

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

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34

35 **Abstract**

36

37 The aim of this project was to establish past variations in 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 (30° S), for geochemical and biological analyses, including the following:
41 sensitive redox trace elements, biogenic opal, total organic carbon (TOC), diatoms, and
42 stable isotopes of organic carbon and nitrogen. Three remarkable periods were
43 established with different environmental conditions and productivities: (1) > cal BP
44 6600, (2) cal BP 4500–1800, and (3) cal BP 100 to the present (CE 2015). The first
45 period was characterized by a remarkably higher productivity (higher diatom
46 abundances and opal) in which large fluxes of organic compounds were also inferred
47 from the accumulation of elements, such as Ba, Ca, Ni, Cd, and P in the sediments.
48 Meanwhile, significantly reduced conditions at the bottom of the bays were suggested
49 based on the large accumulation of Mo, Re, and U, showing a peak at cal BP 6600 when
50 sulfidic conditions could have been present. According to the pollen moisture index,
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
53 southerly western winds, emulating the La Niña-like conditions, as has been described
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
56 identified between Cal BP 3400 and 4000, although lower amounts of diatom (valves g⁻¹)
57 and nutrient-type metal accumulations were evident. Anoxic conditions at the bottom
58 of the bays changed to an almost stable sub-oxic condition during this time interval. The
59 third period was marked by intense oxygenation after cal BP 1800, as observed by a
60 drastic change in the accumulation of U, Mo, and Re. This was followed by a return to
61 more reduced conditions over the past two centuries, characterized by a small
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
64 humidity conditions were established after cal BP 6600 to the present. We suggest that
65 the oxygenation might be associated with a weak effect from the oxygen minimum zone
66 over the shelf and intensified El Niño activity, introducing oxygenated waters to the
67 coastal zones through the propagation of equatorial waves and establishment of

68 conditions that reduced the primary productivity from the mid-Holocene toward the
69 beginning of the modern era.

70 Keywords: paleoproductivity, paleoredox, trace metals, diatoms, opal, organic carbon,
71 Coquimbo, SE Pacific

72

73 **1. Introduction**

74

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
78 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
81 intensity and, therefore, the productivity (Ruttland and Fuenzalida, 1991; Blanco et al.,
82 2002). Other climate patterns demonstrate impacts at longer timescales (inter-annual,
83 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
86 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
92 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
94 are required for the development of the primary productivity.

95 Off Coquimbo (30° S), there is normally semi-permanent and intense upwelling forced
96 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
98 Niño, the intensities of the mean winds alongshore are reduced (conversely, during La
99 Niña) (Rahn and Garreaud, 2013), impacting the upper circulation of the ocean and
100 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.,

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).
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).
119 North-central Chile is a semi-arid zone that does not receive large fluvial contributions,
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
127 Coquimbo (30° 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
135 determined the enrichment/depletion of elements to establish the primary prevailing

136 environmental conditions during the sedimentation of particulate matter (Böning et al.,
137 2009). In addition, we considered the diatom assemblages with biogenic opal as a
138 measurement of siliceous export production, total organic carbon (TOC), and stable
139 isotopes to identify variations in the organic fluxes to the bottom. Moreover, pollen
140 grains were used to identify environmental conditions based on the climate relationship
141 of the main vegetation formations in north-central Chile. Based on our records we were
142 able to identify wet/dry intervals, periods with high/low organic fluxes to the sediments,
143 which are related to changes in primary production, and changes in the redox conditions
144 at the bottom, which in turn, have been associated with the main climatic conditions
145 described for the Holocene in this region.

146

147 **2. Study area**

148 The Coquimbo area (29–30 °S), in the southern limit of the north-central Chilean
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.

153 The Tongoy and Guanaqueros bays are located in the southern edge of a broad
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
156 that are predominant in the region. Tongoy Bay is a narrow marine basin (10 km at its
157 maximum width) with a maximum depth of approximately 100 m. To the northeast lies
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
161 (Moraga-Opazo et al., 2011; Rahn and Garreaud, 2013) that reach concentrations of
162 approximately 20 mg m⁻³ (Torres and Ampuero, 2009). In the shallow waters of Tongoy
163 Bay, the high primary productivity results in high TOC in the water column, allowing
164 for the deposition of fine material to the bottom; TOC rises concurrently with periods of
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
167 related to lower sea levels, resulting from annual local wind cycles at a regional meso-
168 scale (Gallardo et al., 2017). Oceanographic time series indicate that transition times
169 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/boya->
171 tongoy). The spatial and temporal variability of these processes is still under study. In
172 addition, oceanic variability along the western coast of South America is influenced by
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
175 al., 2002; Ramos et al., 2008).

176 Sedimentological studies are scarce with regard to the north-central shelf of Chile. A
177 few technical reports indicate that sediments between 27° S and 30° S are composed of
178 very fine sand and silt with relatively low organic carbon content (< 3 and ~5%), except
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
182 rainfall in the zone (0.5–80 mm y⁻¹; Montecinos et al., 2016, Fig. 1). Freshwater
183 discharges are represented by creeks, which receive the drainage of the coastal range
184 forming wetland areas in the coast and even small estuaries, such as Pachingo, located
185 south of Tongoy (Fig. 1). These basins cover ~300 and 487 km², respectively. The water
186 volume in the estuaries is maintained by the influx of seawater mixed with the
187 groundwater supply. Normally, a surface flux to the sea is observed. Freshwater
188 discharges only occur through dry creeks that drain water during high rainfall periods in
189 the coastal zone (Dirección General de Aguas, 2011).

190

191 **3. Materials and methods**

192 **3.1. Sampling**

193 Sediment cores were retrieved from two bays in the Coquimbo region: Bahía
194 Guanaqueros (core BGGC5; 30° 09' S, 71° 26' W; 89 m water depth) and Bahía Tongoy
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
201 and biogenic opal concentrations, C and N stable isotope signatures ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$), and
202 TOC content. The samples first were kept frozen (–20° C) and then freeze-dried before
203 laboratory analyses.

204

205 **3.2. Geochronology (^{210}Pb and ^{14}C)**

206 A geochronology was established combining ages estimated from $^{210}\text{Pb}_{\text{xs}}$ activities
207 suitable for the last 200 years and radiocarbon measurements at selected depths for
208 older ages. The quantification of ^{210}Pb activities was performed through the alpha
209 spectrometry of its daughter ^{210}Po following the procedure of Flynn (1968). The
210 (unsupported) activities of $^{210}\text{Pb}_{\text{xs}}$ were determined as the difference between the ^{210}Pb
211 and ^{226}Ra activities measured in some intervals of the sediment column. Meanwhile,
212 ^{226}Ra was measured by gamma spectrometry at the Laboratoire Géosciences of the
213 Université de Montpellier (France). Standard deviations (SD) of the ^{210}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 $^{210}\text{Pb}_{\text{xs}}$ and ^{14}C measurements and from Bacon age–depth
222 modeling open source software (Blaauw and Christen, 2011), considering the Marine
223 curve ^{13}C (Reimer et al., 2013) (Fig. 4) and a reservoir deviation from the global mean
224 reservoir age of 441 ± 35 y. This was estimated subtracting the ^{14}C age value
225 corresponding at the historical dates 1828 AD and 1908 AD (499 ± 24 and 448 ± 23 ^{14}C
226 y, respectively, Reimer et al., 2013) from the apparent ^{14}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 ($\text{SI} \times 10^{-8}$) 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
234 each sample.

235 The grain size was determined using a Mastersizer 2000 laser particle analyzer (Hydro
236 2000–G, Malvern) in the Sedimentology Laboratory at Universidad de Chile. Skewness,

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

239

240 **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
243 OREME/AETE regional facilities). The analysis considered reference materials (UBN,
244 BEN, and MAG1) that had an accuracy higher than $\pm 5\%$; the analytical precisions were
245 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
247 occurring during the course of an analytical session. The analytical precisions attained
248 were between 1% and 3%.

249 TOC and stable isotope ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) analyses were performed at the Institut für
250 Geographie, Friedrich Alexander Universität (FAU) Erlangen-Nürnberg, Germany
251 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 δ notation as the deviation relative to international standards (Vienna Pee Dee
254 Belemnite for $\delta^{13}\text{C}$ and atmospheric N_2 for $\delta^{15}\text{N}$); thus, $\delta^{13}\text{C}$ or $\delta^{15}\text{N} = [(\text{R sample}/\text{R}$
255 $\text{standard}) - 1] \times 10^3$, where R is $^{13}\text{C}/^{12}\text{C}$ or $^{15}\text{N}/^{14}\text{N}$, respectively. The typical precision
256 of the analyses was $\pm 0.1\text{‰}$ for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$.

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
261 biogenic opal were expressed by multiplying the Si (%) by 2.4 (Mortlock and Froelich,
262 1989). The analytical precision was $\pm 0.5\%$. Accumulation rates were determined based
263 on the sediment mass accumulation rates and amount of opal for each core section in %.

