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#### Vegetation and geochemical responses to Holocene rapid 1

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

#### Hondera record 3

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#### 15 Abstract.

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

#### 1. Introduction

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- 37 Southern Spain has been the location for a number of recent studies detailing past vegetation and former
- 38 climate of the region (Carrión et al., 2001, 2003, 2007, 2010; Carrión, 2002; Combourieu Nebout et al.,
- 39 2009; Jiménez-Espejo et al., 2008; Martín-Puertas et al., 2008, 2010; Fletcher et al., 2010; Nieto-Moreno

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Discussion started: 3 May 2018

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40 et al., 2011, 2015; Rodrigo-Gámiz et al., 2011; Moreno et al., 2012 Jiménez-Moreno et al., 2015). These 41 studies have documented that the western Mediterranean area has been very sensitive to short-term 42 climatic fluctuations throughout the Holocene (e.g., Fletcher and Sánchez-Goñi, 2008; Combourieu 43 Nebout et al., 2009; Fletcher et al., 2010; Jiménez-Moreno et al., 2013). However, a subset of recent 44 studies have attempted to determine how Mediterranean alpine environments have been affected by 45 Holocene climate change through the study of sedimentary records from high elevation wetlands in the 46 Sierra Nevada (Anderson et al., 2011; García-Alix et al., 2012, 2013; Jiménez-Moreno and Anderson, 47 2012; Jiménez-Moreno et al., 2013; Jiménez-Espejo et al., 2014; Ramos-Román et al., 2016; García-Alix 48 et al., 2017). These alpine lake and bog records show minimal anthropic influence because they are 49 usually elevational higher than major regional Late Holocene human landscape modification. This allows 50 for a potentially clearer climatic signal to be determined from these sites. However, even though human 51 impact is less important at high-elevations, the impacts of human activities has been reconstructed from 52 these Late Holocene sedimentary records (Anderson et al., 2011; García-Alix et al., 2012, 2013; 2017). 53 Recent 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 56 lakes, where Saharan dust is especially important in conditioning plankton communities from oligotrophic 57 lakes (Morales-Baquero et al., 2006a, 2006b; Mladenov et al., 2008; Pulido-Villena et al., 2008b; Reche 58 et al., 2009). Although this eolian signal has been occasionally recorded in the sedimentary sequences 59 from the Sierra Nevada lakes (Jimenez-Espejo et al., 2014; García-Alix et al., 2017), the record of 60 inorganic nutrients in Saharan dust input in past lake geochemistry has remained elusive. This study 61 investigates a multiproxy sediment core record from Laguna Hondera (LH), located in the Sierra Nevada 62 range with three main goals: (1) identifying and characterizing climatic variability during the Holocene, 63 focusing on vegetation changes, eolian input and runoff sediments variations; (2) understanding the 64 Saharan dust influence in past lake sedimentation and geochemistry, and (3) investigating the 65 anthropogenic impact in the area.

### 2. Study Area

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

# 2.1. Regional Climate and Vegetation

- 76 Mediterranean climate characterises southern Iberia, with a marked seasonal variation between warm and
- dry summers and cool and humid winters (e.g. Lionello et al., 2006). Overprinting this general climate is

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Discussion started: 3 May 2018

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- 78 the influence of the North Atlantic Oscillation (NAO) (Trigo et al., 2004; Trouet et al., 2009). Southern
- 79 Iberia is also characterized by strong altitudinal contrasts, which in turn controls the precipitation
- 80 patterns, with mean annual values ranging from <400mm yr -1 to >1400 mm yr -1 in the southeast desert
- 81 lowlands and the southwest highland, respectively (Jiménez-Moreno et al., 2013 and references therein),
- demonstrating the complexity of climate regime in this area.
- 83 As with most mountainous regions, species and species groupings in the Sierra Nevada are distributed
- 84 with respect to elevation, depending on the temperature and rainfall gradients (e.g., El Aallali et al., 1998;
- Valle, 2003). Above 2800 m the crioromediterranean flora occurs as tundra-like open grassland. The
- oromediterranean belt (1900 -2800 m), mostly includes dwarf *Juniperus* (juniper), xerophytic shrublands
- and pasturelands and Pinus sylvestris and P. nigra. The supramediterranean belt (~1400 1900 m) is
- 88 characterized by mixed deciduous and evergreen forest species (i.e., evergreen and deciduous Quercus,
- 89 with *Pinus spp.* and others). Mesomediterranean vegetation (600 1400 m), includes sclerophyllous
- 90 shrublands and evergreen Quercus woodlands. The natural vegetation has been strongly altered by human
- 91 activities and cultivation in the last centuries, increasing significantly the abundance of *Olea* (olive), due
- 92 to cultivation at lower altitudes (Anderson et al., 2011, and references therein), and *Pinus* due to
- 93 reforestation primarily at higher elevations (Valbuena-Carabaña, 2010).

