

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 Coquimbo (30°S) Region –located in the north-Central Chilean Coast– is  
38 characterized by relative dry summers and a short rainfall period during winter months.  
39 The wet-winter climate results from the interactions between the Southern Westerly  
40 Winds and the South Pacific Subtropical Anticyclone (SPSA). Inter-annual climate  
41 trends are mostly associated with El Niño-Southern Oscillation (ENSO), which  
42 produces high variability in precipitation. With the aim of establishing past variations of  
43 the main oceanographic and climatic features in the north-central Chilean coast, we  
44 analyzed recent sedimentary records of a transitional semi-arid ecosystem susceptible to  
45 environmental forcing conditions. Sediment cores were retrieved from two bays,  
46 Guanaqueros and Tongoy (29–30°S), for geochemical and biological analyses  
47 including: sensitive redox trace elements, biogenic opal, total organic carbon (TOC),  
48 diatoms, stable isotopes of organic carbon and nitrogen. Three remarkable periods were  
49 established, with different environmental conditions and productivities: (1) > cal BP  
50 6500, (2) cal BP 6500 – cal BP 1700 and (3) cal BP 1700 towards the present (CE  
51 2015). The first period was characterized by a remarkably higher productivity (higher  
52 diatom abundances and opal) when a large fluxes of organic compounds was also  
53 inferred from the accumulation of elements such as Ba, Ca, Ni, Cd and P in the  
54 sediments. At the same time, suboxic-anoxic conditions at the bottoms were suggested  
55 by the large accumulation of Mo, Re and U, showing a peak at cal BP 6500 when  
56 sulfidic conditions could have been established. This was also identified as the driest  
57 interval according to the pollen moisture index, although it could be extended until cal  
58 BP 5500. These conditions should be associated to an intensification of the SPSA and a  
59 stronger SWW, emulating La Niña-like conditions as has been described for the SE  
60 Pacific during the early Holocene, which in this case extends until the mid-Holocene.  
61 During most of the second period, lower productivity was observed. However, a small  
62 increment was identified between Cal BP 4500 and 1700 although low amounts of  
63 diatom (valves g<sup>-1</sup>) and nutrient-type metal accumulations were observed, contrasting  
64 with the first period when high opal accumulations and diatom abundances were  
65 synchronized. Oxygen conditions at the bottoms change to an almost stable sub-oxic  
66 condition during this time interval. The third period is marked by an intense  
67 oxygenation after cal BP 1700, as observed by a change in the accumulation of U, Mo  
68 and Re. In Addition, a small productivity rise after cal BP ~130 towards recent times

69 was observed, as suggested by opal accumulations but no increment in diatom  
70 abundance. Overall, lower primary productivity, higher oxygenation at bottoms and  
71 higher humidity conditions were established after cal BP 6500 and towards the present.  
72 We suggest that the oxygenation might be associated with intensified El Niño activity or  
73 similar conditions that introduce oxygenated waters to coastal zones by the propagation  
74 of waves of equatorial origin. This oxygenation is changing the original extent of the  
75 accumulation of elements sensitive to redox changes in sediments, even under the  
76 prevalence of high productivity and sub-oxic conditions.

77

78 Keywords: paleoproductivity, paleoredox, trace metals, diatoms, opal, organic carbon,  
79 Coquimbo, SE-Pacific

80

## 81 **1. Introduction**

82

83 Mean climatic conditions at the SE Pacific are modulated by the dynamic of the  
84 Southern Pacific Subtropical Anticyclone (SPSA) and the Humboldt Current System.  
85 The SPSA has seasonal, decadal and inter-decadal variability modulating the strength of  
86 the southern westerly winds (SWW) and hence the main oceanographic feature of the  
87 Eastern boundary margin, the upwelling, influencing the biogeochemical processes  
88 related to the inputs of nutrient and biological productivity. Seasonal variations produce  
89 periods of intense upwelling when the SPSA is stronger, while the opposite is true when  
90 it is weak (Croquette et al, 2007). The coastal wind pattern produced alongshore varies  
91 along the SE Pacific showing lower seasonality between 18°–30°S, and producing a  
92 semi-permanent upwelling (Pizarro et al., 1994; Figueroa and Moffat, 2000). This  
93 system is highly affected by the inter-annual variability imposed by El Niño Southern  
94 Oscillation (ENSO), with impacts on the wind intensity. The upwelling brings nutrient-  
95 poor waters during the warm phase, while the opposite happens during the cold phase  
96 (Ruttland and Fuenzalida, 1991; Blanco et al., 2002). Other climate patterns—namely the  
97 Pacific Decadal Oscillation (PDO) and the Southern Annular Mode (SAM)—operate on  
98 a much longer time scale (inter-annual, decadal, inter-decadal) modifying the strength  
99 and the position of the SWW, and thereby producing cold/warm periods and  
100 intense/weak upwelling (Ancapichún and Garcés-Vargas, 2015). In addition, the austral  
101 insolation influences the extent of the Antarctic sea ice and the Hadley cell, which act as  
102 important forces to the latitudinal displacement of the ITCZ (Inter-tropical Convergence  
103 Zone; Kaiser et al., 2008, and reference there in). These fluctuations produce humid and  
104 arid conditions along the SE Pacific where the wind's intensity remains the key factor  
105 for the upwelling's strength and, therefore, for the supply of nutrients to the photic zone,  
106 all of which are required for development of primary productivity.  
107 Off Coquimbo (30°S), there is normally a semi-permanent and intense upwelling forced  
108 by local winds, strongly influenced by topographic features (Figueroa and Moffat,  
109 2000) and ENSO variability (Escribano et al., 2004). During El Niño, mean winds  
110 alongshore reduce their intensity and the South East Pacific anticyclone weakens.  
111 Conversely, during La Niña mean winds alongshore increase their intensity and the  
112 anticyclone is reinforced (Rahn and Garreaud, 2013). This has an impact on the upper  
113 circulation of the ocean affecting oxygenation and the strength of upwelling. The high  
114 productivity that takes place close to the coast during normal periods (Escribano et al.,

115 2004 and references therein) maintains a zone of low dissolved oxygen content along  
116 the margin reinforcing the oxygen minimum zone (OMZ). This zone develops along the  
117 North and South Pacific Ocean and its intensity, thickness, and temporal stability vary  
118 as a function of latitude (Helly and Levin, 2004, Ulloa et al., 2012). To the north (e.g.  
119 21°S) and off Peru, the OMZ occurs permanently, and can extend into the euphotic  
120 zone. In the case of northern Chile and southern Peru, there is no significant interface  
121 with the benthic environment due to the presence of a narrow continental shelf (Helly  
122 and Levin, 2004). The OMZ dynamic off Coquimbo has not been studied in detail, but a  
123 seasonal intrusion of low oxygen waters to the coast has been observed (Gallardo et al.,  
124 2017). During the 97-98 El Niño event, the oxygenation of bottoms was clearly detected  
125 in north (23°S) and south-central Chile (36°S) (Ulloa et al., 2001; Sellanes et al., 2007;  
126 Gutiérrez et al., 2006), changing the normal suboxic conditions at the bottom, the  
127 normal composition of macrofauna and related geochemical characteristics of the  
128 sediments that have implications that persist for many years after the event (Sellanes et  
129 al., 2007; Gutiérrez et al., 2006).

130 These changes in primary productivity and oxygenation at the bottom can be observed  
131 in sedimentary records which respond to the amount of organic carbon that has settled  
132 on the bottom and to the diagenetic reactions during organic matter remineralization.  
133 Trace elements are commonly used as indicators of these processes, observed as  
134 element enrichment or depletion. It is driven by organic matter fluxes and redox  
135 conditions that modify the original extension of metal enrichment, which depend on the  
136 oxygen content during early diagenesis in the upper sediment layers and overlying  
137 water (Nameroff et al., 2002; Zheng et al., 2002; McManus et al., 2006; Siebert et al.,  
138 2003). Therefore they are a useful tool to establish temporal changes in primary  
139 productivity and also to establish changes in the oxygenation at the bottom on  
140 sedimentary records.

141 Our work focuses on the past variations of the environmental conditions deduced from  
142 marine sedimentary records of inorganic and organic proxies over the last ~8000 years  
143 BP, obtained from a transitional semi-arid ecosystem off central Chilean coast (30°S),  
144 close to Lengua de Vaca point, the most relevant upwelling area of Chile's northern  
145 margin (Shaffer et al., 1999; Thiel et al., 2007). We considered redox trace element  
146 measurements that respond to local hypoxia (U, Mo and Re) as well as nutrient-type  
147 elements, which follow the organic fluxes to the sediments (Ba, Ni Cu, P) (Tribovillard,  
148 2006). Additionally, we measured Fe and Mn which play a key role in adsorption-

149 desorption and scavenging processes of dissolved elements in bottom waters and  
150 sediments, and we measured Ca, K and Pb used to assess terrigenous inputs by coastal  
151 erosion, weathering and eolian transport, which is also true for Fe and Mn (Calvert and  
152 Pedersen, 2007). Ca accumulation depends, in turn, on carbonate productivity and  
153 dissolution, which has been used as a paleoproductivity proxy (Paytan, 2008; Govin et  
154 al., 2012). We determined the enrichment/depletion of elements to establish the main  
155 environmental conditions prevailing during the sedimentation of the particulated  
156 material (Böning et al., 2009). In addition, we considered the diatoms assemblages with  
157 biogenic opal as a measurement of siliceous export production, TOC and stable isotopes  
158 to identify variations in the organic fluxes to the bottoms. Moreover, pollen grains were  
159 used to identify environmental conditions based on the climate relationship of the main  
160 vegetation formations in North-Central Chile. Based on our records we were able to  
161 identify wet/dry intervals, periods with high/low organic fluxes to the sediments related  
162 to changes in primary production, and changes in the redox conditions at the bottoms.

163

## 164 **2. Study area**

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

170 The Tongoy and Guanaqueros bays are located in the southern edge of a broad  
171 embayment between small islands to the north (29°S; Choros, Damas and Chañaral) and  
172 Lengua de Vaca Point to the south (30°S) (Fig. 1), protected from predominant  
173 southerly winds. Tongoy Bay is a narrow marine basin (10 km at its maximum width)  
174 with a maximum depth of ~100 m. To the northeast lies Guanaqueros Bay, a smaller  
175 and shallower basin. High wind events evenly distributed throughout the year promote  
176 an important upwelling center at Lengua de Vaca Point, developing high biomass along  
177 a narrow coastal area (Moraga-Opazo et al., 2011; Rahn and Garreaud, 2013), and  
178 reaching maximum concentrations of ~20 mg m<sup>-3</sup> (Torres and Ampuero, 2009). In the  
179 shallow waters of Tongoy Bay, the high primary productivity results in high TOC in the  
180 water column allowing for the deposition of fine material to the bottom; TOC rises  
181 concurrently with the periods of low oxygen conditions (Fig. 3; Muñoz et al.,  
182 unpublished data). Recent oceanographic studies indicate that low dissolved oxygen

183 water intrusions from the shelf (Fig. 2) seem to be related to lower sea levels resulting  
184 from local wind annual cycles at a regional meso-scale (Gallardo et al., 2017). The  
185 spatial and temporal variability of these processes is still under study.  
186 Sedimentological studies are scarce in Chile's northern-central shelf. A few technical  
187 reports indicate that sediments between 27°S and 30°S are composed of very fine sand  
188 and silt with relatively low organic carbon content (<3 and ~5%), except in very limited  
189 coastal areas where organic material accounts for approximately ~16% (Muñoz,  
190 unpublished data; FIP2005-61 Report, www.fip.cl). Coastal weathering is the main  
191 source of continental input due to scarce river flows and little rainfall in the zone (0.5 to  
192 ~80 mm yr<sup>-1</sup>; Montecinos et al., 2016, Fig.1). Freshwater discharges are represented by  
193 creeks, which receive the drainage of the coastal range forming wetland areas in the  
194 coast and even small estuaries, such as Pachingo, located south of Tongoy (Fig. 1).  
195 These basins cover ~300 and 487 km<sup>2</sup>, respectively. The water volume in the estuaries  
196 is maintained by the influx of seawater mixed with groundwater supply. No surface flux  
197 to the sea is observed. Therefore, freshwater discharge occurs only during high rainfall  
198 periods in the coastal zone (DGA, 2011), which normally takes place during El Niño  
199 years when higher runoff has been recorded in the area during the austral winter (Valle-  
200 Levinson et al., 2000; Garreaud et al., 2009). Under this scenario, marine sediments are  
201 often highly influenced by primary production in the water column, and therefore,  
202 sedimentary records can reveal past variability in primary production and in the  
203 oceanographic conditions over the shelf, which ultimately respond to major atmospheric  
204 patterns.

