Core	Depth (cm)	Age from CRS model (AD) <sup>a</sup>	Age years BP <sup>b</sup>	<sup>14</sup> C age Marine 13.14	<sup>14</sup> C age BP from foram.	DR
BGGC5	10.5	1828	122	499±24	940±25	441±35
BTCG8	5.5	1908	42	448±23	890±15	442±27

<sup>&</sup>lt;sup>a</sup>Anno Domini

Table 3. Concentration of elements in the Pachingo wetland sediments, considered as lithogenic background for the study area. The values correspond to mean concentrations in the surface sediments (0–3 cm).

Element	Metal/Al x 10 <sup>3</sup>	S
Ca	686.5	139.3
Fe	591.3	84.5
P	8.6	0.7
Sr	5.7	0.6
Ba	5.6	0.1
Cu	0.258	0.019
Ni	0.174	0.005
U	0.020	0.003
Mo	0.020	0.003
Cd	0.0021	0.0003
Re	0.00004	0.00001

<sup>&</sup>lt;sup>b</sup>Before present=1950

Table 4. Spearman rank order correlations for geochemical data. Significant values > 0.8 are indicated in bold.

BGGC	25															
	Al	P	K	Ca	Mn	Fe	Ni	Cu	Mo	Cd	Re	Sr	U	Ba	Opal	TOC
Al	1.00	-0.62	0.49	-0.48	0.64	0.60	-0.75	0.56	-0.10	-0.73	-0.08	-0.33	0.08	0.49	-0.52	-0.44
P		1.00	-0.31	0.37	-0.45	-0.56	0.56	-0.57	0.01	0.61	-0.11	0.39	-0.12	-0.20	0.49	0.24
K			1.00	-0.24	0.90	0.83	-0.29	0.47	0.28	-0.42	0.33	-0.12	0.50	0.26	-0.25	-0.19
Ca				1.00	-0.47	-0.50	0.44	-0.64	0.23	0.59	0.39	0.92	0.30	-0.60	0.18	0.32
Mn					1.00	0.94	-0.51	0.68	-0.01	-0.68	0.07	-0.32	0.24	0.43	-0.39	-0.31
Fe						1.00	-0.49	0.81	0.03	-0.70	0.11	-0.40	0.23	0.36	-0.37	-0.21
Ni							1.00	-0.51	0.49	0.91	0.35	0.25	0.26	-0.70	0.72	0.64
Cu								1.00	-0.12	-0.71	-0.06	-0.61	0.00	0.31	-0.39	-0.07
Mo									1.00	0.50	0.88	0.10	0.91	-0.48	0.33	0.36
Cd										1.00	0.36	0.42	0.27	-0.67	0.70	0.54
Re											1.00	0.27	0.92	-0.50	0.16	0.38
Sr												1.00	0.24	-0.36	0.05	0.17
U													1.00	-0.39	0.10	0.29
Ba														1.00	-0.30	-0.59
Opal															1.00	0.35
TOC																1.00
BTGC	8															
BTGC	8 Al	P	K	Ca	Mn	Fe	Ni	Cu	Мо	Cd	Re	Sr	U	Ba	Opal	TOC
Al		-0.19	-0.17	<b>Ca</b> -0.37	<b>Mn</b> -0.02	-0.03	-0.39	-0.04	-0.39	<b>Cd</b> 0.02	<b>Re</b> -0.13	<b>Sr</b> -0.58	<b>U</b> -0.19	<b>Ba</b> 0.07	-0.41	
Al P	Al		-0.17 0.23	-0.37 0.00	-0.02 0.43	-0.03 0.28	-0.39 0.58	-0.04 0.23	-0.39 0.37	0.02 0.13	-0.13 -0.04	-0.58 0.30	-0.19 0.14	0.07 -0.14	-0.41 0.56	TOC -0.29 0.13
Al P K	Al	-0.19	-0.17	-0.37 0.00 -0.02	-0.02 0.43 0.54	-0.03 0.28 0.41	-0.39	-0.04 0.23 0.22	-0.39 0.37 -0.11	0.02	-0.13	-0.58 0.30 0.19	-0.19 0.14 -0.28	0.07 -0.14 0.28	-0.41 0.56 0.26	TOC -0.29 0.13 0.20
Al P K Ca	Al	-0.19	-0.17 0.23	-0.37 0.00	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27	-0.39 0.58 0.43 0.00	-0.04 0.23 0.22 -0.23	-0.39 0.37 -0.11 0.39	0.02 0.13 0.05 0.01	-0.13 -0.04 -0.04 0.33	-0.58 0.30 0.19 0.50	-0.19 0.14 -0.28 0.47	0.07 -0.14 0.28 -0.34	-0.41 0.56 0.26 0.20	TOC -0.29 0.13 0.20 0.34
Al P K Ca Mn	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64	-0.04 0.23 0.22 -0.23 0.01	-0.39 0.37 -0.11 0.39 0.05	0.02 0.13 0.05 0.01 0.33	-0.13 -0.04 -0.04 0.33 0.15	-0.58 0.30 0.19 0.50 0.32	-0.19 0.14 -0.28 0.47 -0.02	0.07 -0.14 0.28 -0.34 0.24	-0.41 0.56 0.26 0.20 0.32	TOC -0.29 0.13 0.20 0.34 0.00
Al P K Ca Mn Fe	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71	-0.39 0.37 -0.11 0.39 0.05 -0.40	0.02 0.13 0.05 0.01	-0.13 -0.04 -0.04 0.33 0.15 -0.67	-0.58 0.30 0.19 0.50 0.32 -0.37	-0.19 0.14 -0.28 0.47 -0.02 -0.62	0.07 -0.14 0.28 -0.34 0.24 0.13	-0.41 0.56 0.26 0.20 0.32 0.14	TOC -0.29 0.13 0.20 0.34
Al P K Ca Mn Fe	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56	0.02 0.13 0.05 0.01 0.33 -0.48 0.20	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b>	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45
Al P K Ca Mn Fe Ni	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56 -0.25	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64 -0.22	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19 -0.61	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16 -0.10	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21	-0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37
