



26     **ABSTRACT**

27     We analyze the geochemistry of Rano Aroi mire record (Easter Island) using bulk  
28 peat composition (C, N, S) and stable isotopes ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ,  $\delta^{34}\text{S}$ ) and major, minor and  
29 trace elemental composition obtained by ICP-AES (Al, Ti, Zr, Sc, V, Y, Fe, Mn, Th, Ba,  
30 Ca, Mg and Sr). Peat geochemistry and the pollen record are used to reconstruct the  
31 environmental changes during the last 70 kyr BP. Principal component analysis on ICP-  
32 AES data revealed that three main components account for the chemical signatures of  
33 the peat. The first component, characterized by lithogenic elements (combined signal of  
34 V, Al, Sc, Y, Cr, Cd, Ti, Zr and Cu), evidences long-term changes in the basal fluxes of  
35 mineral material into the mire. This component, in combination with stable isotopes and  
36 pollen data suggests a link between soil erosion and vegetation cover changes in the  
37 Rano Aroi watershed. The second component is identified by the signal of Fe, Mn, Th,  
38 Ba, Zr and Ti, and is indicative of strong runoff events during enhanced precipitation  
39 periods. The third component (tied mainly to Ca, Sr and Mg) reflects a strong peat  
40 oxidation event that occurred during an arid period with more frequent droughts,  
41 sometime between 39 and 31 kyr BP. Correlation coefficients and a multiple regression  
42 model (PCR analysis) between peat organic chemistry and the principal components of  
43 ICP-AES analysis were calculated. Isotope chemistry of the peat organic matter further  
44 contributes to define Rano Aroi environmental history:  $\delta^{13}\text{C}$  data corroborates a  
45 vegetation shift documented by the palynological record from  $\text{C}_4$  to  $\text{C}_3$  between 55 and  
46 45 kyr cal BP; the  $\delta^{15}\text{N}$  record identifies periods of changes in mire productivity and  
47 denitrification processes, while the  $\delta^{34}\text{S}$  peat signature indicates a marine origin of S  
48 and significant diagenetic cycling. The geochemical and environmental evolution of  
49 Rano Aroi mire is coherent with the regional climatic variability and suggests that  
50 climate was the main forcing in mire evolution during the last 70 kyr BP. The coupling

51 of geochemical and biological proxies improves our ability to decipher depositional  
52 processes in tropical and subtropical peatlands and to use these sequences for  
53 paleoenvironmental and paleoclimate reconstructions.

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## 78 **1. INTRODUCTION**

79 Peatlands are paleoenvironmental archives capable of registering atmospheric,  
80 hydrological and ecological changes in the past (Jackson and Charman, 2010).  
81 Researchers have traditionally studied peat accumulation and decay dynamics (Clymo,  
82 1984), and attempted to understand the contribution of these organic soils to the global  
83 carbon cycle (Gorham, 1991), as well as to reconstruct paleoecological changes using  
84 macrofossils, pollen (Barber et al., 2003; Birks and Birks, 2006) and charcoal remains  
85 (Whitlock and Larsen, 2001). In more recent times, inorganic geochemical proxies from  
86 peat sequences have increasingly been used to obtain high-resolution climatic and  
87 environmental reconstructions. For example, wind regime variability has been inferred  
88 from changes in the chemical concentrations of dust particles trapped in the peat  
89 (Kylander et al., 2005; Martínez Cortizas et al., 2002, 2007a; Shotyk, 1996; Shotyk et  
90 al. 2001;), and changes in vegetation cover and wet to dry transitions from geochemical  
91 compositions of mires (Kylander et al., 2013; Muller et al., 2008).

92 A complementary biogeochemical approach to mire studies focuses on the  
93 characterization of the peat organic matter, through the determination of isotopic  
94 signatures ( $\delta D$ ,  $\delta^{13}C$ ,  $\delta^{15}N$ ,  $\delta^{18}O$ ,  $\delta^{34}S$ ) or its molecular composition (Buurman et al.,  
95 2006; Hong et al., 2001; Kaal et al., 2007; Loisel et al., 2010; Schellekens et al., 2011;  
96 Tillman et al., 2010;). The  $\delta^{13}C$  has been applied on bulk peat or isolated compounds as  
97 a tool to explore the origin of the carbon ( $C_3$ ,  $C_4$  plants or aquatic origin) because  
98 photosynthesis fractionation signatures are commonly preserved (Meyers, 2003).  
99 Moreover,  $\delta^{13}C$  together with  $\delta D$  and  $\delta^{18}O$  can track hydrologic changes such as wet to  
100 dry transitions or changes in the precipitation-evaporation balance (Hong et al., 2001).

101 Stable isotopes can also be valuable indicators of organic matter origin and decay ( $\delta^{13}\text{C}$ ,  
102  $\delta^{15}\text{N}$ ) (Aucour et al., 1999; Talbot and Johannessen, 1992), and redox changes ( $\delta^{15}\text{N}$ ,  
103  $\delta^{34}\text{S}$ ) (Jędrysek and Skrzypek, 2005; Nývák et al., 1999 and Talbot and Johannessen,  
104 1992).

105 The majority of mire studies focus mainly on climate or environmental  
106 reconstructions using peat cores from ombrotrophic boreal and temperate mires of the  
107 Northern Hemisphere (Chambers and Charman, 2004; Clymo et al., 1984; Gorham and  
108 Janssens, 2005; Jackson and Charman, 2010; Shotyk, 1996). While receiving increasing  
109 attention during the last decades, peatlands in the Southern Hemisphere remain  
110 substantially less explored. Few studies have attempted to reconstruct environmental  
111 changes on tropical and subtropical mires (Dommain et al., 2011; Kylander et al., 2007;  
112 Muller et al., 2008; Page et al., 2010; Weiss et al., 2002). This study presents organic  
113 and inorganic biogeochemical data, including pollen analysis from the oldest Southern  
114 Hemisphere peat deposit studied to date (Easter Island), to track the environmental  
115 changes of the last ~70 kyr BP. While numerous studies have revealed the  
116 environmental changes on Easter Island using lacustrine sediments (Azizi and Flenley,  
117 2008; Cañellas-Boltà et al., 2012; Cañellas-Boltà et al., 2013; Flenley and King, 1984;  
118 Flenley et al., 1991; Horrocks et al., 2012a, Horrocks et al., 2012b; Mann et al., 2008;  
119 Sáez et al., 2009), fewer works have been carried out on the Easter Island peat  
120 sequences. The Rano Aroi peat record has been analyzed using pollen (Flenley et al.,  
121 1991, Peteet et al., 2003) and XRF core scanner and stable isotope data (Margalef et al.,  
122 2013). Margalef et al. (2013) combined facies and macrofossil descriptions, bulk peat  
123 total carbon (TC), and total nitrogen (TN) and  $\delta^{13}\text{C}$  data with XRF core scanner data  
124 (Ca, Fe and Ti elements) data to reconstruct environmental history of the site at a  
125 millennial time scale. However, to fully reveal the complex interactions and processes

126 controlling the geochemical signatures at Rano Aroi such as soil dust, flood events,  
127 droughts and redox changes, more comprehensive geochemical analyses are needed.  
128 Therefore, in this paper, we analyzed bulk peat samples to obtain absolute  
129 concentrations of sixteen elements (Al, Fe, Ti, Ca, Mg, Sr, Y, Zr, Ba, Sc, V, Cr, Mn, Cu,  
130 Cd, Th). This geochemical dataset is complemented with TC, TN, TS content and the  
131 isotopic composition ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ,  $\delta^{34}\text{S}$ ) of the peat and pollen data. Only a few previous  
132 studies are based on such a broad dataset including chemical and biological data from  
133 the same peat record (Muller, 2006). This comprehensive approach combining inorganic  
134 and organic geochemistry reinforced by pollen analysis allows us to establish links  
135 between vegetation changes, mineral inputs and biogeochemical processes within peat  
136 as a response to autogenic and external forcing. The results improve our understanding  
137 of tropical and subtropical peat dynamics and the environmental and climatic history of  
138 Easter Island since MIS 4 (~70 cal kyr BP).