# 94 2.2. Laguna Hondera

- 95 Laguna Hondera (hereafter LH; 2899 masl; 37°02.88'N, 3°17.66'W, Fig. 1) is a small and shallow lake
- 96 located at the lowest elevation of a set of lakes locally named Cañada de Siete Lagunas, a glacial valley
- 97 between two of the highest peaks of the mountain range in the Iberian Peninsula: Alcazaba (3366m) and
- 98 Mulhacén (3479m). LH has a large catchment area (154.6 ha) compared with previously studied Sierra
- 99 Nevada wetlands (Laguna de Río Seco, LdRS, 9.9 ha; Borreguil de la Caldera, BdlC, 62 ha; Morales-
- 100 Baquero et al., 1999; Ramos-Román et al., 2016). The lake was reduced to a little pond in the deepest
- area of the basin when cored in September 2012, with a maximum depth of only a few centimetres.
- 102 LH presently occurs in the crioromediterranean vegetation belt (2800 m) (El Aallali et al., 1998; Valle 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-
- 108 03 (83cm) was selected for a multiproxy study because it was the longest core. Cores were wrapped with
- 109 tin 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 sediment features described. The magnetic susceptibility
- was measured every 0.5 cm with a Bartington MS2E meter in SI units (x 10-4) along the entire LH 12-03
- 112 core (Fig. 2). The sediment cores were subsampled every 1 cm for different analyses, i.e., one portion for
- pollen and another for geochemical analysis.

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Discussion started: 3 May 2018

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- 114 The age model was built using seven AMS radiocarbon dates from vegetal remains (Table 1; Fig. 2) by
- means of Clam software (Blaauw, 2010; version 2.2), which used the IntCal13 curve for radiocarbon age
- 116 calibration (Reimer et al., 2013). The smooth spline approach was chosen (Fig. 2). The sediment
- 117 accumulation rate (SAR) was calculated with the average rate from the Clam smooth spline output (Fig.
- 118 2).

### 119 3.2. Pollen

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

#### 130 3.3. Geochemical analyses

- 131 An X-Ray fluorescence (XRF) Avaatech core scanner®, located at the University of Barcelona, was used
- 132 to measure light and heavy elements in the LH 12-03 core. An X-ray current of 650 μA, a 10 second
- 133 count time and 10 kV X-ray voltage was used for measuring light elements, whereas 1700 µA X-ray
- 134 current, 35 second count time and 30 kV X-ray voltage was used for heavy elements. Sampling interval
- for these analyses was every 0.5 cm. For our study only three elements (K, Ca and Ti) have been
- 136 considered with enough counts to be representative.
- 137 Chemical composition was also determined on discrete samples every 2 cm. Prior to analysis, the samples
- 138 were dried in an oven and digested with  $HNO_3$  and HF. Inductively coupled plasma-optical emission
- 139 spectrometry (ICP-OES; Perkin-Elmer optima 8300) was used for major element analysis. Blanks and
- 140 international standards were used for quality control the analytical accuracy was higher than ± 2.79%
- and 1.89% for 50 ppm elemental concentrations of Al and Ca, respectively, and better than  $\pm$  0.44% for 5
- ppm elemental concentrations of K.
- 143 Trace element analysis was performed with an inductively coupled plasma mass spectrometry (ICP-MS;
- 144 Perkin Elmer Sciex Elan 5000). Samples were measured in triplicate through spectrometry using Re and
- 145 Rh as internal standards. The instrumental error is 2% for elemental concentrations of 50 ppm (Bea,
- 146 1996). All analyses were performed at the Instrumentations Center for Scientific Research (CIC),
- 147 University of Granada, Spain.

# 148 3.4. Mineralogical analyses

- 149 Morphological and compositional analyses were performed using scanning electron microscopy (SEM)
- 150 with an AURIGA model microscope (Carl Zeiss SMT) coupled with energy-dispersive X-ray

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Discussion started: 3 May 2018

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- 151 microanalysis (EDX) and Electron Backscatter Diffraction (EBSD) mode, also at the CIC (University of
- 152 Granada, Spain). Mineral grains were analysed to determine provenance, in particular those from eolian
- 153 origin.

### 154 3.5 Statistical Analysis

- 155 Principal components analysis (PCA) was run on the geochemical dataset using the PAST software
- 156 (Hammer et al., 2001). PCA finds hypothetical variables (components) accounting for as much as
- possible of the variance in multivariate data (Davis, 1986; Harper, 1999). The elements used in the PCA
- 158 were standardized by subtracting the mean and dividing by the standard deviation (Davis, 1986). Pb was
- 159 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).

#### 4. Results

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### 4.1. Lithology and magnetic susceptibility

- 163 The LH 12-03 sediment core consists primarily of peat in the upper ~60 cm, with mostly sand and clay
- 164 layers below (Fig. 2). Positive MS peaks coincide with the grey clay intervals between 58-72 cm. Peat
- 165 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 76-80 cm, a
- sandy oxidized interval occurs.

