1 Vegetation and geochemical responses to Holocene rapid

2 climate change in Sierra Nevada (SE Iberia): The Laguna

3 Hondera record

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15 Abstract.

16 High-altitude peat bogs and lacustrine records are very sensitive to climate changes and atmospheric dust

17 input. Recent studies have shown a close relationship between regional climate aridity and enhanced

18 eolian input to lake sediments. However, changes in regional-scale dust fluxes due to climate variability

- 19 at short-scales and how alpine environments were impacted by climatic- and human-induced
- 20 environmental changes are not completely understood.
- 21 Here we present a multi-proxy (palynological, geochemical and magnetic susceptibility) lake sediment 22 record of climate variability in the Sierra Nevada (SE Iberian Peninsula) over the Holocene. Magnetic 23 susceptibility and geochemical proxies obtained from the high mountain lake record of Laguna Hondera 24 evidence humid conditions during the Early Holocene, while a trend towards more arid conditions is 25 recognized since ~7000 cal yr BP, with enhanced Saharan eolian dust deposition until Present. This trend 26 towards enhanced arid conditions was modulated by millennial-scale climate variability. Relative humid 27 conditions occurred during the Iberian Roman Humid Period (2600-1450 cal yr BP) and predominantly 28 arid conditions occurred during the Dark Ages and the Medieval Climate Anomaly (1450-650 cal yr BP). 29 The Little Ice Age (650-150 cal yr BP) is characterized in the LH record by an increase in runoff and a 30 minimum in eolian input. In addition, we further suggest that human impact in the area is noticed through 31 the record of Olea cultivation, Pinus reforestation and Pb pollution during the Industrial Period (150 cal yr 32 BP-Present). Furthermore, we estimated that the correlation between Zr and Ca concentrations stands for 33 Saharan dust input to the Sierra Nevada lake records. These assumptions support that present-day
- 34 biochemical observations, pointing to eolian input as the main inorganic nutrient source for oligotrophic
- 35 mountain lakes, are comparable to the past record of eolian supply to these high-altitude lakes.

36 1. Introduction

The southern Iberian Peninsula has been the location for a number of recent studies detailing past vegetation
and former climate of the region (Carrión et al., 2001, 2003, 2007, 2010; Carrión, 2002; Combourieu
Nebout et al., 2009; Jiménez-Espejo et al., 2008; Martín-Puertas et al., 2008, 2010; Fletcher et al., 2010;

40 Nieto-Moreno et al., 2011, 2015; Rodrigo-Gámiz et al., 2011; Moreno et al., 2012 Jiménez-Moreno et al., 41 2015). Some of these studies have also documented that the western Mediterranean area has been very 42 sensitive to short-term climatic fluctuations throughout the Holocene (e.g., Fletcher and Sánchez-Goñi, 43 2008; Combourieu Nebout et al., 2009; Fletcher et al., 2010; Jiménez-Moreno et al., 2013). However, a 44 subset of recent studies have attempted to determine how Mediterranean alpine environments have been 45 affected by Holocene climate change through the study of sedimentary records from high elevation 46 wetlands in the Sierra Nevada (Anderson et al., 2011; García-Alix et al., 2012, 2013; Jiménez-Moreno and 47 Anderson, 2012; Jiménez-Moreno et al., 2013; Jiménez-Espejo et al., 2014; Ramos-Román et al., 2016; 48 García-Alix et al., 2017). These alpine lake and bog records show minimal anthropic influence because 49 they are usually elevational higher than major regional Late Holocene human landscape modification. This 50 allows for a potentially clearer climatic signal to be determined from these sites. Even though human impact 51 is less important at high-elevations, the impacts of human activities has also been reconstructed from these 52 Late Holocene sedimentary records (Anderson et al., 2011; García-Alix et al., 2012, 2013; 2017, 2018). 53 Several studies have highlighted the role of atmospheric mineral dust deposition in marine (Pulido-Villena 54 et al., 2008a) and terrestrial (Morales-Baquero et al., 1999; Ballantyne et al., 2011) ecosystem fertilization 55 through major micronutrients supply. Similar results have been described in the Sierra Nevada alpine lakes,

- where Saharan dust is especially important in conditioning plankton communities from oligotrophic lakes
 (Morales-Baquero et al., 2006a, 2006b; Mladenov et al., 2008; Pulido-Villena et al., 2008b; Reche et al.,
- 58 2009). Although this eolian signal has been occasionally recorded in the sedimentary sequences from the
- 59 Sierra Nevada lakes (Jimenez-Espejo et al., 2014; García-Alix et al., 2017), the record of inorganic nutrients
- 60 in Saharan dust input in past lake geochemistry has remained elusive. This study investigates a multiproxy
- 61 sediment core record from Laguna Hondera (LH), located in the Sierra Nevada range with two main goals:
- 62 (1) identifying and characterizing climatic variability during the Holocene, focusing on vegetation changes,
- 63 eolian input and runoff sediments variations; and (2) understanding the Saharan dust influence in past lake
- 64 sedimentation and geochemistry.

65 2. Study Area

66 Sierra Nevada is the highest mountain range in the southern Iberian Peninsula. Bedrock of the high 67 elevations of the Sierra Nevada is mostly composed of metamorphic rocks, principally mica schists 68 (Castillo Martín, 2009). During the late Pleistocene, the Sierra Nevada was one of the southernmost 69 mountains to support alpine glaciers and its last advance was recorded during the Little Ice Age (LIA; 70 Palma et al., 2017; Oliva et al., 2018). Subsequently to the melting of ice at the end of the Last Glacial 71 Maximum, wetlands and small lakes formed in the glacial cirque basins, which occur between 2451 and 72 3227 masl (Schulte, 2002; Castillo Martín, 2009; Palma et al., 2017). Several alpine wetland and lakes have 73 been studied in this area during the last few years as shown in Figure 1.

74 2.1. Regional Climate and Vegetation

75 Mediterranean climate characterises southern Iberia, with a marked seasonal variation between warm and

- 76 dry summers and cool and humid winters (e.g. Lionello et al., 2006). Overprinting this general climate is
- 77 the influence of the North Atlantic Oscillation (NAO) (Trigo et al., 2004; Trouet et al., 2009). Southern

- 78 Iberia is also characterized by strong altitudinal contrasts, which in turn control the precipitation patterns,
- 79 with mean annual values ranging from <400 mm yr⁻¹ to >1400 mm yr⁻¹ in the southeast desert lowlands
- 80 and the southwest highland, respectively (Jiménez-Moreno et al., 2013 and references therein).
- As with most mountainous regions, species and species groupings in the Sierra Nevada are distributed with
 respect to elevation, depending on the temperature and rainfall gradients (e.g., El Aallali et al., 1998; Valle,
- 83 2003). Above 2800 masl the crioromediterranean flora occurs as tundra-like open grassland. The
- 84 oromediterranean belt (1900-2800 masl) mostly includes dwarf *Juniperus* (juniper), xerophytic shrublands
- 85 and pasturelands and *Pinus sylvestris* and *P. nigra*. The supramediterranean belt (~1400-1900 masl) is
- 86 characterized by mixed deciduous and evergreen forest species (i.e., evergreen and deciduous *Quercus*,
- 88 shrublands and evergreen *Quercus* woodlands. The natural vegetation has been strongly altered by human

with Pinus spp. and others). Mesomediterranean vegetation (600-1400 masl) includes sclerophyllous

- 89 activities and cultivation in the last centuries, increasing significantly the abundance of *Olea* (olive), due to
- 90 cultivation at lower altitudes (Anderson et al., 2011, and references therein), and *Pinus* due to reforestation
- 91 primarily at higher elevations (Valbuena-Carabaña, 2010).

92 2.2. Laguna Hondera

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- 93 Laguna Hondera (hereafter LH; 2899 masl; 37°02.88'N, 3°17.66'W, lake surface: 0.0053 km²; maximum depth: 0.8 m; Morales-Baquero et al., 1999; Fig. 1) is a small and shallow lake located at the lowest 94 95 elevation of a set of lakes locally named Cañada de Siete Lagunas, a glacial valley between two of the 96 highest peaks of the mountain range in the Iberian Peninsula: Alcazaba (3366 masl) and Mulhacén (3479 97 masl). LH has a large catchment area of 1.546 km², which is much larger than previously studied sites in 98 the region (Laguna de Río Seco, LdRS, 0.099 km²; Borreguil de la Caldera, BdlC, 0.62 km²; Morales-99 Baquero et al., 1999; Ramos-Román et al., 2016; Fig 1 for locations). The lake was reduced to a little pond 100 in the deepest area of the basin when cored in September 2012, with a maximum depth of only a few
- 101 centimetres.
- 102 LH presently occurs in the crioromediterranean vegetation belt (2800 masl) (El Aallali et al., 1998; Valle
- 103 et al., 2003). The bedrock in the LH basin consists in Paleozoic and Precambrian mica schist with disthene
- and staurolite of the lower part of the Caldera Formation (Díaz de Federico et al., 1980).