## 139 **2. STUDY SITE**

140 Easter island (27° 07'S, 109° 22'W), known as Rapa Nui in the local language, is a  
141 small volcanic island situated on the edge of South Pacific Convergence Zone (SPCZ),  
142 Intertropical Convergence Zone (ITCZ) and South Pacific Anticyclone (SPA), the three  
143 main features that determine the South Pacific climatic configuration (Fig. 1). The  
144 climate is subtropical, with monthly average temperatures between 18 (August) and  
145 24°C (February) and an extremely variable annual precipitation ranging from 500 to  
146 1800 mm.

147 There are three permanent water bodies on the island, two lakes (Rano Raraku and  
148 Rano Kao) and a mire (Rano Aroi) formed in a volcanic crater. The smooth slopes of the  
149 inner part of the volcanic cone constitute the catchment (15.82 ha). The crater itself is  
150 near the highest summit of the island, Mauna Terevaka (511 m a.s.l), composed by

151 highly porfiric olivinic tholeiite, hawaiite, and basaltic lava flows (Baker 1974,  
152 González-Ferran et al., 2004) and covered by andosols. The surface vegetation of the  
153 mire is characterized by *Scirpus californicus*, *Polygonum acuminatum*, *Asplenium*  
154 *polydon* var. *squamulosum*, *Vittaria elongata* and *Cyclosorus interruptus*, while the  
155 surrounding area is covered by grasslands and a small eucalyptus forest planted during  
156 the 1960s (Rull et al., 2010a). Rano Aroi is a minerotrophic fen, fed by rainfall and  
157 groundwater; hydrogeological and isotopic studies confirm that the system represents a  
158 perched spring connected to the main island aquifer (Herrera and Custodio, 2008;  
159 Margalef et al., 2013).

### 160 **3. METHODOLOGY**

161 In March 2006, a 14 m deep peat core (ARO 06 01) was collected in eleven sections  
162 from the central part of the mire with a UWITEC<sup>®</sup> corer, a modular percussion piston  
163 coring system. The first two meters of the sequence were not kept to avoid potential  
164 anthropic remobilization as described in the central part of the mire previously (Flenley  
165 and King, 1984; Flenley et al., 1991). The core sections were sealed, packed,  
166 transported to the laboratory and stored at 4 °C until sampling. Core sections were split  
167 longitudinally, imaged and the peat facies were described.

#### 168 **3.1 Geochemical analyses**

##### 169 *On the core sections*

170 The core was sampled every 5 cm for total carbon, nitrogen and sulphur (TC, TN,  
171 TS) and stable isotope ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ,  $\delta^{34}\text{S}$ ) analyses. The 218 samples were dried at 60°C  
172 over 48 hours, frozen with liquid nitrogen and ground in a ring mill. Total C, TN and  
173 their stable isotopes ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) were analyzed using a Finnigan delta Plus EA-CF-  
174 IRMS spectrometer, and  $\delta^{34}\text{S}$  measurements were performed using a Finnigan MAT  
175 CHN-IRMS Finnigan DeltaPlus XP (precision of 0.2‰ and 0.3‰ respectively). Both

176 instruments are located at the Serveis Científico-Tècnics (SCT) of the Universitat de  
177 Barcelona (UB).

178 To investigate the mineralogy and size of the mineral grains present in the levels with  
179 higher inorganic content (Margalef et al., 2013), three representative samples were  
180 selected; a level rich in Ca and Fe (4.8 m depth), and two silty levels with high values of  
181 Ti and Fe (8.3 and 10.55 m depth). These samples were dried and mineral grains were  
182 density separated in purified water. The grains were carefully attached to stubs using a  
183 conductive bioadhesive tape. A morphological description using a FEI ESEM-EDS in  
184 the *low vacuum mode* (around 0.5 torr) and *high vacuum mode* ( $< 10^{-4}$  torr) was made  
185 on the selected samples in the SCT-UB. Stubs were carbon-coated before being  
186 observed in *high vacuum mode*. Secondary and backscattered electron images and  
187 Energy-dispersive X-ray spectroscopy (EDS) were used systematically to characterize  
188 the mineral grains.

189 All geochemical sample preparation for Inductively Coupled Plasma-Atomic  
190 Emission Spectrometry (ICP-AES) was performed under clean laboratory conditions  
191 using acid cleaned labware. Each 250 mg of sample was digested using a mixture of  
192  $\text{HNO}_3/\text{HBF}_4$  as described by Krachler et al. (2002). The microwave program included a  
193 40 minute several-stage ramp to 200°C where samples were held for 20 minutes. The  
194 remaining solution was then transferred to Savillex vessels and evaporated on a hotplate  
195 at 50°C. Thereafter  $\text{H}_2\text{O}_2$  was added and allowed to react for half a day. Samples were  
196 then sonicated and evaporated at 50°C. This was followed by a 2 day closed vessel  
197 digestion at 90°C with  $\text{HNO}_3$ . After sonication and evaporation at 50°C an additional  
198 cycle of  $\text{H}_2\text{O}_2$  was made. Samples were once again evaporated and then taken up in 1%  
199  $\text{HNO}_3$  for analysis. The samples were analyzed for elemental concentrations using a  
200 Varian Vista AX ICP-AES at the Department of Geological Sciences, Stockholm



201 University, Sweden.

202 A suite of twenty elements were acquired including Al, Ba, Ca, Cd, Cr, Cu, K, Fe, Li,  
203 Mg, Mn, Na, Sc, Sr, Ti, Th, V, Y, Zn and Zr. To date, there is no certified reference  
204 material for peat that offers a wide range of elemental data. There is however a peat  
205 reference material NIMT/UOE/FM/001 that was tested and analyzed by Yafa et al.  
206 (2004). The analytical performance was thus assessed using this material as well as  
207 NIST SRM-2711a Montana Soil and NIST SRM-2586 Trace Elements in Soil. Five  
208 procedural replicates were made of each reference material. For NIMT/UOE/FM/001,  
209 our peat reference material, most elemental recoveries were high, ranging from 99% to  
210 119%. Cadmium however had even higher recoveries (128%) while low recoveries  
211 were recorded for Ti (63%). For the other two reference materials, both soils, recoveries  
212 were much lower. For NIST SRM-2586 recoveries range from 70-91% for Al, Ba, Ca,  
213 Mg, Sc, Sr and Y and 53-66% for Cd, C, Fe, Mn, V and Zn. Again Ti had exceptionally  
214 low recoveries (44%) while Th had concentrations double the recommended values. For  
215 NIST SRM 2711a recoveries ranged between 64-84% with the exception of Ti (42%)  
216 and Th again had concentrations double the recommended values. Replicates of the  
217 reference materials were generally within 20% or better of each other. Procedural blanks  
218 for were below 3% of the average sample concentrations for the majority of the  
219 elements. Slightly higher blanks were found for Al and Mn (<5%), Ba and Cr (<12%)  
220 and Zr (<32%).

221 While the original intention was to make a total digestion of the peat samples, the  
222 reference materials suggest that this may not be the case. Although the recoveries for the  
223 peat reference material are high, the recoveries for the reference soils are much lower.  
224 Nonetheless, in a paleoenvironmental context the relative changes of the elements can  
225 still reveal past changes. For this reason all elements, even those with lower (Ti, for

226 example) or higher recoveries (Th, for example) or potentially high blanks (Zr, for  
227 example) were included in our interpretation. Some elements were removed from the  
228 dataset because of low concentrations in the peat samples (Li and K), contamination  
229 during sampling (Zn) or because the concentrations were close to the analytical  
230 detection limits (Na). In the case of Th and Zr particularly, both elements present high  
231 communalities in the PCA, suggesting that their signals are not just analytical noise, and  
232 their behavior has a coherent paleoenvironmental significance.

