

Dear Editor;

We have carefully reviewed the previous comments to the second revision to find out which arguments had not been dealt with. We addressed all of them except for the one related to making changes to the organization of the discussion. In this sense, the resulting manuscript was very redundant and there were extensive explanations that in no way helped to improve it. Therefore, this version has been restructured according to the previous suggestions. We arranged the discussion by periods and all the sections were shortened. The manuscript now contains 22 pages, without references, tables and figures.

The referee indicated that the discussion jumps from discussing terrigenous inputs to primary productivity in the next section, without explaining why these are important points in this area. We tried to improve these issues in the discussion, but in the understanding that while the relationship between these aspects is a known process, there is a need to explain the sedimentary records. We also made a sound description of the study area, highlighting the relevance of the continental inputs, the effects of the El Niño events, and the productivity of the area. In addition, the introduction describes the main climatic and oceanographic characteristics of the area and how they could influence sedimentary records, to finalize with the objectives. Thus, we really focused our efforts on the discussion trying to avoid large explanations based on text book knowledge.

Is important to mention that we found that our records make a quite good description of the climatic trends previously illustrated for the Holocene in the region. The climatic interpretations provide a point of view that helps understand the prevalence of the environmental conditions in the last 8000 years. These clearly point to a change to more frequent El Niño events that will have an impact not only on climate and primary productivity, but also on bottom oxygenations.

We look forward to a positive assessment by the reviewers and for the manuscript to be accepted for publication.

Sincerely yours,

Práxedes Muñoz

On behalf of all authors

Referee answer

I think the Authors improved the content of the manuscript in comparison to the previous version. Unfortunately, I am not satisfied with the changes performed, especially as the authors avoided to apply a lot of suggestions/did not apply them thoroughly or even reported to have incorporated them but actually haven't.

Answer: I don't fully agree with this comment. All changes suggested were incorporated. This can be corroborated by comparing the original manuscript with the one with track changes, but the reorganization of the discussion in a timeline was not properly done. Otherwise, we added new references; the introduction was changed completely; we reduced the extension of the methods and added several comments that extended the discussion and the length of the manuscript. This resulted in redundant explanations of our main results which could be explained by the same environmental processes. It was too ambitious to provide a deep analysis of each proxy and still maintain the focus of the study.

Although the discussion gained important content the authors did not clean the text of textbook knowledge, more precisely it is not advisable to explain all proxies you used, or compare your data to, in detail within the discussion. This should either be moved into the introductions/methods or be removed completely. Furthermore, both reviewers found the discussion confusing and unstructured, just adding more text does not help the readability of the text, the discussion must be shortened and restructured in some way. If the authors do not follow the suggestion of the reviewers they could have changed the structure differently. The content of paragraphs and sections are still not connected, the discussion jumps from discussing terrigenous inputs to primary productivity in the next section without explaining why these are important points in this area or have any train of thought.

Answer: In this version we reorganized the discussion, highlighting three main periods when we observed important oceanographic changes based on productivity proxies and redox sensitive elements to interpret the main prevailing oceanographic conditions. Later, in section 5.3, we explain these variations according to climatic interpretations based mainly on pollen records that are consistent with the information reported for this region. In addition, we reduced the abstract, the methodology, the results and the discussion. To reduce the length, we avoided detailed explanations of each proxy focusing the discussion on the main interpretations of the proxies. Some paragraphs were reduced and moved to the introduction and the study site sections; others were totally removed. Some points about discrepancies between cores and proxies were discussed in previous version but now they were explained in more detail, for example in lines 485-492, 572-578, 681-692.

I do not advise publication of this manuscript before it was not shortened to less than 25 pages, 20 would be better and should be sufficient to discuss the main points of the presented data.

Answer: The paper was reduced from 30 pages (970 lines) to almost 22 pages (728 lines), without references and figures.

1 **Reconstructing past variations in environmental conditions and paleoproductivity**
2 **over the last ~8000 years off north-central Chile (30° S)**

3

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34

35 **Abstract**

36

37 The aim of this project was to establish past variations of the main oceanographic and
38 climatic features of a transitional semi-arid ecosystem in the north-central Chilean coast.
39 We analyzed recent sedimentary records retrieved from two bays, Guanaqueros and
40 Tongoy (29–30°S), for geochemical and biological analyses including: sensitive redox
41 trace elements, biogenic opal, total organic carbon (TOC), diatoms, stable isotopes of
42 organic carbon and nitrogen. Three remarkable periods were established, with different
43 environmental conditions and productivities: (1) > cal BP 6500, (2) cal BP 6500 – cal
44 BP 1700 and (3) cal BP 1700 towards the present (CE 2015). The first period was
45 characterized by a remarkably higher productivity (higher diatom abundances and opal)
46 when large fluxes of organic compounds were also inferred from the accumulation of
47 elements such as Ba, Ca, Ni, Cd and P in the sediments. At the same time, suboxic-
48 anoxic conditions at the bottoms were suggested by the large accumulation of Mo, Re
49 and U, showing a peak at cal BP 6500, when sulfidic conditions could have been
50 established. This was also identified as the driest interval according to the pollen
51 moisture index. These conditions should be associated to an intensification of the SPSA
52 and a stronger SWW, emulating La Niña-like conditions as has been described for the
53 SE Pacific during the early Holocene, which in this case extends until the mid-
54 Holocene. During most of the second period, lower productivity was observed.
55 However, a small increment was identified between Cal BP 4500 and 1700, although
56 low amounts of diatom (valves g⁻¹) and nutrient-type metal accumulations were
57 observed. Oxygen conditions at the bottoms change to an almost stable sub-oxic
58 condition during this time interval. The third period is marked by an intense
59 oxygenation after cal BP 1700, as observed by a change in the accumulation of U, Mo
60 and Re. In Addition, a small productivity rise after cal BP ~130 towards recent times
61 was observed, as suggested by opal accumulations but no increment in diatom
62 abundance. Overall, lower primary productivity, higher oxygenation at bottoms, and
63 higher humidity conditions were established after cal BP 6500 and towards the present.
64 We suggest that the oxygenation might be associated with an intensified El Niño
65 activity or similar conditions that introduce oxygenated waters to coastal zones by the
66 propagation of waves of equatorial origin, and establishing conditions that have reduced
67 the primary productivity from the mid Holocene toward the beginning of modern era.

Comentario [P1]: Abstract was shortened

68 Keywords: paleoproductivity, paleoredox, trace metals, diatoms, opal, organic carbon,
69 Coquimbo, SE-Pacific

70

71 **1. Introduction**

72

73 The mean climatic conditions at the SE Pacific are modulated by the dynamic of the
74 Southern Pacific Subtropical Anticyclone (SPSA) and the Humboldt Current System.
75 The SPSA has seasonal, decadal, and inter-decadal variability modulating the strength
76 of the southern westerly winds (SWW) and hence, the main oceanographic feature of
77 the Eastern boundary margin, the upwelling, influencing the biogeochemical processes
78 related to the inputs of nutrient and biological productivity. Seasonal variations produce
79 periods of intense upwelling when the SPSA is stronger, while the opposite is true when
80 it is weak (Croquette et al, 2007). The coastal wind pattern produced alongshore varies
81 along the SE Pacific showing lower seasonality between 18°–30°S, and producing a
82 semi-permanent upwelling (Pizarro et al., 1994; Figueroa and Moffat, 2000). This
83 system is highly affected by the inter-annual variability imposed by El Niño Southern
84 Oscillation (ENSO), with impacts on the wind intensity. The upwelling brings nutrient-
85 poor waters during the warm phase, while the opposite happens during the cold phase
86 (Rutland and Fuenzalida, 1991; Blanco et al., 2002). Other climate patterns –namely
87 the Pacific Decadal Oscillation (PDO) and the Southern Annular Mode (SAM)– operate
88 on a much longer time scale (inter-annual, decadal, inter-decadal) modifying the
89 strength and the position of the SWW, and thereby producing cold/warm periods
90 affecting mainly winter precipitations during positive/negative trends of SAM and
91 leading to intense/weak upwelling (Quintana and Aceituno, 2012; Ancapichún and
92 Garcés-Vargas, 2015). In addition, the orbitally induced austral insolation influences the
93 extent of the Antarctic sea ice and the Hadley cell, which act as important forces to the
94 latitudinal displacement of the ITCZ (Inter-tropical Convergence Zone; Kaiser et al.,
95 2008, and references therein). These fluctuations produce humid and arid conditions
96 along the SE Pacific where the wind's intensity remains the key factor for the
97 upwelling's strength and, therefore, for the supply of nutrients to the photic zone, all of
98 which are required for the development of the primary productivity.

99 Off Coquimbo (30°S), there is normally a semi-permanent and intense upwelling forced
100 by local winds, strongly influenced by topographic features (Figueroa and Moffat,
101 2000) and ENSO variability (Schaffer et al., 1997; Escribano et al., 2004). During El

102 Niño, mean winds alongshore reduce their intensity and the South East Pacific
103 anticyclone weakens. Conversely, during La Niña mean winds alongshore increase their
104 intensity and the anticyclone is reinforced (Rahn and Garreaud, 2013). This has an
105 impact on the upper circulation of the ocean affecting the oxygenation of the water
106 column and the strength of upwelling. The high productivity that takes place close to
107 the coast during normal periods (Escribano et al., 2004 and references therein)
108 maintains a zone of low dissolved oxygen content along the margin reinforcing the
109 oxygen minimum zone (OMZ). This zone develops along the North and South Pacific
110 Ocean and its intensity, thickness, and temporal stability vary as a function of latitude
111 (Helly and Levin, 2004, Ulloa et al., 2012). To the north (e.g. 21°S) and off Peru, the
112 OMZ occurs permanently, and can extend into the euphotic zone. In the case of northern
113 Chile and southern Peru, there is no significant interface with the benthic environment
114 due to the presence of a narrow continental shelf (Helly and Levin, 2004). The OMZ
115 dynamic off Coquimbo has not been studied in detail, but a seasonal intrusion of low
116 oxygen waters to the coast has been observed (Gallardo et al., 2017). During the 97-98
117 El Niño event, the oxygenation of bottoms was clearly detected in north (23°S) and
118 south-central Chile (36°S) (Ulloa et al., 2001; Gutiérrez et al., 2006; Sellanes et al.,
119 2007), changing the normal suboxic conditions at the bottom, the normal composition
120 of macrofauna and related geochemical characteristics of the sediments having
121 implications that persist for many years after the event (Gutiérrez et al., 2006; Sellanes
122 et al., 2007).

123 These changes in primary productivity and oxygenation at the bottom can be observed
124 in sedimentary records which respond to the amount of organic carbon that has settled
125 on the surface sediments and to the diagenetic reactions during organic matter
126 remineralization. Trace elements are commonly used as indicators of these processes,
127 observed as element enrichment or depletion. They are driven by organic matter fluxes
128 and redox conditions that modify the original extension of metal enrichment, which
129 depend on the oxygen content during early diagenesis in the upper sediment layers and
130 overlying water (Nameroff et al., 2002; Zheng et al., 2002; McManus et al., 2006;
131 Siebert et al., 2003). Therefore, they are a useful tool to establish temporary changes in
132 primary productivity and also to establish changes in the oxygenation at the bottom on
133 sedimentary records.