#### 4.2. Chronology and sedimentation rate

- The age –model of LH 12-03 documents that the record spans the last 10800 cal yr BP (Table 1; Fig. 2).
- 170 Sediment accumulation rates (SAR) were calculated using the average rate from the Clam smooth spline
- output (Fig. 2). The SAR below ~39 cm is very constant, varying between 0.049 and 0.061 mm yr<sup>-1</sup>. The
- 172 SAR increases exponentially to 0.098 mm yr<sup>-1</sup> at 22 cm, 0.167 mm yr<sup>-1</sup> at ~9 cm and 0.357 mm yr<sup>-1</sup> at
- 173 the core top. Accordingly with the model age and the SAR, resolution of pollen analysis varies between
- 174 ~40 years per sample in the top of the core and ~120 years per sample in the lower part. The resolution of
- 175 the geochemical analysis on discrete samples changes between 100-400 years per sample, but the
- 176 geochemical XRF core scanning resolution ranges between 15-100 years per sample, providing higher
- 177 resolution than geochemical data on discrete sample. The MS analyses resolution variates between 15-
- 178 100 years per sample.

### 4.3. Pollen

- 180 Fifty distinct pollen taxa were recognized, but only those with abundance higher than 1% are included in
- the pollen diagram (Fig. 3). Five pollen zones for the LH 12-03 record are identified, using variation in
- pollen species plotted in Figure 3 and a cluster analysis run through the program CONISS (Grimm, 1987).
- Zone LH-1 (core bottom-7000 cal yr BP) is defined by only three samples, due to the low preservation of
- pollen in this interval. Pollen in this zone is dominated by an alternation between Asteraceae and *Pinus*
- 185 (Fig. 3). Arboreal pollen (AP), composed primarily of Pinus, but also Quercus, reaches its maximum

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Discussion started: 3 May 2018

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- occurrence (90%) at ~7000 cal yr BP. The highest occurrence of Onagraceae pollen (~10%) takes place in
- this zone, and Caryophyllaceae reaches high values during this zone (~10%) as well. Only minor
- amounts of graminoids (Poaceae and Cyperaceae) occur during this period.
- 189 Zone LH-2 (~7000-4000 cal yr BP) is characterized by high percentages of tree species, primarily Pinus,
- 190 at the beginning of the zone (~90%), decreasing to ~55% at the upper part of the zone, with a minimum
- 191 (~30%) at 5000 cal yr BP. Quercus increases from ~2% at the beginning of the zone to ~10% at the end.
- The highest percentages of Betula pollen (~5%) in the record occurs at this time. Asteraceae pollen (~5-
- 193 30%) is less than in LH-1, but Poaceae increases from <5% at the opening of the zone to >25%.
- 194 Caryophyllaceae and Onagraceae continue to show relatively high values in this zone (~5% and ~6%,
- respectively). Cyperaceae occurs in high percentages (15%).
- 2008 Zone LH-3 (~ 4000-2600 cal yr BP) is defined primarily by a great increase in Poaceae pollen (to ~60%)
- 197 (Fig. 3). Other important herbs and shrubs include Asteraceae (5-15%) and Caryophyllaceae (~5%).
- 198 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 Onagraceae disappears altogether (Fig. 3).
- 201 Zone LH-4 (~ 2600-1450 cal yr BP) pollen assemblages show high variability in this zone. Pinus variates
- between ~80% to ~3% from the onset to the end of the zone. Aquatic pollen such as Cyperaceae (~15%)
- increases. On the other hand, an increase in herbs as Asteraceae (~5-70%) occurs along the zone, Poaceae
- variates between ~7-12%.
- 205 Zone LH-5 (~ 1450-600 cal yr BP) is characterized by an increase in herbaceous pollen, led by Poaceae
- 206 (~35% maximum during this zone), Asteraceae (~60% maximum during this zone after ~1000 cal yr BP)
- and Artemisia (~10%), and with the resulting decrease in AP. Since this zone to the present Quercus is
- the major component of AP instead of *Pinus*. Cyperaceae also shows a decrease, and Ranunculaceae
- 209 reaches ~ 5%.

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- 210 Zone LH-6 (~ 600 cal yr BP-present) is divided in two subzones. LH-6A (~ 600- 150 cal yr BP)
- documents an increase in Olea (~6%), Poaceae (20%), Caryophyllaceae (7%) and Artemisia (~2-20%).
- 212 Pinus (~2%) and Asteraceae (~60%) decrease in this period. Aquatic and wetland pollen show a rise
- 213 (Cyperaceae ~30%, Rannunculaceae ~10%). LH-6B (~ 150 cal yr BP-present) depicts a further increase
- 214 in Olea (~25%), Poaceae (~40%) and Artemisia (~10%).

# 4.4. Sediment composition

- 216 Results of the geochemistry are described following the pollen zonation previously defined (see above).
- 217 The XRF-scanning method relies on determining the relative variations in elements. Nevertheless the
- 218 presence of major variations in organic matter or carbonates makes it important to normalize the
- 219 measured count in order to obtain an environmentally relevant signal (Löwemark et al., 2011).
- 220 Aluminium and titanium normalizations are commonly used to discern possible fluctuations in the
- 221 lithogenic fraction (enrichment or depletion of specific elements), particularly in the terrigenous
- aluminosilicate sediment fraction (Van der Weijden, 2002; Calvert and Pedersen 2007; Martinez-Ruiz et
- Poor detection of Al can be related to either low Al content, or high organic and water content that

al., 2015). For this study, the XRF data were normalized to Ti since Al counts obtained were very low.

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Discussion started: 3 May 2018

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225 increase radiation absorption and affect the intensity of this light element, among other possibilities

226 (Tjallingii et al., 2007).