### 233 *Rock and soil analyses*

234 Determination of the elemental composition of rocks and surface soil samples from  
235 Rano Aroi and Rano Raraku craters and other parts of the island was performed on 16  
236 samples. All soil samples (5 samples, see supporting information) were obtained from  
237 the surroundings of Rano Raraku crater, while rock samples (11 samples, see supporting  
238 information) were collected around the Rano Aroi, Rano Raraku craters and other parts  
239 of the island. All samples were ground in a ring mill and dried at 60°C for 24 h. Each  
240 0.1 g of sample was then digested using a mixture of 2.5 ml HNO<sub>3</sub>+ 5 ml HF + 2.5 ml  
241 HClO<sub>4</sub> in Teflon tubes at 135°C for 12 h. Finally, 1 ml of HNO<sub>3</sub> and 5 ml of purified  
242 water were added to dilute the sample for analysis. The analyses were carried out with a  
243 high-resolution ICP-MS (HR-ICP-MS), Element XR Thermo Scientific located at the  
244 LabGEOTOP service of the Institute of Earth Sciences Jaume Almera-CSIC  
245 (Barcelona). Twenty-three elements were measured. Results are provided in the  
246 supporting information (Table 1).

### 247 **3.2 Pollen analysis**

248 A subset of 25 samples (3-4 g of wet peat) equally distributed along the record were  
249 extracted from Rano Aroi peat record and processed for pollen analysis using standard  
250 laboratory procedures by Rull et al. (2010b), which include sieving, digestions with

251 KOH, HCl and HF, and acetolysis. As an exotic marker, one spore tablet of *Lycopodium*  
252 (Batch. No. 177745, Lund University, Sweden) per gram was added to each sample  
253 before processing. The slides were mounted in silicone oil (not permanents) to be  
254 counted under optical microscope using x40 and x60 objectives mounted on a ZEISS®  
255 microscope with x10 ocular (ZEISS® Axioplan, ZEISS® AxioStar plus, ZEISS® Axio  
256 Scope A1 and ZEISS® Axio Lab. A1).

257 Pollen counting was carried out until at least a total of 200 pollen grains, excluding  
258 spores and aquatic and semi-aquatic plants except for samples with very low pollen  
259 concentration. Fern spores are presented as a sum of all spore types. The pollen results  
260 are presented in percentages excluding spores and aquatic plants. Spores and aquatic  
261 plants are represented as a percentage respect the pollen sum. The pollen zonation was  
262 performed using psimpoll 4.26 (Bennett, 2002) by optimal splitting (Bennett, 1996).  
263 The counting of pollen grains and spores was carried out by comparisons with reference  
264 books and atlases for pollen as Hesse et al. (2009); Heusser (1971); Hoeve and  
265 Hendriksse 1998; Tryon and Lugardon (1991); Reille (1992); and the online pollen and  
266 spore atlas APSA (2007).

267 Many questions about former flora ecology and distribution of Easter Island remain  
268 unsolved, despite that several works have tried to reconstruct vegetation changes from  
269 pollen analyses on lake sediments (Azizi and Flenley, 2008; Butler et al., 2004; Dumont  
270 et al., 1998; Cañellas-Boltà et al., 2013; Flenley and King, 1984; Flenley et al., 1991;  
271 Gossen, 2007) or from macrofossil remains on lacustrine records or archeological sites  
272 (Cañellas-Boltà et al., 2012; Dumont et al., 1998; Mann et al., 2008; Orliac and Orliac,  
273 1998; Orliac, 2000; Peteet et al., 2003). As an important number of flora species are  
274 now extinct from the island and the native Easter Island flora has been intensely  
275 perturbed (Dubois et al., 2013; Rull et al., 2010a), it is not possible to reconstruct

276 paleoenvironments from pollen analyses using local modern analogues. For this reason,  
277 a comparison with other islands (Juan Fernández, Hawaii, Rapa Iti) is useful in order to  
278 reconstruct vegetation changes observed in the pollen analysis (see section 4.4 for more  
279 information).

### 280 **3.3 Age-depth model**

281 An age model was built from 19 radiocarbon AMS dates measured from pollen  
282 concentrates in the Poznan Radiocarbon Laboratory (Poland) (see Margalef et al., 2013  
283 for full details). Pollen enrichment process followed a classical treatment (Faegri &  
284 Iversen, 1989; Moore et al., 1991) modified by Rull et al. (2010b). The AMS ages were  
285 calibrated using CALIB 6.02 software, and the INTCAL 98 curve (Reimer et al., 2004)  
286 and CalPal (Danzeglocke, 2008) for samples older than 20,000 radiocarbon yr BP. The  
287 age model was built by simple linear interpolation between the radiocarbon dates as  
288 described in Margalef et al. (2013).

### 289 **3.4 Statistical analysis**

290 Principal component analysis (PCA) was applied in order to reduce the large ICP-  
291 AES dataset to a smaller number of variables. These principal components (PC) could  
292 then be interpreted in terms of geochemical and environmental processes. SPSS version  
293 22 statistical software was used to perform the PCA including varimax rotation over the  
294 previously logged (ln) and standardized ICP-AES dataset (Al, Ba, Ca, Cd, Cr, Cu, Fe,  
295 Mg, Mn, Sc, Sr, Ti, V, Y and Zr).

296 Correlation coefficients between stable isotopes, organic matter elemental  
297 composition and principal components scores were then calculated. To complement the  
298 integrative study of all proxies, multiple regression models (stepwise regression mode)  
299 using the scores of the extracted principal components (PCR analysis) were obtained for  
300 TC, TN, TS,  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ,  $\delta^{34}\text{S}$  using SPSS version 22 software.

## 301 4. RESULTS

### 302 4.1 Peat facies and age model

303 The Rano Aroi core (ARO 06 01) is made up of radicle peat as defined by Succow  
304 and Joosten, (2001), consisting of fine roots (diameter < 1mm) with <10% comprising  
305 larger remains, from Cyperaceae, Poaceae and Polygonaceae (Margalef et al., 2013).  
306 Based on plant type and remain size, geochemistry and degree of peat decomposition, 4  
307 peat facies have been defined (Margalef et al. 2013). **Facies A** (reddish peat) consists of  
308 coarse plant remains, with very high C/N (43-111) ratios and  $\delta^{13}\text{C}$  values between -21‰  
309 and -26‰. **Facies B** (granulated muddy peat) is a light brown peat with low mineral  
310 content, made up of coarse to mid-sized organic fragments, mostly roots and rootlets.  
311 This facies is characterized by high C/N (41-85) ratios and  $\delta^{13}\text{C}$  values ranging from -  
312 14‰ to -26‰. **Facies C** (organic mud) is found as centimeter thick layers interbedding  
313 Facies B. These layers have high mineral contents, high TN (1.08-1.76%) and relatively  
314 light  $\delta^{13}\text{C}$  values (-14‰ to -22‰). **Facies D** consists of dark and fine-grained peat and  
315 is highly decomposed (pictures of the described facies can be found in Margalef et al.,  
316 2013).

317 The age model revealed large changes in peat accumulation rates and the occurrence  
318 of a long hiatus in the sequence (see supporting information, Table 2). (1) The  
319 bottommost part of the sequence (8.75 m -13.9 m) was accumulated prior to 55 kyr BP  
320 and, in consequence, it is beyond the limits of radiocarbon dating. Because the  
321 accumulation rate between 55 and circa 40 cal kyr BP (3.7–8.72 m) is almost constant  
322 and the peat facies are the same until the base of the sequence, we make the initial  
323 assumption that accumulation rates were also relatively constant for the bottommost  
324 part of the sequence. Consequently, this part of the sequence has been dated by  
325 extrapolation using the accumulation rate of the upper section (3.7–8.72 m) of the core

326 sequence (see Margalef et al., 2013 for further details). (2) A sharp unconformity occurs  
327 at 4.25 m (39 cal kyr BP), where highly oxidized peat (Facies D) is overlaid by much  
328 less decomposed peat (Facies A). Dating results, the peat facies D and the  
329 biogeochemistry data suggest that during a certain period of time after 39 kyr cal BP the  
330 wetland underwent a long-term drought (with likely associated peat decomposition and  
331 loss) resulting in a subaerial exposure (Margalef et al., 2013). (3) Above the clear  
332 unconformity, our age model shows extraordinary low peat accumulation rates, and  
333 16,000 years are represented by 104 cm (between 3.27 m and 4.31 m, see table 2 from  
334 supporting information).