264

265 **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⁻¹ 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,
280 with the assistance of the Heusser (1973) pollen catalog. A total of 100–250 terrestrial
281 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
283 spores). Pollen percentage diagrams and zonation were generated using the Tilia
284 software (Grimm, 1987).

285 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)
288 value for this index point corresponds to relatively wetter (drier) conditions.

289

290 **4. Results**

291 **4.1. Geochronology**

292 The activity of ²¹⁰Pb_{xs} (unsupported) was obtained from the surface to a depth of 8 cm
293 in the two cores, with an age of ~AD 1860 at 8 cm in both (Table S1). Greater surface
294 activities were obtained for core BGGC5 (13.48 ± 0.41 dpm g⁻¹) than core BTGC8 (5.80
295 ± 0.19 dpm g⁻¹), showing an exponential decay with depth (Fig. 4). A recent
296 sedimentation rate of 0.11 ± 0.01 cm y⁻¹ was estimated.

297 The age–depth model provided a maximum age of cal BP 7990 for core BGGC5 and cal
298 BP 8012 for core BTGC8 (Fig. 4). A mean sedimentation rate of 0.026 ± 0.012 cm y⁻¹
299 was estimated for core BGGC5, with a period of relatively low values (< 0.01 cm y⁻¹)
300 between cal BP 240 and 1500 and between cal BP ~5000 and 6400. This variation in the
301 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
303 estimates the accumulation rates before and after the hiatus not auto-correlated,

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

320

321 **4.2. Geophysical characterization**

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

338 ($> 1 \text{ g cm}^{-3}$) (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,
341 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
344 at core BTGC8 where it was only observed during the last ~ 2000 years.

345

346 **4.3. Biogenic components**

347 **4.3.1. Siliceous microfossils and biogenic opal**

348 The total diatom abundance fluctuated between 5.52×10^5 and 4.48×10^7 valves g^{-1} at
349 core BGGC5. This abundance showed good correlation with biogenic opal content at
350 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
354 ($< 2 \times 10^5$ valves g^{-1} and $< 3\%$, 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,
358 diatoms were the most important assemblage of siliceous microfossils (96%), followed
359 by sponge spicules (3%). The contributions of phytoliths and chrysophyte cysts was less
360 than 2% at core BGGC5. *Ch RS* was dominant in the diatom assemblage ($\sim 90\%$; Fig. 6)
361 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
363 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.

365

366 **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 $\sim 1.5\%$ at core BTGC8 (Figs. 5a, 5b).
369 Furthermore, $\delta^{13}\text{C}$ 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 $\delta^{15}\text{N}$
371 from the deeper sections to the surface of the core ($< 7\text{‰}$ to $> 10\text{‰}$). This increase

372 was less evident at core BGGC5, with values of ~9‰ at depth to > 10‰ at the surface
373 (Figs. 5a, 5b). The reduced TOC content was related to the slightly higher $\delta^{13}\text{C}$ values
374 (approximately -20‰) in both cores.

375

376 **4.3.3. Pollen record**

377 Initial surveys at core BTGC8 (Tongoy Bay) revealed extremely low pollen
378 abundances, which hampered further palynology work. A comprehensive pollen
379 analysis was conducted only for core BGGC5 (Guañaqueros Bay). The pollen record
380 of core BGGC5 consisted of 29 samples shown in Fig. 7. The record was divided into
381 five general zones following visual observations of changes in the main pollen types
382 and was also assisted by CONISS cluster analysis.

383 Zone BG-1 (cal BP 7990–7600): This zone is dominated by the herbaceous taxa
384 Chenopodiaceae, *Leucheria*-type, Asteraceae subfamily (subf.) Asteroideae, and
385 Apiaceae with overall high values for the wetland genus *Typha* spp.

386 Zone BG-2 (cal BP 7600–6700): This zone is also dominated by Chenopodiaceae,
387 *Leucheria*-type, and Asteraceae subf. Asteroideae. In addition, other non-arboreal
388 elements, such as *Ambrosia*-type, Poaceae, Brassicaceae, and *Chorizanthe* spp.,
389 increase considerably.

390 Zone BG-3 (cal BP 6700–3500): This zone is marked by a steady decline in
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.

397 Zone BG-5 (cal BP 50–Present): The upper portion of the record is dominated by
398 Asteraceae subf. Asteroideae and Poaceae and is marked by higher amounts of
399 Geraniaceae, Asteraceae subf. Mutisieae, Myrtaceae, and *Q. saponaria*. Additionally,
400 this zone includes introduced pollen types such as *Rumex* spp. and *Pinus* spp. The
401 latter is not shown in Fig. 7 because its abundance was minimal.

402 Overall, the most distinctive trend revealed by core BGGC-5 is a long-term decline in
403 Chenopodiaceae and higher amounts of Euphorbiaceae and Asteraceae subf.

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

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

407

408 **4.4. Trace element distributions**

409 Trace element distributions are shown in Figs. 8a and 8b for Guanaqueros (BGGC5)
410 and Tongoy Bays (BTGC8), respectively. We use Al as a normalizing parameter for
411 the enrichment/depletion of elements due to its conservative behavior. The elements
412 are presented as metal/Al ratios. Trace metals are sensitive to the presence of oxygen
413 (U, Re, Mo), showing an increasing metal/Al ratio from the base of core BGGC5 (cal
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
418 BTGC8 increase over time; however, the peak was observed at cal BP ~1000 for U
419 and Re and at cal BP 6000 for Mo, with a second minor peak at cal BP 3400. Iron
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
422 core (BTGC8). Instead, core BTGC8 showed higher values before cal BP 6400. In
423 both cores, Fe increased over the past ~80 years, whereas no clear trend could be
424 established for Mn. In general, metal/Al values were higher at core BGGC5.

425 A second group of elements (metal/Al ratio), including Ca, Ni, Cd, and P (related to
426 primary productivity and organic fluxes), showed a pattern similar to that of Mo/Al of
427 core BGGC5, i.e., increasing values from cal BP ~7990, reaching the highest values
428 near cal BP 6600–7000; afterwards, the values followed a constant reducing trend
429 toward the present. Otherwise, Cu/Al (a nutrient-type element) showed a different
430 pattern, similar to the Fe/Al distribution, with a maximum value at cal BP 3500–3800
431 and a conspicuous upward trend over the past ~80 years. A third group, consisting of
432 Ba and Sr, exhibited a similar pattern but smoother, showing the maximum values
433 before cal BP 6600. At BTGC8 core, a less clear pattern was demonstrated. Ca, Ni,
434 Cd, and P ratios at core BTGC8 showed only slightly decreasing values and very low
435 peak values compared with core BGGC5; however, Ni/Al showed increasing
436 concentrations over the past 80 years, which was not observed at core BGGC5.
437 Metal/Al ratios of Ba and Sr showed no substantial variation in time. In general, all the

438 elemental concentrations were lower than at core BGGC5 and presented similar long-
439 term reduction patterns toward the present, except for Cu, Ni, and Fe.
440 The authigenic enrichment factor (EF) of elements was estimated as: $EF = (Me/Al)_{\text{sample}}$
441 $/ (Me/Al)_{\text{detrital}}$, where $(Me/Al)_{\text{sample}}$ is the bulk sample metal (Me) concentration
442 normalized to the Al content, and the denomination “detrital” indicates a lithogenic
443 background (Böning et al., 2009). Detrital ($[Me]_{\text{detrital}}$ and $[Al]_{\text{detrital}}$) concentrations were
444 established considering the local metal abundance, which is more accurate than using
445 mean Earth crust values (Van der Weijden, 2002). We used average element
446 concentrations on surface sediments (0–3 cm) of the Pachingo wetland (Table 3). The
447 values suggest a large enrichment of nutrient-type elements in a period prior to
448 cal BP 6600, following the trend of the Me/Al ratios, except for Ba and Fe, which did
449 not show authigenic enrichment. The EFs exhibited a sharp decrease in enrichment in
450 recent times after cal BP 80 (Fig. 9).

451

452 **5. Discussion**

453 **5.1. Sedimentary composition of the cores: terrestrial *versus* biogenic inputs**

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

466 The isotopic variations in $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ did not clearly establish differences between
467 the sediments of the two bays; however, minor differences in $\delta^{15}\text{N}$ would indicate a
468 greater influence of the upwelling nutrient supply and OMZ on the shelf, resulting in a
469 $\delta^{15}\text{N}$ of 9–10‰ in the Guanaqueros Bay, values which are slightly higher than that in
470 the Tongoy Bay sediments (Figs. 5a, 5b). This isotopic composition corresponds with

471 that of NO_3^- in the upwelling waters (De Pol-Holz et al., 2007) in the range of those
472 measured at north-central Chile (~11‰; Hebbeln et al., 2000, De Pol-Holz et al.,
473 2007, 2009). This is due to the isotopic fractionation of NO_3^- during nitrate reduction
474 within the OMZ, which leaves remnant NO_3^- enriched in ^{15}N (Sigman et al., 2009;
475 Ganeshram et al., 2000 and references therein). This is particularly relevant because it
476 demonstrates the relevance of OMZ over the shelf sediments off Coquimbo at shallow
477 depths and the influence of the poleward undercurrent from the Perú OMZ (Mollier-
478 Vogle et al., 2012). At sediment core BTGC8, lower values (< 8‰) measured at
479 greater depths within the core should account for a mix with isotopically lighter
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
482 sediments of which have $\delta^{15}\text{N}$ of 2–6‰ (Muñoz et al., data will be published
483 elsewhere). This suggests that Tongoy sediments contain a combination with
484 continental material (Fig. 5b).