227 Since data spacing is different between the analyses on discrete samples and the XRF scanner, a linear

228 interpolation was performed with the purpose of equalizing the space of the different time series (150-300

229 years). Afterwards, the mobile average was worked out along the time series (taking into account the 5

230 nearest points) in order to easily identify trends by means of smoothing out data irregularities. The

231 obtained data were compared, and both XRF-scanner and discrete sample data showed a good correlation.

As a consequence, the geochemical proxies displayed higher time resolution than the discrete samples

233 (Table 2). Discrete sample and XRF data results are described together in order to simplify this section

234 (Fig. 4).

235 Zone LH-1 (core bottom- ~7000 cal yr BP) is typified by maximum values of K/Al and K/Ti ratios,

236 coinciding with the lowest values in Ca/Al, Ca/Ti and Zr/Al ratios. Pb/Al data show a stable pattern

during this interval. Nevertheless, between 10000-9000 cal yr BP and ~8200 cal yr BP the trends were

238 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.

240 Zone LH-2 (~7000-4000 cal yr BP) shows a decreasing trend in K/Al and K/Ti ratios, while an increasing

trend in Zr/Al, Ca/Al and Ca/Ti occurred. The Pb/Al ratio remains constant throughout this zone.

Zone LH-3 (~4000-2600 cal yr BP) documents an increase in Zr/Al, Ca/Al and Ca/Ti ratios, which

243 reaches a maximum at ~2600 cal yr BP. A K/Al and K/Ti minima occurs between ~3000 and ~2600 cal

yr BP. The Pb/Al ratio shows a positive peak at ~2800 cal yr BP.

Zone LH-4 (~2600-1450 cal yr BP) is characterized by low Ca/Al, Ca/Ti and Zr/Al ratios, with relatively

246 high K/Al and K/Ti ratios. The Pb/Al ratio shows a flat pattern, increasing at ~1500 cal yr BP.

Zone LH-5 (~1450- 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 period.

249 Zone LH-G6 (~ 650 cal yr BP- present) is divided in two subzones. During the LH-G6a subzone, low

250 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. LH-G6b is characterized by Zr/Al, Ca/Al,

252 Ca/Ti, K/Ti and Pb/Al maxima. Lower K/Al ratio occurs in this zone.

253 Several studies have demonstrated that PCA analysis of geochemical data can elucidate the importance of

254 different geochemical components driving the environmental responses in marine and lacustrine records

255 (Bahr et al., 2014; Yuan, 2017). We performed a PCA analysis of the LH geochemical data, which

256 yielded two significant components (Fig. 5). The first principal component (PC1) describes 58% of the

257 total variance. The main negative loadings for PC1 are Rb, Ba, Al, K, Ca, Mg and Sr, while large positive

258 loadings correspond to Zr and Rare Earth Elements (REE). The second principal component (PC2)

259 explains 17% of the total variance. The main negative loading for PC2 are Fe, Ca, Zr, Mg and Lu.

Positive loads correspond to Al, K, Ba, Sr and other elements.

261 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.

Other minerals such as zircon, rounded quartz and monazite were also identified (Fig.6).

Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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#### 264 5. Discussion

265 Pollen and geochemical proxies have been widely used for reconstructing vegetation changes and 266 environmental and climate variations in southern Iberia (e.g. Carrión, 2002; Sánchez-Goñi and Fletcher, 267 2008; Anderson et al., 2011; Nieto-Moreno et al., 2011; Jiménez-Moreno et al., 2012; Moreno et al., 268 2012; Fletcher and Zielhofer, 2013; Jiménez-Espejo et al., 2014; Ramos-Román et al., 2016). Variations 269 in the occurrences of arboreal taxa such as Pinus and other mesic species (e.g, Betula, Quercus), 270 indicating relative humid and warm conditions, and xerophytic species (e.g., Poaceae, Asteraceae, 271 Amaranthaceae, Artemisia), representing aridity, have been useful for reconstructing relative humidity 272 changes in southern Iberian (e.g. Carrión et al., 2001, 2007, 2010; Anderson et al., 2011; Jiménez-Moreno 273 et al., 2012, 2013, 2015; Ramos-Román et al., 2016). 274 Over 75% of the total geochemical data variance is explained by the PC1 and PC2 (Fig. 5). We interpret 275 the results of PC1 as resulting from certain sorting between heavy minerals (positive loading; Zr and 276 REE) vs. clay minerals and feldspars (negative loadings; K, Al and Ca). The drainage basin is composed 277 mainly by mica schist, consequently enhanced in K-rich minerals such as mica and feldspar (Díaz de 278 Federico et al., 1980). PC1 points to a sorting between heavy minerals (enriched in Zr and REE) and clays 279 and feldspars (enriched in K and Al) (Fig. 5a), probably linked to physical weathering within the basin 280 and to resulting runoff until final deposition in the lake. 281 On the other hand, we interpret the results of PC2 as differentiating autochthonous elements (positive 282 loadings) vs. Saharan allochthonous input (negative loadings). In the first case, due to the abundance of 283 mica schist within the LH drainage basin (Díaz de Federico et al., 1980), the K/Al and K/Ti ratios are 284 interpreted as detrital products, and thus a proxy of runoff. In the second case, PC2 negative loading Zr, 285 Ca, Mg and Fe (Fig. 5b) grouped elements that are coherent with Saharan input composition (dolomite, 286 iron oxides and heavy minerals) (Ávila, 1997; Morales-Baquero et al., 2006b; Pulido-Villena et al., 287 2007). In addition, Ca shows a strong positive correlation with Zr since 6300 cal yr BP (r =0.57; p<0.05) 288 supporting an eolian origin of the Ca in LH sediments. For instance, enrichment in heavy minerals such 289 as zircon and palygorskite has previously been used as an eolian proxy in the western Mediterranean (e.g., 290 Combourieu Nebout et al., 2002, Rodrigo-Gámiz et al., 2011, 2015). High concentrations of Ca in other lacustrine systems is usually associated with biogenic sources when anti-correlated with terrigenous 291 292 elements (Yuan, 2017). Nevertheless, elevated Ca in the LH record is linked with detrital elements, as 293 shown by PC1, where Ca is associated with K and Al (Fig. 5a). For these reasons Ca/Al and Ca/Ti ratios 294 are used in LH as eolian input proxies. 295 Elemental ratio variations, such as the ratios K/Al and K/Ti indicating fluvial input and ratios Zr/Al or 296 Zr/Th indicating aridity and eolian input, have been previously interpreted from Alboran Sea records as 297 well as in southern Iberia (Martín-Puertas et al., 2010; Nieto-Moreno et al., 2011, 2015; Rodrigo-Gámiz 298 et al., 2011; Jiménez-Espejo et al., 2014; Martínez-Ruiz et al., 2015; García-Alix et al., 2017). Thus, the 299 integration of both palynological data and geochemical ratios used as detrital input from LH have allowed 300 the reconstruction of the palaeoclimate and palaeoenvironmental history in Sierra Nevada during the