## 335 ***4.2 Geochemistry on peat cores***

### 336 *4.2.1 Organic geochemistry*

337 Total C concentrations are highly variable but without any specific trend, ranging  
338 from 40% to 70% (Fig. 2).  $\delta^{13}\text{C}$  shows constant values around -14 ‰ between 14 m and  
339 9 m shifting gradually to values around -26‰ from 9 m to 6 m, and staying around  
340 these more negative values for the uppermost five meters of the sequence. In addition to  
341 the long-term trend, the  $\delta^{13}\text{C}$  curve also shows high-frequency changes (dips) and  
342 significantly lower values for Facies C between 11 m and 6 m (Fig 2 and 6; Margalef et  
343 al., 2013). Total N contents range between +0.5% and +1.75%. C/N ratios are between  
344 33 and 111.  $\delta^{15}\text{N}$  values oscillate between -2.03 and +8.87‰. Peat sections with the  
345 heaviest isotopic composition, above +8‰, are found at 12.4 m and 10.7 m depth and  
346 above 6 ‰ at 3.40 m depth (Fig. 2).

347 Total S values are low (less than 1%) and show a decreasing trend from the bottom  
348 of the core to 6 m depth, followed by a slight increase from this depth to the top of the  
349 core. On the other hand, the  $\delta^{34}\text{S}$  record can be divided into two main sections: below 8  
350 m the average value is  $+18.04 \pm 0.88$  ‰ and above this depth it is  $+19.33 \pm 1.60$ ‰. The

351 shift between the two sections occurs at 7.7-7.2 m. (Fig. 2).

#### 352 *4.2.1 Mineral grain found on peat cores geochemistry*

353 Most of the mineral grains observed under electronic microscope showed evidence of  
354 transport or advanced weathering. At 8.3 m and 10.55 m depth small grains (30-500  
355  $\mu\text{m}$ ) were rutile, ilmenite and quartz, while the bigger grains ( $\geq 500 \mu\text{m}$ ) were composed  
356 of inosilicates (pyroxenes), plagioclases and quartz. The sample from 4.8 m depth  
357 showed smaller grains (10-20  $\mu\text{m}$ ) composed of iron, magnesium and aluminum oxides,  
358 quartz and organic compounds bound to Ca, Br and Mg. Some of these organic  
359 compounds were large up to 200  $\mu\text{m}$ .

#### 360 *4.3 Rock and soil analyses*

361 The elemental composition of selected rock and soil samples from the Rano Aroi  
362 watershed and other areas of the island was analyzed to investigate the origin of the  
363 inorganic fraction arriving to the mire. The ICP-MS analyses show that basalt, hawaiiite  
364 and tholeiite rocks are especially rich in  $\text{Fe}_2\text{O}_3$  (16-22%) and  $\text{TiO}$  (2.5-4.4%) (see  
365 supporting information Table 1) as documented by previous petrographic studies (Baker  
366 et al., 1974). The other major components are found in the following concentrations:  
367  $\text{Al}_2\text{O}_3$ : 16-22%,  $\text{MnO}$ : 0.3-0.5 % and  $\text{CaO}$ : 3.3-9%. Minor and trace elements showed  
368 the following ranges: Sc: 24-39  $\mu\text{g g}^{-1}$ , V: 170-500  $\mu\text{g g}^{-1}$ , Co: 40-163  $\mu\text{g g}^{-1}$ , Cr: 1-60  
369  $\mu\text{g g}^{-1}$ , Ni: 2.2-29.5  $\mu\text{g g}^{-1}$ , Cu: 16-112  $\mu\text{g g}^{-1}$ , Zn: 110-185  $\mu\text{g g}^{-1}$ , Sr: 76-280  $\mu\text{g g}^{-1}$ , Y:  
370 40-85  $\mu\text{g g}^{-1}$ , Zr: 276-497  $\mu\text{g g}^{-1}$ , Ba: 107-202  $\mu\text{g g}^{-1}$ , Th: 2.4-5.2  $\mu\text{g g}^{-1}$  (Table 1 of  
371 supporting information). The rock samples around Rano Aroi are particularly enriched  
372 in Ti, Al, Sc, V, Cr, Ni, Cu and depleted in Y, Zr, and Ba, when compared to samples  
373 from other areas of the island. On the other hand, the Easter Island andosols sampled for  
374 this study were especially enriched in Al, Cr, Y, Zr and depleted in Mg, Ca, Sc, V, Co,  
375 Ni, Cu, Zn, and Sr compared to rock samples.

376 **4.4 Peat ICP-AES data and PCA**

377 A PCA was performed using the suite of selected elements from the bulk peat  
378 analysis. Three components explaining 85% of the matrix variance were identified. The  
379 absolute concentration variations for elements representative of these PCs are shown in  
380 Fig. 2. The results of the PCA are presented in terms of the factor loading of each  
381 element in the extracted PCs by showing the fractionation of the communalities (i.e., the  
382 proportion of the variation of each variable explained by each PC) (Fig. 3) and by the  
383 depth records of the PCs factor scores (Fig. 4). The first component (PC1, 40.8% of the  
384 variance) is tied mainly to V, Al, Sc, Cr, Cd and Y (with positive loadings between 0.71  
385 and 0.93, Al shown in Fig. 2) and to a lesser extent to Ti, Zr, Cu and Mn (Ti and Zr  
386 shown in Fig. 2, communalities shown in Fig. 3). The second component (PC2, 23.1%  
387 of the variance) is characterized by large positive loadings (between 0.69 and 0.93) of  
388 Th, Fe, Mn and Ba (Fe shown in Fig. 2) with significant contributions from Ti, Zr and  
389 Sr (Ti and Zr shown in Fig. 2, communalities shown in Fig. 3). And the third component  
390 (PC3, 21.3% of the variance) is characterized by large positive loadings (between 0.87  
391 and 0.94) of Mg, Ca and Sr and moderate ones of Cu and Ba (Ca shown in Fig. 2,  
392 communalities shown in Fig. 3).

393 PC1 shows high scores in the older section of the peat record (>55.5 kyr BP; 9  
394 m). A clear shift from higher to lower values is observed between 55.5 and 41.5 cal kyr  
395 BP (9-5.16 m), followed by a rapid increase until 31cal kyr BP (3.76 m) and  
396 stabilization thereafter (Fig. 4). PC2 variability shows a peaky pattern. From the bottom  
397 of the core to 6.1 m PC2 scores present peaks at 10.6 m, 10.01 m, 8.7 m and 7.73 m.  
398 From 6.1 m to 4.8 m values become high and quite stable, then start to decline gradually  
399 until 2.96 m (Fig. 4). Two prominent peaks occur in this last interval (4.8-2.96 m): at  
400 4.05 m and at 3.45 m, the latter being the maximum of the entire record. At the upper



401 part of the sequence, from 2.96 m to 2.40 m PC2 shows a progressive increase. The PC3  
402 scores do not show a clear trend at the bottom of the sequence. From 10.49 m to 8.15 m  
403 there is a clear declining trend, and a marked see-saw pattern until 5.79 m. Scores  
404 rapidly increase from 5.79 m to 5.41 m, defining a broad peak up to 4.25 m. From 4.25  
405 m to 3.16 m PC3 scores decline irregularly, but a clear peak stands out in this interval at  
406 3.86 m. The uppermost part of the sequence presents an increasing trend, (Fig. 4).

#### 407 **4.5 Pollen record**

408 In this work, only the most abundant taxa: Poaceae, *Arecaceae* , Asteraceae,  
409 *Coprosma* and Cyperaceae pollen types together with fern spores sum have been used.  
410 The following taxons are included as “others”: *Triumfetta*, *Acalypha*, *Trema*  
411 (*Ulmaceae*), *Pinus*, *Macaranga*, *Sapinus*, *Plantago* plus indeterminate and unknown  
412 pollen types.