134 Our work focuses on the past variations of the environmental conditions deduced from
135 marine sedimentary records of inorganic and organic proxies over the last ~8000 years

136 BP, obtained from a transitional semiarid ecosystem off the central Chilean coast
137 (30°S), close to Lengua de Vaca point, the most relevant upwelling area of Chile's
138 northern margin (Shaffer et al., 1999; Thiel et al., 2007). We considered redox trace
139 element measurements that respond to local hypoxia (U, Mo and Re), as well as
140 nutrient-type elements that follow the organic fluxes to the sediments (Ba, Ni Cu, P)
141 (Tribovillard, 2006). Additionally, we measured Fe and Mn which play a key role in
142 adsorption-desorption and scavenging processes of dissolved elements in bottom waters
143 and sediments, and we measured Ca, K and Pb used to assess terrigenous inputs by
144 coastal erosion, weathering and eolian transport, which is also true for Fe and Mn
145 (Calvert and Pedersen, 2007). Ca accumulation depends, in turn, on carbonate
146 productivity and dissolution, which has been used as a paleoproductivity proxy (Paytan,
147 2008; Govin et al., 2012). We determined the enrichment/depletion of elements to
148 establish the main environmental conditions prevailing during the sedimentation of
149 particulate matter (Böning et al., 2009). In addition, we considered the diatom
150 assemblages with biogenic opal as a measurement of siliceous export production, TOC,
151 and stable isotopes to identify variations in the organic fluxes to the bottoms. Moreover,
152 pollen grains were used to identify environmental conditions based on the climate
153 relationship of the main vegetation formations in North-Central Chile. Based on our
154 records we were able to identify wet/dry intervals, periods with high/low organic fluxes
155 to the sediments related to changes in primary production, and changes in the redox
156 conditions at the bottoms.

157

158 **2. Study area**

159 The Coquimbo area (29-30°S) –in the southern limit of the northern-central Chilean
160 continental margin– constitutes a border area between the most arid zones of northern
161 Chile (Atacama Desert) and the more mesic Mediterranean climate in central Chile
162 (Montecinos et al., 2016). Here, the shelf is narrow and several small bays trace the
163 coast line.

164 The Tongoy and Guanaqueros bays are located in the southern edge of a broad
165 embayment between small islands to the north (29°S; Choros, Damas and Chañaral) and
166 Lengua de Vaca Point to the south (30°S) (Fig. 1), protected from predominant
167 southerly winds. Tongoy Bay is a narrow marine basin (10 km at its maximum width)
168 with a maximum depth of ~100 m. To the northeast lies Guanaqueros Bay, a smaller
169 and shallower basin. High wind events evenly distributed throughout the year promote

170 an important upwelling center at Lengua de Vaca Point, developing high biomass along
171 a narrow coastal area (Moraga-Opazo et al., 2011; Rahn and Garreaud, 2013), and
172 reaching maximum concentrations of $\sim 20 \text{ mg m}^{-3}$ (Torres and Ampuero, 2009). In the
173 shallow waters of Tongoy Bay, the high primary productivity results in high TOC in the
174 water column allowing for the deposition of fine material to the bottom; TOC rises
175 concurrently with the periods of low oxygen conditions (Fig. 2; Muñoz et al.,
176 unpublished data). Recent oceanographic studies indicate that low dissolved oxygen
177 water intrusions from the shelf (Fig. 3) seem to be related to lower sea levels resulting
178 from annual local wind cycles at a regional meso-scale (Gallardo et al., 2017).
179 Oceanographic time series indicate that transition times develop in short periods due to
180 changes in the directions and intensities of the winds along the coast, with a strong
181 seasonality (<http://www.cdom.cl/boyas-oceanograficas/boya-tongoy>). The spatial and
182 temporal variability of these processes is still under study. In addition, oceanic
183 variability along the western coast of South America is influenced by equatorial Kelvin
184 waves on a variety of timescales, from intra-seasonal (Shaffer et al., 1997) and seasonal
185 (Pizarro et al., 2002; Ramos et al., 2006), to inter-annual (Pizarro et al., 2002; Ramos et
186 al., 2008). Coastal-trapped Kelvin waves originating from the equator can propagate
187 along the coast, modifying the stability of the regional current system and the
188 pycnocline, and triggering extra-tropical Rossby waves (Pizarro et al., 2002; Ramos et
189 al., 2006; 2008). This oceanographic feature will change the oxygen content in the bays
190 with major impacts on redox-sensitive elements in the surface sediments.
191 Sedimentological studies are scarce in Chile's northern-central shelf. A few technical
192 reports indicate that sediments between 27°S and 30°S are composed of very fine sand
193 and silt with relatively low organic carbon content (<3 and ~5%), except in very limited
194 coastal areas where organic material accounts for approximately ~16% (Muñoz,
195 unpublished data; FIP2005-61 Report, www.fip.cl). Coastal weathering is the main
196 source of continental input due to scarce river flows and little rainfall in the zone (0.5 to
197 $\sim 80 \text{ mm yr}^{-1}$; Montecinos et al., 2016, Fig.1). Freshwater discharges are represented by
198 creeks, which receive the drainage of the coastal range forming wetland areas in the
199 coast and even small estuaries, such as Pachingo, located south of Tongoy (Fig. 1).
200 These basins cover ~ 300 and 487 km^2 , respectively. The water volume in the estuaries
201 is maintained by the influx of seawater mixed with groundwater supply. No surface flux
202 to the sea is observed. Therefore, freshwater discharge occurs only during high rainfall
203 periods in the coastal zone (DGA, 2011), which normally takes place during El Niño

Comentario [P2]: This paragraph was moved from the discussion, is relevant to understand the oxygen variability in the area

204 years when higher runoff has been recorded in the area during the austral winter (Valle-
205 Levinson et al., 2000; Montecinos and Aceituno, 2003; Garreaud et al., 2009). Under
206 this scenario, marine sediments are often highly influenced by primary production in the
207 water column, terrestrial runoff, and therefore, sedimentary records can reveal past
208 variability in primary production and in the oceanographic conditions over the shelf,
209 which ultimately respond to major atmospheric patterns in the region.

210

211 **3. Materials and methods**

212 **3.1. Sampling**

213 Sediment cores were retrieved from two bays in the Coquimbo region: Bahía
214 Guanaqueros (core BGGC5; 30°09' S, 71°26' W; 89 m water depth) and Bahía Tongoy
215 (core BTGC8; 30°14' S, 71°36' W; 85 m water depth) (Fig. 1.), using a gravity corer
216 (KC-Denmark) during May 2015, on board the L/C Stella Maris II owned by the
217 Universidad Católica del Norte. The length of the cores was 126 cm for BGGC5 and 98
218 cm for BTGC8.

219 Subsequently, the cores were sliced into 1-cm sections and subsamples were separated
220 for grain size measurements, magnetic susceptibility, trace elements, biogenic opal, C
221 and N stable isotope signatures ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$), and TOC analyses. The samples were first
222 kept frozen (-20°C) and then freeze-dried before laboratory analyses.

223

224 **3.2. Geochronology (^{210}Pb and ^{14}C)**

225 Geochronology was established combining ages estimated from $^{210}\text{Pb}_{\text{xs}}$ activities
226 suitable for the last 200 years and radiocarbon measurements at selected depths for
227 older ages. ^{210}Pb activities were quantified through alpha spectrometry of its daughter
228 ^{210}Po following the procedure of Flynn (1968). $^{210}\text{Pb}_{\text{xs}}$ (unsupported) activities were
229 determined as the difference between ^{210}Pb and ^{226}Ra activities measured in some
230 intervals of the sediment column. ^{226}Ra was measured by gamma spectrometry at the
231 Laboratoire Géosciences of the Université de Montpellier (France). Standard deviations
232 (SD) of the ^{210}Pb inventories were estimated propagating counting uncertainties
233 (Bevington and Robinson, 1992) (Table S1, supplementary data). The ages were based
234 on the Constant Rate of Supply Model (CRS, Appleby and Oldfield, 1978).

235 Radiocarbon measurements were performed on a mix of planktonic foraminifer species
236 in core BGGC5 whereas the benthic foraminifer species *Bolivina plicata* was selected
237 for core BTGC8 (Table 1). The samples were submitted to the National Ocean Sciences

238 AMS Facility (NOSAMS) of the Woods Hole Oceanographic Institution (WHOI). The
239 time scale was obtained according to the best fit of ages obtained from $^{210}\text{Pb}_{\text{xs}}$ and ^{14}C
240 (Fig. 4), using the CLAM 2.2 software and using the Marine curve 13C (Reimer et al.,
241 2013). A reservoir deviation from the global mean reservoir age (DR) of 441 ± 35 years
242 was considered, established according Sabatier et al. (2010). This was estimated
243 subtracting the ^{14}C age value corresponding at the historical dates 1828 AD and 1908
244 AD (499 ± 24 and 448 ± 23 ^{14}C yr, respectively, Reimer et al., 2013) from the apparent
245 ^{14}C age of foraminifers measured at depths of 5 and 10 cm for cores BTGC8 and
246 BGGC5, respectively (Sabatier et al., 2010; Table 2).

247

248 **3.3. Geophysical characterization**

249 Magnetic susceptibility ($\text{SI} \times 10^{-8}$) was measured with a Bartington Susceptibility Meter
250 MS2E surface scanning sensor at the Sedimentology Laboratory at Centro Eula,
251 Universidad de Concepción. Mean values from three measurements were calculated for
252 each sample.

253 Grain size was determined using a Mastersizer 2000 laser particle analyzer, coupled to a
254 Hydro 2000–G Malvern in the Sedimentology Laboratory of Universidad de Chile.

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

257

258 **3.4. Chemical analysis**

259 Trace element analyses were performed by ICP-MS (Inductively Coupled Plasma-Mass
260 Spectrometry) using an Agilent 7700x at Université de Montpellier (OSU
261 OREME/AETE regional facilities). The analysis considered reference materials (UBN,
262 BEN and MAG1) obtaining an accuracy higher than $\pm 5\%$; the analytical precisions were
263 between 1% and 3%. Internal standardizations with In and Bi were used to deconvolve
264 mass-dependent sensitivity variations of both matrix and instrumental origin, occurring
265 during the course of an analytical session. The analytical precisions attained were
266 between 1% and 3%.

267 TOC and stable isotope ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) analyses were performed at the Institut für
268 Geographie, Friedrich Alexander Universität (FAU) Erlangen-Nürnberg, Germany
269 using a Carlo Erba elemental analyzer NC2500 and an isotope-ratio-mass spectrometer
270 (Delta Plus, Thermo-Finnigan) for isotopic analysis. Stable isotope ratios are reported in
271 the δ notation as the deviation relative to international standards (Vienna Pee Dee

Comentario [P3]: This paragraph was shortened (16 lines were eliminated).

272 Belemnite for $\delta^{13}\text{C}$ and atmospheric N_2 for $\delta^{15}\text{N}$, so $\delta^{13}\text{C}$ or $\delta^{15}\text{N} = [(\text{R sample/R}$
273 standard) - 1] $\times 10^3$, where R is $^{13}\text{C}/^{12}\text{C}$ or $^{15}\text{N}/^{14}\text{N}$, respectively. Typical precision of
274 the analyses was $\pm 0.1\%$ for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$.

Comentario [P4]: This paragraph was shortened, 5 lines were eliminated.