205

### 206 **3. Materials and methods**

#### 207 **3.1. Sampling**

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

214 Subsequently, the cores were sliced into 1-cm sections and subsamples were separated  
215 for grain size measurements, magnetic susceptibility, trace elements, biogenic opal, C

216 and N stable isotope signatures ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ), and TOC analyses. The samples were first  
217 kept frozen ( $-20^\circ\text{C}$ ) and then freeze-dried before laboratory analyses.

218

### 219 **3.2. Geochronology ( $^{210}\text{Pb}$ and $^{14}\text{C}$ )**

220 Geochronology was established combining ages estimated from  $^{210}\text{Pb}_{\text{xs}}$  activities  
221 suitable for the last 200 years and radiocarbon measurements at selected depths for  
222 older ages.  $^{210}\text{Pb}$  activities were quantified through alpha spectrometry of its daughter  
223  $^{210}\text{Po}$  following the procedure of Flynn (1968).  $^{210}\text{Pb}_{\text{xs}}$  (unsupported) activities were  
224 determined as the difference between  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$  activities measured in some  
225 intervals of the sediment column.  $^{226}\text{Ra}$  was measured by gamma spectrometry at the  
226 Laboratoire Géosciences of the Université de Montpellier (France). Standard deviations  
227 (SD) of the  $^{210}\text{Pb}$  inventories were estimated propagating counting uncertainties  
228 (Bevington and Robinson, 1992) (Table S1, supplementary data). The ages were based  
229 on the Constant Rate of Supply Model (CRS, Appleby and Oldfield, 1978).

230 Radiocarbon measurements were performed on a mix of planktonic foraminifera species  
231 in core BGGC5 whereas the benthic foraminifera species *Bolivina plicata* was selected  
232 for core BTGC8 (Table 1). The samples were submitted to the National Ocean Sciences  
233 AMS Facility (NOSAMS) of the Woods Hole Oceanographic Institution (WHOI). The  
234 time scale was obtained according to the best fit of ages obtained from  $^{210}\text{Pb}_{\text{xs}}$  and  $^{14}\text{C}$   
235 (Fig. 4), using the CLAM 2.2 software and using the Marine curve 13C (Reimer et al.,  
236 2013). A reservoir deviation from the global mean reservoir age (DR) of  $441 \pm 35$  years  
237 was considered, established according Sabatier et al. (2010). This was estimated  
238 subtracting the  $^{14}\text{C}$  age value corresponding at the historical dates 1828 AD and 1908  
239 AD ( $499 \pm 24$  and  $448 \pm 23$   $^{14}\text{C}$  yr, respectively, Reimer et al., 2013) from the apparent  
240  $^{14}\text{C}$  age of foraminifers measured at depths of 5 and 10 cm for cores BTGC8 and  
241 BGGC5, respectively (Sabatier et al., 2010; Table 2).

242

### 243 **3.3. Geophysical characterization**

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

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



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

252

### 253 **3.4. Chemical analysis**

254 Trace element analyses were performed by ICP-MS (Inductively Coupled Plasma-Mass  
255 Spectrometry) using an Agilent 7700x at Université de Montpellier (OSU  
256 OREME/AETE regional facilities). Sediment samples and geochemical reference  
257 materials (UBN, BEN and MAG1) were dissolved using a concentrated mix of acids  
258 (HF-HNO<sub>3</sub>-HClO<sub>4</sub>) in Savillex screw-top Teflon beakers at 120°C. Final solution  
259 considered the addition of a known weight of internal standard solution consisting of 1  
260 ppb of In and Bi. Internal standardization used ultra-pure solution enriched in In and Bi,  
261 both elements whose natural abundances in geological samples do not contribute  
262 significantly to the added internal standard. This is used to deconvolve mass-dependent  
263 sensitivity variations of both matrix and instrumental origin, occurring during the course  
264 of an analytical session.

265 Mean metal concentrations for the analyzed samples were determined by external  
266 calibrations prepared daily from multi- and mono-elemental solutions, with  
267 concentrations in the range of 0.05–10 ppb for trace elements and of 1–10 ppm for  
268 major elements (Ca, K). Polyatomic interferences were controlled by running the  
269 machine at an oxide production level <1%. The analytical precisions attained by this  
270 technique were between 1% and 3% and accuracy better than ±5%.

271 TOC and stable isotope ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) analyses were performed at the Institut für  
272 Geographie, Friedrich Alexander Universität (FAU) Erlangen-Nürnberg, Germany  
273 using a Carlo Erba elemental analyzer NC2500 and an isotope-ratio-mass spectrometer  
274 (Delta Plus, Thermo-Finnigan) for isotopic analysis. Carbon and nitrogen contents were  
275 determined from the peak-area-versus-sample-weight ratio of each individual sample  
276 and calibrated with the elemental standards cyclohexanone-2,4-dinitrophenylhydrazone  
277 (C<sub>12</sub>H<sub>14</sub>N<sub>4</sub>O<sub>4</sub>) and atropine (C<sub>17</sub>H<sub>23</sub>NO<sub>3</sub>) (Thermo Quest). A laboratory-internal organic  
278 standard (Peptone) with known isotopic composition was used for final isotopic  
279 calibrations. Stable isotope ratios are reported in the  $\delta$  notation as the deviation relative  
280 to international standards (Vienna Pee Dee Belemnite for  $\delta^{13}\text{C}$  and atmospheric N<sub>2</sub> for  
281  $\delta^{15}\text{N}$ ), so  $\delta^{13}\text{C}$  or  $\delta^{15}\text{N}$  = [(R sample/R standard) - 1] x 10<sup>3</sup>, where R is <sup>13</sup>C/<sup>12</sup>C or  
282 <sup>15</sup>N/<sup>14</sup>N, respectively. Typical precision of the analyses was ±0.1‰ for  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ .

283

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

291

### 292 **3.5. Microfossils analyses**

293 Qualitative abundances of siliceous microfossils were carried out every centimeter  
294 following the Ocean Drilling Program (ODP) protocol described by Mazzullo et al.  
295 (1988), with this information were selected some sections every ~4, 8 and 12 cm for  
296 BGGC5 and at ~6 cm for BTGC8 for quantitative abundances of microfossils (diatoms,  
297 silicoflagellates, sponge spicules, crysophyts and phytoliths). Briefly ~ 0.5 g of freeze-  
298 dried sediment was treated according to Schrader and Gersonde (1978) for siliceous  
299 microfossils. Siliceous microfossils were identified and counted under an Olympus  
300 CX31 microscope with phase contrast. 1/5 of the slides were counted at 400X for  
301 siliceous microfossils and one transect at 1000x was counted for *Chaetoceros* resting  
302 spores (*Ch.* resting spores). Two slides per sample were counted; the estimated counting  
303 error was 15%. Total diatom abundances are given in valves  $g^{-1}$  of dry sediments.

304 Pollen analysis was conducted following the standard methodology for sediment  
305 samples (Faegri and Iversen, 1989). The samples were mounted with liquid glycerol and  
306 sealed permanently with paraffin wax. Pollen identification was conducted under a  
307 stereomicroscope at 400 fold magnification with the assistance of the Heusser (1973)  
308 pollen catalogue. A total of 100-250 terrestrial pollen grains were counted on each  
309 sample depending on their abundance. Pollen percentage for each taxon was calculated  
310 from the total sum of terrestrial pollen. The percentage of aquatic pollen and fern spores  
311 was calculated based on the total terrestrial sum plus their respective group. Pollen  
312 percentage diagrams were generated using the Tilia software (E. Grimm, Illinois State  
313 Museum, Springfield, IL. USA). The diagram was divided into “zones” based on the  
314 identification of the most important changes in pollen percentage and assisted by a  
315 cluster ordination (CONISS) performed by the same software.

316 We further summarize pollen-based precipitation trends by calculating a Pollen  
317 Moisture Index (PMI), which is defined as the normalized ratio between Euphorbiaceae  
318 (wet coastal scrubland) and Chenopodiaceae (arid scrubland). Thus, positive (negative)  
319 values of this index indicate the relative expansion (reduction) of coastal scrubland  
320 under relatively wetter (drier) conditions.

321

## 322 **4. Results**

### 323 **4.1. Geochronology**

324  $^{210}\text{Pb}_{\text{xs}}$  (unsupported activity) was obtained from the surface at a depth of 8 cm in the  
325 two cores, with an age of  $\sim$  AD 1860 at 8 cm in both (Table S1). Greater surface  
326 activities were obtained for core BGGC5 ( $13.48 \pm 0.41$  dpm  $\text{g}^{-1}$ ) compared to core  
327 BTGC8 ( $5.80 \pm 0.19$  dpm  $\text{g}^{-1}$ ), showing an exponential decay with depth (Fig. 4). A  
328 recent sedimentation rate of  $0.11 \pm 0.01$  cm  $\text{yr}^{-1}$  was estimated.

329 The age model provided a maximum age of cal BP 8210 for core BGGC5, and cal  
330 BP 7941 for core BTGC8 (Fig. 4). A mean sedimentation rate of  $0.02$  cm  $\text{yr}^{-1}$  was  
331 estimated for core BGGC5, with a period of relative low values ( $0.01$  cm  $\text{yr}^{-1}$ ) between  
332 cal BP  $\sim$ 4000 and 6000. For BTGC8, sedimentation rates were less variable and around  
333  $0.013$  cm  $\text{yr}^{-1}$  in the entire core. An age reserve estimation following the method by  
334 Sabatier et al. (2010) resulted in  $441 \pm 35$  and  $442 \pm 27$  years for BGGC5 and BTGC8  
335 cores, respectively (Table 2). These values were close to the global marine reservoir and  
336 higher than other estimations along the Chilean margin at shallower depths ( $146 \pm 25$   
337 years at  $< 30$  water depth; Carré et al., 2016; Merino-Campos et al., 2018). Our coring  
338 sites are deeper ( $\sim 90$  m water depth) and influenced by upwelled water from Lengua de  
339 Vaca Point, which could explain such differences. However, moderate differences were  
340 observed between models using both reservoir values. Thus, our estimations were based  
341 on two pre-bomb values established with  $^{210}\text{Pb}$  measured in sediments and  $^{14}\text{C}$  in  
342 foraminifers, used for the age modeling.

343

### 344 **4.2. Geophysical characterization**

345 Sediments retrieved from the bays showed fine grains within the range of very fine sand  
346 and silt in the southern areas. There, grain size distribution was mainly unimodal, very  
347 leptokurtic, better sorted and skewed to fine grain when compared to sediments from  
348 the northern areas. Sediment cores obtained from the northern areas were sandy (coarse  
349 sand and gravel), with abundant calcareous debris. Longer cores of soft sediment were

350 retrieved at the southernmost areas (BGGC5 and BTGC8), where the silty component  
351 varied between 40 % and 60 % (Fig. 1 and 5a,b). The clay component was very low at  
352 both cores (<2%). The sediment's color ranged from very dark grayish brown to dark  
353 olive brown (2.5Y 3/3–3/2) in Guanaqueros Bay (BGGC5) and from dark olive gray to  
354 olive gray (5Y 3/2–4/2) in Tongoy Bay (BTGC8). Visible macro-remains (snails and  
355 fish vertebrae) were found, as well as weak laminations at both cores. The magnetic  
356 susceptibility showed higher values close to the surface, up to 127 SI x10<sup>-8</sup> at BGGC5,  
357 and relative lower values (85 SI x10<sup>-8</sup>) at BTGC8. At greater depths, however, the  
358 values were very constant, around 5–8 x10<sup>-8</sup> SI at BGGC5 core and around 12–20 x10<sup>-8</sup>  
359 SI at BTGC8 core. In both cores, susceptibility rises substantially in the last century  
360 (Figs. 5a, 5b). Lower bulk densities were estimated at core BGGC5 (0.7–0.9 g cm<sup>-3</sup>),  
361 compared with core BTGC8 (>1 g cm<sup>-3</sup>) (Fig. 5a, 5b). In line with this, mean grain size  
362 amounted to 60–80 µm in Guanaqueros Bay (BTGC8), compared to 50–60 µm in  
363 Tongoy Bay (BGGC5). Both cores were negatively skewed, with values of -1 to -1.2 at  
364 BGGC5, and -1 to -2.5 at BTGC8. Minor increases towards coarser grain size were  
365 observed in the last ~1000 years, especially in Tongoy Bay (BTGC8). In both cases,  
366 grain size distributions were strongly leptokurtic. Ca/Fe ratio also reduced in time,  
367 except at core BTGC8 where it was only observed during the last ~2000 years.