485 Thus, cores BGGC5 and BTGC8 in the Guanaqueros and Tongoy Bays record the
486 variability in oceanographic conditions; however, in the Tongoy core, the
487 concentration of oceanographic proxies is diluted owing to the input of terrigenous
488 material. This helps to decipher the climatic variability, considering that the main
489 input of clastic material to the area takes place during major flooding events.
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,
492 making the sedimentary records of these sites complementary.

493

494 **5.2. Temporal variability of primary productivity and the oxygenation of bottoms**

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

505 sediments in less anoxic environments with moderate productivity (Torres et al., 1996;
506 Dymon et al., 1992) as is the case with our study site (Gross Primary Productivity
507 = 0.35 to 2.9 g C m⁻¹d⁻¹; Daneri et al., 2000). This leads to a negative correlation with
508 TOC (-0.59; Table 4), owing to the remobilization of Ba under anoxic conditions
509 before cal BP 6600. Meanwhile, the P distribution showed a trend similar to that of
510 TOC and the other elements related to the organic fluxes to the bottom (Ni, Cd),
511 although with a lower correlation (~0.6). This is consistent with the distributions
512 observed for U, Re, and Mo at core BGGC5, which indicate that anoxic or suboxic
513 conditions were developed from cal BP 7990 to 1800 but were stronger before cal BP
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
519 related to oxygen changes (Calvert and Pedersen, 2007, and references therein).

520 Otherwise, the accumulation of P depends on the deposition rate of organic P (dead
521 plankton, bones, and fish scales) on the bottom and is actively remineralized during
522 aerobic or anaerobic bacterial activity. P and TOC showed a declining trend toward
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
525 proxies could be explained by the higher remineralization of organic material settled at
526 the bottom due to higher oxygen availability, as shown by U, Mo, and Re distributions
527 (Figs. 8a, 8b). However, the lower $\delta^{15}\text{N}$, depending on the denitrification process, is
528 similar to that at deeper environments in the zone (De-Pol Holz et al., 2009),
529 suggesting the influence of the reductive environment of OMZ over the shelf. The
530 influence of the primary productivity on oxygen consumption at the bottom over time
531 would be secondary in this system, which is considered to be moderated in
532 productivity compared with upwelling centers in north and south Chile.

533 Productivity reconstructions were based on the qualitative relative abundances of
534 diatom and sponge spicules, quantitative diatom counts (valves g⁻¹), and biogenic opal
535 content only in core BGGC5, since core BTGC8 registered low valve counts (< 1% in
536 relative diatom abundance). However, in both cores, diatom assemblages were
537 represented mainly by *Ch. RS*, which are used as upwelling indicators (Abrantes 1988,
538 Vargas et al., 2004). The downcore siliceous productivity based on opal distribution

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

544 At first period (> cal BP 6600), the opal accumulation rate was remarkably high,
545 amounting to $\sim 35 \pm 18 \text{ g m}^{-2} \text{ y}^{-1}$ (range: 16–119 $\text{g m}^{-2} \text{ y}^{-1}$, Fig. 9) when *Chaetoceros*
546 spores were predominant, indicating an intensification in upwelling. During this
547 period, all metal proxies suggest that primary productivity increases before cal BP
548 6600, owing to the high concentrations and major enrichment of Ni, Ca, and P that
549 occurred in this period, concomitant with higher opal accumulation within the
550 sediments (Fig. 6 and 9). From these elements, Ni is the best indicator of organic
551 sinking flux related with diatom productivity in organic-rich upwelling sediments
552 (Böning et al., 2015), which helps to sustain our statement. In addition, the authigenic
553 enrichments of Cd were very high (> 100, Fig. 9) resulting in high Cd/U ratios (> 2;
554 Fig. 9), indicative for anoxic conditions as this ratio could vary between 0.2 and 2,
555 from suboxic to anoxic environments (Nameroff et al., 2002). The Cd accumulation in
556 this period was higher than that reported for a highly productive zone off Concepción
557 in periods of high organic carbon accumulation in the sediments (~ 5 , Muñoz et al.,
558 2012). Additionally, the high enrichment of Mo (~ 20) indicates the prevalence of
559 anoxic conditions at the bottom in this period due to the control by sulfide
560 concentrations (Huerta-Diaz and Morse, 1992; Chaillou et al., 2002; Nameroff et al.,
561 2002; Sundby et al., 2004, Tribovillard et al., 2004). Our low U/Mo ratio (~ 0.3 , Fig. 9)
562 corroborates this assumption, as similar to those values reported today at shallower
563 anoxic zones off Perú interrupted by seasonal oxygenation (McManus et al., 2006;
564 Sholz et al., 2011, Salvattecchi et al., 2016; Vergara et al., 2016). This is similar to our
565 shelf, notwithstanding the prevalence of very reduced conditions within the sediments.
566 The enhanced reduced conditions in this period, probably sulfidic, favor the
567 accumulation of Mo and Cd over that of U, occurring in anoxic environments where
568 the chemocline is close to the water–sediment interface or above it, allowing the
569 formation of authigenic Mo that exceed the U uptake within the sediments (Algeo and
570 Tribovillard, 2009 and references therein). Re is enriched in less reduced conditions
571 than Mo, resulting in the lowest Re/Mo in this period (Fig. 9). This is congruent with
572 the environmental conditions at the bottom in zones of high productivity and intense

573 upwelling, where sulfidic conditions are developed owing to oxygen consumption in
574 the shallower zones and linked to the OMZ, as occur at northern Chilean regions,
575 where the main productivity is developed over the narrow shelf. Thus, the high
576 productivity before cal BP 6600 could result from a more intense upwelling that
577 generated permanent reduced conditions that became very anoxic at the bottom in this
578 period. Even so, the low oxygen conditions prevailed in the subsequent periods but
579 were less intense than before.

580 After cal BP 6400 until 4500 we obtained little information owing to a gap in the
581 sedimentary record, which made it difficult to visualize changes in the oxygenation
582 and productivity proxies in this interval. However, in the next period (cal BP 4500–
583 1800), we observe that the opal accumulation was lower than in the previous recorded
584 period, $12 \pm 4 \text{ g m}^{-2} \text{ y}^{-1}$ (range: $6\text{--}20 \text{ g m}^{-2} \text{ y}^{-1}$, peaking at cal BP 3400–4000; Fig. 9),
585 which is partially consistent with nutrient-type element distributions and element
586 enrichment (Fig. 8a, Fig. 9). Fe clearly shows higher values at approximately cal BP
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
590 lower organic matter deposition into the sediments during this period (Fig. 8a). Thus, a
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)
593 where lower accumulations of nutrient-type elements were also observed at cal BP
594 3600–4000 and cal BP 2600 than at cal BP 6200 (Muñoz et al., 2012). However, low
595 oxygen conditions within the sediments were maintained, which could be more related
596 to the manifestation of the OMZ close to the coast, favoring Mo and Re accumulation
597 until cal BP 1700–1800 (Fig. 8a). Lower Cd/U ratios (~ 1 ; Fig. 9) were estimated,
598 suggesting higher variations in the primary productivity but with moderate changes in
599 the oxygen conditions at the bottom. High Re/Mo and U/Mo ratios could indicate a
600 shift toward less reduced conditions but still anoxic, since U, Re, and Mo are highly
601 enriched (6, 20, and 15, respectively; Fig. 9). U and Re accumulations occur in
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
604 to lower productivity, producing low oxygen consumption and a less sulfidic
605 environment along the central-Chilean margin ($30\text{--}36^\circ \text{ S}$), which is in agreement with
606 the lower biogenic opal flux and diatom abundance after cal BP 6600 (Figs. 6, 9).