275 Biogenic opal was estimated following the procedure described by Mortlock and
276 Froelich (1989). The analysis was done by molybdate-blue spectrophotometry (Hansen
277 and Koroleff, 1999) conducted at the laboratories of Marine Organic Geochemistry and
278 Paleoceanography, University of Concepción, Chile. Values are expressed as biogenic
279 opal by multiplying the Si (%) by 2.4 (Mortlock and Froelich, 1989). Analytical
280 precision was $\pm 0.5\%$. Accumulation rates were determined based on sediment mass
281 accumulation rates and amount of opal at each core section in %.

282

283 3.5. Microfossils analyses

284 Qualitative abundances of siliceous microfossils were carried out every one centimeter
285 following the Ocean Drilling Program (ODP) protocol, described by Mazzullo and
286 Graham (1988). This information was used to select some sections every ~4, 8 and 12
287 cm for BGGC5 and at ~6 cm for BTGC8, for quantitative abundances of microfossils
288 (diatoms, silicoflagellates, sponge spicules, crysophytes and phytoliths). Roughly ~ 0.5 g
289 of freeze-dried sediment was treated according to Schrader and Gersonde (1978) for
290 siliceous microfossils. They were identified and counted under an Olympus CX31
291 microscope with phase contrast. 1/5 of the slides were counted at 400X for siliceous
292 microfossils and one transect at 1000x was counted for *Chaetoceros* resting spores (*Ch.*
293 resting spores). Two slides per sample were counted; the estimated counting error was
294 15%. Total diatom abundances are given in valves g^{-1} of dry sediments.

295 Pollen analysis was conducted following the standard pollen extraction methodology
296 (Faegri and Iversen, 1989). The identification was conducted under a stereomicroscope,
297 with the assistance of the Heusser (1973) pollen catalogue. A total of 100-250 terrestrial
298 pollen grains were counted in each sample. Pollen percentage for each taxon was
299 calculated from the total sum of terrestrial pollen (excluding aquatic taxa and fern
300 spores). Pollen percentage diagrams and zonation were generated using the Tilia
301 software (Grimm, 1987).

Comentario [P5]: This paragraph was shortened, 6 lines were eliminated.

302 We further summarize pollen-based precipitation trends by calculating a Pollen
303 Moisture Index (PMI), which is defined as the normalized ratio between Euphorbiaceae
304 (wet coastal scrubland) and Chenopodiaceae (arid scrubland). Thus, positive (negative)
305 values of this index point to relatively wetter (drier) conditions.

306

307 **4. Results**

308 **4.1. Geochronology**

309 $^{210}\text{Pb}_{\text{xs}}$ (unsupported activity) was obtained from the surface at a depth of 8 cm in the
310 two cores, with an age of ~ AD 1860 at 8 cm in both (Table S1). Greater surface
311 activities were obtained for core BGGC5 ($13.48 \pm 0.41 \text{ dpm g}^{-1}$) compared to core
312 BTGC8 ($5.80 \pm 0.19 \text{ dpm g}^{-1}$), showing an exponential decay with depth (Fig. 4). A
313 recent sedimentation rate of $0.11 \pm 0.01 \text{ cm yr}^{-1}$ was estimated.

314 The age model provided a maximum age of cal BP 8210 for core BGGC5, and cal
315 BP 7941 for core BTGC8 (Fig. 4). A mean sedimentation rate of 0.02 cm yr^{-1} was
316 estimated for core BGGC5, with a period of relative low values (0.01 cm yr^{-1}) between
317 cal BP ~4000 and 6000. For BTGC8, sedimentation rates were less variable and around
318 0.013 cm yr^{-1} in the entire core. An age reservoir estimation following the method by
319 Sabatier et al. (2010) resulted in 441 ± 35 and 442 ± 27 years for BGGC5 and BTGC8
320 cores, respectively (Table 2). These values were close to the global marine reservoir and
321 higher than other estimations along the Chilean margin at shallower depths (146 ± 25
322 years at < 30 water depth; Carré et al., 2016; Merino-Campos et al., 2018). Our coring
323 sites are deeper (~90 m water depth) and influenced by upwelled water from Lengua de
324 Vaca Point, which could explain such differences. However, moderate differences were
325 observed between models using both reservoir values. Thus, our estimations were based
326 on two pre-bomb values established with ^{210}Pb measured in sediments and ^{14}C in
327 foraminifers, used for the age modeling.

328

329 **4.2. Geophysical characterization**

330 Sediments retrieved from the bays showed fine grains within the range of very fine sand
331 and silt in the southern areas. There, grain size distribution was mainly unimodal, very
332 leptokurtic, better sorted and skewed to fine grain when compared to sediments from
333 the northern areas. Sediment cores obtained from the northern areas were sandy (coarse
334 sand and gravel), with abundant calcareous debris. Longer cores of soft sediment were
335 retrieved at the southernmost areas (BGGC5 and BTGC8), where the silty component
336 varied between 40 % and 60 % (Fig. 1 and 5a,b). The clay component was very low at
337 both cores ($< 2\%$). The sediment's color ranged from very dark grayish brown to dark
338 olive brown (2.5Y 3/3–3/2) in Guanaqueros Bay (BGGC5) and from dark olive gray to
339 olive gray (5Y 3/2–4/2) in Tongoy Bay (BTGC8). Visible macro-remains (snails and

340 fish vertebrae) were found, as well as weak laminations at both cores. The magnetic
341 susceptibility showed higher values close to the surface, up to $127 \text{ SI} \times 10^{-8}$ at BGGC5,
342 and relative lower values ($85 \text{ SI} \times 10^{-8}$) at BTGC8. At greater depths, however, the
343 values were very constant, around $5\text{--}8 \times 10^{-8} \text{ SI}$ at BGGC5 core and around $12\text{--}20 \times 10^{-8}$
344 SI at BTGC8 core. In both cores, susceptibility rises substantially in the last century
345 (Figs. 5a, 5b). Lower bulk densities were estimated at core BGGC5 ($0.7\text{--}0.9 \text{ g cm}^{-3}$),
346 compared with core BTGC8 ($>1 \text{ g cm}^{-3}$) (Fig. 5a, 5b). In line with this, mean grain size
347 amounted to $60\text{--}80 \mu\text{m}$ in Guanaqueros Bay (BTGC8), compared to $50\text{--}60 \mu\text{m}$ in
348 Tongoy Bay (BGGC5). Both cores were negatively skewed, with values of -1 to -1.2 at
349 BGGC5, and -1 to -2.5 at BTGC8. Minor increases towards coarser grain size were
350 observed in the last ~ 1000 years, especially in Tongoy Bay (BTGC8). In both cases,
351 grain size distributions were strongly leptokurtic. Ca/Fe ratio also reduced in time,
352 except at core BTGC8 where it was only observed during the last ~ 2000 years.

353

354 4.3. Biogenic components

355 4.3.1. Siliceous microfossils and biogenic opal

356 Total diatom abundance fluctuated between 5.52×10^5 and 4.48×10^7 valves g^{-1} at core
357 BGGC5. Total diatom abundance showed a good correlation with biogenic opal content
358 at BGGC5 ($R^2 = 0.52$, $P < 0.5$), with values raising from 72 cm to the bottom of the core,
359 corresponding to cal BP 5330, and reaching their highest values before cal BP 6500. On
360 the contrary, diatom abundance and biogenic opal were much lower at core BTGC8 (< 2
361 $\times 10^5$ valves g^{-1} and $< 3\%$, respectively). Here, the siliceous assemblage was almost
362 completely conformed by *Chaetoceros* resting spores (RS) (Fig. 6).

363 A total of 135 and 8 diatom taxa were identified in cores BGGC5 and BTGC8
364 respectively, where core BTGC8 registered very low diatom abundances. In general,
365 diatoms were the most important assemblage of siliceous microfossils (96 %), followed
366 by sponge spicules (3 %). The contribution of phytoliths and chrysophyte cysts was less
367 than 2 % at core BGGC5. *Chaetoceros* (RS) dominated diatom assemblage ($\sim 90\%$; Fig.
368 6), and included the species *C. radicans*, *C. cinctus*, *C. constrictus*, *C. vanheurckii*, *C.*
369 *coronatus*, *C. diadema*, and *C. debilis*. Other upwelling group species recorded (mainly
370 at core BGGC5) were: *Skeletonema japonicum*, and *Thalassionema nitzschioides* var.
371 *nitzschioides* (Table S2). Other species range from $\sim 0.3\%$ to 6 % of the total
372 assemblage.

373

Comentario [P6]:

This paragraph was shortened, 6 lines were eliminated.

374 **4.3.2. TOC and stable isotopes distribution**

375 Consistent with opal and diatoms, core BGGC5 showed higher values of TOC
376 (between 2 % and 5 %) compared with less than ~1.5 % at core BTGC8 (Fig. 5a,b).
377 Furthermore, $\delta^{13}\text{C}$ was slightly higher at core BTGC8 (-20 ‰ to -21 ‰) compared
378 with core BGGC5 (-21 ‰ to -22 ‰), the former is also showing slightly higher values
379 of $\delta^{15}\text{N}$ from the deeper sections to the surface of the core (<7 ‰ to >10 ‰). This
380 increase was less evident at core BGGC5, with values of ~9 ‰ at depths to >10 ‰ on
381 the surface (Fig. 5a,b). The reduced TOC content was related to slightly higher $\delta^{13}\text{C}$
382 values (~ -20 ‰) in both cores.

383

384 **4.3.3. Pollen record**

385 Initial surveys at core BTGC8 (Tongoy Bay) revealed extremely low pollen
386 abundances which hampered further palynology work. A comprehensive pollen
387 analysis was only conducted for core BGGC5 (Guañeros Bay). The pollen record
388 of core BGGC5 consisted of 29 samples shown in Figure 7. The record was divided
389 into five general zones following visual observations of changes in the main pollen
390 types and also assisted by CONISS cluster analysis.

391 Zone BG-1 (cal BP 8200 – 7600): This zone is dominated by the herbaceous taxa
392 Chenopodiaceae, *Leucheria*-type, Asteraceae subfamily (subf.) Asteroideae, Apiaceae
393 with overall high values for the wetland genus *Typha* spp.

394 Zone BG-2 (cal BP 7600 – 6500): This zone is also dominated by Chenopodiaceae,
395 *Leucheria*-type and Asteraceae subf. Asteroideae. In addition, other non-arboreal
396 elements such as *Ambrosia*-type, Poaceae, Brassicaceae and *Chorizanthe* spp. expand
397 considerably.

398 Zone BG-3 (cal BP 6500 – 3400): This zone is marked by a steady decline in
399 Chenopodiaceae and *Leucheria*-type, and by the expansion of several other
400 herbaceous elements, such as Euphorbiaceae, *Baccharis*-type and Brassicaceae.

401 Zone BG-4 (cal BP 3400 – 120): This zone is mostly dominated by Ast. subf.
402 Asteroideae, and marked by the decline of Chenopodiaceae and *Leucheria*-type. Other
403 coastal taxa –such as Euphorbiaceae, *Baccharis*-type, Asteraceae subf.

404 Chichorioideae, *Quillaja saponaria*, Brassicaceae and *Salix* spp.– also expand in this
405 zone.

406 Zone BG-5 (cal BP 120 – -60): The upper portion of the record is dominated by
407 Asteraceae subf. Asteroideae and Poaceae, and marked by higher amounts of
408 Geraniaceae, Asteraceae subf. Mutisieae, Myrtaceae and *Q. saponaria*. Additionally,
409 this zone includes introduced pollen types such as *Rumex* spp. and *Pinus* spp. The
410 latter is not shown in the diagram of Figure 8 because its abundance was minimal.
411 Overall, the most distinctive trend revealed by core BGGC-5 is a long-term reduction
412 in Chenopodiaceae and higher amounts of Euphorbiaceae and Asteraceae subf.
413 Asteroideae. Along with these changes, a further expansion of several other pollen
414 representative of the coastal shrub land vegetation began at about cal BP 6500.