105 3. Methods

106 **3.1.** Core sampling, lithology and chronology

- 107 Six sediment cores were recovered from LH with a Livingstone piston corer in September 2012. LH 12-03
- 108 (83 cm) was selected for a multi-proxy study because it was the longest core. Cores were wrapped with tin
- 109 foil and plastic film and transported to Universidad de Granada, where they were stored at 4°C.
- 110 Core LH 12-03 was split longitudinally and the sediments were described. Magnetic susceptibility was
- 111 measured every 0.5 cm with a Bartington MS2E meter in SI units (x 10⁻⁴) (Fig. 2). The sediment cores were
- subsampled every 1 cm for several analyses, including pollen and geochemistry.
- 113 The age model was built using seven AMS radiocarbon dates from vegetal remains (Table 1; Fig. 2) by
- 114 means of Clam software (Blaauw, 2010; version 2.2), which used the IntCal13 curve for radiocarbon age

115 calibration (Reimer et al., 2013). A smooth spline approach was chosen (Fig. 2). The sediment accumulation

rate (SAR) was calculated with the average rate from the Clam smooth spline output (Fig. 2).

117 3.2. Pollen

118 Pollen analysis was performed on 1 cm³ of sample collected at regular 1 cm interval throughout the first 62 119 cm of the core. Older sediments (from 62 to 82 cm depth) were barren in pollen, and only one interval at 120 73 cm could be studied (Fig. 2). Pollen extraction included HCl and HF treatment, sieving, and the addition 121 of Lycopodium spores for calculation of pollen concentration (modified from Faegri and Iversen, 1989). 122 Sieving was done using a 10 µm nylon sieve. The resulting pollen residue was suspended in glycerine and 123 mounted on microscope slides. Slides were analysed at 400x magnification counting a minimum of 300 124 pollen grains, not including the local aquatic species Cyperaceae, Ranunculaceae and Typha. An overview 125 of pollen taxa with abundances >1% for core LH 12-03 is plotted using the Tilia software (Grimm, 1993) 126 in Figure 3. The pollen zonation was delimitated visually by a cluster analysis constrained by age of taxa 127 abundance >1% using CONISS software (Grimm, 1987) (Fig. 3). Olea was differentiated from others 128 Oleaceae, such as Phillyrea, based on the thicker intine and higher size of reticulum in polar vision (Beug, 129 2004).

130 3.3. Geochemical analyses

131 X-ray fluorescence (XRF) Avaatech core scanner®, located at the University of Barcelona, was used to 132 measure light and heavy elements in the LH 12-03 core. An X-ray current of 650 μ A, a 10 second count 133 time and 10 kV X-ray voltage was used for measuring light elements, whereas 1700 μ A X-ray current, 35 134 second count time and 30 kV X-ray voltage was used for heavy elements. Sampling interval for these 135 analyses was every 0.5 cm. For our study only three elements (K, Ca and Ti) have been considered with 136 enough counts to be representative.

137 Inductively coupled plasma-optical emission spectrometry (ICP-OES; Perkin-Elmer optima 8300) was 138 used for major element analysis on discrete samples every 2 cm. Prior to analysis, the samples were dried 139 in an oven and digested with HNO₃ and HF. Blanks and international standards were used for quality 140 control, the analytical accuracy was higher than $\pm 2.79\%$ and 1.89% for 50 ppm elemental concentrations 141 of Al and Ca, respectively, and better than $\pm 0.44\%$ for 5 ppm elemental concentrations of K.

- 142 Trace element analysis was performed with an inductively coupled plasma mass spectrometry (ICP-MS;143 Perkin Elmer Sciex Elan 5000). Samples were measured in triplicate through spectrometry using Re and
- 144 Rh as internal standards. The instrumental error is 2% for elemental concentrations of 50 ppm (Bea, 1996).
- 145 Both ICP-OES and ICP-MS analyses were performed at the Centre for Scientific Instrumentation (CIC),
- 146 University of Granada, Spain.

147 3.4. Mineralogical analyses

- 148 Morphological and compositional analyses were performed using scanning electron microscopy (SEM)
- 149 with an AURIGA model microscope (Carl Zeiss SMT) coupled with energy-dispersive X-ray microanalysis
- 150 (EDX) and Electron Backscatter Diffraction (EBSD) mode, also at the CIC (University of Granada, Spain).
- 151 Mineral grains were analysed to determine provenance, in particular those from eolian origin.

152 3.5 Statistical Analysis

- 153 R-mode principal components analysis (PCA) was run on the geochemical dataset using the PAST software
- 154 (Hammer et al., 2001). PCA identifies hypothetical variables (components) accounting for as much as
- 155 possible of the variance in multivariate data (Davis, 1986; Harper, 1999). The elements used in the PCA
- 156 were standardized by subtracting the mean and dividing by the standard deviation (Davis, 1986). Pb was
- 157 not included in the PCA analysis due to its anthropogenic origin from mining and industrial pollution during
- the latest Holocene in this area (García-Alix et al., 2013).

159 **4. Results**

160 4.1. Lithology and magnetic susceptibility

161 The LH 12-03 sediment core consists primarily of peat in the upper ~60 cm, with mostly sand and clay

162 layers below (Fig. 2). Positive MS peaks coincide with the grey clay intervals between 58 and 72 cm. Peat

- 163 intervals coincide with relatively low MS values. For example, a minimum in MS occurs at 36-48 cm depth,
- related with a peaty interval with root remains. Near the bottom of the core, between 76 and 80 cm, a sandy
- 165 oxidized interval occurs.

166 4.2. Chronology and sedimentation rate

167 The age model of LH 12-03 documents that the record spans the last 10800 cal yr BP (Table 1; Fig. 2). 168 Sediment accumulation rates (SAR) were calculated using the average rate from the Clam smooth spline 169 output (Fig. 2). The SAR below ~39 cm is very constant, varying between 0.049 and 0.061 mm yr ⁻¹. The 170 SAR increases exponentially to 0.098 mm yr $^{-1}$ at 22 cm, 0.167 mm yr $^{-1}$ at ~9 cm and 0.357 mm yr $^{-1}$ at the 171 core top. Accordingly with the model age and the SAR, resolution of pollen analysis varies between ~40 172 years per sample in the top of the core and ~ 120 years per sample in the lower part. The resolution of the 173 geochemical analysis on discrete samples changes between 100 and 400 years per sample, but the 174 geochemical XRF core scanning resolution ranges between 15 and 100 years per sample, providing higher 175 resolution than geochemical data on discrete sample. The MS analyses resolution variates between 15 and 176 100 years per sample.

177 **4.3. Pollen**

178 Fifty distinct pollen taxa were recognized, but only those with abundance higher than 1% are included in 179 the pollen diagram (Fig. 3). Four pollen zones for the LH 12-03 record are identified, using variation in 180 pollen species plotted in Figure 3 and a cluster analysis run through the CONISS software (Grimm, 1987). 181 Zone LH-1 (core bottom-2600 cal yr BP) is subdivided in two subzones. Subzone LH-1A (bottom-4000 cal 182 yr BP) is defined by the alternation between Arboreal Pollen (AP) and herbs. AP is composed primarily of 183 Pinus, but also Quercus. During the interval from ~9500 to ~7000 cal yr, BP only three samples were 184 analysed, due to the low preservation of pollen in this interval. Pollen in this period is dominated by an 185 alternation between Asteraceae (3-60%) and Pinus (5-90%) (Fig. 3). The highest occurrence of Onagraceae 186 (~10%) is identified in this subzone, and Caryophyllaceae reach high values (~10%) as well. Only minor 187 amounts of graminoids (Poaceae and Cyperaceae) occur during this period.

- 188 Between ~7000 to ~4000, *Pinus* pollen decreases from 90 % to ~55%, with a minimum (~30%) at 5000 cal
- 189 yr BP. *Quercus* increase from ~2% to ~10%. The highest percentages of *Betula* (~5%) in the record occurs
- 190 at this time. Asteraceae pollen decreases (~5-30%), but Poaceae increase from <5% at the opening of the

subzone to >25%. Cyperaceae occur in high percentages (15%).