607 Slight increasing values of Re/Mo ratios until ~cal BP 3500 suggest a decreasing trend
608 in the reduced conditions, which became stronger after cal BP 1800. This time was
609 also highlighted in the sedimentary records off Concepcion shelf (36° S, Muñoz et al.,
610 2012) showing maximum enrichment of U and Cr near cal BP 1800, both indicating
611 less reduced conditions toward the present compared with previous periods. After this
612 age, no comparison could be made owing to a discontinuity in the sedimentary records
613 off Concepción. Notwithstanding, the suboxic conditions have prevailed until today at
614 Central Chile, where the oxygenation seems has been stronger off Coquimbo. It could
615 be caused by eddies related to the instabilities of the Peru Undercurrent (Vergara et al.,
616 2016), which seem to start operating more frequently from cal BP 1800 to the present.
617 After this age to cal BP 100, higher productivities were not found, and a second
618 discontinuity (cal BP 1500–240) impeded environmental reconstructions, with the
619 very low estimated sedimentation rate hindering the realization of sufficient time
620 resolution for the proxies in this interval. After cal BP ~100 to recent times (CE 2015)
621 (third period mentioned before), the productivity increased substantially, deduced
622 from the rise in opal accumulations toward the present (mean opal value of $21 \pm 18 \text{ g}$
623 $\text{m}^2 \text{y}^{-1}$, range: $8\text{--}34 \text{ g m}^2 \text{y}^{-1}$; Fig. 9); however, this corresponded with lower diatom
624 abundances, which were observed from cal BP 1800 to the present (range: $0.5\text{--}4.9$
625 $\times 10^6 \text{ valves g}^{-1}$; Fig. 6). This is likely caused by the fact that only a few sections of the
626 core in this interval were analyzed for diatoms, leading to a low resolution for this
627 measurement in the most recent period. Another possibility is that the opal flux was
628 overestimated owing to the fact that the flux calculations were based on recent
629 sedimentation rates, an estimation that tends to be higher than at longer timescales
630 (Sadler et al., 1999). However, the slight increase in the Cd/U ratio and P and Ni
631 enrichment could suggest an increase in the primary productivity and organic fluxes to
632 the bottom in more recent times. In addition, the main trend established before and
633 after the hiatus indicates an increase in the marine productivity, which would not be as
634 high as in the first period (before cal BP 6600). After cal BP 1800, there is an evident
635 change to a less reduced environment toward the present, suggesting a more
636 oxygenated bottom environment concurrent with a reduction in primary productivity,
637 except for the last 100–130 years. Contrary to other metals, there is a conspicuous
638 upward trend for Cu/Al, Fe/Al, and Mn/Al in recent times, which is consistent with the
639 decreasing trend of EFs of Re, U, and Mo (Fig. 8a, 8b, Fig. 9); these estimations
640 would not be influenced by the sedimentation rates but rather the presence of oxygen.

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

652 We suggest that higher productivity in the last 100 years has occurred in a more
653 oxygenated environment, which is actually contradictory. We assume that episodic
654 oxygenation changes the original extent of the accumulation of these sensitive redox
655 trace element accumulations because of their remobilization to soluble forms (Morford
656 and Emerson, 1999; Morford et al., 2009). The main processes involved in the OMZ
657 ventilation at longer timescales are related to El Niño (Vergara et al., 2016 and
658 references therein); thus the increased frequency and intensity of El Niño would result
659 in a mean effect, which is observed as a gradual change in metal enrichment over time.
660 Several observations made at the central Peruvian and south-central Chilean coasts
661 (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
663 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
665 effects of which can be observed several months later. Other oxygenation mechanisms
666 can result from coastal-trapped Kelvin waves originating from the equator and
667 propagating along the coast, which modify the stability of the regional current system
668 and the pycnocline and trigger extra-tropical Rossby waves (Pizarro et al., 2002;
669 Ramos et al., 2006; 2008). This oceanographic feature may have operated more
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.

672

673 **5.3. Main climatic implications**

674 According to paleoenvironmental records, the past climate and oceanographic
675 variability have been interpreted mainly based on the past variability in the intensity of
676 the SWW and latitudinal position of the ITCZ (Veit et al., 1996; Hebbeln et al., 2002;
677 Lamy et al., 1999; Maldonado and Villagrán, 2002). The ITCZ movements from the
678 northernmost or southernmost latitudinal position depend on the different phases of
679 ENSO and PDO variability (Yang and Oh, 2020), as the main regulators of the climate
680 at the centennial and decadal scales. This has an impact over relevant oceanographic
681 characteristics, such as sea surface temperature (SST), upwelling, and accordingly,
682 productivity at the SE Pacific. We established marked differences in paleo-productivity
683 proxies and paleo-oxygenation in the last ~8000 years (Figs. 6, 8), indicating that high
684 marine productivity prevailed during our first period (cal BP 8000–6600), according to
685 what was established for central Chile between 10 and 5 ky owing to sustained mean La
686 Niña-like conditions associated with the cold phase of the PDO (positive phase) (De
687 Pol-Holz et al., 2006; Kaiser et al., 2008; Lamy et al., 2010), concomitantly with
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
691 productivity records associated with low oxygen conditions at the bottom, both reaching
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
694 margin, this period has also been described as being drier with the dominance of La
695 Niña-like conditions according to the northerly position of both westerlies and the ITCZ
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
699 of Pacific from central Chile to the Galapagos (Carré et al., 2012). For central Chile, the
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
702 Villagrán, 2002, 2006), characterized by reduced rainfall but intense coastal humidity,
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
705 et al., 2012) and La Niña-like conditions, which explains the main variability of the
706 SPSA (Ancapichún and Garcés-Vargas, 2015). Others have suggested a reduced ENSO
707 variance during the early and mid-Holocene (Rein et al., 2005), which indicates a less

708 frequent or less intense warm anomaly related to a Central Pacific (CP)-mode ENSO,
709 producing moderate El Niño events at the CP and strong La Niña off Peru (Carré et al.,
710 2014, Mollier-Voguel et al., 2019). This was favorable for upwelling and primary
711 productivity development along the Chilean and Peruvian margin. In addition,
712 Braconnot et al. (2012) indicated that this lower ENSO was linked to fresh water
713 melting that counteracted the insolation regime, pointing a more limited cold–dry period
714 between 6700–7500 years ago, which matches our records of maximum productivity
715 (Figs. 6, 9) concomitantly with the lowest bottom oxygen conditions and indicates a
716 greater influence of the OMZ over the shelf at the central-Chilean margin.

717 After the maximum productivity recorded, a decreasing trend occurred under warm and
718 humid climatic conditions, which would be because of an enhancement in regional
719 precipitation in the northern margin of SWW (Jenny et al., 2003; Maldonado and
720 Villagrán, 2006), consistent with the southern movement of the ITCZ, leading to wetter
721 climatic conditions in the southern tropics regions (Koutavas and Lynch-Stieglitz,
722 2004). A gradual rise in K/Ca, Fe, Al, and Pb distributions was observed in our cores
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
725 and Lam, 2015; Saito et al., 1992; Xu et al., 2015). This indicated that the precipitation
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
728 Joanides, 2012; Carré et al., 2014). It is also consistent with the pollen records of central
729 Chile, which suggest an arid phase from cal BP 6200 until cal BP 4200 (Maldonado and
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
732 dry/humid periods were identified after cal BP 6600. Mostly, a gradual increase in
733 humidity and a weakening in paleo-productivity proxies after cal BP 4500 (Figs. 8, 9)
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
736 Villagrán, 2002, 2006). In general, this is a period of increased ENSO variability (from
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
739 insolation described for the region at the mid- and late Holecene (Haug et al., 2001;
740 Toth et al., 2012; Carré et al., 2014). This is consistent with the occurrence of alluvial
741 episodes in the area caused by more frequent or heavier rainfall events over time,

742 related to intensified westerlies and increased El Niño events observed from Peru to
743 central Chile (Jenny et al., 2002; 2003; Rein et al., 2005; Sandweiss et al., 2007; Ortega
744 et al., 2012; Ortega et al., 2019). A consequence is greater continental inputs, as
745 suggested by our sedimentary records in agreement with the pollen moisture index that
746 indicated more humid conditions through the mid-Holocene to the present. This was
747 concomitant with greater oxygenation at the bottom and reduced primary productivity.
748 Nonetheless, between cal BP 4500 and 3000, a slight increase in diatom abundance and
749 opal concentrations was observed, along with a slight accumulation in nutrient elements
750 (Ni, Cd, Fe, and Ca concentrations; Fig. 8). Small increases in the organic carbon flux
751 and Cd/U ratios (Fig. 5, 9) suggest that the increase in primary productivity could be
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
755 proxies.

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 central Chilean margin, which varied little until cal BP 1800 (Figs. 8, 9; see U, Mo, and
759 Re). Actually, the periodicity of El Niño was similar between cal BP 5000 and cal BP
760 3000 and lower than modern times (Sandweiss et al., 2007), supporting the observation
761 of relatively low variability of the oxygen proxies in the sediments dependent on the
762 OMZ influence over the shelf. This implies that the upper limit location of the OMZ did
763 not drastically change during most of the mid- and late Holocene. Contrary to our
764 observations, the sediments at the Peruvian shelf were less reduced in the late-mid
765 Holocene than at the present, which was due to a deepening in the OMZ by the
766 increased advection of waters enriched in oxygen from the Equatorial Undercurrent and
767 the shifting of the OMZ center toward the Chilean margin (Mollier et al., 2019).
768 Therefore, the enhanced oxygenation of Peru and OMZ deepening translated into a
769 decrease in the oxygen conditions off north-central Chile. This period is followed by an
770 increased El Niño frequency that has been consistent with the intensification and
771 frequency of flooding events recorded in Peru and central Chile in the last ~2000 years
772 (Rein et al., 2005; Sandweiss et al., 2007; Jenny et al., 2002; Toth et al., 2012), which is
773 concomitant with the drastic oxygenation at the bottom observed in our records after cal
774 BP 1800. In this regard, the oxygen variation at the bottom would be related to a less
775 intense effect of the OMZs over the shelf at the central Chilean margin during the warm

776 El Niño-like phases, owing to a deepening of the oxycline (and vice versa during La
777 Niña). These tend to be associated with low productivity and, in turn, a reduction in the
778 organic fluxes and oxygen consumption during organic matter diagenesis.