368

### 369 **4.3. Biogenic components**

#### 370 **4.3.1. Siliceous microfossils and biogenic opal**

371 Total diatom abundance fluctuated between 5.52 x10<sup>5</sup> and 4.48 x10<sup>7</sup> valves g<sup>-1</sup> at core  
372 BGGC5. Total diatom abundance showed a good correlation with biogenic opal content  
373 at BGGC5 (R<sup>2</sup> =0.52, P<0.5), with values raising from 72 cm to the bottom of the core,  
374 corresponding to cal BP 5330, and reaching their highest values before cal BP 6500. On  
375 the contrary, diatom abundance and biogenic opal were much lower at core BTGC8 (< 2  
376 x10<sup>5</sup> valves g<sup>-1</sup> and <3%, respectively). Here, the siliceous assemblage was almost  
377 completely conformed by *Chaetoceros* resting spores (RS) (Fig. 6).

378 A total of 135 and 8 diatom taxa were identified in cores BGGC5 and BTGC8  
379 respectively, where core BTGC8 registered very low diatom abundances. In general,  
380 diatoms were the most important assemblage of siliceous microfossils (96 %), followed  
381 by sponge spicules (3 %). The contribution of phytoliths and chrysophyte cysts was less  
382 than 2 % at core BGGC5. *Chaetoceros* (RS) dominated diatom assemblage (~90 %; Fig.  
383 6), and included the species *C. radicans*, *C. cinctus*, *C. constrictus*, *C. vanheurckii*, *C.*

384 *coronatus*, *C. diadema*, and *C. debilis*. Other upwelling group species recorded (mainly  
385 at core BGGC5) were: *Skeletonema japonicum*, and *Thalassionema nitzschioides* var.  
386 *nitzschioides* (Table S2). Freshwater diatoms (*Diploneis papula*, *Cymbella tumida*,  
387 *Fragilaria capucina*, *Diatoma elongatum*) and non-planktonic diatoms (*Cocconeis*  
388 *scutellum*, *C. costata* and *Gramatophora angulosa*) accounted for ~0.1–5 %; while the  
389 group of coastal planktonic diatoms accounted for ~0.3–6 % of the total assemblage.  
390 The main planktonic diatoms were (*Rhizosolenia imbricata*, and *Thalassiosira*  
391 *eccentrica*). Oceanic-warm diatoms (*Roperia tessellata*, *Th. nitzschioides* var *inflatula*)  
392 and the tytoplanktonic diatom group were rare, with less than 1 %.

393

#### 394 **4.3.2. TOC and stable isotopes distribution**

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

403

#### 404 **4.3.3. Pollen record**

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

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

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

417 considerably.  
418 Zone BG-3 (cal BP 6500 –3400): This zone is marked by a steady decline in  
419 Chenopodiaceae and *Leucheria*-types, and by the expansion of several other  
420 herbaceous elements, such as Euphorbiaceae, *Baccharis*-type and Brassicaceae.  
421 Zone BG-4 (cal BP 3400 – 120): This zone is mostly dominated by Ast. subf.  
422 Asteroideae, and marked by the decline of Chenopodiaceae and *Leucheria*-type. Other  
423 coastal taxa –such as Euphorbiaceae, *Baccharis*-types, Asteraceae subf.  
424 Chichorioideae, *Quillaja saponaria*, Brassicaceae and *Salix* spp.– also expand in this  
425 zone.  
426 Zone BG-5 (cal BP 120 – -60): The upper portion of the record is dominated by  
427 Asteraceae subf. Asteroideae and Poaceae, and marked by higher amounts of  
428 Geraniaceae, Asteraceae subf. Mutisieae, Myrtaceae and *Q. saponaria*. Additionally,  
429 this zone includes introduced pollen types such as *Rumex* spp. and *Pinus* spp. The  
430 latter is not shown in the diagram of Figure 8 because its abundance was minimal.  
431 Overall, the most distinctive trend revealed by core BGGC-5 is a long-term reduction  
432 in Chenopodiaceae and higher amounts of Euphorbiaceae and Asteraceae subf.  
433 Asteroideae. Along with these changes, a further expansion of several other pollen  
434 representative of the coastal shrub land vegetation began at about cal BP 6500.

435

#### 436 **4.4. Trace element distributions**

437 Trace element distributions are shown in figures 8a and 8b for Guanaqueros (BGGC5)  
438 and Tongoy Bays (BTGC8), respectively. We use Al as a normalizing parameter for  
439 enrichment/depletion of elements due to its conservative behavior. The elements are  
440 presented as metal/Al ratios. Trace metals are sensitive to the presence of oxygen (U,  
441 Re, Mo) showing an increasing metal/Al ratio from the base of core BGGC5 (cal BP  
442 ~8210) up to cal BP 6500. After this peak, ratios showed a slight increase towards cal  
443 BP 1700, close to the beginning of the recent era, followed by a sharp reduction until  
444 present. Similarly, metal ratios at core BTGC8 increase over time, yet the peak was  
445 observed at cal BP ~1000. The exception to this trend was Mo, which reached a  
446 maximum value up to cal BC 6500 and then reduced steadily into the present.  
447 Additionally, metal/Al values were higher at core BGGC5. Iron revealed a clear  
448 upward trend around cal BP 3300 – 3500 at core BGGC5, which was not clearly  
449 observed at the Tongoy core. Instead, core BTGC8 showed peak Fe values around cal

450 BP 6500 – 7800; in both cores Fe increased in the past 130 years. No clear trend could  
451 be established for Mn.

452 A second group of elements (metal/Al ratios), including Cd, Ni and P (related to  
453 primary productivity and organic fluxes), showed a pattern similar to that of Mo/Al  
454 towards the bottom of core BGGC5, i.e. increasing values from cal BP ~8000 reaching  
455 highest values around cal BP 6500; after that the values followed constant reductions  
456 towards the present. A third group, consisting of Ba, P and Ca, exhibited a less clear  
457 pattern. Cd/Al and Ni/Al ratios at core BTGC8 showed only slightly decreasing  
458 values, and very low peak values compared to core BGGC5. The same pattern is  
459 observed for other elements. Metal/Al ratios for Ba, Ca and P were lower and  
460 presented a long-term reduction pattern towards the present.

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

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

476

## 477 **5. Discussion**

### 478 **5.1. Sedimentary composition of the cores: terrestrial *versus* biogenic inputs**

479 The sediments in the southern zones of the bays are a sink of fine particles transported  
480 from the north and the shelf (Fig. 5a, 5b), and respond to water circulation in the  
481 Guanaqueros and Coquimbo Bays (Fig. 1). Both have been described as bipolar, i.e.  
482 two counter-rotating gyres moving counterclockwise to the north and clockwise to the  
483 south (Valle-Levinson and Moraga, 2006). This is the result of the wind's

484 predominant direction and a coastline shape delimited by two prominent points to the  
485 north and south. Circulation in Tongoy Bay (the southernmost bay of the system)  
486 shows a different pattern due to its northern direction compared to Guanaqueros Bay,  
487 which opens to the west. The cyclonic recirculation in Tongoy Bay seems to be part of  
488 a gyre larger than the Bay's circulation (Moraga-Opazo et al., 2011) (Fig. 1). This  
489 could explain the differences in the distribution and composition of sediment particles  
490 between both Bays. In Tongoy Bay, there is less organic carbon accumulation ( $< 3 \text{ g}$   
491  $\text{m}^{-2} \text{ yr}^{-1}$ ), siliceous microfossils and pollen (Figs. 5b, 6 and 7). Similarly, in  
492 Guanaqueros Bay TOC contents are only slightly higher ( $> 2 \%$ ), especially between  
493 cal BP 3700 and 4000 and before cal BP 6500 ( $\sim 4 \%$ ), but with greater accumulation  
494 rates of about 7 and 16  $\text{g m}^{-2}\text{yr}^{-1}$ , respectively (Fig. 5a). However, these sediments  
495 contain enough microfossils to establish differences in primary productivity periods  
496 and also provide a pollen record evidencing prevailing environmental conditions.  
497 Stable isotopes measured in the study area were in the range of marine sedimentary  
498 particles for southern oceans at low and mid-latitudes ( $\delta^{13}\text{C}$ ;  $-20 \text{‰} - -24 \text{‰}$ ;  
499 Williams 1970; Rau et al., 1989; Ogrinc et. al. 2005), and slightly lower than the TOC  
500 composition in the water column ( $-18 \text{‰}$ , Fig. 3). This suggests that the organic  
501 particles that settle on the bottom are a more refractory material (C/N: 9–11),  
502 remineralized during particle transportation and sedimentation. This results in lighter  
503 isotopic compositions, especially at core BTGC8. Furthermore,  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  in  
504 settled particles have higher negative values in surface sediments due to a preferential  
505 decomposition of molecules rich in  $^{13}\text{C}$  and  $^{15}\text{N}$ , resulting in lighter isotope values and  
506 higher C/N ratios in sediments than in suspended particles (Fig. 3, 5a, 5b). However,  
507 this is also due to the stronger diagenetic reactions observed near the bottom layer  
508 (Nakanishi and Minagawa, 2003). Thus, these sediments are composed by winnowed  
509 particles transported by water circulating over the shelf, and the isotopic variations  
510 should not clearly establish the contribution of terrestrial inputs.  
511 Otherwise, the isotopic composition of upwelled  $\text{NO}_3^-$  (De Pol-Holz et al., 2007) could  
512 influence the variability of  $\delta^{15}\text{N}$ . Values for  $\delta^{15}\text{N}$  in northern and central Chile are in  
513 the range of those measured at the BGGC5 core ( $\sim 11 \text{‰}$ ; Hebbeln et al., 2000, De Pol-  
514 Holz et al., 2007), resulting from the isotopic fractionation of  $\text{NO}_3^-$  during nitrate  
515 reduction within OMZ, leaving a remnant  $\text{NO}_3^-$  enriched in  $^{15}\text{N}$  (Sigman et al., 2009;  
516 Ganeshram et al., 2000 and references therein). In this case, BGGC5 core sediments



517 represent the effect of the upwelling's nutrient supply and the influence of OMZ on  
518 the shelf, resulting in  $\delta^{15}\text{N}$  of 9 – 10 ‰. At sediment core BTGC8, lower values  
519 (< 8 ‰) measured at greater depths within the core should account for the mix with  
520 isotopically lighter terrestrial organic matter (Sweeney and Kaplan, 1980) due to its  
521 vicinity to a small permanent wetland in the southern side of Tongoy Bay. The  
522 material collected at Pachingo wetland showed  $\delta^{15}\text{N}$  of 1 – 8 ‰ (Muñoz et al., data  
523 will be published elsewhere) in the range of sedimentary environments influenced by  
524 terrestrial runoff (Sigman et al., 2009). Likewise, in most cases lower TOC is  
525 consistent with lighter isotope  $\delta^{15}\text{N}$  values, and also with higher C/N ratios, suggesting  
526 a combination with continental material (Fig. 5b).

527 MS measurements revealed lower values in both cores (BGGC5:  $5 - 8 \times 10^{-8}$  SI;  
528 BTGC8:  $12 - 20 \times 10^{-8}$  SI), except during the last ~200 years (CE 1800), when it  
529 reaches higher values substantially similar to those observed in the Pachingo wetland  
530 ( $40 - \sim 200 \times 10^{-8}$  SI; unpublished data), in the southern area of Tongoy Bay, pointing  
531 to an increase in flooding events in the last 200 years. Magnetite has a strong  
532 response to magnetic fields and its concentration is considered proportional to  
533 magnetic susceptibility (Dearing, 1999), but suffers post-depositional  
534 transformations (alteration of magnetite minerals) and can be diluted by biogenic  
535 components (carbonates, silicates), altering the MS intensity in areas with high  
536 organic accumulation rates (Hatfield and Stoner, 2013). This, however, is not the  
537 case of our cores where low sedimentation rates were estimated ( $0.01 - 0.02 \text{ cm yr}^{-1}$ )  
538 and the MS should be mainly accounting for the particles' source. The higher MS  
539 measurements on surface sediments would indicate a greater contribution of  
540 terrestrial material. The area is surrounded by several creeks that are only active  
541 during major flooding events, with greater impacts on Tongoy compared to  
542 Guanaqueros Bay. There has been a considerable increment in the contribution of  
543 terrigenous material in Tongoy Bay, in recent times (Ortega et al., 2019), which is  
544 diluting organic proxy records and increasing the grain size. Our records indicate a  
545 slight increase in mean grain size in both bays, supported also by a slight reduction in  
546 the Ca/Fe ratio pointing to a higher Fe input from continental erosion (Fig. 5a, 5b).  
547 Furthermore, lower concentrations of Ca in the deepest part of both cores to the  
548 surface was interpreted as a declining primary productivity (Keshav and Achyuthan,  
549 2015; Sun et al., 2016); however, higher concentrations were measured in core

550 BGGC5 compared with core BTGC8, where more terrigenous influence is being  
551 suggested. The slight rise of K/Ca ratio in time –from the bottom to the surface–  
552 should also be interpreted as a slight growth of the continental input, since K is  
553 related to siliciclastic material from coastal erosion, and from fluvial and  
554 groundwater inputs. However, the variation of Ca was larger (Fig.6a, 6b), resulting in  
555 higher K/Ca ratios on the surface. This suggests that the continental input has not  
556 changed much in time –at a millennial scale– but rather that primary productivity has  
557 declined (Fig. 5a, 5b).