Al P K Ca Mn Fe Ni Cu	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68 0.45	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56 0.59	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21 0.58	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37
Al P K Ca Mn Fe Ni Cu Mo Cd	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56 -0.25	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56 0.59	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64 -0.22 0.66 0.39	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19 -0.61 0.69 0.52	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16 -0.10 -0.41	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21 0.58 0.10	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37 0.30 -0.12
Al P K Ca Mn Fe Ni Cu Mo Cd	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56 -0.25	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68 0.45	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56 0.59	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64 -0.22 0.66 0.39 0.53	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19 -0.61 0.69	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16 -0.10	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21 0.58 0.10 0.13	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37 0.30 -0.12 0.17
Al P K Ca Mn Fe Ni Cu Mo Cd Re Sr	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56 -0.25	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68 0.45	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56 0.59	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64 -0.22 0.66 0.39	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19 -0.61 0.69 0.52 <b>0.83</b> 0.58	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16 -0.10 -0.41	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21 0.58 0.10 0.13	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37 0.30 -0.12 0.17 0.23
Al P K Ca Mn Fe Ni Cu Mo Cd Re Sr U	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56 -0.25	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68 0.45	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56 0.59	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64 -0.22 0.66 0.39 0.53	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19 -0.61 0.69 0.52 <b>0.83</b>	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16 -0.10 -0.41 0.11 -0.16 -0.13 -0.19	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21 0.58 0.10 0.13 0.52 0.21	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37 0.30 -0.12 0.17 0.23 0.00
Al P K Ca Mn Fe Ni Cu Mo Cd Re Sr U Ba	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56 -0.25	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68 0.45	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56 0.59	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64 -0.22 0.66 0.39 0.53	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19 -0.61 0.69 0.52 <b>0.83</b> 0.58	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16 -0.10 -0.41 0.11 -0.16 -0.13	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21 0.58 0.10 0.13 0.52 0.21 -0.28	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37 0.30 -0.12 0.17 0.23 0.00 -0.42
Al P K Ca Mn Fe Ni Cu Mo Cd Re Sr U	Al	-0.19	-0.17 0.23	-0.37 0.00 -0.02	-0.02 0.43 0.54 -0.33	-0.03 0.28 0.41 -0.27 0.21	-0.39 0.58 0.43 0.00 0.64 0.13	-0.04 0.23 0.22 -0.23 0.01 0.71 0.24	-0.39 0.37 -0.11 0.39 0.05 -0.40 0.56 -0.25	0.02 0.13 0.05 0.01 0.33 -0.48 0.20 -0.68 0.45	-0.13 -0.04 -0.04 0.33 0.15 -0.67 0.25 -0.56 0.59	-0.58 0.30 0.19 0.50 0.32 -0.37 0.64 -0.22 0.66 0.39 0.53	-0.19 0.14 -0.28 0.47 -0.02 -0.62 0.19 -0.61 0.69 0.52 <b>0.83</b> 0.58	0.07 -0.14 0.28 -0.34 0.24 0.13 -0.16 -0.10 -0.41 0.11 -0.16 -0.13 -0.19	-0.41 0.56 0.26 0.20 0.32 0.14 <b>0.80</b> 0.21 0.58 0.10 0.13 0.52 0.21	TOC -0.29 0.13 0.20 0.34 0.00 0.10 0.45 0.37 0.30 -0.12 0.17 0.23 0.00