779 After cal BP 1800, few records were obtained until cal BP 130, when we observed the
780 restoration of more reduced conditions, although lower than during previous periods.
781 This corresponds to the time of Peruvian upwelling shift due to the northward
782 displacement of the ITCZ to the modern position and the enhancement of the Walker
783 circulation (Gutiérrez et al., 2009), which establishes an intensification of the upwelling
784 in the eastern Pacific; consequently, an increase in the primary productivity, producing
785 high demand for oxygen during organic matter remineralization, as observed today,
786 which leads to stronger oxygen consumption in the northern part of the eastern margin.

787

788 **6. Conclusions**

789 Our results suggest that the geochemistry and sedimentary properties of the coastal
790 shelf environments in north-central Chile have changed considerably during the
791 Holocene period, suggesting two relevant changes in the redox conditions and
792 productivity, which point to a more reducing environment and higher productivity
793 around cal BP 6600. Afterwards, a less reducing environment along with decreasing
794 trends in primary productivity and increased humid conditions occurs with time. The
795 oxygenation of the surface limit of the OMZ has been proposed as the main
796 mechanism that controls the reduced conditions over the shelf and slope sediments
797 during the mid-Holocene, which mainly affected the Peruvian margin closed to the
798 OMZ edge. This led to contrasting conditions in the central-Chilean margin where the
799 most reduced conditions were observed, which was maintained with low variability
800 until cal BP 1800. After this age, the OMZ expression over the shelf was weak,
801 returning to more reduced conditions in recent times (two last centuries), similar to the
802 Peruvian margin but weaker at north-central Chile.

803 The northward shifts of the SWW belt, in addition to an increased frequency in El Niño
804 events, have been proposed as the main drivers for climatic conditions during this
805 period. These elements have introduced high variability in the primary productivity
806 during this time interval. This also impacted the accumulation of organic matter due to
807 an intensification of its remineralization, showing a decreasing trend in the buildup of
808 nutrient-type elements and organic carbon burial rates toward the present. Otherwise,
809 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.

820

821 **7. References**

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1302

1303 **Author contributions:** PM prepared the manuscript with contributions from all co-
1304 authors. PM, LR and LD developed the proposal and conducted field work. AM, KA
1305 and MR assisted in field work in different campaigns. All authors participated in
1306 different laboratory work and data analysis, PM, LD and KA conducted metal and
1307 radioisotope analyses. MR analyzed physical data and graphs editing. MS helped with
1308 alpha counting on prepared samples. CM ran stable isotope and TOC analysis. LR, CL,

1309 PC, GS and KL assisted in specimen identifications of foraminifers and diatoms. AM
 1310 and IJ identified pollen and assisted with the age modeling. GV analyzed physical
 1311 properties of the sediments and contributed to writing and editing the manuscript.

1312

1313 **Competing interests.** The authors declare that they have no conflict of interest.

1314

1315 **Tables**

1316

1317 Table 1. Radiocarbon dates for BGGC5 and BTGC8 sediment cores collected from
 1318 mixed planktonic foraminifera and monospecific benthic foraminifera (*Bolivina*
 1319 *plicata*), respectively. The ¹⁴C-AMS was performed at NOSAM-WHOI. The lab code
 1320 and conventional ages collected from each core section are indicated. For error
 1321 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
Planktonic foraminifera							
BGGC5							
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 foraminifera							
BTGC8							
5-6	<i>Bolivina plicata</i>	4.2	OS-130657	0.8953	0.0017	890	15
20-21	<i>Bolivina plicata</i>	7.7	OS-123670	0.7337	0.0021	2,490	25
30-31	<i>Bolivina plicata</i>	13.0	OS-123671	0.6771	0.0016	3,130	20
40-41	<i>Bolivina plicata</i>	11.0	OS-123672	0.6507	0.0019	3,450	25
50-51	<i>Bolivina plicata</i>	8.7	OS-123673	0.5877	0.0014	4,270	20
60-61	<i>Bolivina plicata</i>	13.0	OS-123674	0.5560	0.0018	4,720	25
71-72	<i>Bolivina plicata</i>	10.0	OS-123675	0.4930	0.0013	5,680	20
80-81	<i>Bolivina plicata</i>	7.3	OS-123676	0.4542	0.0012	6,340	20
90-91	<i>Bolivina plicata</i>	6.8	OS-123677	0.4259	0.0015	6,860	30
96-97	<i>Bolivina plicata</i>	6.8	OS-123678	0.3903	0.0013	7,560	25

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1326 Table 2. Reservoir age estimation considering the ^{210}Pb age determined with the CRS
 1327 model (McCaffrey and Thomson, 1980) at selected depth sections of the core, as
 1328 compared with ^{14}C ages (y BP) from the marine 13.14 curve (Reimer et al., 2013),
 1329 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

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1331

1332 Table 3. Concentration of elements in the Pachingo wetland sediments, considered as
 1333 lithogenic background for the study area. The values correspond to mean concentrations
 1334 in the surface sediments (0–3 cm).

Element	Metal/Al x 10 ³	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

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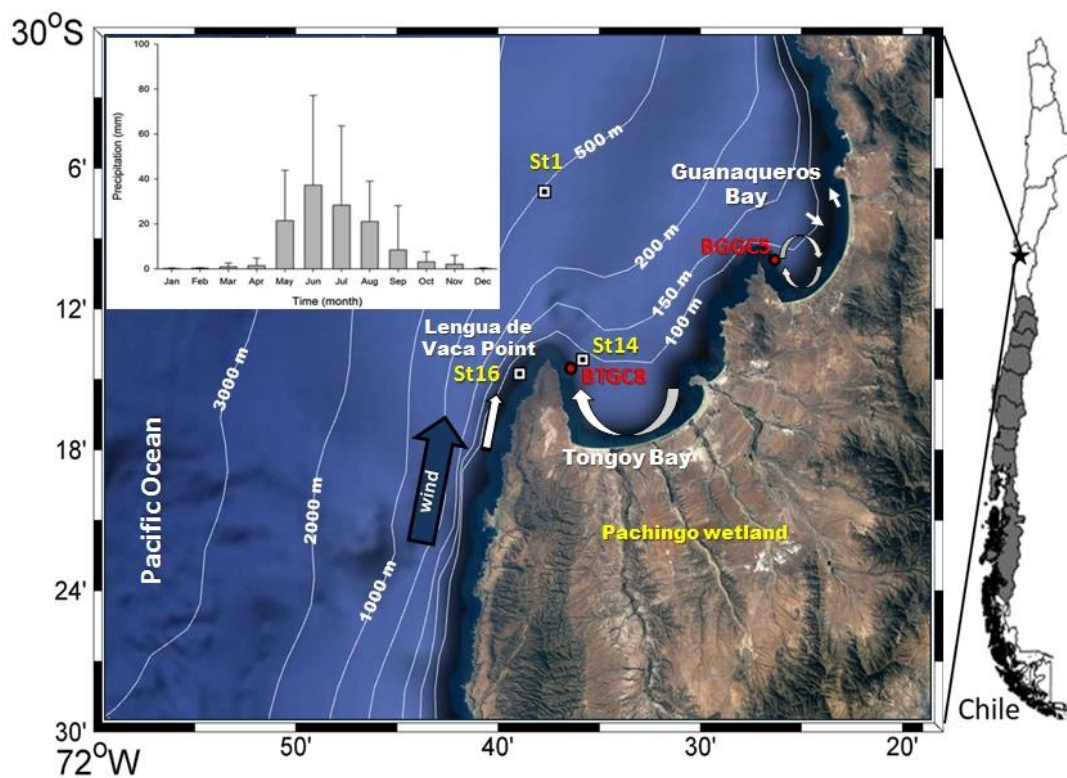
1343 Table 4. Spearman rank order correlations for geochemical data. Significant values