413 Asteraceae (arbustive types) are present in Hawaii, Rapa and Juan Fernandez Island  
414 although nowadays these are not found on Easter Island, (Brown, 1935; Flenley et al.,  
415 1991, Zizka, 1991). In Hawaii these small trees form scarce-forested landscapes in  
416 between true forest and bare lava flows (Flenley, 1991). Native and endemic Poaceae  
417 are mainly present in meadows and open landscapes. Some biogeographical and  
418 ecological constraints of the known species are given in Table 3 of the supporting  
419 Information. Based on the endocarp of fossil seeds, an endemic species of *Arecaceae*  
420 tree has been proposed: *Paschalococos disperta* (Dransfield et al., 1984). This endemic  
421 species is extremely similar to *Jubaea chilensis* (Dransfield et al., 1984, Flenley et al.,  
422 1991) and is presented as an emblematic case of extinction. *Coprosma* is a genus of  
423 flowering plants (small trees) present on many Pacific sites (New Zealand, Hawaii,  
424 Borneo, Rapa, Juan Fernández). It is nowadays completely extinct from Easter Island  
425 but found in ancient lake and peat sediments (Flenley, 1991; Horrocks et al., 2013).

426 Cyperaceae is another abundant pollen type predominantly representing taxa found in  
427 moist and waterlogged parts of grassland meadows or peatlands (supporting  
428 information, Table 3). Finally, fern spores also constitute an important part of Rano Aroi  
429 pollen record, in contrast to the low dominance of fern spores observed in Rano Raraku  
430 lake sediment cores (Cañellas-Boltà et al., 2013; Flenley et al., 1991).

431 Two significant zones were obtained (Fig.5) over the complete pollen dataset:

432 Zone I (13.9-7.48 m depth, 71-48.3 kyr BP) is characterized by the dominance of  
433 Poaceae (70-90%) with an important contribution of *Arecaceae*, *Coprosma* pollen and  
434 fern spores.

435 Zone II (7.48-2.35 m depth, 48.3-8.5 kyr BP) is defined by an increase of Asteraceae  
436 at the expense of Poaceae and *Arecaceae* pollen. The appearance of a higher amount of  
437 Cyperaceae, probably from the mire itself, is remarkable. The elevated number of fern  
438 spores indicates that these might have been growing in the vicinity of the mire.  
439 Cyperaceae as other aquatic and semiaquatic plants can be useful to determine local  
440 conditions because they grow specifically in moist and flooded areas. Because of the  
441 strong local signal they are not included in the sum to perform percentages.

442 This Zone has been divided in three subzones in the following manner:

443 -Zone IIa (7.48-5.8 m depth, 48.3-43 kyr BP): this subzone is characterized by  
444 very high percentages of Asteraceae pollen (up to 70.5% at 6.15 m depth) while Poaceae  
445 pollen percentage remains lower than in Zone I. Fern spores start to be more abundant,  
446 together with Cyperaceae that depict increasing percentages from bottom to top of the  
447 zone.

448 -Zone IIb: (5.8-3.4 m depth, 43-20.6 kyr BP). The subzone consists of a pollen  
449 assemblage with a lower percentage of Asteraceae and a higher contribution of  
450 *Arecaceae*, Cyperaceae and fern spores. This subzone also contains a hiatus at 4.25 m

451 (see section 4.1).

452 - Zone IIc (3.4-2.35 m depth, 20.6-8.5 kyr BP) is characterized by a considerable  
453 reduction in Asteraceae and the dominance of Cyperaceae and ferns.

#### 454 ***4.6 Correlation between inorganic and organic peat chemistry***

455 In the PCA described above we did not include the variables that characterize the  
456 chemical nature of the organic matter, because our main objective was to determine the  
457 chemical nature of the minerogenic fraction in the peat. The next step is to correlate the  
458 first three PC with the chemical composition of the peat organic matter (TC, TN, TS,  
459 and the isotopic composition) to look for covariance and to investigate the underlying  
460 processes controlling the changes in peat inorganic and organic matter, and how  
461 external and internal (i.e. postdepositional) processes have affected both the inorganic  
462 and the organic chemistry. The largest correlation values (Table 1) were found between  
463 PC1 scores and TN, C/N ratios,  $\delta^{15}\text{N}$ ,  $\delta^{13}\text{C}$  ( $r$  are 0.66, -0.62, 0.65 and 0.59  
464 respectively). Lower, but significant correlations ( $p < 0.01$ ) were found between PC1 and  
465 TS ( $r = -0.45$ ), PC2 and  $\delta^{13}\text{C}$  ( $r = -0.31$ ), and PC3 and TC, C/N ratios and TS ( $r$  are 0.30,  
466 0.32 and 0.35 respectively, Table 1).

467 This relatively weak correlation structure may indicate that peat organic chemistry  
468 was mainly controlled by factors other than those governing the observed changes in the  
469 inorganic chemistry. But it may also reflect that there was no single dominant control  
470 and that all or a combination of factors is responsible for peat geochemistry. To check  
471 the latter possibility we performed a multiple regression analysis using as input  
472 variables the three extracted principal components. Such a multiple regression models is  
473 called principal components regression, PCR (see for example Desta Fekedulegn et al.,  
474 2002). The results of the PCR models can be found in Table 2. As expected, the models  
475 show larger correlation coefficients. Nevertheless, TC and  $\delta^{34}\text{S}$  show a low ( $R = 0.35$

476 and  $R = 0.49$ , respectively) and TS only a moderate correlation ( $R = 0.66$ ) with the  
477 principal components, which may be taken as evidence that C and S cycling in Rano  
478 Aroi was not only coupled to the main processes responsible for the changes in the  
479 inorganic chemistry. On the contrary, TN, C/N ratios,  $\delta^{13}\text{C}$ , and  $\delta^{15}\text{N}$  show significantly  
480 larger correlation coefficients ( $R = 0.72\text{-}0.75$ ). For these variables PC1 has the highest  
481 regression coefficients (positive for TN,  $\delta^{13}\text{C}$ , and  $\delta^{15}\text{N}$ , and negative for the C/N ratio;  
482 Table 2) and is thus the most influential. A visual representation of the adjustment of the  
483 predictive models to the original data can be found in the Figure 1 of the supporting  
484 information. For TN and C/N ratios the similarity of the records of observed and  
485 expected values are quite remarkable, while for  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  the model basically  
486 accounts for the long-term changes but not for the short term events. For TS the fitting  
487 between real and expected values is pretty good except for the section comprised  
488 between 6.34 and 5.54.

## 489 **5. DISCUSSION**

### 490 **5.1 Factors controlling peat elemental composition**

491 The PCA performed on the Rano Aroi inorganic elemental dataset identified three  
492 main components that enable an identification of specific variables for each component  
493 (i.e., chemical elements in this case) whose behavior is similar and, thereby, likely  
494 controlled by the same process (Reimann et al., 2008).

#### 495 *5.1.1. Long-term mineral fluxes of very fine particles*

496 PC1 is characterized by large positive loadings ( $>0.7$ ) of typically lithogenic  
497 elements (V, Al, Sc, Y, Ti, Zr) and some metals (Cr, Cd, Cu). These elements are  
498 associated with very fine particulate material and this component can confidently be  
499 related to the deposition of soil dust transported by wind together with contributions  
500 from hydric erosion. Given the isolation of Easter Island, this signal would be mainly

501 dominated by fluxes from the volcanic rocks and soils of the island itself. Elements on  
502 this first component (like Al, Ti, Zr, Cr, Cu and other metals) are enriched in volcanic  
503 soils with increasing degree of pedogenesis (Martínez Cortizas et al., 2007a). Almost  
504 the same association of chemical elements (V, Al, Ti, Sc, Cu) was found by Muller et al.  
505 (2008) in the study of the composition of the Lynch's Crater, a mire in NE Australia  
506 with remarkable similarities with Rano Aroi (both mires are minerotrophic and formed  
507 within a volcanic crater). The weathering of volcanic materials leads to the distribution of  
508 this association of elements in (1) secondary minerals or organo-metallic compounds,  
509 which are poorly crystalline or in (2) primary minerals, which are very resistant to  
510 weathering. Most of these mineral phases are characterized by very fine (probably  $\leq 50$   
511  $\mu\text{m}$ ) particles, which are easily mobilized by eolian or hydric erosion. This is consistent  
512 with what is found after SEM observations on the Rano Aroi record because particle  
513 size (except for Facies C) was dominated by fine silt and clay fractions ( $<30 \mu\text{m}$ ).