## **Figures**

Figure 1. Study area showing the positions of sampling stations. Sediment cores were retrieved from Guanaqueros Bay (BGGC5) and Tongoy Bay (BTGC8) at water depths of 89 and 85 m, respectively. Information of dissolved oxygen in the water column at St1 and St16 and that of suspended organic particles collected at St14 sampling sites was gathered in a previous project (INNOVA 07CN13 IXM-150). Monthly precipitation in mm (bars) (mean  $\pm$  SD; Montecinos et al., 2016). Schematic representation of the circulation in the bays (white arrows) and wind direction (blue arrow) is indicated, as obtained from Valle-Levinson and Moraga-Opazo (2006) and Moraga-Opazo et al. (2011).

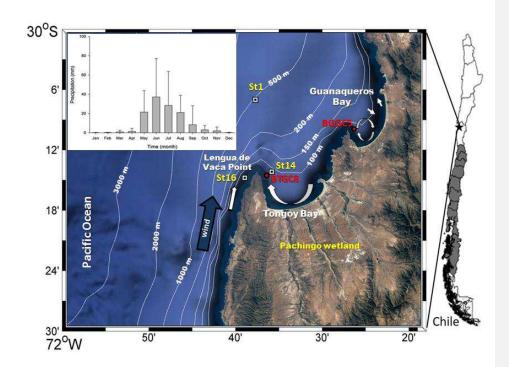


Figure 2. Suspended particulate matter composition (TOC% and  $\delta^{13}C_{org}$ ) measured in the water column between October 2010 and October 2011, at station St14, Tongoy Bay, Coquimbo (30° S).

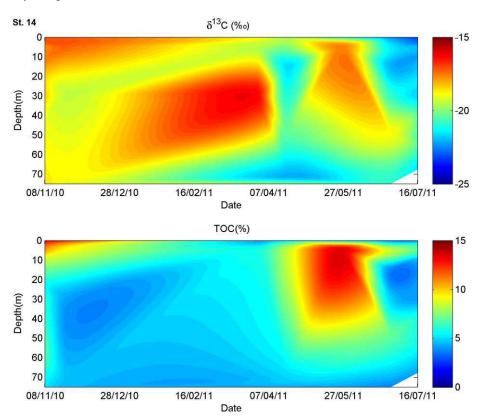


Figure 3. Dissolved oxygen time series in the water column measured between October 2010 and January 2011, at stations St1, St14, and St16 off Tongoy Bay, Coquimbo  $(30^{\circ} \text{ S})$ .

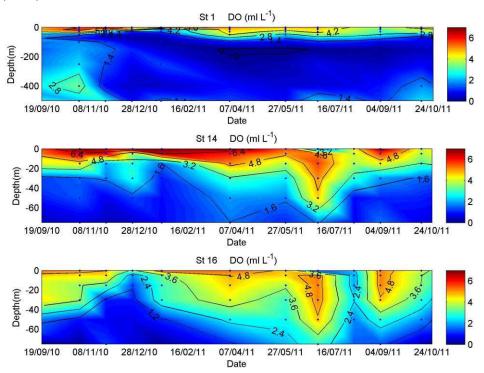


Figure 4. Age model based on <sup>14</sup>C-AMS and <sup>210</sup>Pb measurements. The timescale was obtained according to the Bacon age—depth modeling open source software (Blaauw and Christen, 2011) considering the Marine curve <sup>13</sup>C (Reimer et al., 2013).

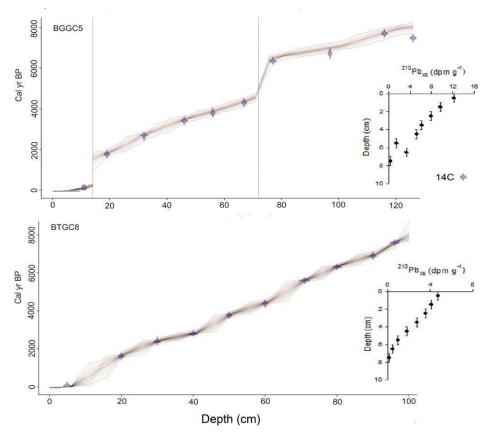
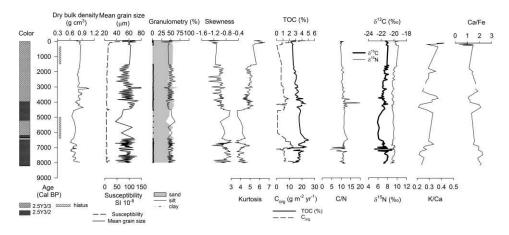