1344 > 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

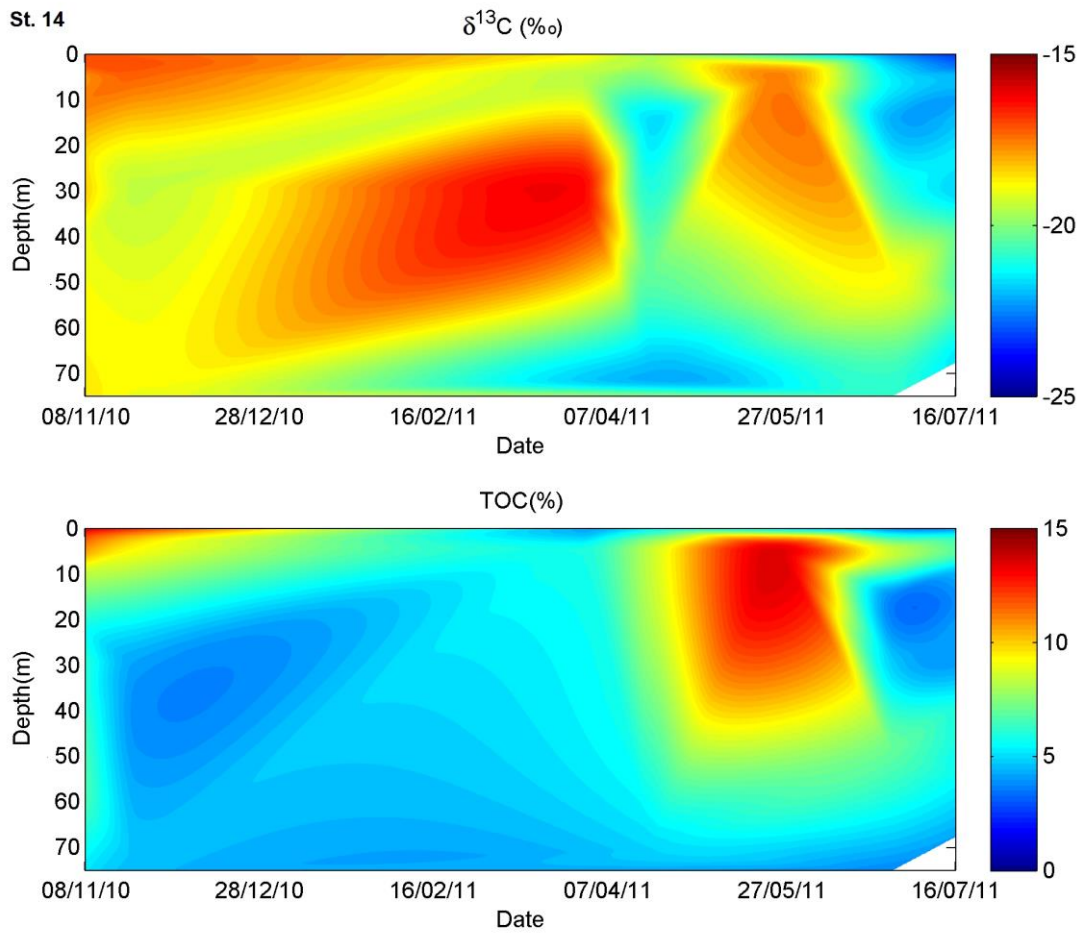
Figures

1345 Figure 1. Study area showing the positions of sampling stations. Sediment cores were
1346 retrieved from Guanaqueros Bay (BGGC5) and Tongoy Bay (BTGC8) at water depths
1347 of 89 and 85 m, respectively. Information of dissolved oxygen in the water column at
1348 St1 and St16 and that of suspended organic particles collected at St14 sampling sites
1349 was gathered in a previous project (INNOVA 07CN13 IXM-150). Monthly
1350 precipitation in mm (bars) (mean \pm SD; Montecinos et al., 2016). Schematic
1351 representation of the circulation in the bays (white arrows) and wind direction (blue
1352 arrow) is indicated, as obtained from Valle-Levinson and Moraga-Opazo (2006) and
1353 Moraga-Opazo et al. (2011).



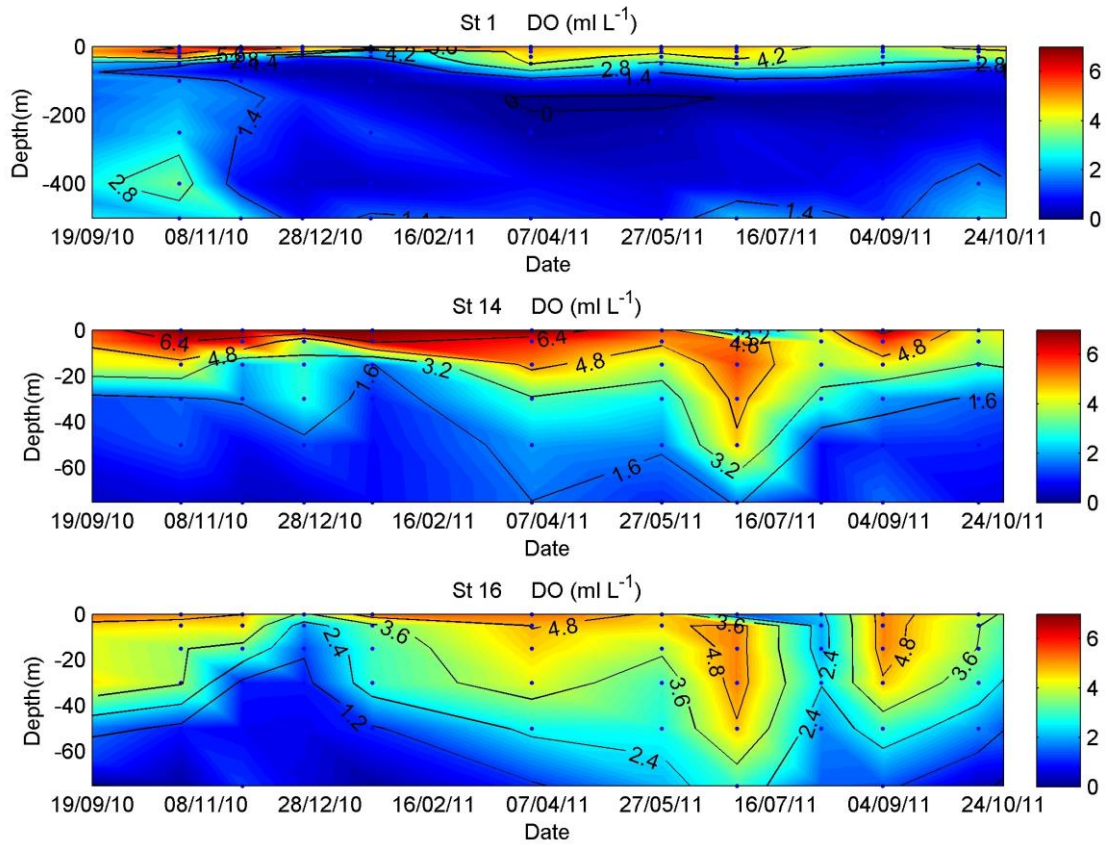
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1359 Figure 2. Suspended particulate matter composition (TOC% and $\delta^{13}\text{C}_{\text{org}}$) measured in
1360 the water column between October 2010 and October 2011, at station St14, Tongoy
1361 Bay, Coquimbo (30° S).



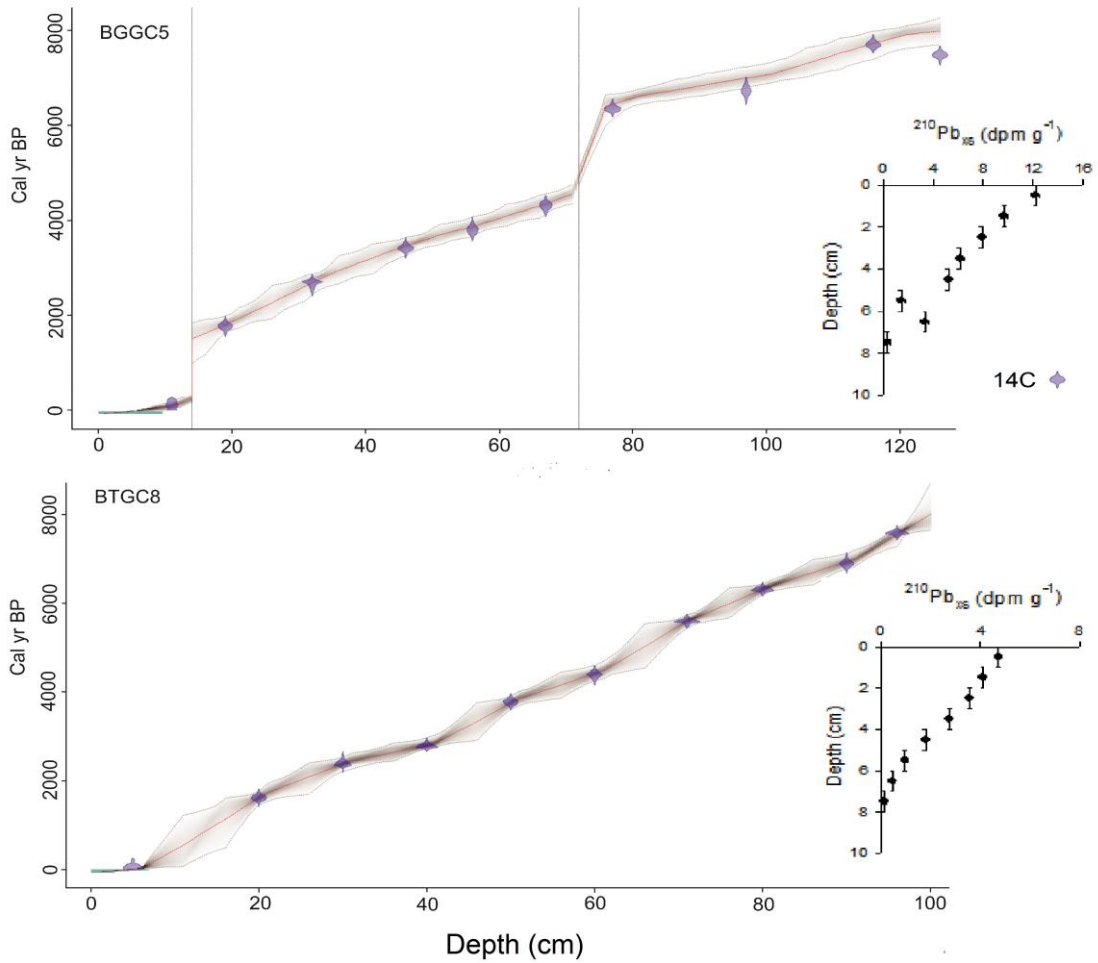
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1373 Figure 3. Dissolved oxygen time series in the water column measured between October
1374 2010 and January 2011, at stations St1, St14, and St16 off Tongoy Bay, Coquimbo
1375 (30° S).