558 Thus, cores BGGC5 and BTGC8 in Guanaqueros and Tongoy Bays are recording the  
559 variability of oceanographic conditions, but in the Tongoy core, the concentration of  
560 oceanographic proxies dilute due to the input of terrigenous material. This helps to  
561 decipher the climatic variability considering that the main input of clastic material to  
562 the area takes place during major flooding events. Additionally, the main circulation  
563 of the bay system leads to favorable conditions for sedimentation and the preservation  
564 of organic marine proxies in the Guanaqueros Bay, hence making the sedimentary  
565 records of these sites complementary.

566

## 567 **5.2. Temporal variability of proxies for primary productivity**

568 Several elements that take part in phytoplankton growth are useful to interpret the  
569 variations in primary productivity with time, as they are preserved in the sediments  
570 under suboxic-anoxic conditions. This produces enrichment over crustal abundance  
571 which distinguishes them from continental inputs. The presence of free dissolved  
572 sulfides produced by sulfate reduction reactions in the diagenesis of organic matter  
573 allows for the precipitation of metals into pore waters (Calvert and Pedersen, 1993;  
574 Morse and Luther, 1999). At the same time, organic matter remineralization releases  
575 ions into pore waters where they could form organic complexes and insoluble metal  
576 sulfides. Conversely, they could be incorporated into pyrite as Cd, Ni and Cu, showing  
577 different degrees of trace metal pyritization (Huerta-Diaz and Morse, 1992). Ca, Sr,  
578 Cd and Ni profiles suggest a lower share of organic deposition over time (Fig. 8a, 8b),  
579 consistent with the slight reduction of TOC content observed in the sediments (Figs.  
580 5a, 5b), and concomitantly with other elements related to organic fluxes to the bottom  
581 and primary productivity. In the case of Ba, it is actively incorporated into  
582 phytoplankton biomass or adsorbed onto Fe oxyhydroxides, increasing the Ba flux  
583 towards the sediments, where it is also released during organic matter diagenesis. Ba is

584 precipitating in microenvironments where Ba-sulfate reaches supersaturation  
585 (Tribovillard et al., 2006 and references therein), but it is dissolved in suboxic-anoxic  
586 environments or where sulfate is significantly depleted (Torres et al., 1996; Dymond  
587 et al., 1992). Therefore, it is better preserved in less anoxic environments with  
588 moderate productivity, expected to be the case of our study site (Gross Primary  
589 Productivity = 0.35 to 2.9 g C m<sup>-1</sup>d<sup>-1</sup>; Daneri et al., 2000). Hence, the slight rise of Ba  
590 from cal BP 4000 to the present (Fig. 8a) is more of a response to a less anoxic  
591 environment than to an increase in primary productivity, and results in a low negative  
592 correlation with TOC (-0.59; Table 5) due to Ba remobilization in anoxic conditions  
593 before cal BP 6500. After this age, the reduction in TOC and other nutrient-type  
594 elements (Ni, Sr, Ca, Cd) into the present is consistent with the rise in oxygen in  
595 bottoms. On the other hand, P distribution showed a trend similar to that of TOC and  
596 other elements related to organic fluxes into the bottom (Ni, Cd), although with a  
597 lower correlation (~0.6). The accumulation of P depends on the deposition rate of  
598 organic P (dead plankton, bones and fish scales) on the bottom, and is actively  
599 remineralized during aerobic or anaerobic bacterial activity. P and TOC showed a  
600 declining trend towards the present, suggesting reducing flux of organic matter over  
601 time, which was also observed for Ni and Cd distributions. Alternatively, reducing  
602 fluxes of organic proxies could be explained by the higher remineralization of organic  
603 material settled on the bottom due to higher oxygen availability (Figs. 8a, 8b).

604 Productivity reconstructions were based on qualitative diatom and sponge spicules  
605 relative abundances, quantitative diatom counts (valves g<sup>-1</sup>) and biogenic opal content  
606 only in core BGGC5, since core BTGC8 registered low valve counts (< 1 % in relative  
607 diatom abundance). However, in both cores diatom assemblages were represented  
608 mainly by *Ch.* resting spores, which are used as upwelling indicators, showing higher  
609 concentrations during periods of high productivity and upwelling (Abrantes 1988,  
610 Vargas et al., 2004). In addition, *Ch.* resting spores are highly silicified and well  
611 preserved in coastal sediments (Blasco et al., 1981). The downcore siliceous  
612 productivity based on opal distribution (Fig. 6) distinguished three main time intervals  
613 of higher productivity: (1) > cal BP 6500, (2) cal BP 1700 – cal BP 4500 and (3)  
614 recent times (CE 2015) – cal BP ~130. The opal accumulation rate in the first interval  
615 was remarkably high, amounting to ~27 ± 13 g m<sup>2</sup> yr<sup>-1</sup> (range: 9 – 53 g m<sup>2</sup>yr<sup>-1</sup>, Table  
616 4), when *Chaetoceros* spores were predominant, indicating an upwelling  
617 intensification; during the second interval, it decreased to ~ 11 ± 4 g m<sup>2</sup> yr<sup>-1</sup> (range: 2 –

618 21 g m<sup>-2</sup>yr<sup>-1</sup>, Table 4). This is partially consistent with nutrient-type element  
619 distributions (Fig. 8a). The third interval accounts for the last ~200 years, when high  
620 opal accumulations and high Cd/U ratios could also be observed increasing towards  
621 the present (mean opal value of 29 ± 14 g m<sup>2</sup> yr<sup>-1</sup>, range: 3 – 40 g m<sup>2</sup> yr<sup>-1</sup>). However,  
622 low diatom abundances were observed (range: 0.5 – 4.9 x10<sup>6</sup> valves g<sup>-1</sup>), probably  
623 because recent sedimentation rates were higher, altering the estimations of opal flux.  
624 Additionally, few sections of the core surface were analyzed for diatoms leading to a  
625 low resolution of this measurement in the most recent period. Cu and Fe also increased  
626 during this period (Fig. 8a), contributing to fertilize the environment and promoting  
627 primary productivity. In this sense, higher productivity in the last 200 years could be  
628 suggested but further investigations are needed. The second time interval with a higher  
629 productivity was not clearly identified in terms of metals, except for Fe, which clearly  
630 shows higher values during this period (Fig. 8a). During the first period, all metal  
631 proxies showed primary productivity increases before cal BP 6500, as indicated by  
632 opal accumulation within the sediments. Here, Cd and U accumulations in the  
633 sediments resulted in high Cd/U ratios, even at core BTGC8 (> 2; Fig. 6), indicating  
634 very low oxygen conditions (Cd/U ratios could vary between 0.2 and 2 from suboxic  
635 to anoxic environment; Nameroff et al., 2002). Lower ratios (< 1; Fig. 6) were  
636 estimated when the opal accumulation was low during the second time interval,  
637 indicating higher variations in primary productivity over time with moderate changes  
638 in oxygen conditions in the bottoms. Furthermore, opal showed good correlations with  
639 Ni and Cd (~0.70; Table 5; Fig. 8a), all of which suggests the relevance of bottom  
640 organic fluxes for the buildup of elements within the sediments, and establishes a clear  
641 period of higher primary productivity around cal BP 6500, when the lowest oxygen  
642 conditions prevailed (Fig. 6).

643

### 644 **5.3. Temporal variability of proxies for bottom water oxygenation**

645 The distributions of U, Re and Mo at core BGGC5 indicate that anoxic or suboxic  
646 conditions were developed from cal BP 8200 to ~ cal BP 1700 (Fig. 8a, 8b). After this  
647 period and into the present, however, a remarkable reduction in their concentration  
648 suggests a more oxygenated bottom environment, concurrent with lower organic  
649 fluxes to the sediments. The Re profile shows the influence of suboxic waters not  
650 necessarily associated with higher organic matter fluxes to the bottom. Since this  
651 element is not scavenged by organic particles, its variability is directly related to

652 oxygen changes (Calvert and Pedersen, 2007, and references therein). Additionally, it  
653 is strongly enriched above crustal abundance under suboxic conditions (Colodner et  
654 al., 1993; Crusius et al 1996), being >10 times at core BGGC5 (Table 4) before cal BP  
655 1700. Similarly, U shows a similar pattern and while organic deposition has an impact  
656 on its distribution (Zheng et al., 2002), it is also related to changes in bottom oxygen  
657 conditions. This is because its shift from a soluble conservative behavior to a non-  
658 conservative and insoluble behavior depends solely on redox potential changes that  
659 occur near the Fe(III) reduction zone (Klinkhammer and Palmer, 1991.).  
660 Molybdenum, which showed higher values at cal BP 6500, also indicates the presence  
661 of sulfidic conditions, as shown by a Re distribution highly enriched under anoxic  
662 environments (Colodner et al., 1993), and by the reduction of Re(VII) to Re(IV),  
663 forming ReO<sub>2</sub> or ReS (Calvert and Pedersen, 2007). The enrichment of Rhenium, U  
664 and Mo is used to decipher the redox condition within the sediments, even in places  
665 with high lithogenic inputs that could obscure the authigenic enrichment of other  
666 elements under similar conditions (Crusius et al., 1996). In both places, the  
667 concentrations of these elements showed values above the crustal abundance,  
668 especially in core BGGC5 (Table 4), with Re and Mo enriching by ~19 and U by ~5,  
669 except in the past ~1700 years when they reduced by half. This suggests that the  
670 presence of anoxic conditions was stronger before cal BP 1700 (based on mean EFs  
671 and Me/Al ratios distribution), with a peak around cal BP 6500 (based on EF<sub>Cd</sub>) and  
672 followed by a less anoxic condition after cal BP ~1700 (Fig. 8a, Table 4). The most  
673 important enrichment was observed for Cd (> 30) that was higher before cal BP 6500  
674 (~140), in agreement with higher opal accumulation and diatom abundance (Fig. 6,  
675 Table 4). The most important enrichment could similarly indicate the sulfidic  
676 condition within the sediments that allows for Cd precipitation. It is also supported by  
677 Mo enrichment (mean EF<sub>Mo</sub>=16.9), since its buildup within the sediments is highly  
678 controlled by sulfide concentrations (Chaillou et al., 2002; Nameroff et al., 2002;  
679 Sundby et al., 2004).

680 Something similar occurs in Tongoy Bay (core BTGC8), but trace metal  
681 concentrations are lower for all elements and also for TOC, suggesting a limited  
682 influence on metal accumulation within the sediments.

683 Thus, these elements suggest anoxic or even sulfidic conditions within the sediments  
684 in both places at around cal BP 6500 – 7200 (Fig. 8a, 8b). After this period, a second  
685 peak but less intense low oxygen condition is observed at the beginning of the recent

686 era (cal BP 1700), continuing with conspicuous oxygenation until present times. This  
687 interpretation –based on the distribution of U, Re and Mo– complements the  
688 observations of nutrient-type elements pointing both to oxygenation changes and to  
689 changes in organic fluxes throughout the sediments. A less prominent accumulation of  
690 nutrient-type elements (Ni, Cd, Ba, Ca and P) would point to lower organic matter  
691 deposition into the sediments but still promoting low oxygen conditions within the  
692 sediments and lower sulfide content over time, which are nevertheless high enough to  
693 sustain Mo accumulation until cal BP 1700. After that, lower Re, U and Mo  
694 accumulation and EFs were observed, suggesting the relevance of bottom oxygenation  
695 (Table 4). This could also explain the conspicuous upward trend of Cu/Al and Fe/Al in  
696 recent times due to the presence of oxides (Fig. 8a, 8b). Apparently, a low level of  
697 dissolved Cu is maintained by the complexation with organic compounds produced by  
698 phytoplankton and Cu adsorption on Fe oxides (Peacock and Sherman, 2004; Vance et  
699 al., 2008; Little et al., 2014), with both processes augmenting Cu in the particulate  
700 phase over surface sediments ( $EF_{Cu}=4.6\pm 0.5$ , Table 4). In our study sites, Fe and Cu  
701 concentrations were higher in surface sediments, probably related to a higher  
702 availability of Fe and Cu in the environment (Fig. 8a, 8b). In turn, this could be  
703 associated with mining activities carried out in the area since the beginning of  
704 cal BP 14 (AD 1936).