415

416 **4.4. Trace element distributions**

417 Trace element distributions are shown in figures 8a and 8b for Guanaqueros (BGGC5)
418 and Tongoy Bays (BTGC8), respectively. We use Al as a normalizing parameter for
419 enrichment/depletion of elements due to its conservative behavior. The elements are
420 presented as metal/Al ratios. Trace metals are sensitive to the presence of oxygen (U,
421 Re, Mo) showing an increasing metal/Al ratio from the base of core BGGC5 (cal BP
422 ~8210) up to cal BP 6500. After this peak, ratios showed a slight increase towards cal
423 BP 1700, close to the beginning of the recent era, followed by a sharp reduction until
424 present. Similarly, metal ratios at core BTGC8 increase over time, yet the peak was
425 observed at cal BP ~1000. The exception to this trend was Mo, which reached a
426 maximum value up to cal BP 6500 and then reduced steadily into the present.
427 Additionally, metal/Al values were higher at core BGGC5. Iron revealed a clear
428 upward trend around cal BP 3300 – 3500 at core BGGC5, which was not clearly
429 observed at the Tongoy core. Instead, core BTGC8 showed peak Fe values around cal
430 BP 6500 – 7800; in both cores, Fe increased in the past 130 years. No clear trend
431 could be established for Mn.

432 A second group of elements (metal/Al ratios), including Ca, Ni, Cd and P (related to
433 primary productivity and organic fluxes), showed a pattern similar to that of Mo/Al of
434 core BGGC5, i.e. increasing values from cal BP ~8000 reaching highest values around
435 cal BP 6500; after that the values followed constant reductions towards the present. A
436 third group, consisting of Ba and Sr, exhibited a less clear pattern. Ca, Ni, Cd and P
437 ratios at core BTGC8 showed only slightly decreasing values, and very low peak
438 values compared to core BGGC5. Metal/Al ratios of Ba and Sr showed no substantial
439 variation in time, except for Ca and Cd in the last 1000–1700 years. In general, all the

440 elements' concentrations were lower and presented a long-term reduction pattern
441 towards the present.

442 An exception to the previously described patterns was Cu/Al, which reach a maximum
443 value at cal BP ~3600–3700 and showed a conspicuous upward trend in the past ~130
444 years. This was also observed at core BTGC8, but with lower concentrations than at
445 core BGGC5.

446 The authigenic enrichment factor of elements was estimated according to: $EF =$
447 $(Me/Al)_{sample} / (Me/Al)_{detrital}$; where $(Me/Al)_{sample}$ is the bulk sample metal (Me)
448 concentration normalized to Al content and the denomination “detrital” indicates a
449 lithogenic background (Böning et al., 2009). Detrital ($[Me]_{detrital}$ and $[Al]_{detrital}$)
450 concentrations were established considering local TM abundance, which is more
451 accurate than using mean Earth crust values (Van der Weijden, 2002). We used average
452 element concentrations on surface sediments (0–3 cm) of the Pachingo wetland (Table
453 3). The values suggest a large enrichment of nutrient-type elements in a period prior to
454 cal BP 6500, following the trend of the Me/Al ratios, except for Ba and Fe which did
455 not show authigenic enrichment. EFs showed a sharp enrichment reduction in recent
456 times after cal BP 130 (Table 4).

457

458 5. Discussion

459 5.1. Sedimentary composition of the cores: terrestrial *versus* biogenic inputs

460 The sediments in the southern zones of the bays are a sink of fine particles transported
461 from the north and the shelf (Fig. 5a, 5b), and respond to water circulation in the
462 Guanaqueros and Coquimbo Bays (Fig. 1) having two counter-rotating gyres moving
463 counterclockwise to the north and clockwise to the south (Valle-Levinson and
464 Moraga, 2006) (Fig. 1). The differences established by the sediment composition of
465 the bays shows that Guanaqueros Bay's sediments better represent the organic carbon
466 flux to the bottoms, with higher accumulation rates (mean value: $16 \text{ g m}^{-2} \text{ yr}^{-1}$) and
467 higher amounts of siliceous microfossils. Furthermore, is it a better zone than Tongoy
468 to identify pollen records (Figs. 5b, 6 and 7). Both areas have sediments composed by
469 winnowed particles, relatively refractory material (C/N: 9–11), which has a slightly
470 lower isotopic composition than the TOC composition in the column water (-18 ‰,
471 Fig. 2), and transported by water circulating over the shelf.

472 The isotopic variations in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ did not clearly establish differences between
473 bays sediments, but minor differences in $\delta^{15}\text{N}$ would point to a greater influence of the

Comentario [P7]: After this line, all paragraphs were modified

474 upwelling's nutrient supply and the OMZ on the shelf, resulting in $\delta^{15}\text{N}$ of 9 – 10 ‰
475 in the Guanaqueros Bay, values which are slightly higher than in the Tongoy Bay
476 sediments. This isotopic composition correspond to that of NO_3^- in upwelled waters
477 (De Pol-Holz et al., 2007) in the range of those measured at northern and central Chile
478 (~11 ‰; Hebbeln et al., 2000, De Pol-Holz et al., 2007), coming from the isotopic
479 fractionation of NO_3^- during nitrate reduction within the OMZ, leaving a remnant NO_3^-
480 enriched in ^{15}N (Sigman et al., 2009; Ganeshram et al., 2000 and references therein).
481 At sediment core BTGC8, lower values (< 8 ‰) measured at greater depths within the
482 core should account for the mix with isotopically lighter terrestrial organic matter
483 (Sweeney and Kaplan, 1980) due to its vicinity to a small permanent wetland in the
484 southern side of Tongoy Bay (Pachingo), the sediments of which have $\delta^{15}\text{N}$ of 1 –
485 8 ‰ (Muñoz et al., data will be published elsewhere), suggesting that Tongoy
486 sediments contain a combination with continental material (Fig. 5b).
487 Thus, cores BGGC5 and BTGC8 in the Guanaqueros and Tongoy Bays are recording
488 the variability of oceanographic conditions, but in the Tongoy core, the concentration
489 of oceanographic proxies dilutes due to the input of terrigenous material. This helps to
490 decipher the climatic variability considering that the main input of clastic material to
491 the area takes place during major flooding events. Additionally, the main circulation
492 of the bay system leads to favorable conditions for sedimentation and the preservation
493 of organic marine proxies in the Guanaqueros Bay, hence making the sedimentary
494 records of these sites complementary.

495

496 **5.2. Temporal variability of primary productivity and the oxygenation of bottoms**

497 Ca, Sr, Cd and Ni profiles suggest a lower share of organic deposition over time (Fig.
498 8a, 8b), consistent with the slight reduction of TOC content observed in the sediments
499 (Figs. 5a, 5b), and concomitantly with other elements related to organic fluxes to the
500 bottom and primary productivity. In the case of Ba, it is actively incorporated into
501 phytoplankton biomass or adsorbed onto Fe oxyhydroxides, increasing the Ba flux
502 towards the sediments. It is better preserved in less anoxic environments with
503 moderate productivity (Torres et al., 1996; Dymond et al., 1992), as is the case of our
504 study site (Gross Primary Productivity = 0.35 to 2.9 g C m⁻¹d⁻¹; Daneri et al., 2000).
505 The maximum Ba concentrations were before cal BP 6500. The same is true for Ca,
506 Cd and Ni, suggesting that the maximum productivity and organic fluxes to the
507 bottoms occurred during this period. After this age, the reduction in TOC and other

Comentario [P8]: We combine the discussion of proxies for oxygenation and productivity. This paragraph start with a short explanation of the profiles.

Comentario [P9]: We analyze the proxies (organics and inorganics) according time (from the past to the present), considering the main periods observed.

508 nutrient-type elements (Ni, Sr, Ca, Cd) into the present is consistent with the rise in
509 oxygen in bottoms. Hence, the slight rise of Ba from cal BP 4000 to the present (Fig.
510 8a) is a response to this less anoxic environment leading to negative correlation with
511 TOC (-0.59; Table 5) due to Ba remobilization in anoxic conditions before
512 cal BP 6500. On the other hand, P distribution showed a trend similar to that of TOC
513 and other elements related to organic fluxes into the bottom (Ni, Cd), although with a
514 lower correlation (~0.6). This is consistent with the distributions observed for U, Re,
515 and Mo at core BGGC5, which indicate that anoxic or suboxic conditions were
516 developed from cal BP 8200 to ~ cal BP 1700, but were stronger before cal BP 6500
517 (Fig. 8a, 8b). After this period and into the present, a remarkable reduction in their
518 concentration suggests a more oxygenated bottom environment, concurrent with lower
519 organic fluxes to the sediments. The Re profile shows the influence of suboxic waters
520 not necessarily associated with higher organic matter fluxes to the bottom. Since this
521 element is not scavenged by organic particles, its variability is directly related to
522 oxygen changes (Calvert and Pedersen, 2007, and references therein). Additionally, it
523 is strongly enriched above crustal abundance under suboxic conditions (Colodner et
524 al., 1993; Crusius et al 1996), being >10 times at core BGGC5 (Table 4) before cal BP
525 1700. In the same manner, U shows a similar pattern and while organic deposition has
526 an impact on its distribution (Zheng et al., 2002), it is also related to changes in bottom
527 oxygen conditions.

528 Otherwise, the accumulation of P depends on the deposition rate of organic P (dead
529 plankton, bones and fish scales) on the bottom, and is actively remineralized during
530 aerobic or anaerobic bacterial activity. P and TOC showed a declining trend towards
531 the present, suggesting reducing flux of organic matter over time, which was also
532 observed for Ni and Cd distributions. Alternatively, reducing fluxes of organic proxies
533 could be explained by the higher remineralization of organic material settled on the
534 bottom due to higher oxygen availability, as shown by U, Mo and Re distributions
535 (Figs. 8a, 8b). To better approach this issue, establishing the variability of primary
536 productivity over time and the environmental factors that facilitate its development is
537 required.

538 Productivity reconstructions were based on qualitative diatom and sponge spicules
539 relative abundances, quantitative diatom counts (valves g^{-1}), and biogenic opal content
540 only in core BGGC5, since core BTGC8 registered low valve counts (< 1 % in relative
541 diatom abundance). However, in both cores diatom assemblages were represented

542 mainly by *Ch.* resting spores, which are used as upwelling indicators (Abrantes 1988,
543 Vargas et al., 2004). The downcore siliceous productivity based on opal distribution
544 (Fig. 6) distinguished three main time intervals of higher productivity, which coincide
545 with the ages highlighted by the distribution of the sedimentary proxies seen
546 previously: (1) > cal BP 6500, (2) cal BP 1700 – cal BP 4500 and (3) recent times (CE
547 2015) – cal BP ~130. The opal accumulation rate in the first interval was remarkably
548 high, amounting to $\sim 27 \pm 13 \text{ g m}^{-2} \text{ yr}^{-1}$ (range: 9 – 53 $\text{g m}^{-2} \text{ yr}^{-1}$, Table 4), when
549 *Chaetoceros* spores were predominant, indicating an upwelling intensification. During
550 the first period, all metal proxies showed primary productivity increases before cal BP
551 6500, as indicated by opal accumulation within the sediments. Here, Cd and U
552 accumulations in the sediments resulted in high Cd/U ratios, even at core BTGC8 (> 2;
553 Fig. 6), pointing to very low oxygen conditions (Cd/U ratios could vary between 0.2
554 and 2 from suboxic to anoxic environment; Nameroff et al., 2002). In addition, during
555 this period the presence of sulfidic conditions is suggested by Cd and Ni enrichments
556 (>140 and 3, respectively, Table 4), since its buildup within the sediments is highly
557 controlled by sulfide concentrations (Chaillou et al., 2002; Nameroff et al., 2002;
558 Sundby et al., 2004), though Mo and Re were not especially high during this period
559 (~17 and ~19, respectively, Table 4), all of which is suggesting an intensification of
560 the upwelling during this time interval.