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1390 Figure 4. Age model based on ^{14}C -AMS and ^{210}Pb measurements. The timescale was
1391 obtained according to the Bacon age–depth modeling open source software (Blaauw and
1392 Christen, 2011) considering the Marine curve ^{13}C (Reimer et al., 2013).

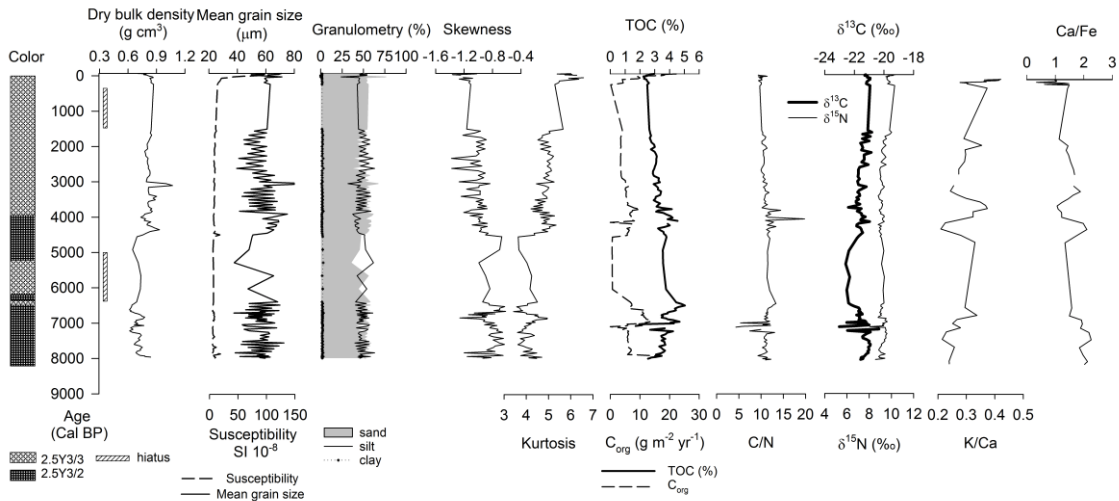


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1405 Figure 5. Characterization of sediment cores retrieved from (a) Guanaqueros Bay
 1406 (BGGC5) and (b) Tongoy Bay (BTGC8), where the color (Munsell chart scale)
 1407 represents the depth, dry bulk density, mean grain size, granulometry (% sand, silt, and
 1408 clay), statistical parameters (skewness, kurtosis), organic components (TOC, C/N ratio,
 1409 stable isotopes $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) and chemical composition (K/Ca, Ca/Fe).