705 At present, the suboxic conditions inside the Bays result from the influence of adjacent  
706 water masses with low oxygen contents related to the oxygen minimum zone (OMZ)  
707 (Fig. 2). These suboxic conditions are centered at ~250 m outside the Bays and keep  
708 low oxygen concentrations below 40 m within the Bays. Oceanographic time series  
709 indicate that transition times develop in short periods due to changes in the directions  
710 and intensities of the winds along the coast, which favors upwelling and thus the entry  
711 of water with low oxygen content to the Bays with a strong seasonality  
712 ([http://www.ceazamet.cl/index.php?pag=mod\\_estacion&e\\_cod=BTG](http://www.ceazamet.cl/index.php?pag=mod_estacion&e_cod=BTG)). Additionally,  
713 oceanic variability along the western coast of South America is influenced by  
714 equatorial Kelvin waves on a variety of timescales, from intra-seasonal (Shaffer et  
715 al., 1997) and seasonal (Pizarro et al., 2002; Ramos et al., 2006) to inter-annual  
716 (Pizarro et al., 2002; Ramos et al., 2008). Coastal-trapped Kelvin waves originating  
717 from the equator can propagate along the coast, modifying the stability of the regional  
718 current system and the pycnocline, and triggering extra-tropical Rossby waves  
719 (Pizarro et al., 2002; Ramos et al., 2006; 2008). This oceanographic feature will

720 changes the oxygen content within the bays with major impacts on redox-sensitive  
721 elements in surface sediments; thus, the increased frequency and intensity of this  
722 variability would result in a mean effect which is observed as a gradual change in  
723 metal contents over time.

724

#### 725 **5.4. Climatic interpretations**

726 The present-day climate of the semi-arid region of Chile is largely influenced by the  
727 position of the Southeast Pacific Subtropical Anticyclone (SPSA) and latitudinal  
728 displacements of the Southern Westerly Winds (SWW). The dynamic of these large-  
729 scale atmospheric systems, from seasonal to decadal timescales, controls the amount of  
730 precipitation that reaches this region. Because the semi-arid region of Chile represents  
731 the northernmost area under the influence of the SWW, precipitation is relatively scarce  
732 and restricted to the austral winter months when SPSA and SWW shift northwards,  
733 bringing precipitation fronts to the semiarid coast and inland (Montecinos and Aceituno,  
734 2003; Quintana and Aceituno, 2012).

735 According to modern climatology, paleoenvironmental records from the semiarid region  
736 have mostly been interpreted to reflect past variability in the intensity and latitudinal  
737 position of the SWW (Veit et al., 1996; Hebbeln et al., 2002; Lamy et al., 1999;  
738 Maldonado and Villagrán, 2002), controlled by the temperature gradient of the ocean's  
739 surface (Lamy et al., 2010), sun variability and orbital forcing (Varma et al., 2012;  
740 Koffman et al., 2014). Thus, at mid-latitudes of the southern hemisphere, early  
741 Holocene has been described as a warm period with summer-like conditions, due to  
742 reduced westerlies in the northern margin associated with a reduced sea surface  
743 temperature gradient between the tropical and subtropical Pacific (Lamy et al., 2010);  
744 period that can be extended until cal BP ~8600–5000 (Kaiser et al., 2008; Ortega et al.,  
745 2012, Maldonado et al., 2016). In particular, pollen records from the southern coastal  
746 areas of Coquimbo (32°S) point to prevailing wet conditions before cal BP 8700, which  
747 brought the expansion of swamp forests areas along the coast followed by a lengthy arid  
748 phase until Cal BP 6200 (Maldonado and Rozas, 2008; Maldonado and Villagrán,  
749 2006). This scenario occurred concomitantly with reduced rainfalls and intense coastal  
750 humidity associated to coastal fogs that frequently occur during the spring by a  
751 strengthening of the SE Pacific Subtropical Anticiclone (Vargas et al., 2006; Garreaud  
752 et al 2008; Ortega et al., 2012). This matches the driest conditions along the entire  
753 record, detected in the first portion of our pollen reconstruction from core BGGC5 in

754 the Guanaqueros Bay (cal BP 8200 – 7600), still suggesting drier but less intense  
755 conditions until cal BP ~5500. This is represented by relatively low values of the Pollen  
756 Moisture Index (Fig. 9). The enhancement of regional precipitation has been observed  
757 after this date in pollen records in the northern margin of SWW (Jenny et al., 2003;  
758 Maldonado and Villagrán, 2006). These findings are also consistent with our Al and Pb  
759 records, elements that are usually considered to be indicators of continental particles  
760 that enter marine waters by fluvial or aerial means (Calvert and Pedersen, 2007; Govin  
761 et al., 2012; Ohnemus and Lam, 2015; Saito et al., 1992; Xu et al., 2015). The trends are  
762 similar to the pollen record, i.e., a gradual rise over time, more clearly from  
763 cal BP ~5000, suggesting enhanced humid conditions during recent periods (Fig.9).  
764 This is also supported by grain size and K/Ca and Fe ratios, known to be indicators of  
765 the changes in terrigenous inputs off the coasts in northern-central Chile (Kaiser et al.,  
766 2008). Such increments over the last ~5000 years point to higher continental inputs  
767 most probably caused by frequent or heavier rainfall events over time, which at present  
768 are an important source of sands and K in the northern Chilean margin. K/Ca and Fe  
769 distributions point to a mean trend towards more humid conditions, consistent with  
770 pollen records at a regional scale (Maldonado & Villagrán 2006); these suggest more  
771 humid conditions from cal BP 5000 with the highest values since cal BP 1700.  
772 Furthermore, a trend towards increasing precipitations is also consistent with the  
773 occurrence of alluvial episodes since cal BP 8600 (Ortega et al., 2012), which following  
774 an increasing trend from the mid-Holocene toward recent times (Orthegea et al., 2019).  
775 The synchronicity of our records between highest productivity and dry conditions that  
776 peak prior to ~cal BP 6500 highlights the role of the SPSA as an important driver of  
777 paleoproductivity changes in the coast of semi-arid Chile which prevailed during the  
778 early portion of the Holocene to the mid-Holocene (considered as cal BP ~6000). The  
779 prevalence of ENSO cold periods between 6700 – 7500 years ago (Carré et al., 2014) or  
780 El Niño weak periods between ~4500 and 8000 ka (Rein et al., 2005) are described for  
781 the Peru margin, which is consistent with our records and points to more favorable  
782 conditions for upwelling strengthening. After this period, a consistent pattern of  
783 increasing humidity and continental discharge over the last ~6000 – 5000 years is  
784 suggested by our pollen and trace element records. The driver of this long-term paleo-  
785 climatic trend seems to be associated with a weak SPSA and a northern position of the  
786 SWW, leading El Niño-like conditions. However, lower ENSO variability has been  
787 reported at cal BP 4000 – 6000 (Koutavas and Joanides, 2012). Others point to cal BP



788 4500 (Carré et al., 2014), which does not match our records. The conditions reported by  
789 these authors resemble cold periods similar to La Niña-like conditions favorable for  
790 upwelling and productivity enhancement, and drier conditions than those recorded  
791 during former periods. Therefore, the subsequent weakening in paleo-productivity  
792 proxies in our records after cal BP 6500 is not consistent with this scenario. By contrast,  
793 a small rise in diatom abundance and opal between cal BP 4500 and 1700, along with  
794 the buildup of Ni, Cd and Ca concentrations (Fig. 8), and small increments in organic  
795 carbon flux and Cd/U (Fig. 5, 6) suggest higher organic flux and productivity but lower  
796 than what was previously observed during cal BP 6500. The slight rise in productivity  
797 indicates a weak upwelling that could be explained by a higher frequency of warm  
798 events when the modern ENSO regime was established between cal BP ~3000 – 4000  
799 (Carré et al., 2014). In this case, Fe increments could play a role in nutrient inputs for  
800 phytoplankton. This has been documented to provide a boost in the primary productivity  
801 discussed in the sedimentary records of the northern Chilean margin (Dezileau et al.,  
802 2004). In our cores, a short-term rise in Fe concentrations is observed between cal BP  
803 ~4000 – 3300 at the Guanaqueros core, whereas persistent high values are recorded in  
804 the Tongoy core between cal BP 6500 – 7800. Both of these rises match periods with  
805 relatively high primary productivity based on diatoms and opal distributions (Figs. 6,  
806 8b), which supports the role of Fe as a driver of coastal productivity in the past.  
807 Additionally, it indicates that an enhanced productivity not only depends on the  
808 upwelling's strength but on the availability of nutrients, since this area shows permanent  
809 upwelling. In this sense, in periods before cal BP 6500 productivity seems to be  
810 controlled mostly by upwelling in more stable climatic conditions, after which local  
811 nutrient inputs play a very important role in the development of primary productivity.  
812 In sum, our records show a regular rise in humid conditions concurrently with a  
813 declining productivity trend over time after Cal BP 6500. Relevant changes in  
814 oceanographic conditions were observed after cal BP 1700, when oxygenation  
815 conditions changed drastically at the bottoms, but no such intense change in  
816 productivity was observed. Studies of coastal upwelling on the central Peruvian and  
817 south central Chilean coasts (12 – 36 °S) reveal that present-day wet/dry variability  
818 associated with El Niño Southern Oscillation have a strong impact on bottom ocean  
819 oxygenation (Escribano et al., 2004; Gutiérrez et al., 2008; Sellanes et al., 2007). In this  
820 regard, OMZs are expected to be less intense during warm El Niño phases and vice  
821 versa. This connection has been observed by recent studies, as warm events in the

822 Tropical Pacific tend to be associated with low productivity and weak OMZ in the  
823 Peruvian coast (Salvatteci et al., 2014)

824 In this case, warm events in the Eastern Pacific could have reduced the ocean's  
825 productivity and the organic fluxes resulting from primary productivity, leading to a  
826 reduction in oxygen consumption during the diagenesis of organic matter. In the light of  
827 these mechanisms, our results suggest more El Niño-like conditions during the latter  
828 part of the Holocene –as has been documented for the SE Pacific (Koutavas et al., 2006,  
829 Carré et al., 2014)– in agreement with pollen moisture index records and metals  
830 described above. According to our records, low oxygen conditions are revealed by  
831 higher Mo, Re and U buildup –and sulfidic conditions when Cd is also higher. On the  
832 contrary, higher oxygenation should reduce their accumulation in sediments. Thus,  
833 more frequent El Niño events during the latter part of the Holocene should be consistent  
834 with a long-term increase in precipitations revealed by the pollen and trace elements  
835 data. This is consistent with productivity records which showed a small enhancement in  
836 the last 200 years observed from organic carbon flux, TOC (%), and opal  
837 reconstruction; furthermore, slight rises in Pb and Fe were observed, suggesting higher  
838 continental inputs during this period. This is also consistent with an increase in human  
839 activities in the area, particularly intense mining activities and changes in land use that  
840 have promoted soil erosion. However, our evidence is still weak to sustain centennial  
841 time scale records since our observations are based on few data from surface sediments.  
842 This is also less consistent with diatom abundance which was low due to the few  
843 records analyzed, in part explaining the inconsistencies between the rise in organic flux  
844 and low diatom abundance. Otherwise, this could be explained by the fact that during  
845 the El Niño conditions, the normal dominance of diatoms is replaced by smaller size  
846 phytoplankton, resulting in a relevant contribution to overall primary production (Iriarte  
847 et al., 2000; Rutlland and Montecino, 2002; Escribano et al., 2004). Other observations  
848 for northern Chile suggest the intensification of coastal southerly winds as the enhanced  
849 solar heat over the land results in the strengthening of upwelling during warmer ENSO  
850 periods. Moreover, this results in a net increase in primary production (Vargas et al.,  
851 2007). If coming along with Fe inputs to the bay system, this could explain productivity  
852 records during present times. In addition, it provides an important clue to the current  
853 climate scenario in which our records seem to be matching.