- 192 The subzone LH-1B (~ 4000-2600 cal yr BP) is defined primarily by a great increase in Poaceae pollen (to
- 193 ~60%) (Fig. 3). Other important herbs and shrubs include Asteraceae (5-15%) and Caryophyllaceae (~5%).
- 194 Other pollen types that increase for the first time in this zone include Ericaceae (~3%), Artemisia (~3%)
- and Ranunculaceae (~2-6%). *Pinus* (~3-25%) and Cyperaceae (0-14%) record a minimum in this zone, and
- 196 Onagraceae disappear altogether (Fig. 3).
- 197 Zone LH-2 (~ 2600-1450 cal yr BP) pollen assemblages show high variability. *Pinus* pollen variates
 198 between ~80% to ~3% from the onset to the end of the zone. Aquatic pollen such as Cyperaceae (~15%)
 199 increases. On the other hand, an increase in herbs such as Asteraceae (~5-70%) occurs along the zone,
 200 Poaceae pollen variates between ~7 and 12%.
- Zone LH-3 (~ 1450-150 cal yr BP) is subdivided in two subzones. Subzone 3A (~1450-600 cal yr BP) is
 characterized by an increase in herbaceous pollen, led by Poaceae (~35% maximum during this zone),
 Asteraceae (~60% maximum during this zone after ~1000 cal yr BP) and *Artemisia* (~10%), with the
 resulting decrease in AP. Since this zone to the Present, *Quercus* pollen is the major component of AP
 instead of *Pinus*. Cyperaceae also show a decrease, and Ranunculaceae reach ~ 5%. Subzone 3B (~600150 cal yr BP) documents an increase in *Olea* (~6%), Poaceae (20%), Caryophyllaceae (7%) and *Artemisia*(~2-20%). *Pinus* (~2%) and Asteraceae (~20%) decrease in this period. Aquatic and wetland pollen show
- **208** a rise (Cyperaceae ~30%, Rannunculaceae ~10%).
- Zone LH-4 (~ 150 cal yr BP-present) depicts a further increase in *Olea* (~25%), Poaceae (~40%) and
 Artemisia (~10%).

211 4.4. Sediment composition

212 The XRF-scanning method relies on determining the relative variations on elements composition. 213 Nevertheless, due to the presence of major variations in organic matter or carbonates it is necessary to 214 normalize the measured count in order to obtain an environmentally relevant signal (Löwemark et al., 215 2011). Aluminium and titanium normalizations are commonly used to discern possible fluctuations in the 216 lithogenic fraction (enrichment or depletion of specific elements), particularly in the terrigenous 217 aluminosilicate sediment fraction (Van der Weijden, 2002; Calvert and Pedersen 2007; Martinez-Ruiz et 218 al., 2015). For this study, the XRF data were normalized to Ti since Al counts obtained were very low. Poor 219 detection of Al can be related to either low Al content, or high organic and water contents that increase 220 radiation absorption and affect the intensity of this light element, among other possibilities (e.g. Tjallingii 221 et al., 2007).

- Since data spacing is different between the analyses on discrete samples and the XRF scanner, a linear interpolation was performed with the purpose of equalizing the space of the different time series (150-300 years). Afterwards, the mobile average was worked out along the time series (taking into account the 5 nearest points) in order to easily identify trends by means of smoothing out data irregularities. The obtained data were compared, and both XRF-scanner and discrete sample data showed a good correlation. As a
 - 6

- 227 consequence, the geochemical proxies displayed higher time resolution than the discrete samples (Table 2).
- 228 Discrete sample and XRF data results are described together in order to simplify this section (Fig. 4).
- 229 The lower part of the core is typified by maximum values of K/Al and K/Ti ratios, coinciding with the
- 230 lowest values in Ca/Al, Ca/Ti and Zr/Al ratios. Pb/Al data show a stable pattern during this interval.
- 231 Nevertheless, between 10000 and 9000 cal yr BP and ~8200 cal yr BP the trends were reversed, with
- relatively low K/Al, low K/Ti and slightly increasing Zr/Al, Ca/Al and Ca/Ti ratios. A positive peak in
 Pb/Al ratio at ~8200 cal yr BP is also observed.
- 234 Between ~7000 and 4000 cal yr BP a decreasing trend in K/Al and K/Ti ratios occurs along with an
- increasing trend in Zr/Al, Ca/Al and Ca/Ti ratios. The Pb/Al ratio remains constant throughout this interval.
 From ~4000 to ~2600 cal yr BP an increase in Zr/Al, Ca/Al and Ca/Ti ratios is documented. A maximum
 in eolian proxies occurs at ~2600 cal yr BP. A K/Al and K/Ti minima occurs between ~3000 and ~2600 cal
- 238 yr BP. The Pb/Al ratio shows a positive peak at ~2800 cal yr BP.
- The interval between ~2600 and ~1450 cal yr BP is characterized by low Ca/Al, Ca/Ti and Zr/Al ratios,
 with relatively high K/Al and K/Ti ratios. The Pb/Al ratio shows a flat pattern, increasing at ~1500 cal yr
 BP.
- The period between ~1450 and ~650 cal yr BP depicts higher ratios of Zr/Al, Ca/Al and Ca/Ti and
 decreasing ratios of K/Al and K/Ti. A somewhat higher Pb/Al ratio is also registered during this interval.
- From ~650 to ~150 low values of Zr/Al and Ca/Ti ratios and minimum values Ca/Al ratio occur. Higher
- K/Al and K/Ti values are also observed. The Pb/Al ratio decreases during this interval. From ~150 to the
 present, an increase in Zr/Al, Ca/Al, Ca/Ti, K/Ti and a Pb/Al maximum occur. Lower K/Al ratio is recorded
 during this period.
- 248 Several studies have demonstrated that PCA analysis of geochemical data can elucidate the importance of
- 249 different geochemical components driving the environmental responses in marine and lacustrine records
- 250 (Bahr et al., 2014; Yuan, 2017). We performed a PCA analysis of the LH geochemical data, which yielded
- two significant components (Fig. 5). The first principal component (PC1) describes 58% of the total
- 252 variance. The main negative loadings for PC1 are Rb, Ba, Al, K, Ca, Mg and Sr, while large positive
- 253 loadings correspond to Zr and Rare Earth Elements (REE). The second principal component (PC2) explains
- 254 17% of the total variance. The main negative loading for PC2 are Fe, Ca, Zr, Mg and Lu. Positive loads
- 255 correspond to Al, K, Ba, Sr and other elements.
- 256 SEM analyses show an alternation between a lithology rich in rock fragments and another rich in organic
- remains. Also, diatom frustules, rich in silica, are particularly abundant since ~6300 cal yr BP to Present.
- 258 Other minerals such as zircon, rounded quartz and monazite were also identified (Fig. 6).

259 5. Discussion

Pollen and geochemical proxies have been widely used for reconstructing vegetation changes andenvironmental and climate variations in southern Iberia (e.g. Carrión, 2002; Sánchez-Goñi and Fletcher,

- 262 2008; Anderson et al., 2011; Nieto-Moreno et al., 2011; Jiménez-Moreno et al., 2012; Moreno et al., 2012;
- 263 Fletcher and Zielhofer, 2013; Jiménez-Espejo et al., 2014; Ramos-Román et al., 2016). Variations in the
- 264 occurrences of arboreal taxa such as *Pinus* and other mesic species (e.g., *Betula*, *Quercus*), indicating
- relative humid and warm conditions, and xerophytic species (e.g., Poaceae, Asteraceae, Amaranthaceae,

266 Artemisia), representing aridity, have been useful for reconstructing relative humidity changes in southern 267 Iberian (e.g. Carrión et al., 2001, 2007, 2010; Anderson et al., 2011; Jiménez-Moreno et al., 2012, 2013, 268 2015; Ramos-Román et al., 2016). Pinus reach percentages over 80% in our record. This bisaccate pollen 269 grain is favoured by wind transport and has a larger dispersal area than other tree species, and sometimes 270 might be overrepresented (Poska and Pidek, 2010; Pérez-Díaz et al., 2016). Nevertheless, LH is located at 271 2899 masl only 99 m above treeline and the upper boundary of the oromediterranean belt (1900-2800 masl) 272 where Pinus sylvestris is the main tree specie (El Aallali et al., 1998; Valle, 2003). Therefore, this 273 apparently anomalous high concentration of *Pinus* may be caused by an upward migration of the 274 oromediterranean belt and treeline towards higher elevations and around the LH during warmer and more 275 humid periods, which could have been overstated due to its high pollen-production and dispersal. Therefore, 276 *Pinus* seems to be mostly recording a regional climatic signal, without allocthonous influence.