514 Therefore, PC1 would track the long-term *background* fluxes of inorganic particulate  
515 material coupled to soil pedogenesis and erosion (and factors affecting both). Fine  
516 airborne dust particles are enriched in many elements compared to coarser ones  
517 (Schuetz, 1989), and these chemical processes lead to potential physical and chemical  
518 fractionation during dust transport, which seems to be more intense at short distances  
519 from the source area and attenuates during long-range transport, as the grain size of the  
520 dust decreases and homogenizes. The elemental composition reflected by PC1 is  
521 consistent with these chemical enrichment processes associated to dust input variability.

#### 522 *5.1.2 Strong runoff events and coarser detrital input*

523 The second component, PC2, is characterized by large positive loadings of Fe, Mn  
524 and Ba and moderate positive loadings of Ti and Zr. These elements are associated to  
525 coarser particles entering the mire. Non-systematic Scanning Electron Microscopy

526 (SEM) observations of the peat layers corresponding to high PC2 scores showed an  
527 abundance of sand (50-600  $\mu\text{m}$ ) and coarse silt (20-50  $\mu\text{m}$ ) particles.

528 Iron and Mn are elements which can show a strong redox behavior, tending to be  
529 depleted under anoxic conditions due to the mobility of their reduced forms and  
530 accumulated under oxidizing conditions in peatlands (Chesworth et al., 2006;  
531 Steinmann and Shotyk, 1997). Furthermore, PC2 variability (and the elemental profiles,  
532 Figure 2) does not show a long-term trend as documented for redox sensitive elements  
533 (i.e. Fe or Mn) in the Lynch's Crater record (Muller et al., 2008). However, the Rano  
534 Aroi record has rather a peaky pattern resembling an "event signal". Despite its  
535 potential mobility, Fe has been found to be immobile in certain peatlands (Muller et al.,  
536 2008; Weiss et al., 2002) and previously formed Fe oxides/hydroxides were found to be  
537 stable in lake sediments even under anoxic conditions (Gälman et al., 2009).  
538 Additionally, in most soils developed on volcanic rocks such as those in the Rano Aroi  
539 catchment, Fe is largely hosted by primary minerals and the Fe that is released during  
540 weathering accumulates as non-crystalline or poor crystalline Fe forms (i.e. ferrihydrite,  
541 Fe-organic matter associations) and secondary Fe phases (oxides and hydroxides)  
542 (García-Rodeja et al., 2007). Barium, Ti, Th and Zr have only one oxidation state and  
543 are not sensitive to redox changes, and major hosting minerals of Ba (barite, witherite),  
544 Ti (ilmenite, rutile) and Zr (zircon, baddeleyite) are highly resistant to weathering. Thus,  
545 in Rano Aroi PC2 is not likely to reflect diagenetic changes associated to changes in  
546 redox conditions. Instead, while PC1 reflects the long-term particulate terrigenous input  
547 into Rano Aroi, PC2 most likely represents strong, highly erosive runoff events capable  
548 of transporting solid particles by suspension or eventually traction to the center of the  
549 mire. Peaks in PC2 scores coincide with the presence of Facies C, organic mud, which  
550 has been interpreted as being representative of wet events and higher water table levels

551 in the mire (Margalef et al., 2013).

### 552 *5.1.3 Post-depositional enrichments*

553 PC3 is characterized by large positive loadings of Ca, Sr, and Mg. These elements  
554 are transported to the mire included in very fine soil particles (as primary minerals such  
555 as plagioclase), but because they are also highly mobile as ions, they are transported to  
556 the mire as dissolved species, too. Due to their chemical mobility, groundwater can also  
557 greatly contribute to their distribution, sometimes by diffusion from the underlying  
558 sediments as a result of the chemical dissolution of Ca-bearing minerals (Shotyk et al.,  
559 2002). And, as essential nutrients, they are also subjected to intense biocycling.

560 The most prominent feature of the PC3 record is the maximum values (Fig. 4, Fig. 6,  
561 Fig. 7) attained between ca. 42-39 kyr BP (4.25 m and 5.41 m depth) coinciding with  
562 facies D (highly decomposed peat, Margalef et al., 2013). Age model, geochemistry and  
563 the sharp discontinuity described at the uppermost limit of facies D suggest that there  
564 was a loss (i.e. erosion) of previously accumulated peat layers. This would also mean  
565 that the geochemical features exhibited below this sedimentary hiatus (between 5.41 m  
566 and 4.25 m depth) were acquired thousand years later than accumulation, as diagenetic  
567 changes caused by peat aerial exposure. Given this circumstance the most likely  
568 explanation for enrichment on facies D was an intensive use of these elements as  
569 bio nutrients by the plant community because a more intensive input of Ca, Mg and Sr  
570 as a solute form would require a wetter climate. The lowering of the mire water table,  
571 due to drought phases, is known to accelerate peat decomposition (Ise et al., 2008) and  
572 produce an enrichment of certain elements in the peat (Biester et al., 2012; Martínez  
573 Cortizas et al., 2007b). The increase of Ca –among other elements- at surface levels  
574 when peat growth stagnates has been explained by Damman et al. (1992) as an effect of  
575 the detritus cycle. At Lynch's Crater, Ca, Sr and Mg were also enriched in the more

576 decomposed peats (Muller et al. 2008). Another example of similar chemical  
577 enrichments comes from a Canadian mire composed of a 153-cm-thick layer of  
578 ombrotrophic, moderately decomposed peat overlying highly humified, minerotrophic  
579 peat: Ca was enriched 10 fold; Mg 2-3 fold; Fe (3 fold) in the highly decomposed peat  
580 (Zoltai and Johnson, 1985). In Rano Aroi the maximum concentrations for Ca, Mg and  
581 Sr in the c. 42-31 kyr BP (6-4 m depth) peat are around 5 fold, 3 fold and 2.5 fold higher  
582 than average concentrations in the peat deposited ca. 55-42 kyr BP (8-6 m).

## 583 ***5.2 Linking the organic and inorganic peat chemistry***

### 584 *5.2.1 PC1 and organic chemistry*

585 The long-term variability of PC1 is similar to the long-term evolution of the  $\delta^{13}\text{C}$   
586 record from 55.5 to 43.9 kyr BP (9-6 m, Fig. 6) where both curves have a declining  
587 trend. The  $\delta^{13}\text{C}$  trend shows a shift from values typical of  $\text{C}_4$  plant types to lighter ones  
588 (characteristic of  $\text{C}_3$  type) and suggests a change in the peat forming plant community  
589 (Margalef et al., 2013). This synchronicity between  $\delta^{13}\text{C}$  and PC1 reveals an intimate  
590 relation between soil evolution and vegetation cover (see section 5.3).

591 The TN content decreases in the upper part of the sequence (Fig. 6) suggesting a link  
592 with soil evolution (PC1) and vegetation cover ( $\delta^{13}\text{C}$ ).  $\delta^{15}\text{N}$  (Fig. 2) can provide  
593 information on organic matter origin, nitrogen fixation ( $\delta^{15}\text{N} = 0$  to  $+3$  ‰) or plant  
594 productivity, but also syndepositional processes such as denitrification ( $\delta^{15}\text{N} \geq +8$  ‰)  
595 (Handley et al., 1999; Meyers and Ishiwatari, 1993; Talbot and Johannessen, 1992). At  
596 Rano Aroi the average  $\delta^{15}\text{N}$  is around  $+2.7$  ‰, which is an isotopic signature typical of  
597 nitrogen fixation. The general  $\delta^{15}\text{N}$  trend correlates directly with PC1 variation, as  
598 denoted by the high PCR regression coefficient ( $R = 0.56$ ). Two hypotheses can be  
599 proposed to explain this relationship: (1) larger inputs of fine or very fine mineral  
600 particles to the mire may have triggered conditions of enhanced productivity and



601 consequently, higher  $\delta^{15}\text{N}$  values; or (2) the vegetation change can entail a differential  
602 fractionation of the peat forming plant remains (Talbot, 2001). However, two prominent  
603 peaks of  $\delta^{15}\text{N}$  at 66.8 kyr BP and 62.4 kyr BP (12.36 m and 11.07 m, Fig. 1 from  
604 supporting information) are not accounted for by the PCR model and thus, they are  
605 apparently not related to the influx of mineral matter or long term shifts (supporting  
606 information, Fig. 1). These very high  $\delta^{15}\text{N}$  values ( $\delta^{15}\text{N} \geq 7 \text{ ‰}$ ) may have been reached  
607 by the preferential loss of light nitrogen through denitrification or ammonization (Talbot  
608 et al., 2001) indicating anoxic phases. Therefore, our results suggest that different  
609 processes could change the isotopic signature over different time scales: long-term  
610 variability related to a shift in the vegetation and short-term variability related to small-  
611 scale events such as a change in the potential redox.