561 In the second interval, opal accumulation decreased to $\sim 11 \pm 4 \text{ g m}^{-2} \text{ yr}^{-1}$ (range: 2 – 21
562 $\text{g m}^{-2} \text{ yr}^{-1}$, peaking at cal BP 3500–4000; Table 4, Fig. 6a), which is partially consistent
563 with nutrient-type element distributions (Fig. 8a). Fe clearly shows higher values
564 around cal BP 3500 (Fig. 8a), helping to boost primary productivity at this time, with a
565 small diatom increase, measured as valves per gram and abundance (%) (Fig. 6a).
566 Other elements showed less prominent accumulations (Ni, Cd, Ba, Ca and P), pointing
567 to lower organic matter deposition into the sediments during this period (Fig. 8a).
568 However, low oxygen conditions within the sediments are maintained, which could be
569 more related to the manifestation of the oxygen minimum zone close to the coast,
570 favoring Mo and Re accumulation until cal BP 1700 (Fig. 8a). Lower Cd/U ratios (~ 1;
571 Fig. 6) were estimated, suggesting higher variations in primary productivity but with
572 moderate changes in oxygen conditions at the bottoms. After cal BP 1700, there is an
573 evident and remarkable reduction in organic fluxes to the sediments and a drastic
574 change to a less reduced environment towards the present, suggesting a more
575 oxygenated bottom environment concurrent with a reduction in primary productivity,

Comentario [P10]: We define the main productivity periods that are related with the other proxies distribution mentioned before.

Comentario [P11]: We highlighted the main observations in the first period.

Comentario [P12]: We continued with the observations done in the second period

Comentario [P13]: And observations in the third period, including the last ~ 130 years

576 except for the last ~130 years, when increasing opal accumulations and Cd/U ratios
577 were estimated towards the present (mean opal value of $29 \pm 14 \text{ g m}^2 \text{ yr}^{-1}$, range: 3 –
578 $40 \text{ g m}^2 \text{ yr}^{-1}$; Fig. 5, 6, Table 4). However, low diatom abundances were observed
579 (range: $0.5 - 4.9 \times 10^6 \text{ valves g}^{-1}$), probably because few sections of the core surface
580 were analyzed for diatoms, leading to a low resolution of this measurement in the most
581 recent period. In addition, the flux calculations were based on recent sedimentation
582 rates, an estimation that tends to be higher than in the millennial time scale,
583 overestimating the opal flux. In part, this explains the inconsistencies found between
584 the rise in organic flux and low diatom abundance.

585 Otherwise, there is a conspicuous upward trend of Cu/Al, Fe/Al and Mn/Al in recent
586 times, consistent with high organic fluxes to the bottoms (Fig. 5a,b; 8a,b) and
587 concomitant with the decreasing trend and lower EFs of Re, U, and Mo (Fig. 8a, 8b,
588 Table 4). This could be due to the presence of particulate forms and oxides formation
589 (Peacock and Sherman, 2004; Vance et al., 2008; Little et al., 2014) occurring in the
590 presence of an oxygenated environment that results in a high metal enrichment of
591 these elements ($EF_{Cu}=4.6\pm 0.5$, Table 4). All of this is suggesting a higher productivity
592 in the last 130 years occurring in a more oxygenated environment, which is actually
593 contradictory. We assume that episodic oxygenation events related to El Niño change
594 the original extent of these sensitive redox trace elements' accumulation because of
595 their remobilization to soluble forms (Morford and Emerson, 1999); therefore, the
596 increased frequency and intensity of El Niño would result in a mean effect which is
597 observed as a gradual change in metal contents over time. Several observations made
598 at the central Peruvian and south-central Chilean coasts ($12 - 36^\circ \text{S}$) reveal that
599 present-day wet/dry variability associated with ENSO has a strong impact on the
600 bottom ocean oxygenation (Escribano et al., 2004; Gutiérrez et al., 2008; Sellanes et
601 al., 2007), suggesting a large increase in oxygen levels at bottoms during El Niño
602 events that change the sediment geochemistry, the effects of which can be observed
603 several months later. Thus, the oceanographic conditions that have prevailed in the
604 past should be determinants not only for productivity but also for oxygen conditions
605 above the bottoms, which is reflected in our sedimentary records.

606

607 **5.3. Main climatic implications**

608 According to modern climatology, paleoenvironmental records from semi-arid regions
609 have been interpreted mainly based on the past variability in the intensity and latitudinal

Comentario [P14]: We modified this section following the same structure than before.

610 position of the SWW (Veit et al., 1996; Hebbeln et al., 2002; Lamy et al., 1999;
611 Maldonado and Villagrán, 2002). This has an impact over relevant oceanographic
612 characteristics such as upwelling and, therefore, productivity. We established marked
613 differences in paleo productivity proxies and paleo-oxygenation in the last 8300 years
614 (Figs. 6, 8); a high marine productivity prevailed during our first period (cal BP 8300 –
615 6500) according to what was established for central Chile between 10 and 5 kyr due to
616 sustained mean La Niña-like conditions (De Pol-Holz et al., 2006; Kaiser et al., 2008),
617 which is caused by reduced ENSO variability and a northward displacement of the
618 ITCZ and implies more permanent southeast trades and hence, the upwelling of rich-
619 nutrient cold waters (Koutavas et al., 2006). Our high productivities occurred
620 concomitantly with low oxygen conditions at bottoms, both reaching a maximum level
621 at cal BP 6500. This corresponds to the highest productive period in the last 8300 years,
622 indicating an intensification of the SPSA and a weakening of SWW. In addition, our
623 pollen records point to the main driest conditions during this period (Fig. 9), which
624 matches with other reports in the area, indicating that an arid phase was developed until
625 cal BP 5700 (Jenny et al 2002, Maldonado and Villagrán, 2006), and which could be
626 extended until cal BP 4200 (Maldonado and Rozas, 2008; Maldonado and Villagrán,
627 2002, 2006). This period was characterized by reduced rainfalls and intense coastal
628 humidity, which have been associated to coastal fogs that frequently occur during the
629 spring due to a strengthening of the SPSA (Vargas et al., 2006; Garreaud et al 2008;
630 Ortega et al., 2012) and La Niña-like conditions, associated with the cold phase of the
631 Pacific Decadal Oscillation, which explains the main variability of the SPSA
632 (Ancapichún and Garcés-Vargas, 2015). Strengthened easterlies favor upwelling and
633 cause SST cooling, also pointing to a northward location of the ITCZ which was
634 suggested for the early-mid Holocene (Kaiser et al., 2008; Lamy et al., 2010). This
635 would be consistent with our records and points to more favorable conditions for
636 upwelling strengthening around cal BP 6500 at central Chile. However, others have
637 suggested a reduced ENSO variance in this period (Rein et al., 2005) linked to fresh
638 water melting that counteracted the insolation regime (Braconnot et al., 2012), but
639 points to a more limited cold-dry period between 6700 – 7500 years ago. This could be
640 due to less frequent or less intense warm anomalies related to a CP mode ENSO, which
641 produce moderate El Niño events at the Central Pacific (CP) and strong La Niña off
642 Peru (Carré et al., 2014), matching our records of maximum productivity.

643 After this date, a decreasing productivity occurred under more warm and humid climatic
644 conditions that would be due to an enhancement of regional precipitation in the northern
645 margin of SWW (Jenny et al., 2003; Maldonado and Villagrán, 2006), consistent with
646 the gradual rise of K/Ca, Fe, Al and Pb distributions in our cores (Fig. 8, 9), usually
647 considered to be indicators of continental input by fluvial or aerial transport (Calvert
648 and Pedersen, 2007; Kaiser et al., 2008; Govin et al., 2012; Ohnemus and Lam, 2015;
649 Saito et al., 1992; Xu et al., 2015). This would contradict a second period of reduced (or
650 weak) ENSO activity reported for cal BP 4500, and also with others who sustain that
651 this weak activity condition took place from cal BP 6000 to 4000 (Koutavas and
652 Joanides, 2012; Carré et al., 2014). It is also consistent with the pollen records of central
653 Chile that suggest an arid phase from cal BP 6200 until cal BP 4200 (Maldonado and
654 Villagrán, 2006). No sharply contrasting dry and cold periods were identified after cal
655 BP 6500, mostly a gradual increase in humidity and a weakening in paleo-productivity
656 proxies (Fig. 8, 9) that would be consistent with the beginning of higher ENSO
657 variability for central Chile after cal BP 5700 (Jenny et al., 2002, Maldonado and
658 Villagrán, 2002, 2006). Nonetheless, the slight rise of diatom abundance and opal
659 concentrations between cal BP 4500 and 3000, along with a slight accumulation of
660 nutrient elements (Ni, Cd, Fe and Ca concentrations; Fig. 8) and small rises in organic
661 carbon flux and Cd/U ratios (Fig. 5, 6), would be related to an increase in continental
662 nutrient inputs that help primary productivity development observed in sedimentary
663 records for the north-central Chilean margin (Dezileau et al., 2004; Kaiser et al., 2008).
664 A peak of La Niña activity around cal BP 3000–4000 has been otherwise proposed for
665 the tropical east Pacific (Toth et al., 2012), which would also explain the increase in the
666 productivity's proxies. This is a period of increased ENSO variability from cal BP~
667 5700, and stronger El Niño events after cal BP 4000–4500, concomitant with the high
668 variability of latitudinal displacements of the ITCZ related to the seasonality of
669 insolation described for the mid and late Holocene period (Haug et al., 2001; Toth et al.,
670 2012; Carré et al., 2014). This is consistent with the occurrence of alluvial episodes in
671 the area caused by more frequent or heavier rainfall events over time, related to
672 intensified Westerlies and increased El Niño events (Jenny et al., 2002; 2003; Ortega et
673 al., 2012; Ortega et al., 2019). This is leading to more humid conditions and greater
674 continental inputs as suggested by our pollen moisture index and sedimentary records.
675 In spite of the dominance of warm events described for this period, they were not strong
676 enough to change the suboxic conditions at the bottoms, which were maintained until

677 cal BP 1700 (Fig. 8; see U, Mo and Re). After that, the drastic oxygenation of the
678 bottoms occur during higher frequency and intensity of flooding events recorded in
679 central Chile in the last 2000 years, consistent with more frequent El Niño events (Jenny
680 et al., 2002, Toht et al., 2012). In this regard, oxygen variations at the bottoms would be
681 related to less intense OMZs during warm El Niño-like phases (and vice versa during
682 La Niña). These tend to be associated with low productivity (Salvatteci et al., 2014),
683 and, in turn, reduce organic fluxes and oxygen consumption during organic matter
684 diagenesis. Thus, more frequent El Niño events in recent times could be the cause for
685 oxygen increments and lower productivity, which has been deduced from very low
686 chlorins (or photosynthetic pigments) sediment records in the past 2000 years (Rein,
687 2007), which is consistent with our observations.