Over 75% of the total geochemical data variance is explained by the PC1 and PC2 (Fig. 5). We interpret
the results of PC1 as resulting from certain sorting between heavy minerals (positive loading; Zr and REE)
vs. clay minerals and feldspars (negative loadings; K, Al and Ca). The drainage basin is composed mainly
by mica schist, consequently enhanced in K-rich minerals such as mica and feldspar (Díaz de Federico et
al., 1980). This sorting between heavy minerals (enriched in Zr and REE) and clays and feldspars (enriched
in K and Al) (Fig. 5a), was probably linked to physical weathering within the basin and to resulting runoff
until final deposition in the lake.

284 On the other hand, we interpret the results of PC2 as differentiating autochthonous elements (positive 285 loadings) vs. Saharan allochthonous input (negative loadings). In the first case, due to the abundance of 286 mica schist within the LH drainage basin (Díaz de Federico et al., 1980), the K/Al and K/Ti ratios are 287 interpreted as detrital products, and thus a proxy of runoff. In the second case, PC2 negative loading Zr, 288 Ca, Mg and Fe (Fig. 5b) grouped elements that are coherent with Saharan input composition (dolomite, 289 iron oxides and heavy minerals) (Ávila, 1997; Morales-Baquero et al., 2006b; Moreno et al., 2006; Pulido-290 Villena et al., 2007). In addition, Ca shows a strong positive correlation with Zr since 6300 cal yr BP (r 291 =0.57; p<0.05) supporting an eolian origin of the Ca in LH sediments. Although we cannot exclude others 292 nearby Ca sources or changes in the source of African dust (Moreno et al., 2006), the 85% of dust reaching 293 south Iberia derives from the Sahara (Morales-Baquero and Pérez-Martínez, 2016; Jiménez et al., 2018). 294 For instance, enrichment in heavy minerals such as zircon and palygorskite has previously been used as an 295 eolian proxy in the western Mediterranean (e.g., Combourieu Nebout et al., 2002, Rodrigo-Gámiz et al., 296 2011, 2015). High concentrations of Ca in other lacustrine systems is usually associated with biogenic 297 sources when anti-correlated with terrigenous elements (Yuan, 2017). Nevertheless, elevated Ca in the LH 298 record is linked with detrital elements, as shown by PC1, where Ca is associated with K and Al (Fig. 5a). 299 Therefore Ca/Al and Ca/Ti ratios are used in the LH record as Saharan eolian input proxies.

Elemental ratio variations, such as the ratios K/Al and K/Ti indicating fluvial input and the ratios Zr/Al or
Zr/Th indicating aridity and eolian input, have been previously interpreted in Alboran Sea marine records
as well as in southern Iberia lake records (Martín-Puertas et al., 2010; Nieto-Moreno et al., 2011, 2015;
Rodrigo-Gámiz et al., 2011; Jiménez-Espejo et al., 2014; Martínez-Ruiz et al., 2015; García-Alix et al.,
2017, 2018). Thus, the integration of both palynological data and geochemical ratios used as detrital input

305 from LH have allowed the reconstruction of the palaeoclimate and palaeoenvironmental history in Sierra

306 Nevada during the Holocene.

307 5.1. Holocene palaeoclimate and palaeoenvironmental history

308 5.1.1. Early and Mid-Holocene humid conditions (10800–7000 cal yr BP)

309 The wettest conditions are recorded during the Early Holocene in Sierra Nevada. This is shown in the LH 310 record by the highest K/Al ratio and MS values, and the low values in Zr/Al, Ca/Al and Ca/Ti ratios, 311 suggesting that runoff dominated over eolian processes at this time (zone LH-1; Fig. 7) and agrees with 312 previous studies in the area (Anderson et al., 2011; Jiménez-Moreno and Anderson, 2012; García-Alix et 313 al., 2012; Jiménez-Espejo et al., 2014). Unfortunately, the pollen record from LH during this interval is 314 insufficient to definitely confirm this interpretation, due to the high detrital sediment composition and low 315 organic content, as shown by the low MS values and low pollen preservation. However, high percentages 316 of AP in two out of three analysed samples suggest humid conditions and high runoff during this period, 317 and maybe an upward migration of the oromediterranean belt inferred by the high *Pinus* percentages.

An Early Holocene humid stage is noticed in other nearly sites, such as the south-faced Laguna de Río Seco (LdRS; Fig. 1) (Anderson et al., 2011), when the highest lake level of the Holocene occurred. This is also coeval with the dominance of arboreal species such as *Pinus* as well as aquatic and wetland plants (Anderson et al., 2011). Low eolian input, noted by geochemical ratios, is also recorded in LdRS during this interval (Jiménez-Espejo et al., 2014). Further indications of elevated humidity come from the northfacing Borreguil de la Virgen (BdlV) (see Fig. 1), which is dominated by an AP assemblage and a high occurrence of aquatic algae *Pediastrum* along with a higher lake level (Jiménez-Moreno et al., 2012).

Although the preponderance of evidence accumulated for the Early Holocene suggests overall humid
conditions, at least three relatively arid periods are identified with the geochemical data in the LH record
(Fig. 7). The first arid period occurred between ~9600 and 9000 cal yr BP, the second occurred ~8200 cal
yr BP and the third around 7500 cal yr BP.

329 The first arid event is characterized in LH by a decrease in K/Al and K/Ti ratios and MS, resulting from the 330 lower runoff input with the concomitant change to a more peaty composition. This event could be correlated 331 with a dryness event recorded in the Siles Lake record (Carrion, 2002) at ~9300 cal yr BP noticed by an 332 increase in *Pseudoschizaea*, which was coeval with a minor decrease in arboreal pollen also recorded in 333 several sites in North Iberia (Iriarte-Chiapusso et al., 2016). At marine site ODP 976 (Fig.1; Combourieu-334 Nebout et al., 2009) a decrease in deciduous Quercus occurred between 9500 and 9200 cal yr BP indicating 335 a rapid excursion towards arid conditions (Fig.7). The speleothem record of Corchia Cave also shows dryer 336 conditions during this interval (Fig. 7; Regattieri et al., 2014) In addition, a decrease in fluvial input in the 337 Southern Alps and an aridification phase in southeastern France and southeastern Iberia has been similarly 338 recorded (Jalut et al., 2000).

The second dry event recorded at ~8200 cal yr BP is depicted in LH record by a negative peak in K/Ti and K/Al ratios, and by the onset of a trend toward peatier lithology as evidenced by the MS profile. This event is not recognized in LH record as clearly as the 9500 cal yr BP and the 7500 cal yr BP dry events. A decrease in *Pinus* percentage is observed in the nearby LdRS (Anderson et al., 2011), while a forest decrease is recorded in the Alboran Sea sites MD95-2043 and ODP 976. In several records from north western Iberia, a decrease in arboreal pollen also occurred at this time (Iriarte-Chiapusso et al., 2016). The 8.2 ka event was the most rapid climate change towards cooler conditions occurred during the
Holocene. It was defined in Greenland ice cores by minimum values in δ18O and affected the North Atlantic
basin and the Mediterranean area (Alley et al., 1997; Rasmussen et al., 2007; Wiersma et al., 2011). Recent
simulations point to a fresh water input in North Atlantic which could slow down the North Atlantic Deep
Water (NADW) formation preventing the heat transport over the north hemisphere (Wiersma et al., 2010,
2011; Young et al., 2013).

351 Another dry event is recorded in LH at ~7500 cal yr BP evidenced by the higher peat content in the 352 sediment, as well as by the lower MS values and a relative minimum in the K/Ti ratio. A relative AP 353 minimum also occurred in LH at this time. This short-live event is depicted sharper than 8200 cal yr BP 354 event in several sites in southern Iberia and Alboran Sea: In the Padul record, located at 725 masl at the 355 lower part of Sierra Nevada a decrease in both evergreen and deciduous Quercus is interpreted as a dry and 356 cold event (Ramos-Román, 2018; Ramos-Román et al., 2018a); forest expansion in Guadiana valley during 357 the early-mid Holocene is interrupted by a xeric shrublands development between 7850 and 7390 cal yr BP 358 (Fletcher et al., 2007); in the Alboran Sea a decrease in deciduous Quercus is registered at site MD95-2043; 359 at site 300G a decrease in winter and summer temperatures is also recorded during this interval (Jiménez-360 Espejo et al., 2008); in lake Pergusa (south Italy) a trend toward arid conditions began at ~7500 cal yr BP 361 (Magny et al., 2012); in Corchia Cave an arid excursion occurred at ~7500 cal yr BP within an overall 362 humid period between 8300 cal yr BP and 7200 cal yr BP (Fig. 7; Regattieri et al., 2014).

363 Importantly, these arid events recorded in LH at 9600 to 9000 cal yr BP and 8200 cal yr BP are coeval with364 the ice-rafted debris events 6 and 5 defined by Bond et al. 1997 in North Atlantic.