Figure 5. Characterization of sediment cores retrieved from (a) Guanaqueros Bay (BGGC5) and (b) Tongoy Bay (BTGC8), where the color (Munsell chart scale) represents the depth, dry bulk density, mean grain size, granulometry (% sand, silt, and clay), statistical parameters (skewness, kurtosis), organic components (TOC, C/N ratio, stable isotopes  $\delta^{15}$ N and  $\delta^{13}$ C) and chemical composition (K/Ca, Ca/Fe).

a)



1416 b)

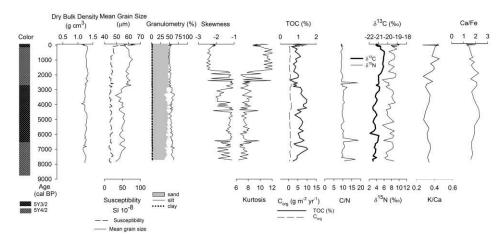
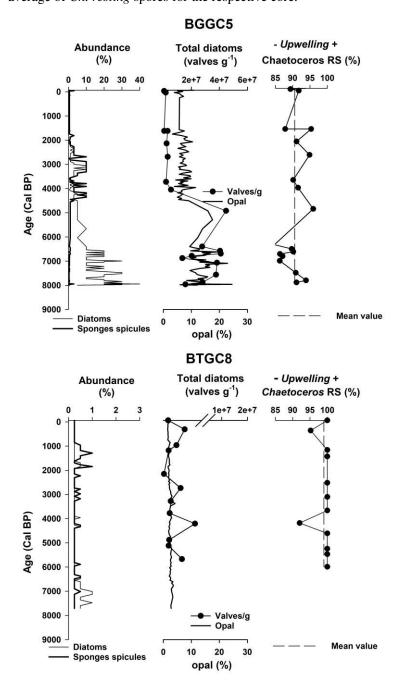


Figure 6. Diatom and sponge spicule relative abundances, total diatom counts (valves g<sup>-1</sup>) and opal (%), opal accumulation (g m<sup>-2</sup> y<sup>-1</sup>), and downcore variations in *Ch*. RS percentages as proxies of upwelling intensity in the BGGC5 and BTGC8 cores (Guanaqueros and Tongoy Bay, respectively). The medium dashed line represents the average of *Ch. resting* spores for the respective core.



## Figure 7. Pollen record in BGGC5 core.

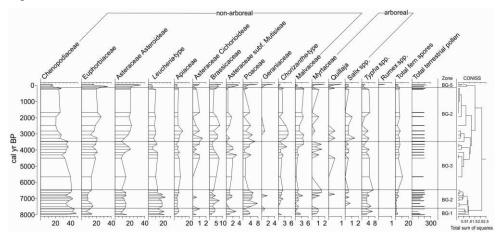
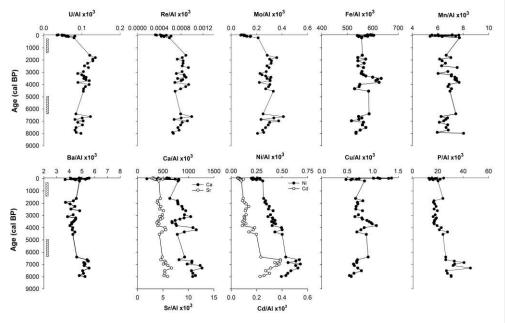


Figure 8. Downcore trace element variations in: (a) Guanaqueros Bay (BGGC5) and (b)
Tongoy Bay (BTGC8), off Coquimbo (30 °S).

1455 a)



1457 b)

1456

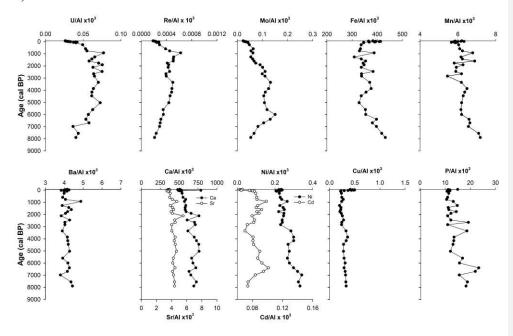


Figure 9. Opal accumulation and authigenic enrichment factor (EF) of trace elements calculated for Guanaqueros Bay (BGGC5 core). Lithogenic background was estimated from the surface sediments of Pachingo wetland cores (see text). Pollen moisture index defined as the normalized ratio between Euphorbiaceae (wet coastal shrub land) and Chenopodiaceae (arid scrubland). Positive (negative) values for this index indicate the relative expansion (reduction) of coastal vegetation under wetter (drier) conditions. Pb and Al distribution at BGGC5 core, representatives of terrigenous input to the bay.