612 Total S (decreasing) and  $\delta^{34}\text{S}_{\text{CDT}}$  (increasing) trends from 55.5 to 43.9 kyr BP (9-6 m)  
613 also show a differential S assimilation and fractionation through time (Fig. 2). The  
614 changes in S cycling, especially  $\delta^{34}\text{S}_{\text{CDT}}$ , seem partially related to PC1 and the shift in  
615  $\delta^{13}\text{C}$  (Table 1). Sulfur is incorporated by plants and bacteria, especially in the form of  
616 organosulfur compounds, which seem to be the dominant S fraction in peat (Novák et  
617 al., 1994 and 1999; Wieder and Lang, 1988). The chemical composition of the Rano  
618 Aroi basin lithology indicates that inorganic S-content is low or negligible and this is  
619 not considered a source of S to the mire (Baker et al., 1974, Margalef et al., 2013).  
620 Because no volcanic eruption has been recorded nor have ash layers have been  
621 described in Rano Aroi and Rano Raraku in late Quaternary sediments (Flenley 1991,  
622 Sáez et al. 2009) the most likely dominant S source is marine sulphate. Variations in TS  
623 and  $\delta^{34}\text{S}_{\text{CDT}}$  can be therefore explained as changes in the loss after early diagenesis  
624 (bacterial sulphate reduction and fixation) that discriminates against the heavier isotope  
625  $^{34}\text{S}$  (see supporting information for additional information about sulphur interpretation).

626      5.2.2 *PC2 and organic chemistry*

627      PCR analyses show that organic matter composition does not significantly  
628 correlate with PC2; however, several cause-effect relationships can be drawn from  
629 the stable isotope, TC, and TN records.  $\delta^{13}\text{C}$  second order changes (i.e. peaks) show  
630 lower values coinciding with Facies C and PC2 peaks from the bottommost part of  
631 the record until 6 m depth (Fig. 6). This relationship between organic matter and  
632 Facies C can be explained by differential fractionation due to moisture changes or a  
633 higher proportion of  $\text{C}_3$  plants during wet events (Margalef et al., 2013). Abrupt  
634 increases of TN also match PC2 peaks (Fig. 6). High TN values together with low  
635 C/N ratios can be attributed to a higher contribution of lacustrine algal material  
636 (low C/N), in contrast to high C/N values that indicate higher proportions of  
637 terrestrial or aquatic plants (*versus* algae) organic matter (Meyers, 1994).

638      5.2.3 *PC3 and organic chemistry*

639      The organic chemistry does not correlate with PC3. Nevertheless, the drought event  
640 may have partially determined the  $\delta^{13}\text{C}$  signature like the lighter ratios on the highly  
641 decomposed part of the record show. The same can be stated for the C/N ratios that are  
642 slightly higher between 5 m and 4.23 m depth (Fig. 2). Total S displays a relative  
643 enrichment between 5 m and 4.23 m depth while the underlying level, from 6.5 m to 5.5  
644 m depth, becomes depleted in S. This pattern could respond to bioaccumulation through  
645 the formation of organic S compounds that are more stable under oxidizing conditions  
646 at the expense of the S released from the layers that remained under reducing conditions  
647 near the watertable interphase (coherently when  $\delta^{34}\text{S}_{\text{CDT}}$  reaches the maximum values).

648      5.3 *Rano Aroi environmental reconstruction: climate, basin and peatland*  
649 *interactions*

650      The Rano Aroi dataset and our multi-proxy approach allow us to reconstruct

651 paleoenvironmental changes considering the intimate interplay between climate forcing,  
652 basin and catchment evolution (soil and vegetation changes) and peat processes.

653 MIS 4 (73.5-59.4 kyr BP) is a period characterized by low southern Pacific SST  
654 temperatures (Kaiser et al., 2005, Pena et al., 2008, Fig. 7H). The Antarctic Circumpolar  
655 Current (ACC) was enhanced and the Southern Westerlies moved equatorward resulting  
656 in sea ice export away from Antarctica (Kaiser et al., 2005). Sea level was globally low,  
657 between 90 and 100 m below the present day (Grant et al., 2012, Fig. 7E), which has  
658 been proposed as an important factor for Easter Island's hydrology and groundwater  
659 levels as well as that of other small islands (Margalef et al., 2013). A lower sea level  
660 would probably also cause lower groundwater levels. This time period sees a complete  
661 dominance of C<sub>4</sub> plant types (mostly Poaceae) on the Rano Aroi basin and Terevaka  
662 area (Fig. 6) and the presence of *Arecaceae* and *Coprosma* taxa on the island. Some  
663 studies have proposed the development of palm tree forests preferentially in the lower  
664 areas of the island (Flenley et al., 1991), although other authors suggest that the  
665 palynological results obtained so far are also coherent with a mosaic vegetation pattern  
666 with forested areas around permanent mires, lakes and the coastline as gallery forests  
667 (Rull et al., 2010a).

668 The prevalence of C<sub>4</sub> plants suggests drier conditions that would lead to a low degree  
669 of pedogenesis in the catchment soils; a scenario that is coherent with the low global  
670 temperatures (Grant et al., 2012; Kaiser et al, 2005; Fig. 7H and E). Additionally,  
671 herbaceous plant dominance may have facilitated higher soil erodibility, either eolian  
672 and/or hydrological. The result was a higher dust flux of typical lithogenic elements and  
673 metals into the mire, as summarized by PC1.

674 Globally warmer conditions heralded the arrival of MIS 3 (59.4-27.8 kyr BP). SST of  
675 mid latitudes of Pacific Ocean increased around 5°C between 62.7 and 61.5 kyr BP

676 (Fig. 7H). This change was coupled to a rapid sea level rise, and between 62.9 and 61.2  
677 kyr BP sea level shifted from -96 m a.s.l to -75 m a.s.l (Grant et al., 2012). In the South  
678 Pacific, atmospheric patterns underwent important reorganizations. A record from the  
679 Cariaco Basin (Peterson et al., 2000, Fig. 7F) indicates that between 61.2 and 59.9 kyr  
680 BP the Intertropical Convergence Zone (ITCZ) was situated in a stable southern  
681 position leading to very dry conditions in Northern Hemisphere tropics. The southern  
682 latitudinal migration of ITCZ therefore leads to the opposite hydrologic trend for the  
683 low latitudes in the Southern Hemisphere (Leduc et al., 2009; Wang et al., 2007,). The  
684 early MIS3 has been characterized in the Rano Aroi record as a humid period, as  
685 expressed by the abrupt events of higher sediment delivery (see section 5.1.2, and Fig. 6  
686 and 7). It is significant that the first important wet event on Easter Island, starting in  
687 Rano Aroi at 61.6 kyr BP is apparently synchronously with important global changes in  
688 (1) sea level rise, (2) the position of the ITCZ and (3) SST.

689 The new warmer and wetter conditions of Early MIS 3 (61.6 to ~40 kyr cal BP) were  
690 linked to an intensification of the degree of pedogenesis, which led to a decrease in the  
691 flux of lithogenic elements to the mire. In parallel to the decline in PC1 values (Fig.  
692 7B), the bulk peat stable isotope data values ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) started a gradual decline at  
693 55 cal kyr BP (Fig. 6). The isotopic change indicates a shift in the vegetation  
694 community forming the peat, which becomes dominated by  $\text{C}_3$  plant types. As stated in  
695 previous sections, pollen data suggests that a complete dominance of Poaceae  
696 (presumably  $\text{C}_4$  species) was replaced by a combination of Asteraceae, *Coprosma*,  
697 Poaceae and ferns between 51 and 48 kyr BP (Fig. 5). The vegetation change around the  
698 mire and the presence of scarce forest, trees and shrubs may have prevented soil erosion  
699 and reduced the fluxes of mineral matter to the mire (PC1). Finally, the expansion of  
700 Cyperaceae (presumably  $\text{C}_3$  species, such as *Scirpus californicus*) probably constituted

701 an important physical barrier during the C<sub>3</sub> dominance period, slowing the runoff input,  
702 except during stronger events (PC2).