365 5.1.2. Mid- and Late Holocene (~7000 cal yr BP-2600 cal yr BP)

366 The Middle and Late Holocene in the southern Iberian Peninsula is characterized by a trend towards more 367 arid conditions (Jalut et al., 2009; Anderson et al., 2011; Rodrigo-Gámiz et al., 2011; Jiménez-Moreno and 368 Anderson, 2012; Jiménez-Espejo et al., 2014). In the LH record an abrupt decrease in the MS values 369 indicates a lithological change to more peaty sedimentation at ~7000 cal yr BP. Similarly, a decrease in the 370 K/Al and K/Ti ratios, points to a transition to less humidity and runoff (Fig. 7). Quercus percentage 371 increases at this time, partially replacing the Pinus, which mainly compose the AP during the record. A 372 progressive increasing trend in eolian input from Sahara (Zr/Al, Ca/Al and Ca/Ti ratios) is observed around 373 5500-6500 cal yr BP (Fig. 7), also pointing to an increase in aridity in the area. This change coincides with 374 regional increases in the Zr/Th ratio (equivalent to Zr/Al ratio) and Artemisia pollen, and with decreases in 375 Betula and Pinus in the LdRS record (Anderson et al., 2011; Jiménez-Espejo et al., 2014), and in Pinus in 376 the BdlV record (Jiménez-Moreno et al., 2012). Rodrigo-Gámiz et al. (2011) and Jiménez-Espejo et al. 377 (2014) observed similar geochemical patterns in western Mediterranean marine records and in LdRS, with 378 a decline in fluvial input, and a decline in surface runoff, respectively. The same pattern is noticed in marine 379 pollen records MD95-2043 and ODP 976 (Fletcher and Sanchez-Goñi, 2008; Combourieu-Nebout et al., 380 2009; Fig. 7). Contemporaneously, aridity is also suggested from speleothem data around the Mediterranean 381 area: At El Refugio cave, a hiatus in the speleothem growing rate occurred between 7300 and 6100 cal year 382 BP (Walczak et al., 2015), which is coeval with a drop in δ 180 in Soreq (Israel) and Corchia (Italy; CC26; 383 Fig. 1 and 7) caves at 7000 cal yr BP (Bar-Matthews et al., 2000; Zanchetta et al., 2007; Regattieri et al.,

2014). Also at ~7000 cal yr BP a decreasing trend in the deciduous/sclerophyllous pollen ratio occurred in
southeastern France and Iberia (Jalut et al., 2000) and at continental sites around the Mediterranean Sea
(Jalut et al., 2009). In addition, very low lake levels were recorded in the Sahara-Sahel Belt (Liu et al.,
2007) and in the Southern Alps (Magny et al., 2002).

388 Enhanced arid conditions are observed in the LH record between 4000 and 2500 cal yr BP, interpreted 389 through a decline in AP and a Poaceae maximum. Also a surface runoff minimum and an increase in eolian 390 input proxies took place between 3500 and 2500 cal yr BP (zone LH-3). In Corchia Cave an arid interval 391 was recorded at ~3100 cal yr BP (Regattieri et al., 2014), coeval with another one observed globally and 392 described by Mayewski et al. (2004) between 3500 and 2500 cal yr BP. Nevertheless, this period is not 393 climatically stable, fluctuations are observed in in K/Ti, K/Al, Ca/Ti, Ca/Al and Zr/Al ratios. Furthermore, 394 peaks in Quercus are recorded in LH, LdlM and ODP 976 sites at ~3900 cal yr BP and ~3100 cal yr BP, 395 when AP in LH decreases (Combourieu-Nebout et al., 2009; Jiménez-Moreno et al., 2013). This fact a 396 priori contradictory, could be explained by altitudinal displacements of the tree taxa such as *Quercus* in the 397 oromediterranean belt due to the climatic variability occurred along this interval (Carrión, 2002). During 398 warmer periods, this species would be displaced towards higher elevation and the influence 399 of Quercus pollen in Sierra Nevada would be larger, this could explain relative higher Quercus percentages 400 in LdlM, LH and also in the ODP 976 record. The same relationship between Quercus and Pinus is 401 observed comparing the BdlC and Padul records, located closely but with large altitude difference (BdlC 402 ~2992 masl; Padul ~725 masl; Ramos-Román, 2018) where is also likely linked to movements in the 403 oromediterranean belt (Ramos-Román, 2018). These altitudinal displacements of the tree taxa have been 404 previously related to temperature changes in others southern Iberian records, suggesting an ecological niche 405 competition between Pinus and Quercus species at middle altitudes (see Carrión et al., 2002 for a revision).

406 5.1.3. Iberian Roman Humid Period (IRHP; ~2600-1450 cal yr BP)

Because there is no consensus in the literature about the chronology for the main climatic stages during the
last 2000 years (Muñoz-Sobrino et al., 2014; Helama et al., 2017), here we follow the chronology proposed
by Moreno et al. (2012): Dark Ages (DA, 1450-1050 cal yr BP); Medieval Climate Anomaly (MCA, 1050650 cal yr BP); and LIA (650-150 cal yr BP). Another climatic stage preceeds the DA – the Iberian Roman
Humid Period (IRHP, 2600-1600 cal yr BP), originally described by Martín-Puertas et al. (2008). However,
in the LH record we have established different IRHP limits (2600-1450), based accordingly to the pollen
zonation (Fig. 3), and coinciding with the DA onset defined by Moreno et al, (2012).

414 The IRHP has been described as the wettest period in the western Mediterranean from proxies determined 415 both in marine and lacustrine records during the Late Holocene (Reed et al., 2001; Fletcher and Sanchez-416 Goñi 2008; Combourieu-Nebout et al., 2009; Martín-Puertas et al., 2009; Nieto-Moreno et al., 2013; 417 Sánchez-López et al., 2016). A relative maximum in AP occurred in the LH record during this time, also 418 indicating forest development and relative high humidity during the Late Holocene in the area (zone LH-419 4; Fig. 7). This is further supported by high K/Al and K/Ti ratios and MS values, indicating high detrital 420 input in the drainage basin, a minimum in Poaceae and low Saharan eolian input (low Ca/Al, Ca/Ti and 421 Zr/Al ratios) (Fig. 7). Fluvial elemental ratios have also shown an increase in river runoff in Alboran Sea 422 marine records (Nieto-Moreno et al., 2011; Rodrigo-Gámiz et al., 2011). This humid period seems to be 423 correlated with a solar maximum (Solanki et al., 2004) and persistent negative NAO conditions (Olsen et

424 al., 2012), which could have triggered general humid conditions in the Mediterranean. However, in the LH 425 record fluctuation in AP between 2300 and 1800 cal yr BP occurred, pointing to arid conditions at that time. 426 This arid event also seems to show up in BdlC, with a decrease in AP between 2400 and 1900 cal yr BP 427 (Ramos-Román et al., 2016) and in Zoñar Lake, with water highly chemically concentrated and gypsum 428 deposition between 2140 and 1800 cal yr BP (Martín-Puertas et al., 2009). In Corchia Cave a rapid 429 excursion towards arid condition is recoded at ~2000 cal yr BP (Regattieri et al., 2014) (Fig.7). As we 430 explained in section 5, the apparently anomalous percentages of Pinus at this time, could be justified by an 431 upward migrations of the oromediterranean forest species triggered by higher temperatures and/or the high 432 pollen-production and dispersal of Pinus. Nevertheless, we cannot exclude others factors that could 433 influence the pollen transport such as the wind energy, mostly controlled by the NAO in the southern Iberia. 434 A persistent negative NAO phase, as occurred during the IRWP (Sánchez-López et al., 2016), would have 435 triggered more humid conditions and higher westerlies influence over southern Europe. The higher 436 occurrence of *Pinus* in the surrounding area due to the favourable climatic conditions, along with the higher 437 wind energy over Sierra Nevada and the characteristics of bissacate pollen, could have overstate the 438 percentages of Pinus in our record.

439 5.1.4. Dark Ages and Medieval Climate Anomaly (DA, MCA; 1450-650 cal yr BP)

Predominantly arid conditions, depicted by high abundance of herbaceous and xerophytic species and an AP minimum in the LH record, are shown for both DA and MCA (zone LH-5; Fig. 7). This is further supported in this record by an increase in Saharan eolian input Ca/Al, Ca/Ti and Zr/Al ratios, and by a decrease in surface runoff, indicated by the K/Al and K/Ti ratios (zone LH-5; Fig. 7). These results from LH agree with climate estimations of overall aridity modulated by a persistent positive NAO phase during this period (Trouet et al., 2009; Olsen et al., 2012), also previously noted by Ramos-Román et al. (2016) in the area (Fig. 7).