#### 5.1. Holocene palaeoclimate and palaeoenvironmental history

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Discussion started: 3 May 2018

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# 303 5.1.1. Early and Mid-Holocene humid conditions (core bottom – ~7000 cal yr BP)

304 The wettest conditions are recorded during the Early Holocene in Sierra Nevada. This is shown in the LH 305 record by the highest K/Al ratio and MS values, and the low values in Zr/Al, Ca/Al and Ca/Ti ratios, 306 suggesting that runoff dominated over eolian processes at this time (zone LH-1; Fig. 7) and agrees with 307 previous studies in the area (Anderson et al., 2011; Jiménez-Moreno and Anderson, 2012; García-Alix et 308 al., 2012; Jiménez-Espejo et al., 2014). Unfortunately, the pollen record from LH during this interval is 309 insufficient to definitely confirm this interpretation, due to the high detrital sediment composition and low 310 organic content, as shown by the low MS values and low pollen preservation. However, high percentages 311 of AP in two out of three analysed samples suggest humid conditions and high runoff during this period. 312 An Early Holocene humid stage is noticed in other nearly sites, such as the south-faced Laguna de Río 313 Seco (LdRS; Fig. 1) (Anderson et al., 2011), when the highest lake level of the Holocene occurred. This is 314 also coeval with the dominance of arboreal species such as Pinus as well as aquatic and wetland plants 315 (Anderson et al., 2011). Low eolian input, noted by geochemical ratios, is also recorded in LdRS during 316 this interval (Jiménez-Espejo et al., 2014). Further indications of elevated humidity come from the north-317 facing Borreguil de la Virgen (BdlV) (see Fig. 1), which is dominated by an AP assemblage and a high 318 occurrence of aquatic algae Pediastrum along with a higher lake level (Jiménez-Moreno et al., 2012). 319 Although the preponderance of evidence accumulated for the Early Holocene suggests overall humid 320 conditions, at least three relatively arid periods are identified with the geochemical data in the LH record 321 (Fig. 7). The first arid period occurred between ~9600-9000 cal yr BP, the second occurred ~8200 cal yr 322 BP and the third around 7500 cal yr BP. 323 The first arid event is characterized in LH by a decrease in K/Al and K/Ti ratios and MS, resulting from 324 the lower runoff input with the concomitant change to a more peaty composition. This event could be 325 correlated with a dryness event recorded in the Siles Lake record (Carrion, 2002) at ~9300 cal yr BP 326 noticed by an increase in Pseudoschizaea, which was coeval with a minor decrease in arboreal pollen also 327 recorded in several sites in North Iberia (Iriarte-Chiapusso et al., 2016). At marine site ODP 976 (Fig.1; 328 Combourieu-Nebout et al., 2009) a decrease in deciduous Quercus occurred between 9500-9200 cal yr 329 BP indicating a rapid excursion towards arid conditions (Fig.7). The speleothem record of Corchia Cave 330 also shows dryer conditions during this interval (Fig. 7; Regattieri et al., 2014) In addition, a decrease in 331 fluvial input in the Southern Alps and an aridification phase in southeastern France and southeastern 332 Iberia has been similarly recorded (Jalut et al., 2000). 333 The second dry event recorded at ~8200 cal yr BP is depicted in LH record by a negative peak in K/Ti 334 and K/Al ratios, and by the onset of a trend toward peatier lithology as evidenced by the MS profile. This 335 event is not recognized in LH record as clearly as the 9500 cal yr BP and the 7500 cal yr BP dry events. A 336 decrease in Pinus percentage is observed in the nearby LdRS (Anderson et al., 2011), while a forest 337 decrease is recorded in the Alboran Sea sites MD95-2043 and ODP 976. In several records from north 338 western Iberia a decrease in arboreal pollen also occurred at this time (Iriarte-Chiapusso et al., 2016). 339 The 8.2 ka event was the most rapid climate change towards cooler conditions occurred during the 340 Holocene. It was defined in Greenland ice cores by minimum values in δ18O and affected the North 341 Atlantic basin and the Mediterranean area (Alley et al., 1997; Rasmussen et al., 2007; Wiersma et al., 342 2011). Recent simulations point to a fresh water input in North Atlantic which could slow down the North

Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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343 Atlantic Deep Water (NADW) formation preventing the heat transport over the north hemisphere

344 (Wiersma et al., 2010, 2011; Young et al., 2013).

> Another dry event is recorded in LH at ~7500 cal yr BP evidenced by the higher peat content in the sediment, as well as by the lower MS values and a relative minimum in the K/Ti ratio. A relative AP minimum also occurred in LH at this time. This short-live event are depicted sharper than 8200 cal yr BP event in several sites in southern Iberia and Alboran Sea: In the Padul record, located at 744 masl at the lower part of Sierra Nevada a decrease in both evergreen and deciduous Quercus is interpreted as a dry and cold event (Ramos-Román et al., in review); forest expansion in Guadiana valley during the earlymid Holocene is interrupted by a xeric shrublands development between 7850-7390 cal yr BP (Fletcher et al., 2007); in the Alboran Sea a decrease in deciduous Quercus is registered at site MD95-2043; at site 300G a decrease in winter and summer temperatures is also recorded during this interval (Jiménez-Espejo et al., 2008); in lake Pergusa (south Italy) a trend toward arid conditions began at ~7500 cal yr BP (Magny et al., 2012); in Corchia Cave an arid excursion occurred at ~7500 cal yr BP within an overall humid period between 8300 cal yr BP and 7200 cal yr BP (Fig. 7; Regattieri et al., 2014).

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357 Importantly, these arid events recorded in LH at 9600-9000 cal yr BP and 8200 cal yr BP are coeval with

358 the ice-rafted debris events 6 and 5 defined by Bond et al. 1997 in North Atlantic.

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

The Middle and Late Holocene in the southern Iberian Peninsula is characterized by a trend towards more arid conditions (Jalut et al., 2009; Anderson et al., 2011; Rodrigo-Gámiz et al., 2011; Jiménez-Moreno and Anderson, 2012; Jiménez-Espejo et al., 2014). In the LH record an abrupt decrease in the MS values indicates a lithological change to more peaty sedimentation at ~7000 cal yr BP. Similarly, a decrease in the K/Al and K/Ti ratios, points to a transition to less humidity and runoff (Fig. 7). Quercus percentages increase at this time, partially replacing the Pinus which mainly compose the AP during the record. A progressive increasing trend in eolian input from Sahara (Zr/Al, Ca/Al and Ca/Ti ratios) is observed around 5500-6500 cal yr BP (Fig. 7), also pointing to an increase in aridity in the area. This change coincides with regional increases in the Zr/Th ratio (equivalent to Zr/Al ratio) and Artemisia pollen, and with decreases in Betula and Pinus in the LdRS record (Anderson et al., 2011; Jiménez-Espejo et al., 2014), and in Pinus in the BdlV record (Jiménez-Moreno et al., 2012). Rodrigo-Gámiz et al. (2011) and Jiménez-Espejo et al. (2014) observed similar geochemical patterns in western Mediterranean marine records and in LdRS, with a decline in fluvial input, and a decline in surface runoff, respectively. The same pattern is noticed in marine pollen records MD95-2043 and ODP 976 (Fletcher and Sanchez-Goñi, 2008; Combourieu-Nebout et al., 2009; Fig. 7). Contemporaneously, aridity is also suggested from speleothem data around the Mediterranean area: At El Refugio cave, a hiatus in the speleothem growing rate occurred between 7300-6100 cal year BP (Walczak et al., 2015), which is coeval with a drop in δ18O in Soreq (Israel) and Corchia (Italy; CC26; 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).

Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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382 Enhanced arid conditions are observed in the LH record between 4000-2500 cal yr BP, interpreted 383 through a decline in AP, a Poaceae maximum and a peak in Artemisia. Also a surface runoff minimum 384 and an increase in eolian input proxies took place between 3500-2500 cal yr BP (zone LH-3). In Corchia 385 Cave an arid interval was recorded at ~3100 cal yr BP (Regattieri et al., 2014), coeval with another one 386 observed globally and described by Mayewski et al. (2004) between 3500-2500 cal yr BP. Nevertheless, 387 this period is not climatically stable, fluctuations are observed in in K/Ti, K/Al, Ca/Ti, Ca/Al and Zr/Al 388 ratios. Furthermore, peaks in Quercus are recorded in LH, LdlM and ODP 976 sites at ~3900 cal yr BP 389 and ~3100 cal yr BP, when AP in LH decreases (Combourieu-Nebout et al., 2009; Jiménez-Moreno et al., 390 2013). This fact a priori contradictory, could be explained by altitudinal displacements of the tree taxa 391 such as Quercus in the oromediterranean belt due to the climatic variability occurred along this interval 392 (Carrión, 2002). During warmer periods, this species would be displaced towards higher elevation and the 393 influence of Quercus pollen in Sierra Nevada would be larger, this could explain relative 394 higher Quercus percentages in LdlM, LH and also in the ODP 976 record. The same relationship 395 between Quercus and Pinus is observed comparing the BdlC and Padul records, located closely but with 396 large altitude difference (BdlC ~2992 masl; Padul ~725 masl; Ramos-Román, 2018) where is also likely 397 linked to movements in the oromediterranean belt (Ramos-Román, 2018). These altitudinal displacements 398 of the tree taxa have been previously related to temperature changes in others southern Iberian records, 399 suggesting an ecological niche competition between Pinus and Quercus species at middle altitudes (see 400 Carrión et al., 2002 for a revision).

## 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, 1050-650 cal yr BP); and LIA (650-100 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).

410 The IRHP has been described as the wettest period in the western Mediterranean from proxies determined 411 both in marine and lacustrine records during the Late Holocene (Reed et al., 2001; Fletcher and Sanchez-412 Goñi 2008; Combourieu-Nebout et al., 2009; Martín-Puertas et al., 2009; Nieto-Moreno et al., 2013; 413 Sánchez-López et al., 2016). A relative maximum in AP occurred in the LH record during this time, also 414 indicating forest development and relative high humidity during the Late Holocene in the area (zone LH-415 4; Fig. 7). This is further supported by high K/Al and K/Ti ratios and MS values, indicating high detrital 416 input in the drainage basin, a minimum in Poaceae and low Saharan eolian input (low Ca/Al, Ca/Ti and 417 Zr/Al ratios) (Fig. 7). Fluvial elemental ratios have also shown an increase in river runoff in Alboran Sea 418 marine records (Nieto-Moreno et al., 2011; Rodrigo-Gámiz et al., 2011). This humid period seems to be 419 correlated with a solar maximum (Solanki et al., 2004) and persistent negative NAO conditions (Olsen et 420 al., 2012), which could have triggered general humid conditions in the Mediterranean. However, in the 421 LH record a decrease in AP between 2300-1800 occurred, pointing to arid conditions at that time. This

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Discussion started: 3 May 2018

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422 arid event also seems to show up in BdlC, with a decrease in AP between 2400-1900 cal yr BP (Ramos-

423 Román et al., 2016) and in Zoñar Lake, with water highly chemically concentrated and gypsum

deposition between 2140-1800 cal yr BP (Martín-Puertas et al., 2009). In Corchia Cave a rapid excursion

425 towards arid condition is recoded at ~2000 cal yr BP (Regattieri et al., 2014) (Fig.7).

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

427 Predominantly arid conditions, depicted by high abundance of herbaceous and xerophytic species and an

428 AP minimum in the LH record, are shown for both DA and MCA (zone LH-5; Fig. 7). This is further

429 supported in this record by an increase in Saharan eolian input Ca/Al, Ca/Ti and Zr/Al ratios, and by a

430 decrease in surface runoff, indicated by the K/Al and K/Ti ratios (zone LH-5; Fig. 7). These results from

431 LH agree with climate estimations of overall aridity modulated by a persistent positive NAO phase during

432 this period (Trouet et al., 2009; Olsen et al., 2012), also previously noted by Ramos-Román et al. (2016)

433 in the area (Fig. 7).

434 Generally arid climate conditions during the DA and the MCA have also been previously described in the

435 LdlM and BdlC records, shown by a decrease in mesophytes and a rise of xerophytic vegetation during

436 that time (Jiménez-Moreno et al., 2013; Ramos-Román et al., 2016). Several pollen records in south and

437 central Iberian Peninsula also indicate aridity during the DA and MCA, for example grassland expanded

438 at Cañada de la Cruz, while in Siles Lake a lower occurrence of woodlands occurred (Carrión, 2002).

439 Also in Cimera Lake low lake level and higher occurrence of xerophytes were recorded (Sánchez-López

et al., 2016). Arid conditions were depicted in Zoñar Lake by an increase in Pistacia and heliophytes (i.e.,

441 Chenopodiaceae) and lower lake level (Martín-Puertas et al., 2010). Similar climatic conditions were

442 noticed in the marine records MD95-2043 and ODP 976 in the Alboran Sea through decreases in forest

443 (Fletcher and Sánchez-Goñi, 2008; Combourieu-Nebout et al., 2009; Fig. 7). Arid conditions in Basa de

la Mora (northern Iberian Peninsula) occurred during this time, characterized by maximum values of

445 Artemisia, and a lower development of deciduous Quercus and aquatic species such as Potamogeton, also

446 indicating low lake water levels (Moreno et al., 2012). Arid conditions were also documented by

geochemical data in marine records from the Alboran Sea (Nieto-Moreno et al., 2013, 2015), in the Gulf

of Lion and South of Sicily (Jalut et al., 2009). Aridity has also been interpreted for central Europe using

449 lake level reconstructions (Magny, 2004) and in speleothems records in central Italy (Regattieri et al.,

450 2014).