688 In recent times, the most extreme and longer ENSO events have been established in the
689 20th century, mostly after the 1940s and characterized by severe floods and droughts
690 linked to global climate change (Gergis and Fowler, 2009). Similarly, warmer periods
691 have been characterized by lower primary productivity and more oxygenated waters
692 over the shelf. However, enhanced solar heat over the land in northern Chile results in
693 the intensification of coastal southerly winds, strengthening upwelling during warmer
694 ENSO periods (Vargas et al., 2007). If this is coming along with Fe inputs to the bay
695 system, it could explain the productivity records during recent times. In addition, during
696 the El Niño conditions, the normal dominance of diatoms is replaced by smaller size
697 phytoplankton, making a relevant contribution to overall primary production (Iriarte et
698 al., 2000; Rutlland and Montecino, 2002; Iriarte and Gozalez, 2004; Escribano et al.,
699 2004) that would change sedimentary diatom records but maintaining organic fluxes to
700 the bottoms.

701

702 **6. Conclusions**

703 The ocean circulation in our study sites seems to impact both places differently,
704 leaving more variable grain compositions and higher TOC contents in the
705 Guanaqueros Bay (core BGGC5) than in the Tongoy Bay (core BTGC8), with the
706 latter increasingly impacted by terrigenous inputs due to the flow of several creeks
707 during major flooding events. Nevertheless, both core records sustain a reduction of
708 organic flux to the bottoms after cal BP ~6500 and into present times. **This** is probably
709 due to higher ENSO variability over time, sustained by an increase in the pollen
710 moisture index, suggesting a long-term rise in precipitation after cal BP 6500. At such

Comentario [P15]: After this line the paragraphs were modified and shortened.

711 point, an overall expansion of the coastal vegetation and larger river runoffs occurred,
712 expanding the grain size of the sediments and increasing the concentrations of
713 elements with an important continental source (Al, Fe, K and Pb). Therefore, eolian
714 and fluvial transportation seems to become relevant after this date to boost
715 phytoplankton when ENSO variability increases and in the face of stronger El Niño
716 events.

717 Our results suggest that the geochemistry and sedimentary properties of coastal shelf
718 environments in north-central Chile have changed considerably during the Holocene
719 period, suggesting two relevant changes in redox conditions and productivity, pointing
720 to a more reducing environment and higher productivity around cal BP 6500. After that,
721 a less reducing environment along with decreasing trends in primary productivity and
722 increased humid conditions in time, were developed until cal BP 1700. The northward
723 shifts of the Southern Westerly Wind belt, in addition to an increased frequency of El
724 Niño events, have been proposed as the main drivers for climatic conditions during this
725 period. These elements have introduced a high variability in primary productivity
726 during this time interval. Additionally, this also impacted the accumulation of organic
727 matter due to an intensification of its remineralization, showing a decreasing trend in the
728 buildup of nutrient type elements and organic carbon burial rates towards the present.
729 The decrease in oxygen content at bottoms was highly influenced during El Niño
730 events, something that seems to have been operating at higher frequencies after cal BP
731 1700, and especially after cal BP 130, when the most extreme events become more
732 frequent.

733 Finally, these changes highlight the sensitivity of these environments to climate
734 variability at different timescales, which is consistent with the description of past
735 regional climatic trends. Based on the dramatic changes observed in the last 1700
736 years, future changes are expected in the ongoing scenario of global warming at
737 unprecedented rates.

738

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1165
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Tables

Table 1. Radiocarbon dates for BGGC5 and BTGC8 sediment cores collected from mixed planktonic foraminifera and monospecific benthic foraminifera (*Bolivina plicata*), respectively. The ^{14}C -AMS was performed at NOSAM-WHOI. The lab code and conventional ages collected from each core section is indicated. For error calculations see <http://www.whoi.edu/nosams/radiocarbon-data-calculations>.

Core identification	Material	Mass (mg)	Lab Code NOSAM	Modern fraction pMC	1σ error	Conventional Age BP	1σ error
BGGC5	Planktonic 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

BTGC8	Benthic 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 (DR) estimation considering the ^{210}Pb age determined with the CRS model (McCaffrey and Thomson, 1980) at a selected depth sections of the core, compared with ^{14}C ages (yr BP) from marine13.14 curve (Reimer et al., 2013), according to Sabatier et al. (2010).

Core	Depth (cm)	Age from CRS model (AD) ^a	Age years BP ^b	^{14}C age Marine 13.14	^{14}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

^aAnno Domini

^bBefore present=1950

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

Element	Metal/Al x 10³	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

Table 4. Mean authigenic enrichment factor (EF) \pm SD of trace elements calculated for Guanaqueros Bay (BGGC5 core). Lithogenic background was estimated from surface sediments of Pachingo wetland cores (see text). Age ranges were based on the variability of diatom abundance (valves g⁻¹).

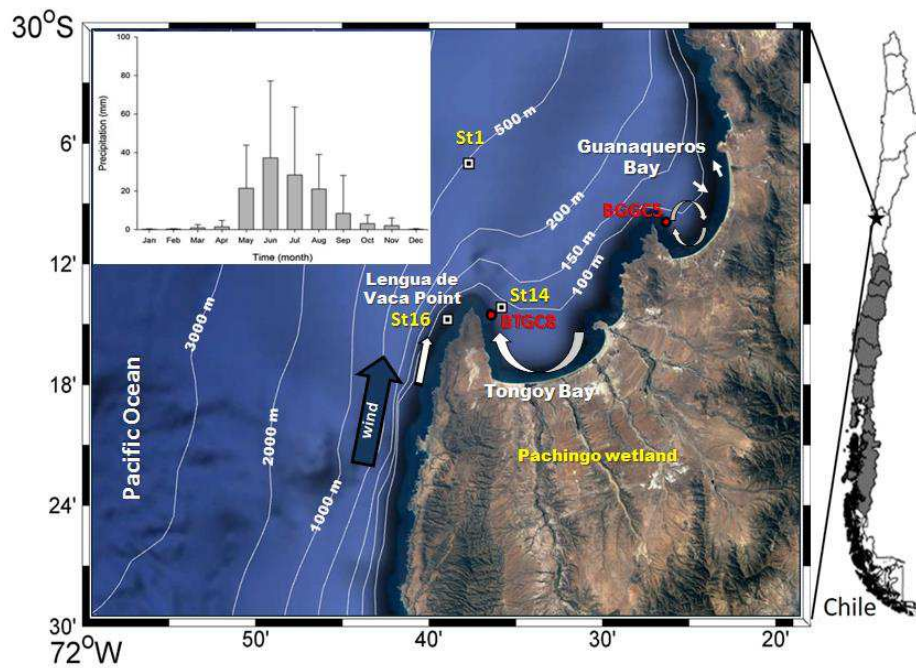
Age range (cal BP)	Diatoms (x10 ⁶) (min-max)	Opal (g m ⁻² yr ⁻¹) (min-max)	EF _U	EF _{Mo}	EF _{Re}	EF _{Fe}	EF _{Ba}	EF _{Cd}	EF _{Ni}	EF _{Cu}	EF _P
-65 – 130	0.5 – 4.9	3 – 40	2.6 \pm 0.7	5.5 \pm 1.3	10.5 \pm 2.0	0.8 \pm 0.1	0.8 \pm 0.1	30.3 \pm 6.3	1.4 \pm 0.2	3.6 ^a \pm 1.3	2.0 \pm 0.4
130 – 1700	0.6 – 1.7	1 – 3	5.6 \pm 1.4	14.5 \pm 3.7	18.4 \pm 3.8	0.9 \pm 0.1	0.8 \pm 0.1	40.6 \pm 3.7	1.9 \pm 0.1	3.0 \pm 0.4	2.4 \pm 0.4
1700 – 4500	1.9 – 5.4	2 – 21	5.5 \pm 0.6	14.5 \pm 1.5	19.8 \pm 2.0	0.9 \pm 0.1	0.8 \pm 0.1	55.1 \pm 12.2	2.3 \pm 0.3	3.1 \pm 0.5	2.2 \pm 0.3
4500 – 6500	2.7 – 4.5	4 – 47	5.1 \pm 0.8	16.9 \pm 3.3	19.5 \pm 3.0	0.9 \pm 0.1	0.9 \pm 0.1	140.1 \pm 46.3	3.4 \pm 0.5	3.1 \pm 0.5	3.2 \pm 0.5
6500 – 8400	15.7 – 41.0	9 – 53	4.5 \pm 0.4	13.9 \pm 2.6	17.9 \pm 2.2	0.9 \pm 0.1	0.9 \pm 0.1	142.5 \pm 24.2	3.4 \pm 0.4	2.5 \pm 0.3	3.9 \pm 0.8

^aMean EF_{Cu} after AD 1936 was 4.6 \pm 0.5

Table 5. Spearman rank order correlations for geochemical data. Significant values >0.8 are indicated in bold.

BGGC5																
	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
BTGC8																
	Al	P	K	Ca	Mn	Fe	Ni	Cu	Mo	Cd	Re	Sr	U	Ba	Opal	TOC
Al	1.00	-0.19	-0.17	-0.37	-0.02	-0.03	-0.39	-0.04	-0.39	0.02	-0.13	-0.58	-0.19	0.07	-0.41	-0.29
P		1.00	0.23	0.00	0.43	0.28	0.58	0.23	0.37	0.13	-0.04	0.30	0.14	-0.14	0.56	0.13
K			1.00	-0.02	0.54	0.41	0.43	0.22	-0.11	0.05	-0.04	0.19	-0.28	0.28	0.26	0.20
Ca				1.00	-0.33	-0.27	0.00	-0.23	0.39	0.01	0.33	0.50	0.47	-0.34	0.20	0.34
Mn					1.00	0.21	0.64	0.01	0.05	0.33	0.15	0.32	-0.02	0.24	0.32	0.00
Fe						1.00	0.13	0.71	-0.40	-0.48	-0.67	-0.37	-0.62	0.13	0.14	0.10
Ni							1.00	0.24	0.56	0.20	0.25	0.64	0.19	-0.16	0.80	0.45
Cu								1.00	-0.25	-0.68	-0.56	-0.22	-0.61	-0.10	0.21	0.37
Mo									1.00	0.45	0.59	0.66	0.69	-0.41	0.58	0.30
Cd										1.00	0.56	0.39	0.52	0.11	0.10	-0.12
Re											1.00	0.53	0.83	-0.16	0.13	0.17
Sr												1.00	0.58	-0.13	0.52	0.23
U													1.00	-0.19	0.21	0.00
Ba														1.00	-0.28	-0.42
Opal															1.00	0.39
TOC																1.00

Figures



1177 Figure 1. Study area showing the position of sampling stations. Sediment cores were
1178 retrieved from Guanaqueros Bay (BGGC5) and from Tongoy Bay (BTGC8) at water

1179 depths of 89 and 85 m, respectively. Information of dissolved oxygen (DO) in the water
1180 column at ST1 and ST16 and of suspended organic particles collected at ST14 sampling
1181 sites was gathered in a previous project (INNOVA 07CN13 IXM-150). Monthly
1182 precipitation in mm (bars) (means \pm SD; Montecinos et al., 2016). Schematic
1183 representation of the bays circulation (white arrows) and wind direction is indicated
1184 (blue arrow) obtained from Valle-Levinson and Moraga-Opazo (2006) and Moraga-
1185 Opazo et al. (2011).