703 The late MIS 3 (40-27.8 cal. kyr BP) has been characterized as a drier phase on  
704 Easter Island (Margalef et al., 2013). Sea level dropped relatively rapidly between 31.4  
705 and 29.4 cal. kyr BP, but a South Pacific thermal response to global cooling was not  
706 recorded until the onset of MIS 2 (Kaiser et al., 2005, Fig. 7H). The Cariaco Basin  
707 record shows that between 35 and 31 cal. kyr BP, the ITCZ was in a northern position  
708 preventing the arrival of strong storms to southern latitudes (Peterson et al., 2000, Fig.  
709 7F). Moreover, several records from South America document intense dry between ca.  
710 41 and ca. 31 cal. kyr BP (Lamy et al., 1998; Stuut and Lamy, 2004) explained by the  
711 southern migration of the Southern Westerlies under the precession forcing (Fig. 7G).  
712 The combination of these regional climate patterns likely led to a long dry period in the  
713 Central Pacific and on Easter Island. The drought started after 39 cal. kyr BP, the age of  
714 the sharp unconformity in the Rano Aroi sequence that separates highly degraded  
715 (below) and fresh peat (above). A lowering of the Rano Aroi water table accelerated  
716 peat decomposition producing an increase in the elements representative of PC3 (Ca, Sr,  
717 and Mg) as a diagenetic imprint (Fig. 7D). The time interval above the discontinuity  
718 depicts net accumulation rates of 0.05 mm/y (Fig. 7A) and represents the reactivation of  
719 peat formation after a long-term pause (probably including erosion) where old carbon  
720 could be incorporated in younger roots and plant remains. Because of this carbon  
721 recycling, the chronology right after the reactivation has to be carefully considered and  
722 the exact date of the reactivation and the amount of peat eroded cannot be properly  
723 determined.

724 Although sea level reached a minimum ca. 23 cal. kyr BP (Grant et al., 2012;  
725 Lambeck and Chappell, 2001; Fig. 7E) and could have negatively affected the

726 groundwater input and the hydrological balance at Rano Aroi, peat formation was active  
727 during the LGM. These can be explained by permanent cold conditions preventing  
728 strong evaporation (Sáez et al., 2009) and a northernmost position of Southern  
729 Westerlies whose influence reached subtropical latitudes during glacial times (Lamy et  
730 al., 1998, Fig. 7G).

731 Rano Aroi peat accumulation reactivated completely by ca. 17.5 kyr BP (the onset of  
732 Termination 1). Sea level started a prominent rise and the Intertropical Convergence  
733 Zone shifted to its furthestmost south position between 21 and 16 kyr BP (Fig. 7F).  
734 During MIS 2 (27.8-14.7 cal. kyr BP), PC3 values remained low, showing no evidence  
735 of a drought period. Conversely, high PC2 events are found during the late glacial at  
736 20.9-19.5 cal. kyr BP and 16.4 cal. kyr BP, likely representing enhanced precipitation  
737 coinciding with HS 2 and 1 (Fig. 7C). Maximum peat accumulation rates (14 cal kyr  
738 BP, Fig. 7A) coincide with the highest rates of sea level rise during deglaciation  
739 (Dickinson, 2001; Hanebuth et al., 2000; Lambeck and Chappell, 2001). Sea level rise  
740 together with warmer SST might have played an important role in the development of  
741 enhanced convection storms.

742 According to several Southern Hemisphere records the Early Holocene was  
743 characterized by a warming (Pena et al., 2008) and SST were maximal at approximately  
744 12 cal kyr BP and generally decreased thereafter until modern SST were reached  
745 (Kaiser et al., 2005; Kaiser et al., 2008). In ARO 06 01 record, only the early Holocene  
746 (11.7-8.5 cal kyr BP) peat remains because the surface levels were rejected to avoid  
747 anthropic remobilization. The most important features characterizing this period are  
748 high PC2 values around 10.2 cal kyr BP indicating strong runoff events, while PC1  
749 points to catchment soil conditions similar to those recorded ca. 48 kyr cal BP indicating  
750 relatively low fluxes of inorganic material under a permanent C<sub>3</sub> plant dominance (as

751 shown by  $\delta^{13}\text{C}$ , Fig. 6 and 7).

## 752 **6. CONCLUSIONS**

753 The organic matter composition (TC, TN, TS,  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ,  $\delta^{34}\text{S}$ ), inorganic  
754 geochemistry and pollen data from Rano Aroi mire provide a coherent reconstruction of  
755 the paleoenvironmental history of Easter Island.

756 Principal components analysis of peat geochemistry reveals that three main  
757 environmental processes have controlled the inorganic elemental composition of the  
758 peat accumulated. (1) The first process, depicted by PC1, reflects changes in the basin  
759 background erosion and transport of the mineral matter as very fine particles into the  
760 mire and it is linked to soil evolution and vegetation shift. In Rano Aroi,  $\delta^{13}\text{C}$  can be  
761 used to infer an important vegetation change from  $\text{C}_4$  to  $\text{C}_3$  plant dominance that  
762 occurred from 55 to 50 kyr BP. The correlation of the  $\delta^{13}\text{C}$  and PC1 records reveals that  
763 vegetation shifts and the evolution of the soils of the mire basin were intimately related  
764 to the rate of allochthonous material transported into the peatland. These environmental  
765 changes also affected the  $\delta^{15}\text{N}$  signal that integrates variability in mire productivity and  
766 redox conditions.  $\delta^{34}\text{S}$  signatures indicate that the S source is primarily marine. The  $\delta^{34}\text{S}$   
767 ratio and TS concentration suggest that S may have been differentially mobilized  
768 depending on vegetation assemblages by sulphate reduction bacteria. (2) The second  
769 process is the occurrence of high precipitation events (identified by the PC2 signal)  
770 related to strong runoff and delivery of large amounts of terrigenous particles coarser  
771 than those mobilized by PC1 process. These events occurred at approximately 60 kyr  
772 BP, 52 kyr cal BP and 42 kyr cal BP. (3) Finally, the third process, illustrated by PC3,  
773 mainly reflects peat oxidation caused by a long-term drought after ca. 39 kyr cal BP.

774 The environmental evolution of Rano Aroi mire, largely driven by hydrological  
775 changes, is coherent with the regional climatic variability described for the last 70 kyr

776 BP. During MIS 4 the Rano Aroi basin was occupied by open grasslands and C<sub>4</sub>  
777 Poaceae dominated the mire owing to the generally cold and relatively dry climate  
778 conditions. MIS 3 was marked by the onset of wet events, which occurred at ca. 60 kyr  
779 BP, 52 kyr cal BP and 42 kyr cal BP.

780 During the first half of MIS 3 and probably driven by the wetter and warmer  
781 conditions, Asteraceae and other small trees became gradually more abundant, forming  
782 scantily wooded areas around Terevaka, while C<sub>3</sub> peat forming plants colonized the Aroi  
783 mire. In contrast, the second half of MIS 3 was drier. A long-term drought led to a water  
784 table drop and enhanced peat mineralization at some time between the 39 and 31 kyr cal  
785 BP. During the MIS 2 and LGM the water table recovered and peat accumulation  
786 resumed under C<sub>3</sub> plant dominance.

787



788 **ACKNOWLEDGEMENTS**

789 This research was funded by the Spanish Ministry of Science and Education through the  
790 projects LAVOLTER (CGL2004-00683/BTE), GEOBILA (CGL2007-60932/BTE) and  
791 CONSOLIDER GRACCIE (CSD2007-00067) and an undergraduate grant JAE (BOE  
792 04/03/2008) to Olga Margalef. We would like to thank CONAF (Chile) and the  
793 Riroroko family for the facilities provided on Easter Island.

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