## 854 **6. Conclusions**

855 Our results suggest that ocean circulation in our study sites seems to impact both  
856 places differently, leaving more variable grain compositions and higher TOC contents  
857 in the Guanaqueros Bay (core BGGC5) than in the Tongoy Bay (core BTGC8), with  
858 the latter increasingly impacted by terrigenous inputs due to the flow of several creeks  
859 during major flooding events. Both core records sustain a reduction of organic flux to  
860 the bottoms after cal BP ~6500 and into present times. This is probably due to more  
861 humid conditions over time, also sustained by ascending ratios of K/Ca, which can be  
862 assumed as a result of higher ENSO variability over time. Some Fe concentrations  
863 increments at cal BP >6500, around cal BP 3000 – 4000, and in the past 200 years are  
864 consistent with increments in primary productivity proxies suggesting their relevance  
865 as nutrient element. However, it also point to inputs by eolian and fluvial transport that  
866 seem to become relevant after cal BP 6500 to boost phytoplankton during less intense  
867 upwelling periods. The last assumption considers that more humid conditions were  
868 favored by a less intense SPSA. Thus, the record of continental proxies suggests a  
869 long-term increase in precipitation, consistent with previous reconstructions in central  
870 Chile. The most distinctive changes were observed after cal BP 6500, when an overall  
871 expansion of the coastal vegetation occurred as a result of a progressive increase in  
872 precipitation and river runoffs, expanding the grain size of the sediments and the  
873 higher concentrations of elements with an important continental source (Al, Fe, K and  
874 Pb).

875 Differences in redox conditions in our records are consistent with less intense upwelling  
876 and more frequent oxygenations of the bottoms occurring during the El Niño-like  
877 conditions. This could be reconstructed from EFs variations and sensitive redox metal  
878 accumulation in the sediments. A clear decreasing trend in Me/Al ratios was apparent,  
879 suggesting less oxygen at the bottoms before the beginning of recent times (cal BP  
880 ~1700), followed by a rapid change to a more oxygenated environment. Oxygen content  
881 in bottom waters was the most relevant factor in sediment metal enrichment above  
882 crustal abundance (highest EFs of U, Mo and Re), since the accumulation of organic  
883 carbon and estimated sedimentation rates were low in the area. Therefore, organic  
884 carbon burial rate is less relevant than oxygen content for the accumulation of metals  
885 within the sediments.

886 Our results suggest that maximum suboxia-anoxia occurred at cal BP ~6500, when  
887 peak U, Mo and Re were recorded, probably in a sulfidic environment.

888 The nutrient-type elements follow a similar trend: lower values at present and higher  
889 ratios around cal BP 6500 (Ca, Ni, P and Cd). Their distribution is consistent with  
890 diatom and opal distributions, showing their dependence on primary productivity and  
891 organic carbon burial rates. If the kinetics reaction is working at low rates for these  
892 elements, they should be highly influenced during oxygenation periods, something that  
893 seems to have been operating at higher frequencies suggesting more frequent El Niño-  
894 like conditions.

895 Increased regional precipitations have been commonly interpreted by a northward shift  
896 of the Southern Westerly Winds belts, yet the higher frequency of El Niño events  
897 more likely introduced a high variability of humidity after cal BP 5000. Thus, the  
898 apparent rise of oxygen conditions at bottoms could have been the result of this  
899 oceanographic feature, which introduced a more oxygenated water mass to the shelf  
900 and bays, temporarily changing the redox conditions in surface sediments and  
901 affecting the sensitive elements to potential redox changes in the environment.  
902 Additionally, this also impacted the accumulation of organic matter due to an  
903 intensification of its remineralization, showing a decreasing trend in the buildup of  
904 nutrient type elements and organic carbon burial rates towards the present.

905 Finally, our results suggest that the geochemistry and sedimentary properties of  
906 coastal shelf environments in north-central Chile have changed considerably during  
907 the Holocene period, suggesting two relevant changes in redox conditions at  
908 cal BP 6500, pointing to a change to a less reducing environment which becomes very  
909 strong after cal BP 2000. In particular, decreasing trends in primary productivity after  
910 cal BP 6500 and increasing trends in oxygenation highlight the sensitivity of these  
911 environments to regional climate changes at different timescales. Future changes are  
912 therefore likely to be expected in the ongoing scenario of environmental changes at  
913 unprecedented rates.

914

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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  $^{14}\text{C}$ -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	1 $\sigma$ error	Conventional Age BP	1 $\sigma$ 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

Table 2. Reservoir age (DR) estimation considering the  $^{210}\text{Pb}$  age determined with the CRS model (McCaffrey and Thomson, 1980) at a selected depth sections of the core, compared with  $^{14}\text{C}$  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) <sup>a</sup>	Age years BP <sup>b</sup>	$^{14}\text{C}$ age Marine 13.14	$^{14}\text{C}$ age BP from foram.	DR
BGGC5	10.5	1828	122	499±24	940±25	441±35
BTCG8	5.5	1908	42	448±23	890±15	442±27

<sup>a</sup>Anno Domini

<sup>b</sup>Before 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).

<b>Element</b>	<b>Metal/Al x 10<sup>3</sup></b>	<b>s</b>
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 diatoms abundance (valves g<sup>-1</sup>).

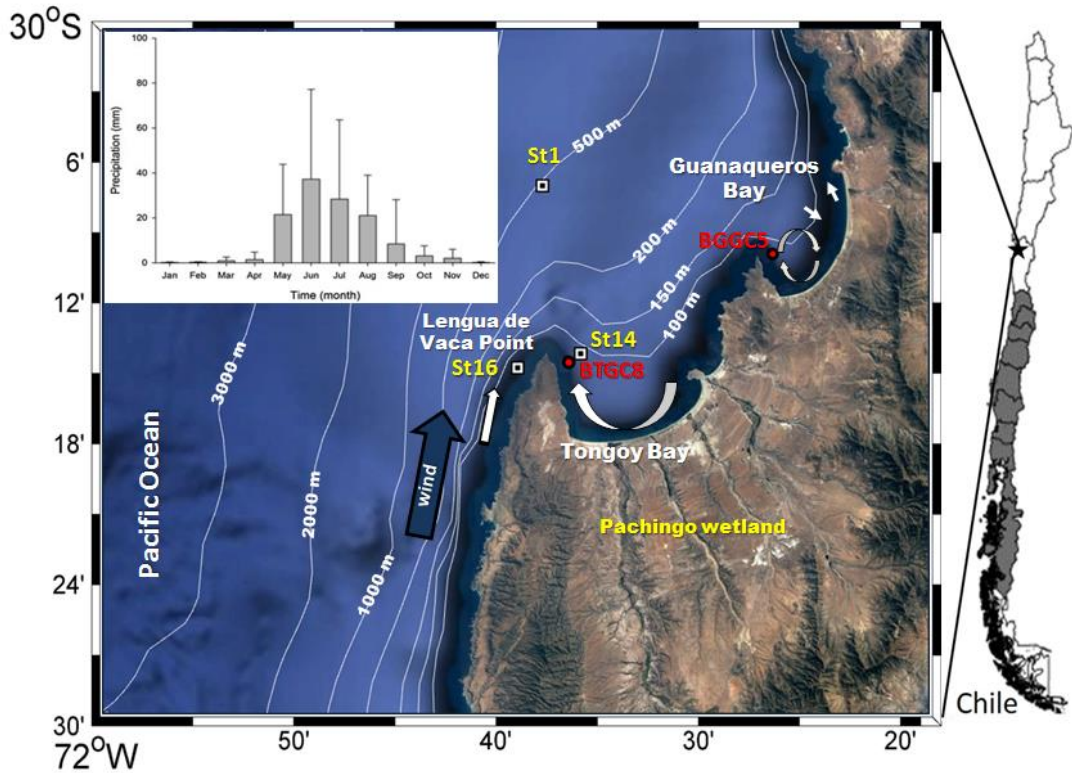
Age range (cal BP)	Diatoms (x10 <sup>6</sup> ) (min-max)	Opal (g m <sup>-2</sup> yr <sup>-1</sup> ) (min-max)	EF <sub>U</sub>	EF <sub>Mo</sub>	EF <sub>Re</sub>	EF <sub>Fe</sub>	EF <sub>Ba</sub>	EF <sub>Cd</sub>	EF <sub>Ni</sub>	EF <sub>Cu</sub>	EF <sub>P</sub>
-65 – 130	0.5 – 4.9	3 – 40	2.6 $\pm 0.7$	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 <sup>a</sup> $\pm 1.3$	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 $\pm 0.8$

<sup>a</sup>Mean EF<sub>Cu</sub> 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.

<b>BGGC5</b>																
	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	<b>0.90</b>	<b>0.83</b>	-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	<b>0.92</b>	0.30	-0.60	0.18	0.32
Mn					1.00	<b>0.94</b>	-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	<b>0.81</b>	0.03	-0.70	0.11	-0.40	0.23	0.36	-0.37	-0.21
Ni							1.00	-0.51	0.49	<b>0.91</b>	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	<b>0.88</b>	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	<b>0.92</b>	-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
<b>BTGC8</b>																
	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	<b>0.80</b>	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	<b>0.83</b>	-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



1370 Figure 1. Study area showing the position of sampling stations. Sediment cores were  
1371 retrieved from Guanaqueros Bay (BGGC5) and from Tongoy Bay (BTGC8) at water  
1372 depths of 89 and 85 m, respectively. Information of dissolved oxygen (DO) in the water  
1373 column at ST1 and ST16 and of suspended organic particles collected at ST14 sampling  
1374 sites was gathered in a previous project (INNOVA 07CN13 IXM-150). Monthly  
1375 precipitation in mm (bars) (means  $\pm$  SD; Montecinos et al., 2016). Schematic  
1376 representation of the bays circulation (white arrows) and wind direction is indicated  
1377 (blue arrow) obtained from Valle-Levinson and Moraga-Opazo (2006) and Moraga-  
1378 Opazo et al. (2011).



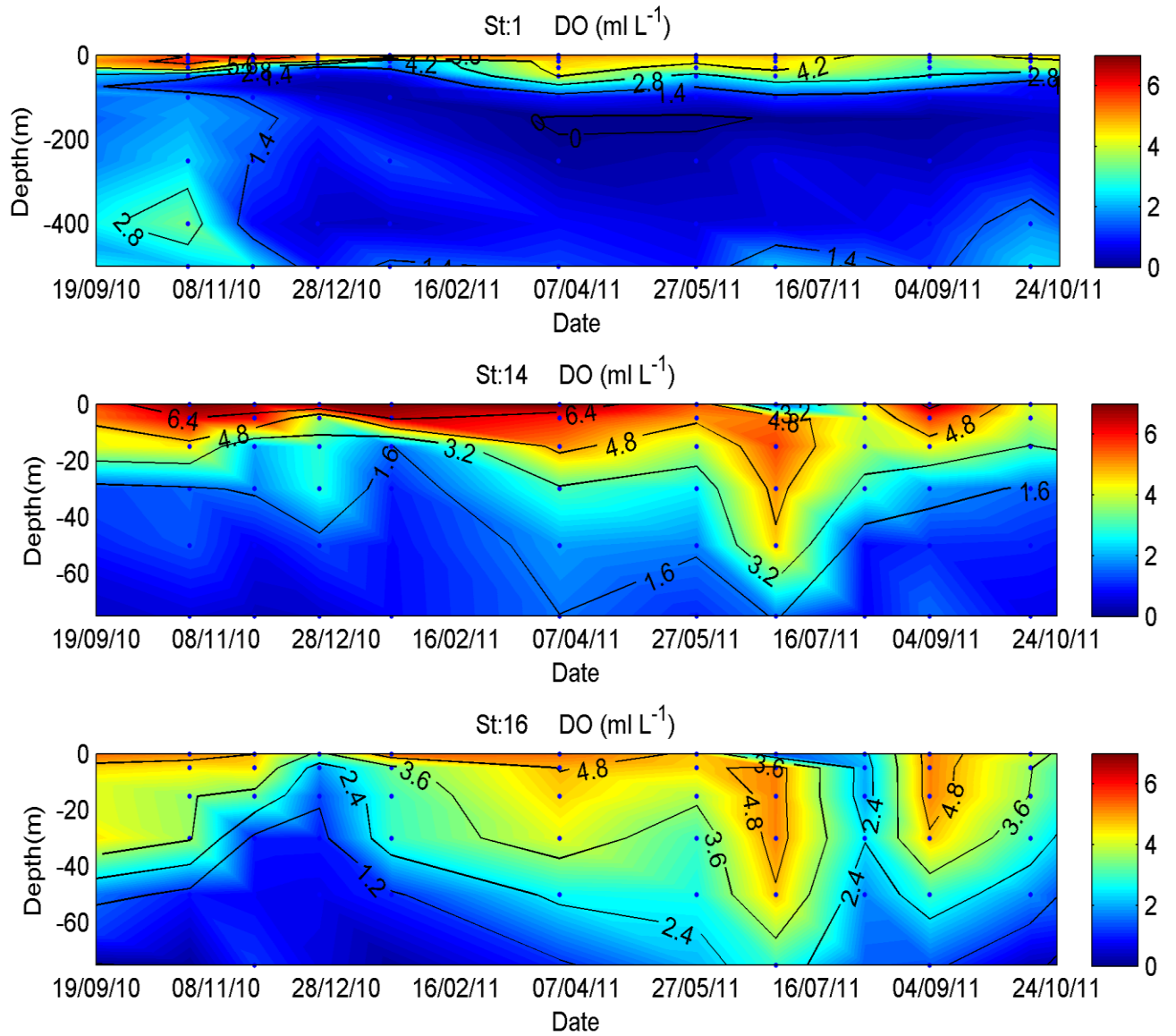


Figure 2. Dissolved Oxygen (DO) time series in the water column measured between October 2010 and January 2011, at stations St1, St14 and St16 off Tongoy Bay, Coquimbo (30°S).