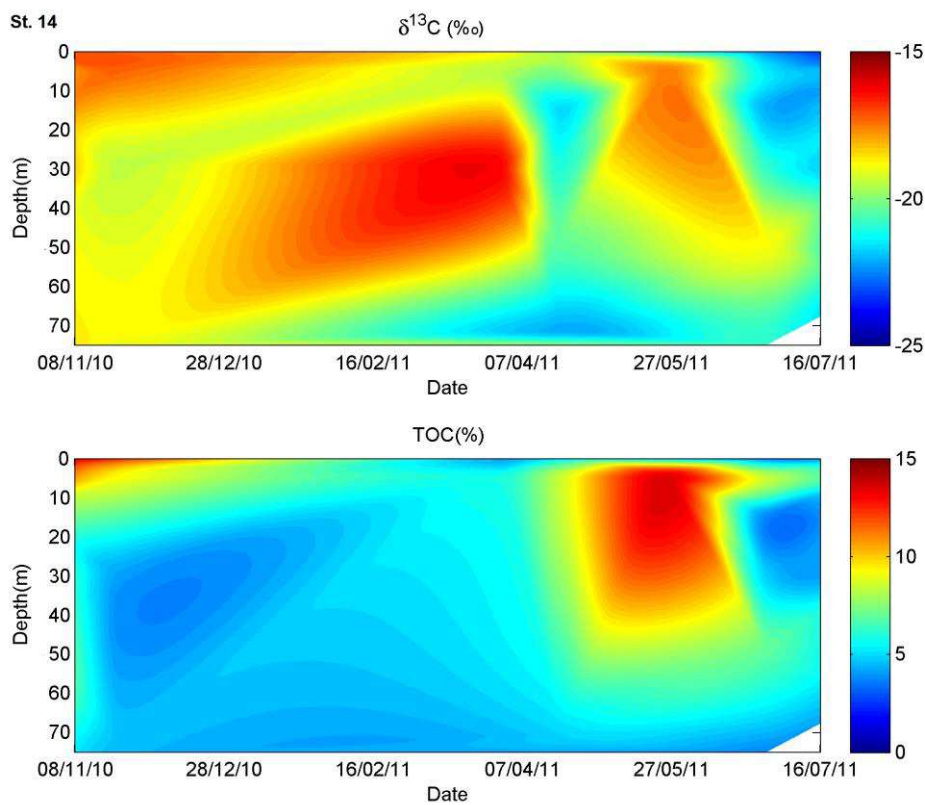


Figure 2. Suspended particulate matter composition (TOC % and $\delta^{13}\text{C}_{\text{org}}$) measured in the water column between October 2010 and October 2011, at station St14, Tongoy Bay, Coquimbo (30°S).

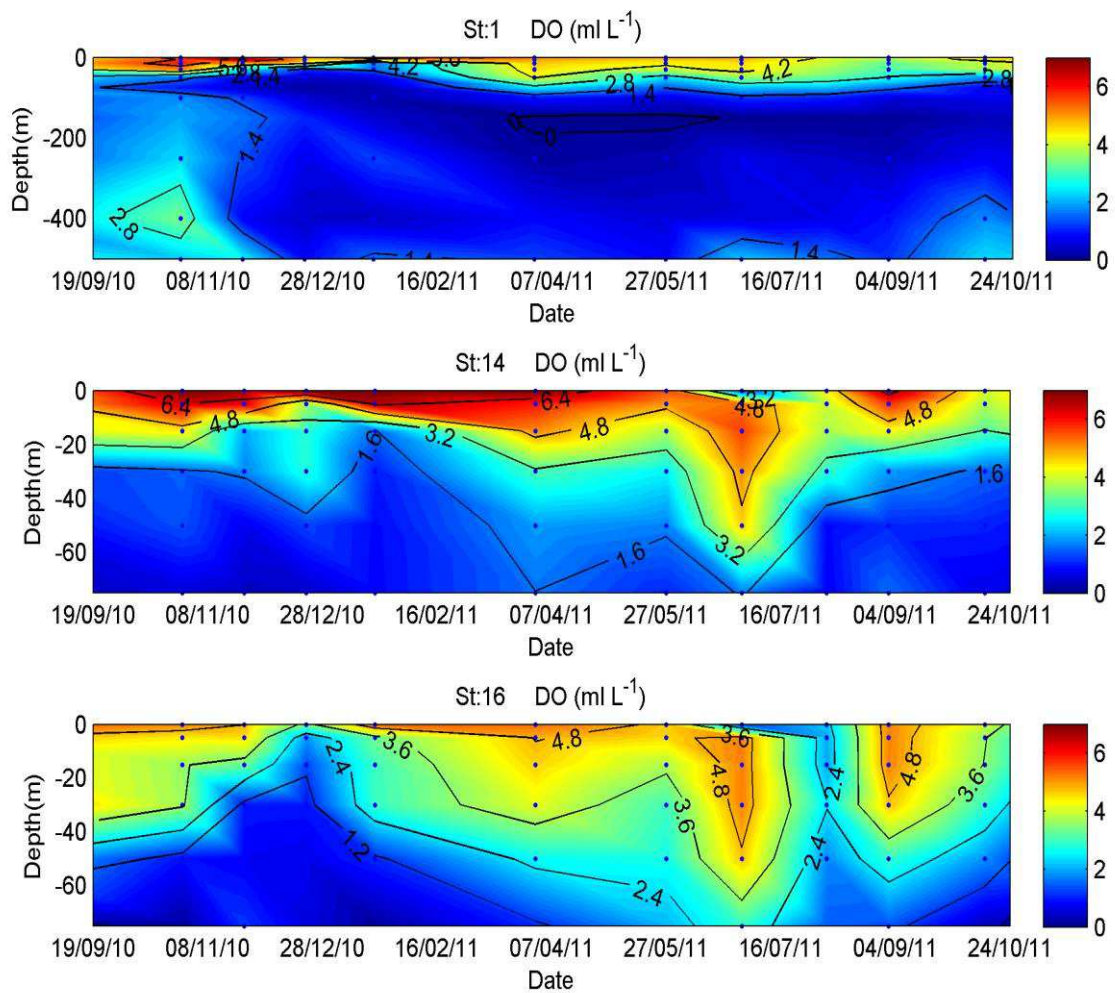


Figure 3. Dissolved Oxygen (DO) time series in the water column measured between October 2010 and January 2011, at stations St1, St14 and St16 off Tongoy Bay, Coquimbo (30°S).

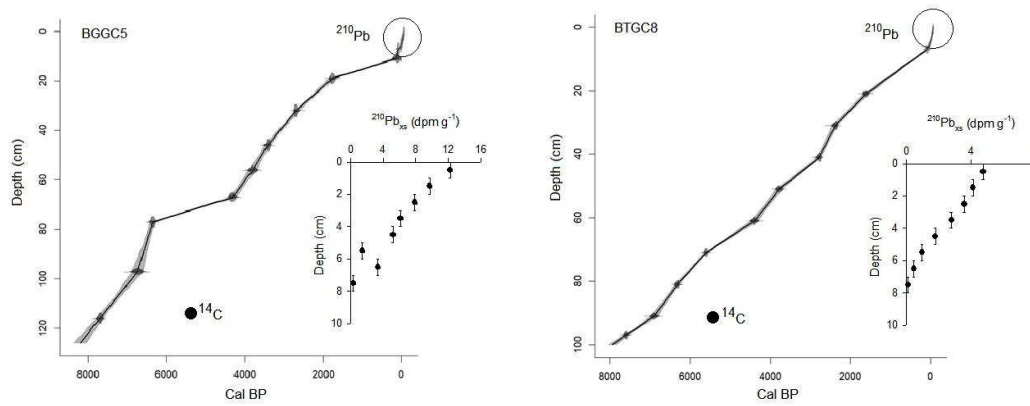
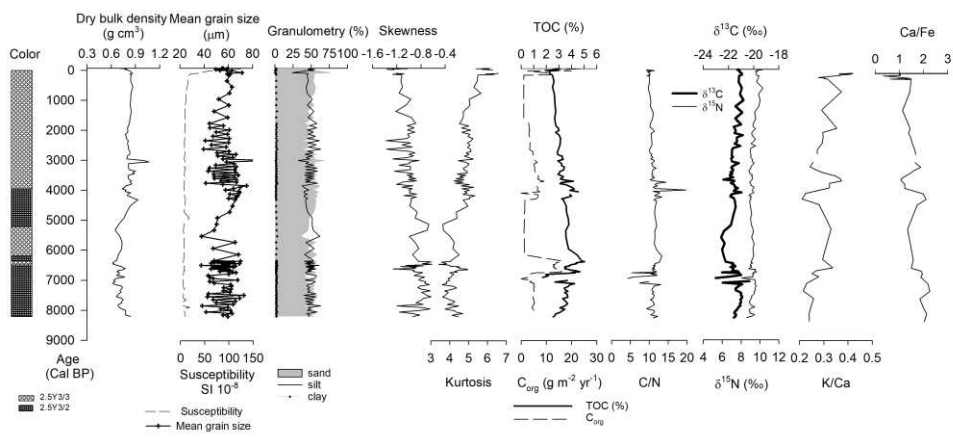


Figure 4. Age model based on ^{14}C AMS and ^{210}Pb measurements. The time scale was obtained according to the best fit of curves of $^{210}\text{Pb}_{\text{xs}}$ and ^{14}C points using CLAM 2.2 software and Marine curve ^{13}C (Reimer et al., 2013).

a) BGGC5



b) BTGC8

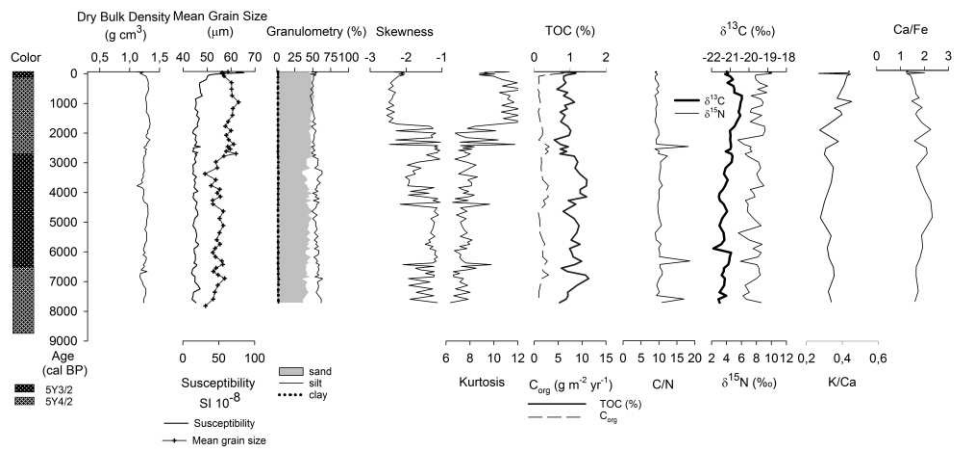


Figure 5. Characterization of sediment cores retrieved from (a) Guanaqueros Bay (BGGC5) and (b) Tongoy Bay (BTGC8), where is shown the color (Munsell chart scale) in depth, dry bulk density, mean grain size, granulometry (% sand, silt and clay), statistical parameters (skewness, kurtosis), organic components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