447 Generally arid climate conditions during the DA and the MCA have also been previously described in the 448 LdlM and BdlC records, shown by a decrease in mesophytes and a rise of xerophytic vegetation during that 449 time (Jiménez-Moreno et al., 2013; Ramos-Román et al., 2016). Several pollen records in south and central 450 Iberian Peninsula also indicate aridity during the DA and MCA, for example grassland expanded at Cañada 451 de la Cruz, while in Siles Lake a lower occurrence of woodlands occurred (Carrión, 2002). Also in Cimera 452 Lake low lake level and higher occurrence of xerophytes were recorded (Sánchez-López et al., 2016). Arid 453 conditions were depicted in Zoñar Lake by an increase in Pistacia and heliophytes (i.e., Chenopodiaceae) 454 and lower lake level (Martín-Puertas et al., 2010). Similar climatic conditions were noticed in the marine 455 records MD95-2043 and ODP 976 in the Alboran Sea through decreases in forest (Fletcher and Sánchez-456 Goñi, 2008; Combourieu-Nebout et al., 2009; Fig. 7). Arid conditions in Basa de la Mora (northern Iberian 457 Peninsula) occurred during this time, characterized by maximum values of Artemisia, and a lower 458 development of deciduous Quercus and aquatic species such as Potamogeton, also indicating low lake 459 water levels (Moreno et al., 2012). Arid conditions were also documented by geochemical data in marine 460 records from the Alboran Sea (Nieto-Moreno et al., 2013, 2015), in the Gulf of Lion and South of Sicily 461 (Jalut et al., 2009). Aridity has also been interpreted for central Europe using lake level reconstructions 462 (Magny, 2004) and in speleothems records in central Italy (Regattieri et al., 2014). Nevertheless, wetter

- 463 conditions were recorded during the DA in some records from northern Iberian Peninsula (Sánchez-López
- 464 et al., 2016). Humid conditions depicted by higher lake level and less salinity occurred in Arreo Lake
- 465 (Corella et al., 2013). In Sanabria Lake, the dominance of planktonic diatom Aulacoseira subborealis is
- 466 interpreted as relative humid conditions at that time (Jambrina-Enríquez et al., 2014). This heterogeneity in
- 467 the climate during the DA is due to the existence of an N-S humidity gradient in the Iberian Peninsula
- 468 (Sánchez-López et al., 2016). Nonetheless, this gradient seems to be more diffuse during the MCA, which
- 469 is characterized as an overall arid period in the entire Iberian Peninsula (Morellón et al., 2012; Sánchez-
- 470 López et al., 2016).

471 5.1.5. Little Ice Age (LIA; 650-150 cal yr BP)

472 The LIA is interpreted as an overall humid period in the LH record. This is indicated by higher AP values 473 than during the MCA, low Saharan dust input (low Ca/Al, Ca/Ti and Zr/Al ratios), a decrease in herbs 474 (Poaceae) and high values in the K/Al and K/Ti ratios indicating enhanced runoff (zone LH-6A; Fig. 7). 475 An increase in fluvial-derived proxies has been previously documented in other Iberian terrestrial records 476 such as Basa de la Mora Lake (Moreno et al., 2012), Zoñar Lake (Martín-Puertas et al., 2010) or Cimera 477 Lake (Sánchez-López et al., 2016) and marine records from the Alboran Sea basin (Nieto-Moreno et al., 478 2011, 2015). Lake level reconstructions in Estanya Lake, in the Pre-Pyrenees (NE Spain), have shown high 479 water levels during this period (Morellón et al., 2009, 2011), supporting our humid climate inferences. 480 Nevertheless, fluctuations in Artemisia during the LIA suggest an unstable period in Sierra Nevada (Fig. 481 8), in agreement with the high variability in *Pinus*, Artemisia, and water availability deduced from recent 482 high-resolution studies in the neighbour BdlC and BdlV records (Ramos-Román et al., 2016; García-Alix 483 et al., 2017). The same pattern occurred in several Iberian records (Oliva et al., 2018), revealing that the 484 LIA was not a climatically stable period and many oscillations at short-time scale occurred.

485 A persistently negative NAO phase, although with high variability, occurred during this period (Trouet et

486 al., 2009), which could explain the overall humid conditions observed in southern Europe. As in the Early
487 Holocene arid events, solar variability has been hypothesized as the main forcing of this climatic event
488 (Bond et al., 2001; Mayewski et al., 2004; Fletcher et al., 2013; Ramos-Román et al., 2016).

489 5.2. Industrial Period (IP; 150 cal yr BP-Present)

490 The IP is characterized by a sharp increase in the Pb/Al ratio in LH record(Fig. 8), suggesting more mining, 491 fossil fuel burning or other human industrial activities (García-Alix et al., 2013, 2017). This is coeval with 492 a rise in AP, which is also related to human activities such as Olea commercial cultivation at lower 493 elevations around Sierra Nevada or Pinus reforestation in the area (Fig. 7 and 8; Valbuena-Carabaña et al., 494 2010; Anderson et al., 2011). The same pattern has also been observed in others records from Sierra Nevada 495 (Jiménez-Moreno and Anderson, 2012; García-Alix et al., 2013; Ramos-Román et al., 2016), in Zoñar Lake 496 and the Alboran Sea records (Martín-Puertas et al., 2010). In addition, a progressively increasing trend in 497 Zr/Al and Ca/Al ratios is observed during the last two centuries, which could be related to increasing local 498 aridity and/or anthropogenic desertification, but also with a change in the origin and/or composition of the 499 dust reaching to the lake (Jiménez-Espejo et al., 2014), likely related to the beginning of extensive 500 agriculture and the concomitant desertification in the Sahel region (Mulitza et al., 2010).

501 5.3 Significance of the eolian record from Laguna Hondera

502 Saharan dust influence over current alpine lake ecosystems is widely known (Morales-Baquero et al., 503 2006a, 2006b; Pulido-Villena et al., 2008b; Mladenov et al., 2011, Jiménez et al., 2018). The most 504 representative elements of Saharan dust in LH record are Fe, Zr and Ca as shown by the PC2 loading (Fig. 505 5), where Ca and Fe directly affect the alpine lake biogeochemistry in this region (Pulido-Villena et al., 506 2006, 2008b, Jiménez et al., 2018). Zirconium is transported in heavy minerals in eolian dust (Govin et al., 507 2012) and has largely been used in the Iberian Peninsula and the western Mediterranean as an indicator of 508 eolian Saharan input (Moreno et al., 2006; Nieto-Moreno et al., 2011; Rodrigo-Gámiz et al., 2011; Jiménez-509 Espejo et al., 2014; Martínez-Ruiz et al., 2015, and references therein). High Zr content has also been 510 identified in present aerosols at high elevations in Sierra Nevada (García-Alix et al., 2017). Considering 511 the low weatherable base cation reserves in LH bedrock catchment area, calcium is suggested to be carried 512 by atmospheric input of Saharan dust into alpine lakes in Sierra Nevada (Pulido-Villena et al., 2006, see 513 discussion; Morales-Baquero et al., 2013). This is the first time that the Ca signal is properly recorded in a 514 long record from Sierra Nevada. This could be explained by higher evaporation rates at this site promoting 515 annual lake desiccation that could prevent Ca water column dissolution and using/recycling by organism, 516 preserving better the original eolian signal. These elements have an essential role as nutrients becoming 517 winnowed and recycled rapidly in the oligotrophic alpine lake ecosystem (Morales-Baquero et al., 2006b). 518 This phenomenon has also been observed in other high-elevation lakes where the phytoplankton is 519 supported by a small and continually recycled nutrient pool (e.g., Sawatzky et al., 2006).

520 The SEM observations further confirm the presence of Saharan dust in the lake sediments from LH and the 521 occurrence of Zircon, the main source of eolian Zr, which is relatively abundant (Fig. 6a). Quartz with 522 rounded morphologies (eolian erosion) are also frequent (Fig. 6b) in the uppermost part of the record as 523 well as REE rich minerals, such as monazite, which is typical from the Saharan-Sahel Corridor area 524 (Moreno et al., 2006) (Fig. 6c). In addition, the fact that the highest correlation between Ca and Zr occurred 525 after ~6300 cal yr BP, (r=0.57 p<0.005) along with the SEM observation and the low availability of Ca in 526 these ecosystems, could suggest that the beginning of Saharan dust arrivals to the lake including both 527 elements took place at this time, giving rise to the present way of nutrient inputs in these alpine lakes 528 (Morales-Baquero et al., 2006b; Pulido-Villena et al., 2006). The onset of Saharan dust input into southern 529 Iberia occurred prior to the end of the African Humid Period (AHP; ~5500 cal yr BP; deMenocal et al., 530 2000), as previously noticed in the nearby LdRS (Jiménez-Espejo et al., 2014) and in Alboran Sea (Rodrigo-531 Gámiz et al., 2011). This could suggest a progressive climatic deterioration in North Africa, which 532 culminated with the AHP demise and the massive Saharan dust input recorded in all records in Sierra 533 Nevada at ~3500 cal yr BP (Fig. 7).