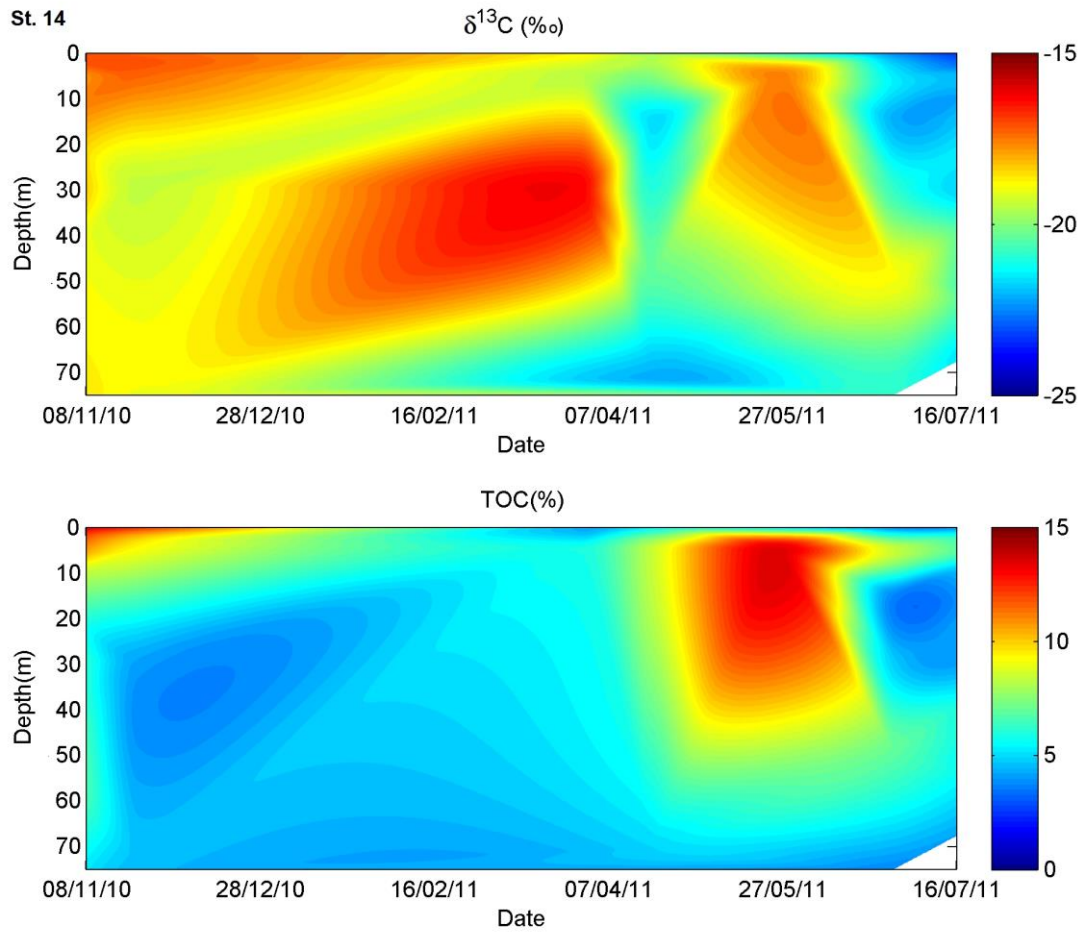


Figure 3. Suspended particulate matter composition (TOC % and  $\delta^{13}\text{C}_{\text{org}}$ ) measured in the water column between October 2010 and October 2011, at station St14, Tongoy Bay, Coquimbo (30°S).

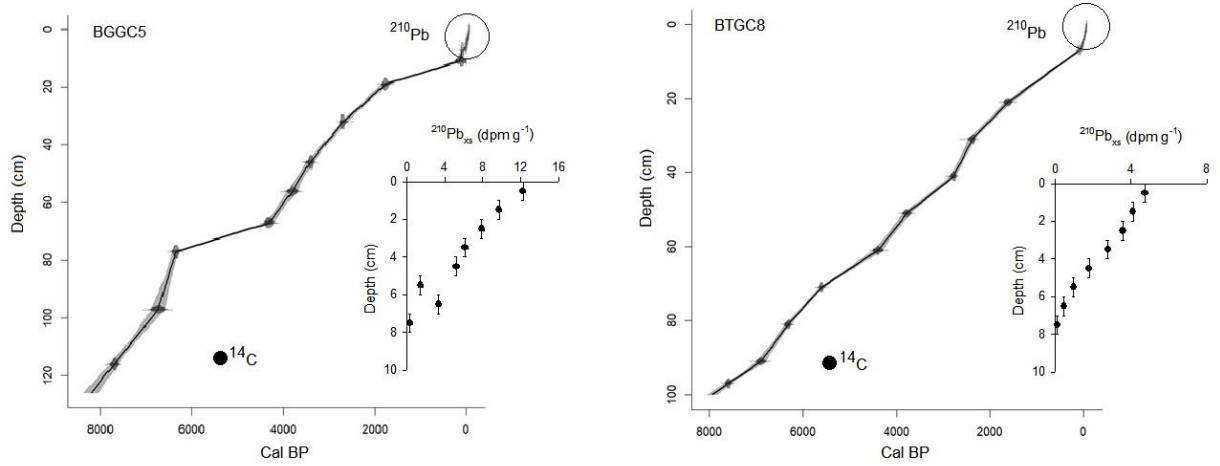
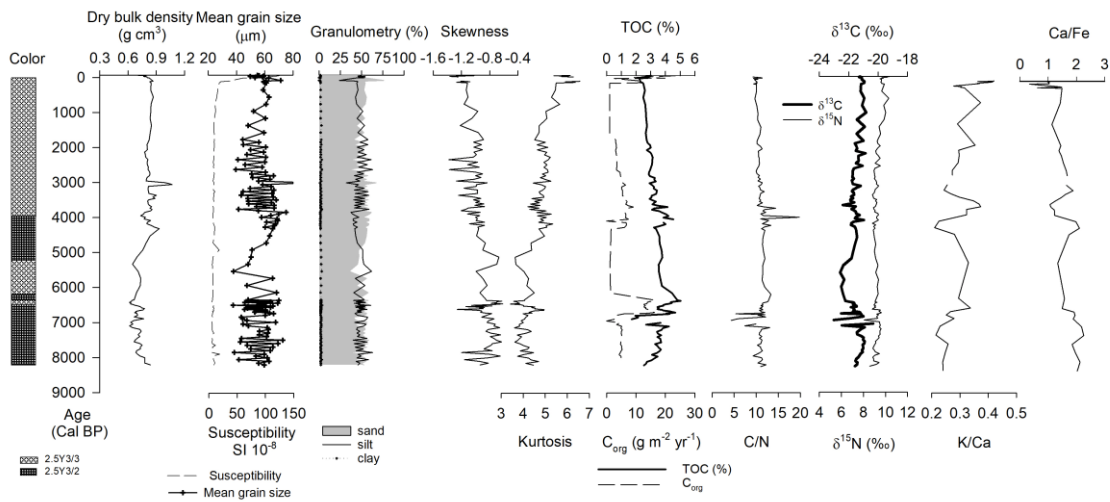


Figure 4. Age model based on  $^{14}\text{C}$ AMS and  $^{210}\text{Pb}$  measurements. The time scale was obtained according to the best fit of curves of  $^{210}\text{Pb}_{\text{xs}}$  and  $^{14}\text{C}$  points using CLAM 2.2 software and Marine curve  $^{13}\text{C}$  (Reimer et al., 2013).

a) BGGC5



b) BTGC8

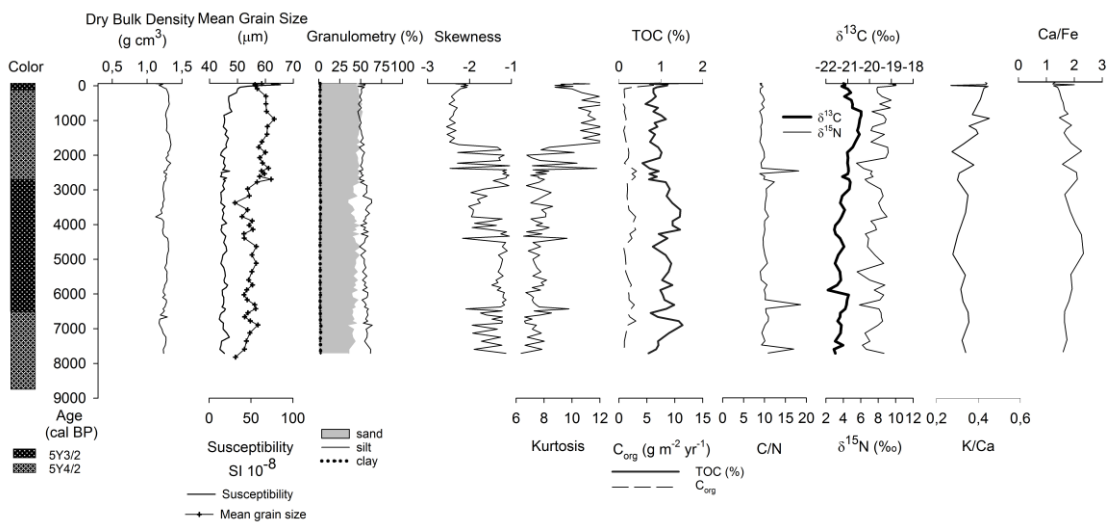


Figure 5. Characterization of sediment cores retrieved from (a) Guanaqueros Bay (BGGC5) and (b) Tongoy Bay (BTGC8). Where is shown the huge (Munsell chart scale) in depth, dry bulk density, mean grain size, granulometry (% sand, silt and clay), statistical parameters (skewness, kurtosis), organic components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