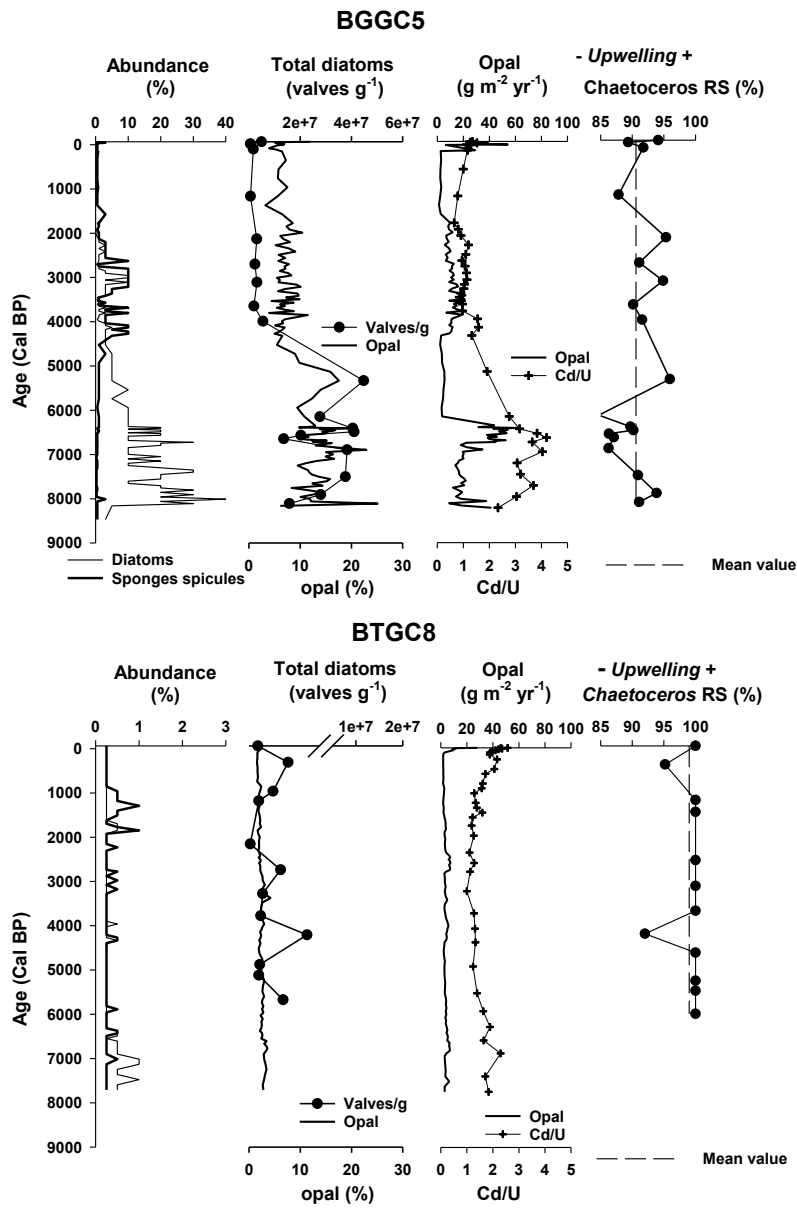


Figure 6. Diatom and sponge spicules' relative abundances, total diatom counts (valves g^{-1}) and opal (%), opal accumulation ($g\ m^{-2}\ yr^{-1}$), and Cd/U ratio, and downcore variations in *Ch.* resting spores percentages as proxy of upwelling intensity in BGGC5 and BTGC8 cores (Guañaqueros and Tongoy Bay, respectively), the medium dash line represents the average of *Ch.* resting spore for the respective core. Cd/U distribution was included as a proxy for redox condition.

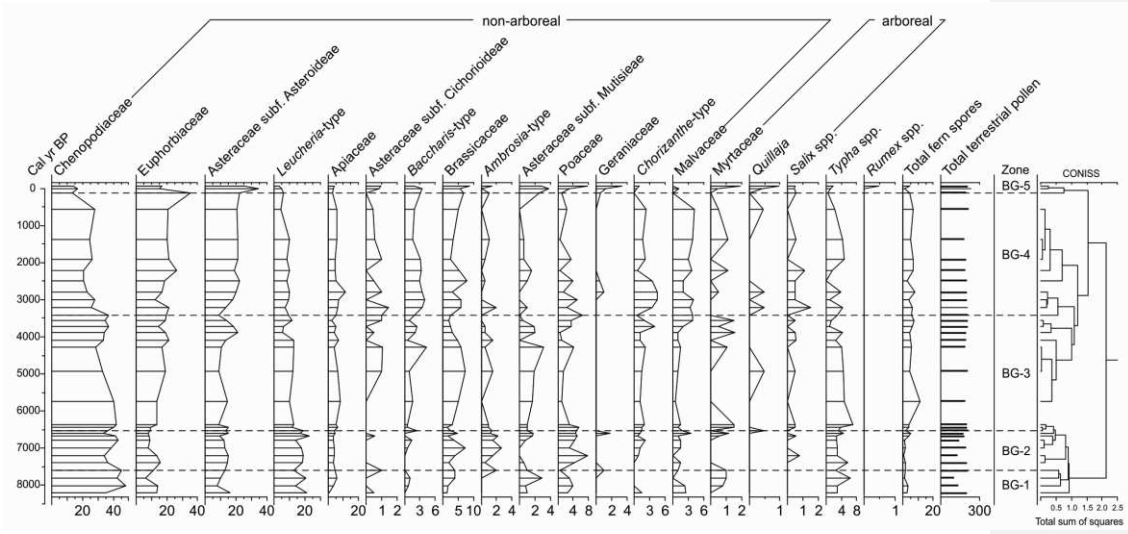
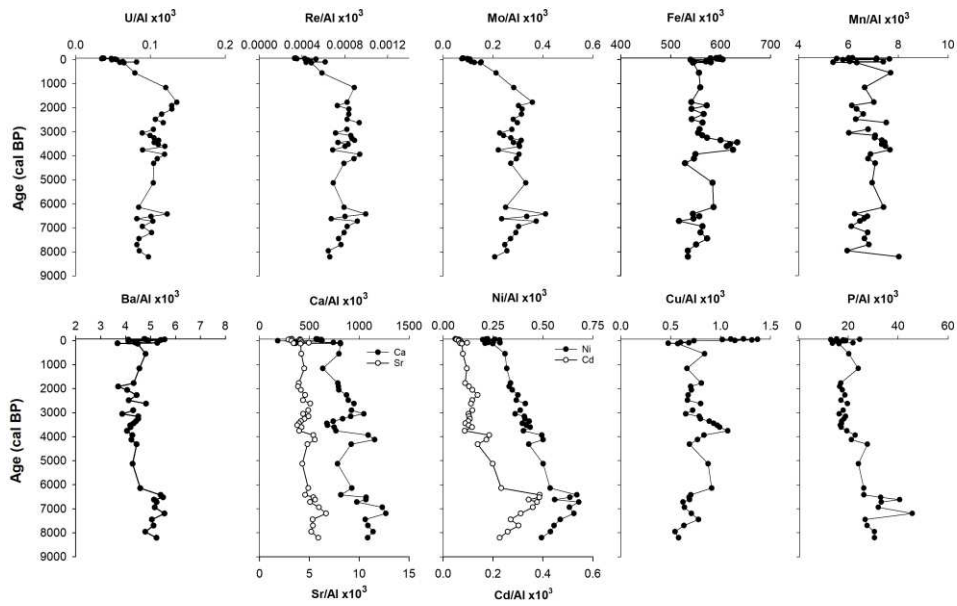


Figure 7. Pollen record in BGGC5 core.

a) BGGC5



b) BTGC8

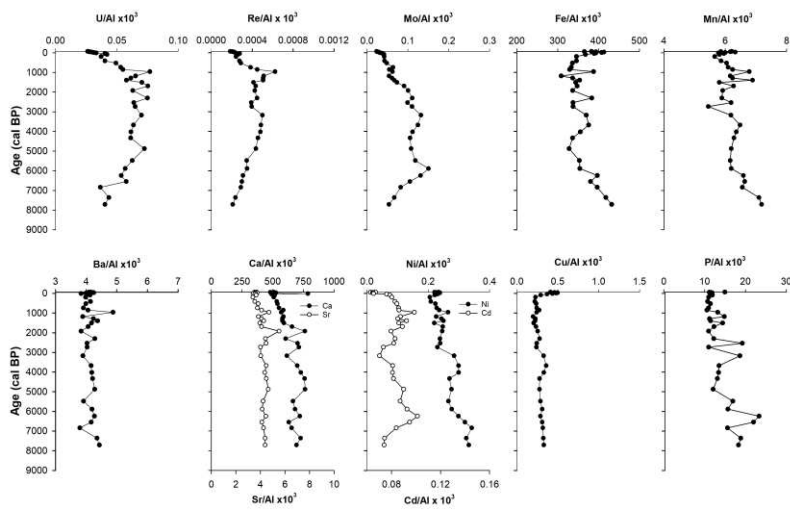


Figure 8. Downcore trace element variations on: (a) Guanaqueros Bay (BGGC5) and (b) Tongoy Bay (BTGC8), off Coquimbo (30°S).

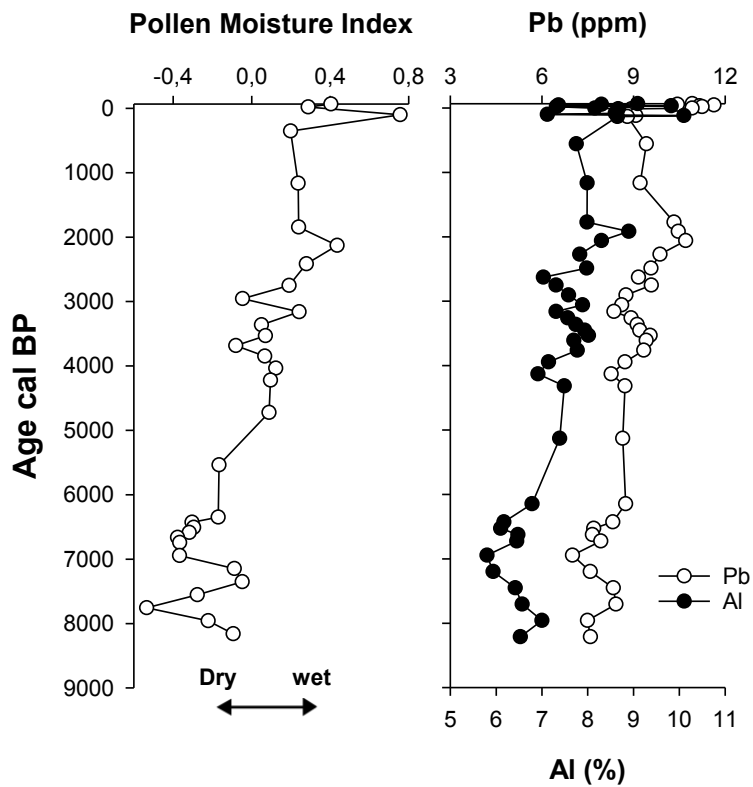


Figure 9. Pollen Moisture Index defined as the normalized ratio between Euphorbiaceae (wet coastal shrub land) and Chenopodiaceae (arid scrubland). Positive (negative) values for this index indicate the relative expansion (reduction) of coastal vegetation under wetter (drier) conditions. Pb and Al distribution at BGGC5 core, representatives of terrigenous input to the bay.