534 6. Conclusions

The multiproxy paleoclimate analysis from LH has allowed the reconstruction of the vegetation and climate evolution in Sierra Nevada and southern Iberia during the Holocene, and the possible factors that have triggered paleoenvironmental changes. Climate during the Early Holocene was predominantly humid, with two relatively arid periods between 10000 and 9000 and at ~8200 cal yr BP, resulting in less detrital inputs and a change to more peaty lithology. The onset of an arid trend took place around 7000 cal yr BP, decreasing the runoff input in the area. A significant increase in eolian-derived elements occurred between
6300 and 5500 cal yr BP, coinciding with the AHP demise. An arid interval is recorded between 4000 and

542 2500 cal yr BP, with a vegetation assemblage dominated by xerophytes.

543 Relative humid conditions occurred in the area between 2500 and 1450 cal yr BP, interrupting the Late 544 Holocene aridification trend. This humid interval was characterized by expansion of forest vegetation, high 545 runoff input, and a more clayey lithology. However, during the DA and the MCA (1450-650 cal yr BP) 546 there was enhanced eolian input and an expansion of xerophytes, indicating increased arid conditions. In 547 contrast, the LIA (650-150 cal yr BP) was characterized by predominant humid conditions as pointed out 548 high runoff and low eolian input. The IP (150 cal yr BP-Present) is characterized in the LH record by the 549 highest values of the Pb/Al ratio, probably indicating fossil fuel burning by enhanced mining and metallurgy 550 industry. The increase in human activities at this time in this area can also be deduced by the expansion of 551 Olea cultivation at lower elevations and Pinus reforestation in the area.

552 Importantly, the LH record shows a unique and exceptional Ca signal derived from eolian input (high Ca-

553 Zr correlation) during the past ~6300 years in Sierra Nevada. The good preservation of the Ca record might

have been favoured by the high evaporation and the low lake depth, which could have prevented Ca column

555 water dissolution and its re-use by organisms. Our record indicate that present-day inorganic nutrient input

- 556 from Sahara was established 6300 yrs ago and lasted until the present, with variations depending on the
- 557 prevailing climate.

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2007.

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Lab Number	Depth (cm)	Dating Method	Age (14C yr BP±1σ)	Calibrated age (cal yr BP)2σ ranges
	0	Present	2012 CE	-63
Poz-72421	7	14C	40±40	29-139
D-AMS 008539	22	14C	1112±32	935-1078
D-AMS 008540	39	14C	2675±30	2750-2809
BETA-411994	44	14C	3350±30	3550-3643
BETA-411995	55.5	14C	5480±30	6261-6318
Poz-72423	57.5	14C	5510±50	6266-6405
Poz-72424	62	14C	6450±50	7272-7433
Poz-72425	74	14C	8620±70	9479-9778

Table 1. Age data for LH 12-03. All ages were calibrated using IntCal13 curve (Reimer et al., 2013) with

⁹⁶⁹ Clam program (Blaauw, 2010; version 2.2).

	Simulation							
Correlation	A		В		С		D	
Ca/Ca	0.63	p<0.01	0.50	p<0.01	0.57	p<0.01	0.54	p<0.01
(XRF)								
K/K (XRF)	0.53	p<0.01	0.64	p<0.01	0.56	p<0.01	0.65	p<0.01

Table 2. Simulation of proxy correlation. A) regular interpolation of 300 years sampling spacing. B) regular interpolation of 300 years sampling spacing and 5 data points moving average. C) regular interpolation of 150 years sampling spacing. D) regular interpolation of 150 years sampling spacing and 5 data point moving average.



1022	Figure 1. (a) Location of the Laguna Hondera (LH) in Sierra Nevada, southern Iberian Peninsula, along
1023	with other nearby records mentioned in the text. (1) El Refugio Cave stalagmite record (Walczak et al.,
1024	2015); (2) ODP 976 pollen record (Combourieu-Nebout et al., 2009); (3) MD95-2043 pollen record
1025	(Fletcher and Sánchez-Goñi, 2008); (4) CC26, Corchia Cave stalagmite record (Zanchetta et al., 2007;
1026	Regattieri et al., 2014). Sierra Nevada north-facing sites are encircled in red, south-facing sites are encircled
1027	in blue (LH: Laguna Hondera, the current study, is shown in bold). LdLM: Laguna de la Mula (Jiménez-
1028	Moreno et al., 2013); BdLV: Borreguil de la Virgen (García-Alix et al., 2012; Jiménez-Moreno and
1029	Anderson, 2012); LdRS: Laguna de Río Seco (Anderson et al., 2011; García-Alix et al., 2013; Jiménez-
1030	Espejo et al., 2014); BdlC: Borreguil de la Caldera (Ramos-Román et al., 2016; García-Alix et al., 2017)
1031	(b) Regional satellite photo of LH. The white line indicates the catchment area. (c) Photo of Laguna
1032	Hondera in September 2012, when the core was taken. Photo taken by Gonzalo Jiménez-Moreno. For the
1033	coloured figure, we refer the reader to the web version of this article.

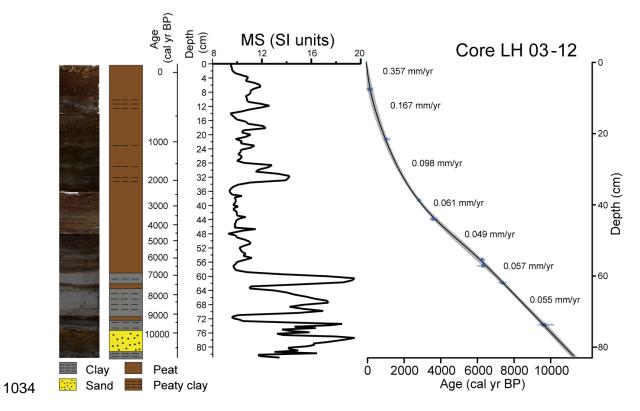


Figure 2. Photo of core LH 12-03, along with the lithology, magnetic susceptibility (MS, in SI units) profile
 and age-depth model. Sediment accumulation rates (SAR in mm yr ⁻¹) are shown between individual
 radiocarbon ages, the grey shadow represent the plus/minus range (see details in text for method of
 construction).

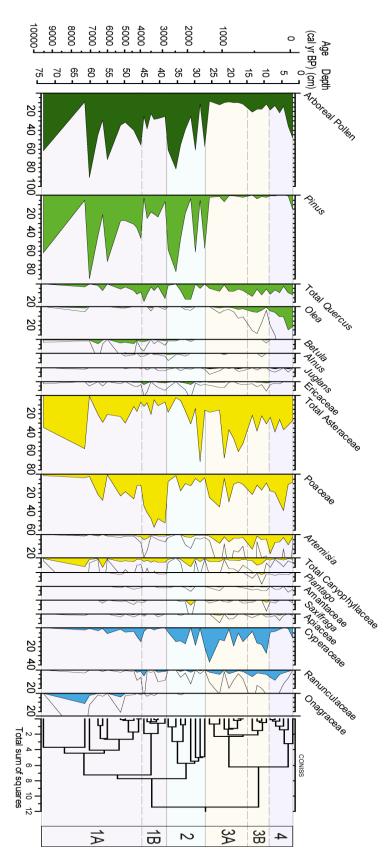


Figure 3. Pollen percentage diagram of the LH 12-03 record showing major selected taxa. Major tree
species are shown in green; shrubs and herbs are shown in yellow; and wetland and aquatic types are in
blue. Pollen was graphed with the Tilia program (Grimm, 1993), and zoned using the CONISS cluster
analysis program (Grimm, 1987).