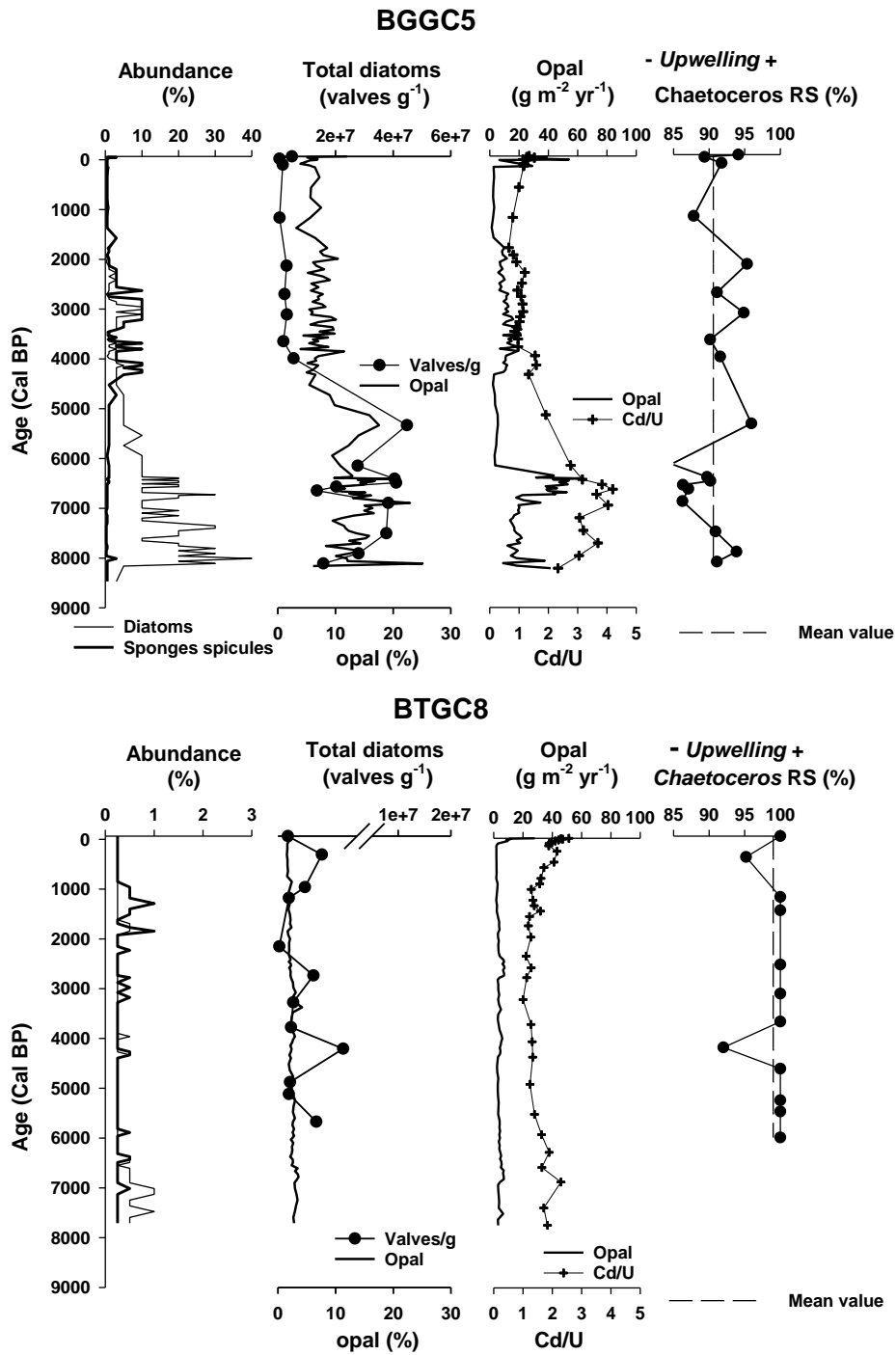


Figure 6. Diatom and sponge spicules relative abundances, total diatom counts (valves g<sup>-1</sup>) and opal (%), opal accumulation (g m<sup>-2</sup> yr<sup>-1</sup>) and Cd/U ratio, and downcore variations in *Ch.* resting spores percentages as proxy of upwelling intensity in BGGC5 and BTGC8 cores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

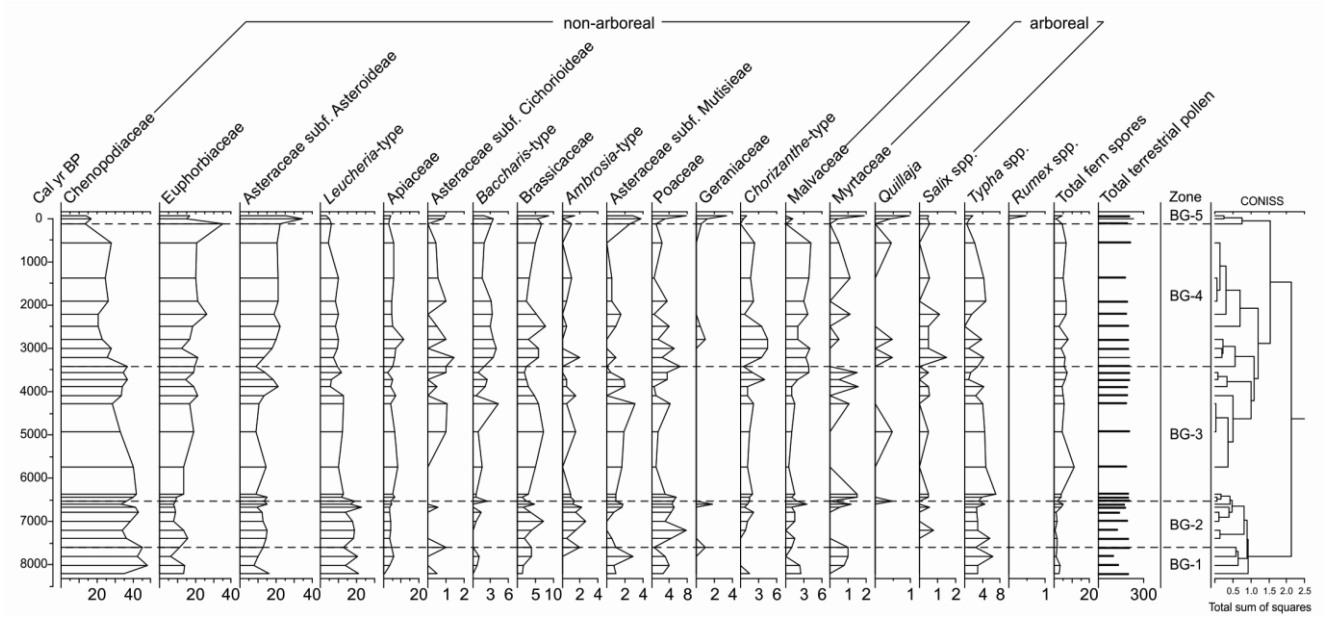
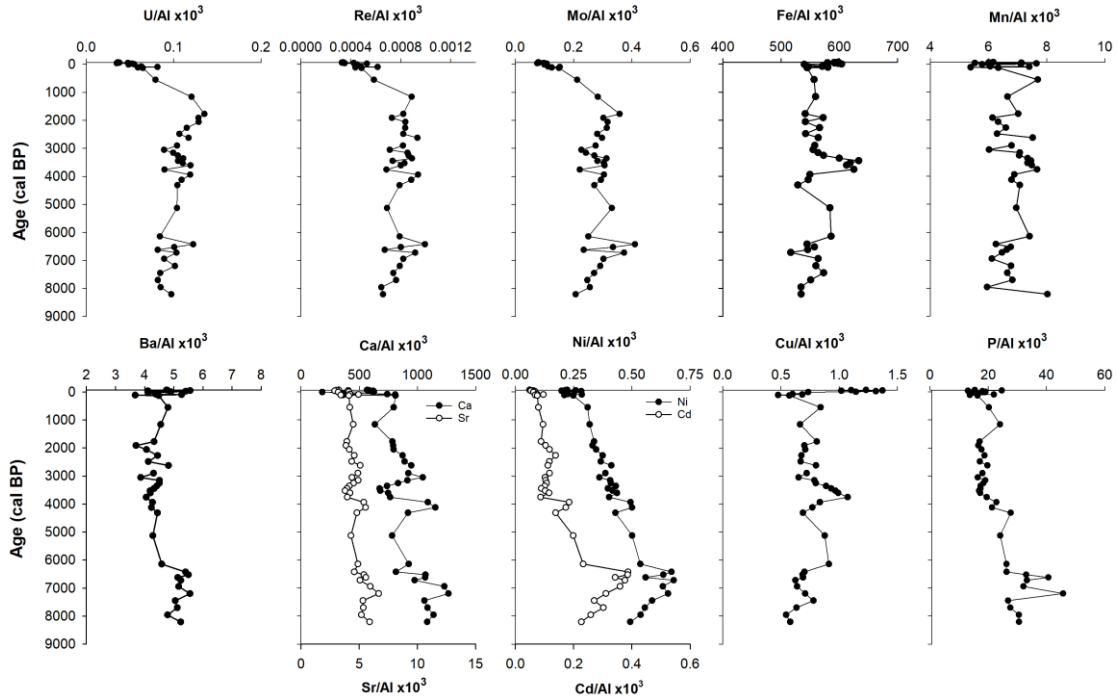


Figure 7. Pollen record in BGGC5 core.

a) BGGC5



b) BTGC8

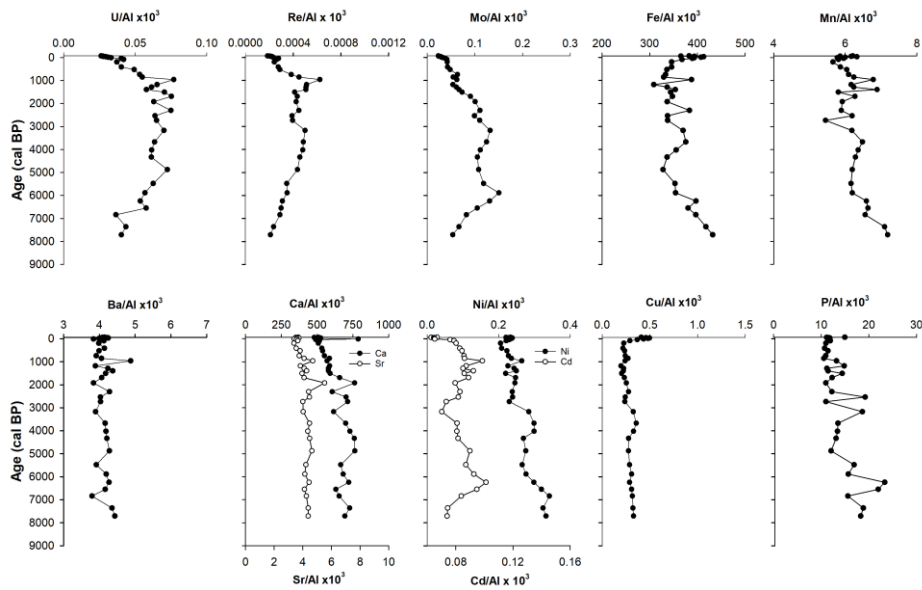


Figure 8. Downcore trace element variations on: (a) Guanaqueros Bay (BGGC5) and (b) Tongoy Bay (BTGC8), off Coquimbo (30°S).

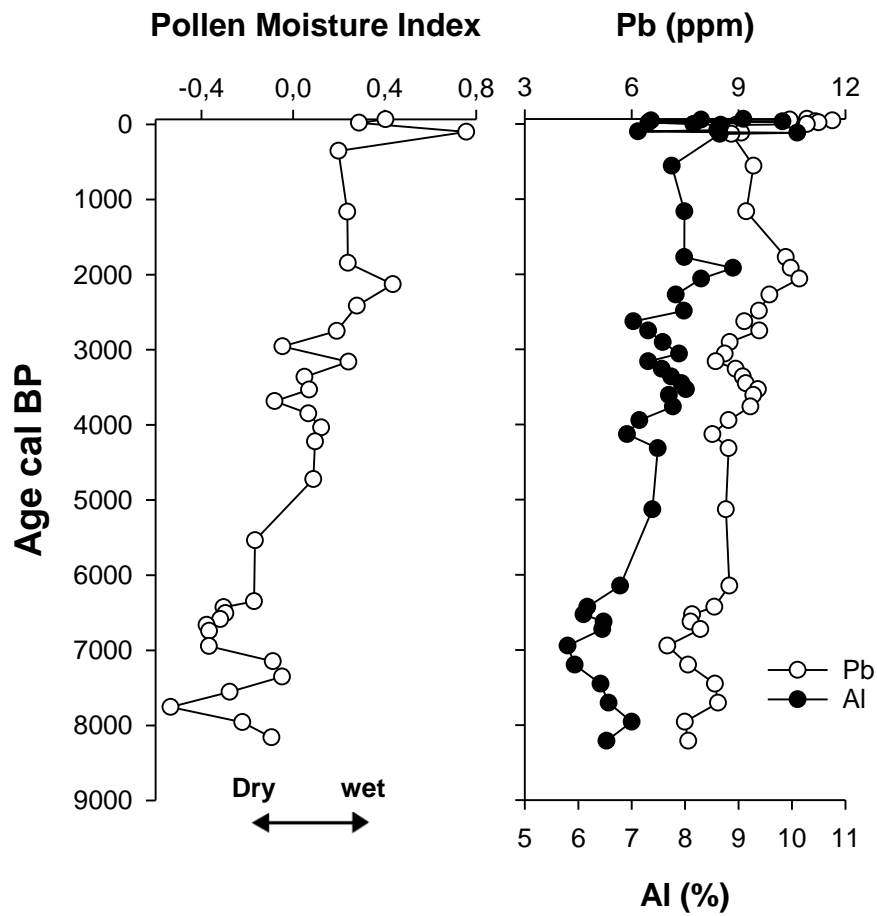


Figure 9. Pollen Moisture Index defined as the normalized ratio between Euphorbiaceae (wet coastal shrub land) and Chenopodiaceae (arid scrubland). Positive (negative) values for this index indicate the relative expansion (reduction) of coastal vegetation under wetter (drier) conditions. Pb and Al distribution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