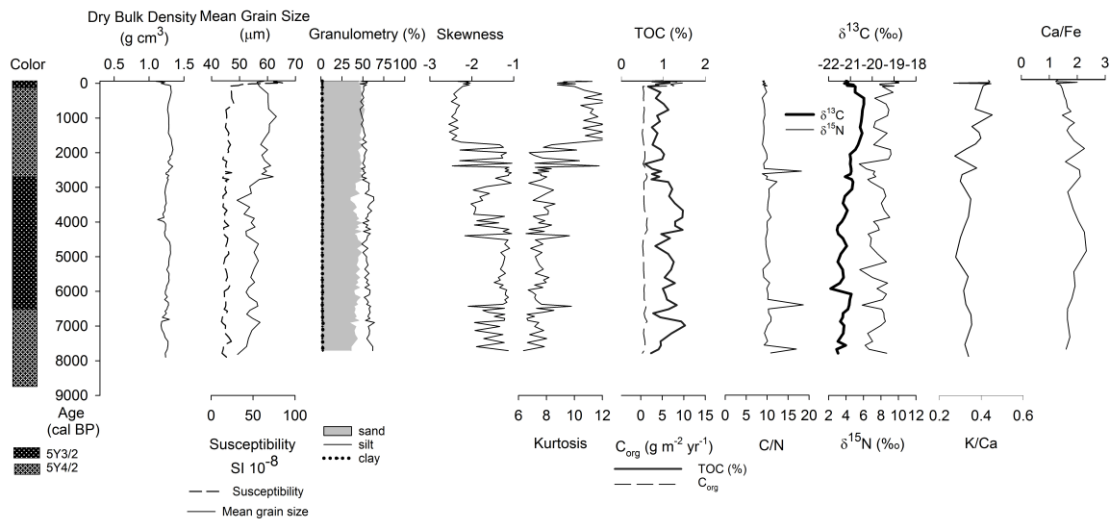
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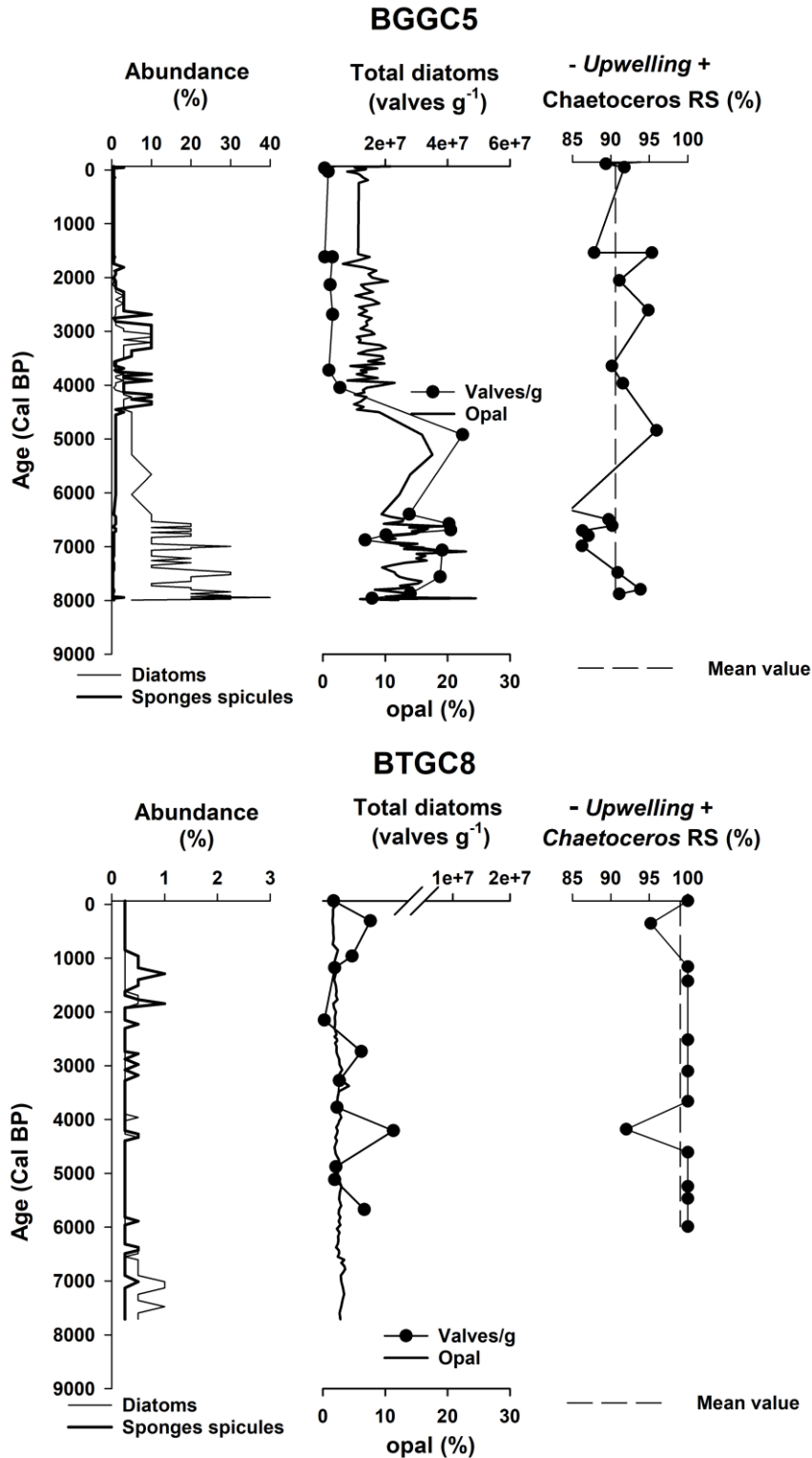
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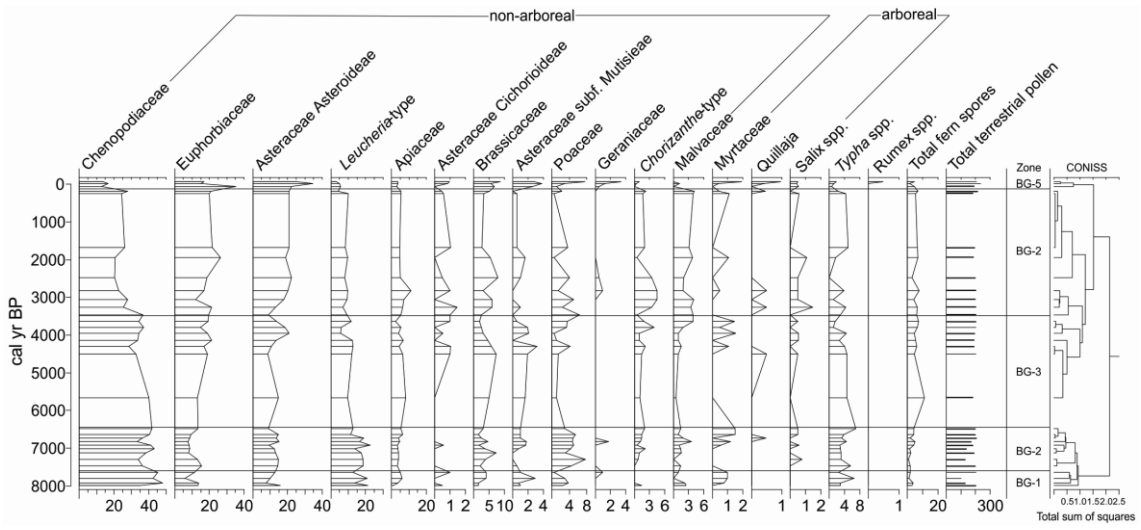
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1420 Figure 6. Diatom and sponge spicule relative abundances, total diatom counts (valves g^{-1}) and opal (%), opal accumulation ($g\ m^{-2}\ y^{-1}$), and downcore variations in *Ch. RS*
 1421 percentages as proxies of upwelling intensity in the BGGC5 and BTGC8 cores
 1422 (Guaanqueros and Tongoy Bay, respectively). The medium dashed line represents the
 1423 average of *Ch. resting* spores for the respective core.
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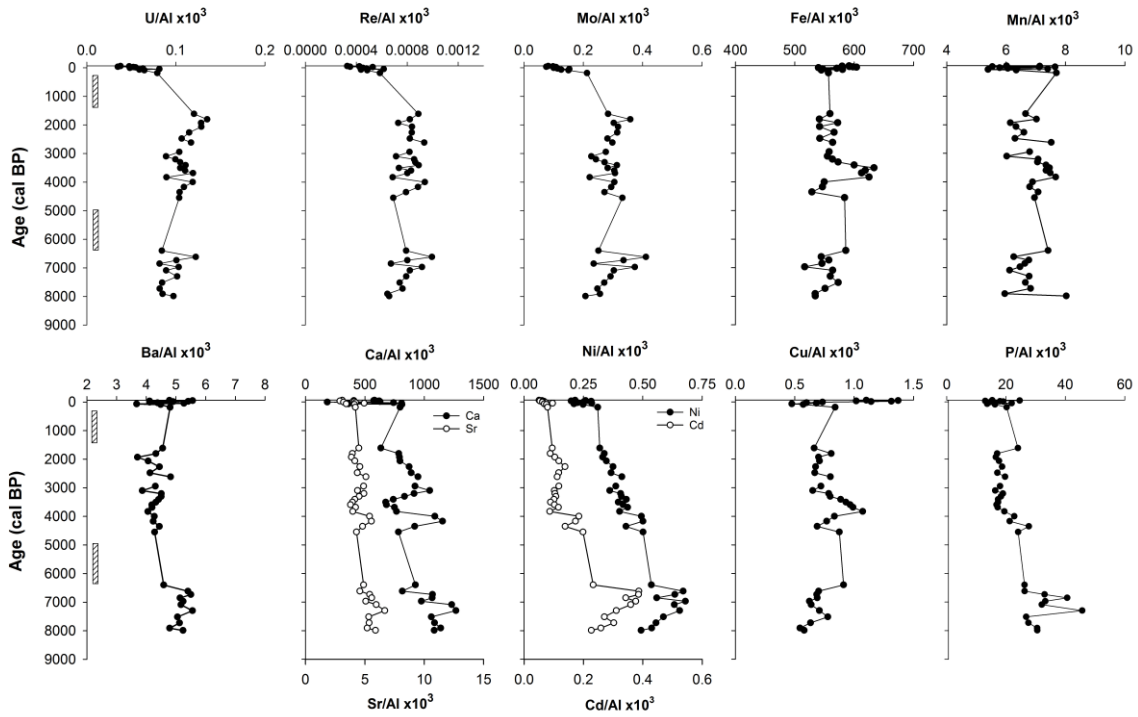
1426 Figure 7. Pollen record in BGGC5 core.



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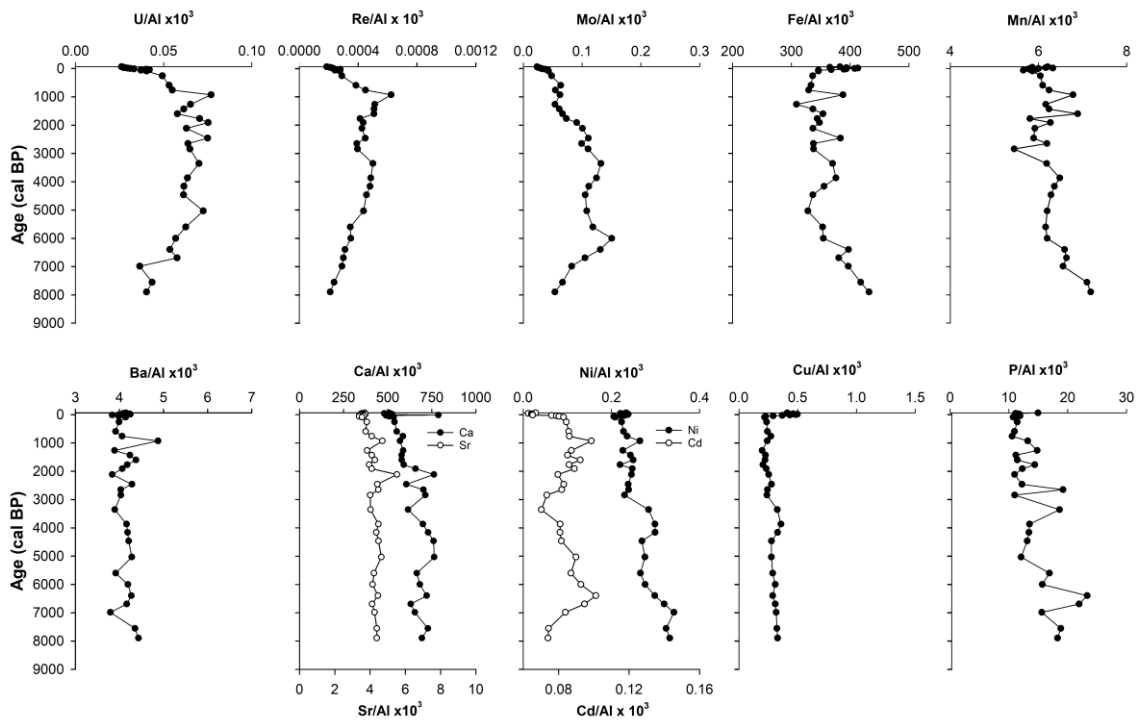
1450 Figure 8. Downcore trace element variations in: (a) Guanaqueros Bay (BGGC5) and (b)
 1451 Tongoy Bay (BTGC8), off Coquimbo (30 °S).

1452 a)



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1454 b)



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1457 Figure 9. Opal accumulation and authigenic enrichment factor (EF) of trace elements
 1458 calculated for Guanaqueros Bay (BGGC5 core). Lithogenic background was estimated
 1459 from the surface sediments of Pachingo wetland cores (see text). Pollen moisture index
 1460 defined as the normalized ratio between Euphorbiaceae (wet coastal shrub land) and
 1461 Chenopodiaceae (arid scrubland). Positive (negative) values for this index indicate the
 1462 relative expansion (reduction) of coastal vegetation under wetter (drier) conditions. Pb
 1463 and Al distribution at BGGC5 core, representatives of terrigenous input to the bay.
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