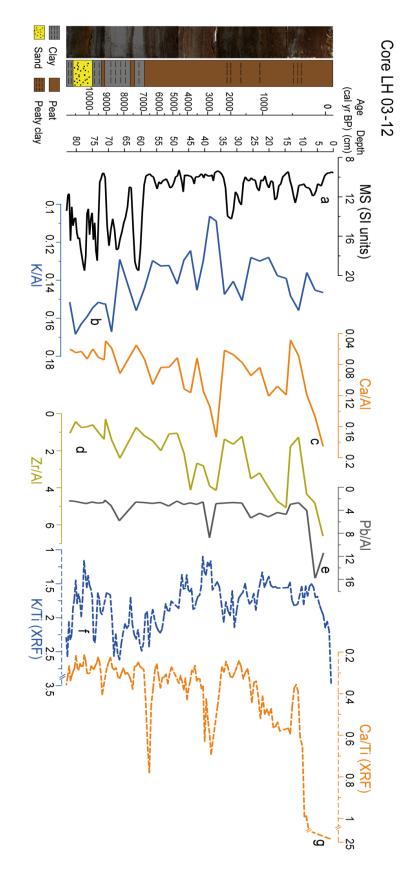
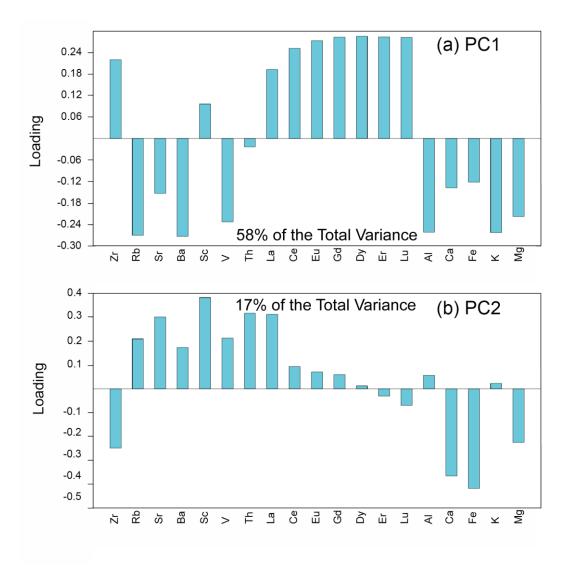


Figure 4. Detailed geochemical diagram of the LH 12-03 record showing the selected proxies: (a) MS; (b)
K/Al; (c) Ca/Al; (d) Zr/Al; (e) Pb/Al; (f) K/Ti (XRF); (g) Ca/Ti (XRF) (MS in SI units, Zr/Al and Pb/Al
scale x 10⁻⁴ and XRF in counts).



1049

1050 Figure 5. Principal Component Analysis (PCA) loadings from selected geochemical elements. (a) PC1,

1051 which describes 58% of total variance; (b) PC2, which describes 17% of total variance.

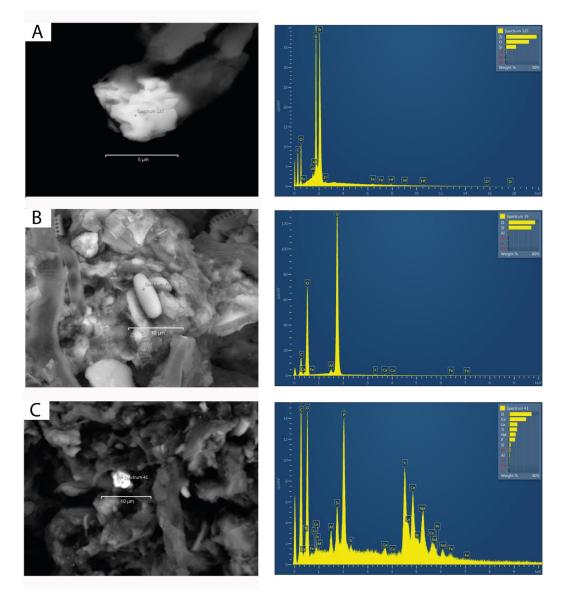




Figure 6. Electron Backscatter Diffraction microphotographs of the Laguna Hondera record with clearer
colours representing heavier minerals. (a) Zircon, with high Zr content (Dr. 01, 4-5 cm); (b) rounded quartz
related with eolian transport (Dr. 01, 2-3 cm); (c) monazite, with high REE content (Dr. 01, 2-3 cm).

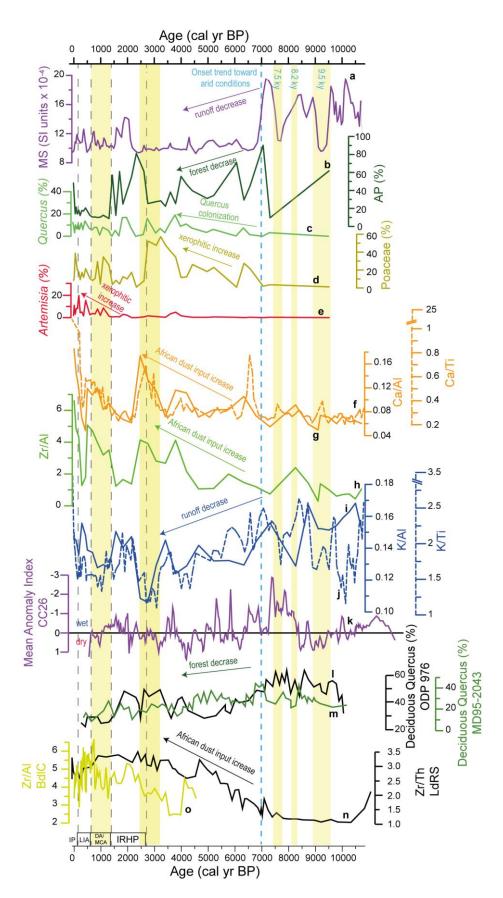


Figure 7. Comparison between the MS data (in SI units x 10⁻⁴), the most important pollen taxa and
geochemical proxies from Laguna Hondera (LH) record, with nearby paleoclimate records. (a) LH
Magnetic Susceptibility (MS) record; (b) Arboreal Pollen (AP) percentage from LH; (c) *Quercus*

- percentage from LH; (d) Poaceae percentage from LH; (e) Artemisia percentage from LH; (f) Ca/Ti (XRF) ratio from LH in dashed line; (g) Ca/Al ratio from LH; (h) Zr/Al ratio from LH; (i) K/Al ratio from LH; (j) K/Ti (XRF) ratio from LH in dashed line; (k) Mean Anomaly Index from CC26 record (Corchia cave; Regattieri et al., 2014); (1) Deciduous Quercus ODP 976 (Alboran Sea; Combourieu-Nebout et al., 2009); (m) Deciduous Quercus MD95-2043 (Alboran Sea; Fletcher and Sanchez-Goñi, 2008); (n) Zr/Th ratio from Laguna de Río Seco (LdRS) (Jiménez-Espejo et al., 2014; García-Alix et al., 2018); (o) Zr/Al ratio from Borreguil de la Caldera (BdlC) (García-Alix et al., 2017; 2018). Yellow bands indicate more arid intervals. Dark dashed lines are used for separating the different Current Era periods: IRHP: Iberian Roman Humid Period; DA: Dark Ages; MCA: Medieval Climate Anomaly; LIA: Little Ice Age; IP: Industrial Period. Blue dashed line indicates the onset of the trend toward arid conditions.

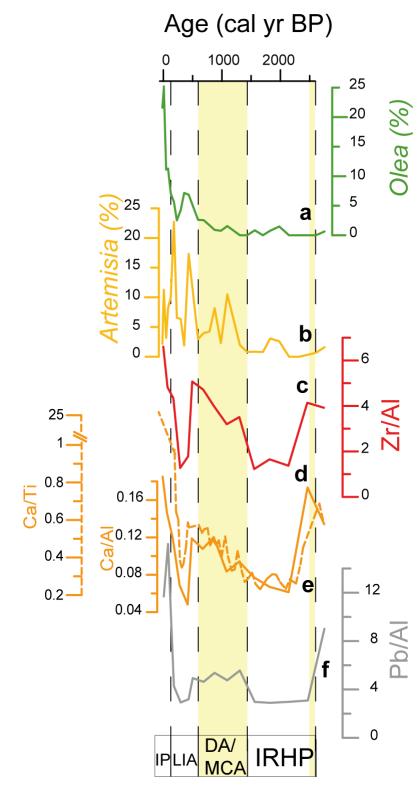


Figure 8. Comparison of geochemical proxies with pollen taxa, related to anthropogenic impact for the last
~2600 cal yr BP. (a) *Olea* percentage from LH; (b) *Artemisia* percentage from LH record; (c) Zr/Al ratio
from LH; (d) Ca/Al ratio from LH; (e) Ca/Ti (XRF) ratio from LH; (f) Pb/Al ratio from LH. Yellow bands
indicate more arid intervals. Dark dashed lines are used for separating the different Current Era periods:
IRHP: Iberian Roman Humid Period; DA: Dark Ages; MCA: Medieval Climate Anomaly; LIA: Little Ice
Age; IP: Industrial Period.