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

452 The LIA is interpreted as an overall humid period in the LH record. This is indicated by higher AP values

453 than during the MCA, low Saharan dust input (low Ca/Al, Ca/Ti and Zr/Al ratios), a decrease in herbs

454 (Poaceae) and high values in the K/Al and K/Ti ratios indicating enhanced runoff (zone LH-6A; Fig. 7).

An increase in fluvial-derived proxies has been previously documented in other Iberian terrestrial records

456 such as Basa de la Mora Lake (Moreno et al., 2012), Zoñar Lake (Martín-Puertas et al., 2010) or Cimera

457 Lake (Sánchez-López et al., 2016) and marine records from the Alboran Sea basin (Nieto-Moreno et al.,
458 2011, 2015). Lake level reconstructions in Estanya Lake, in the Pre-Pyrenees (NE Spain), have shown

459 high water levels during this period (Morellón et al., 2009, 2011), supporting our humid climate

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Discussion started: 3 May 2018

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460 inferences. Nevertheless, recent high-resolution studies in Sierra Nevada (Ramos-Román et al., 2016;

461 García-Alix et al., 2017) and in several Iberian mountains (Oliva et al., 2018) have revealed that LIA was

and not a climatically stable period and many oscillations at short-time scale occurred.

463 A persistently negative NAO phase, although with high variability, occurred during this time period

464 (Trouet et al., 2009), which could explain the overall humid conditions observed in southern Europe. As

465 in the Early Holocene arid events, solar variability has been hypothesized as the main forcing of this

d66 climatic event (Bond et al., 2001; Mayewski et al., 2004; Fletcher et al., 2013; Ramos-Román et al.,

467 2016).

468

# 5.2. Anthropogenic impact in the southern Iberia

469 Previous studies, including the nearby LdRS record in Sierra Nevada, have shown that mining and

470 metallurgy activities commenced by ~4500 cal yr BP in this area (García-Alix et al., 2013, and references

471 therein), as shown by an enhanced Pb/Al ratio since this time. For the LH record, the first clear signal of

472 lead pollution from mining and smeltering occurred around 2800 cal yr BP, coinciding with the Late

473 Bronze Age (LBA) (3200-2800 cal yr BP) and the Early Iron Age (EIA) (2800-2500 cal yr BP) (zone

474 LH-3; Fig. 8). The same signal is also recorded in the nearby LdRS (García-Alix et al., 2013; Fig 8).

475 Many studies, including LdRS, have shown that the IRHP was the most important lead pollution period

476 prior to the IP (Settle and Patterson, 1980). However, the Pb/Al record from LH does not register

477 enhanced pollution at this time. This could be due to a local effect, such as a higher catchment area in LH

478 involving a high runoff input, supported by an increase in the K/Al and K/Ti ratios during this humid

479 period that could have diluted the Pb signal transported by eolian input. Also a regional effect, such as a

480 weaker dust mobilization due to the humid conditions prevailing at this time, or patchy pollution

481 distribution, could explain these diverse records.

482 An increasing trend in Artemisia, which points to a climatic or anthropic aridification, is coeval with

another Pb/Al peak that occurred during the MCA (Fig. 8). Increasing anthropic activities during this time

in the area are justified by the first appearance of coprophilous fungi such as *Sordiales* and *Sporormiella*,

which occurred in BdlC (Ramos-Román et al., 2016) and in LdRS (Anderson et al., 2011), suggesting

486 grazing activity at high altitudes in Sierra Nevada (Anderson et al., 2011; Jiménez-Moreno and Anderson,

487 2012; Ramos-Román et al., 2016). Maxima in Artemisia and coprophilous fungi in Sierra Nevada are also

488 reached during the last 500 years (Anderson et al., 2011; Jiménez-Moreno and Anderson, 2012; Ramos-

489 Román et al., 2016).

495

490 An increase in the Pb/Al ratio is recorded during the IP in the LH record (Fig. 8), suggesting more

491 mining, fossil fuel burning or other human industrial activities. This is coeval with a rise in AP, which is

492 also related to human activities such as Olea commercial cultivation at lower elevations around Sierra

493 Nevada or *Pinus* reforestation in the area (Valbuena-Carabaña et al., 2010; Anderson et al., 2011). The

494 same pattern has also been observed in others records from Sierra Nevada (Jiménez-Moreno and

496 records (Martín-Puertas et al., 2010). In addition, a progressively increasing trend in Zr/Al and Ca/Al

ratios is observed during the last two centuries, which could be related to increasing local aridity and/or

498 anthropogenic desertification, but also with a change in the origin and/or composition of the dust reaching

Anderson, 2012; García-Alix et al., 2013; Ramos-Román et al., 2016), in Zoñar Lake and the Alboran Sea

Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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499 to the lake (Jiménez-Espejo et al., 2014), likely related to the beginning of extensive agriculture and the

500 concomitant desertification in the Sahel region (Mulitza et al., 2010).

501 Therefore, the human impact in LH is mostly remarkable during the last two millennia. The comparison

502 with nearby records such as LdRS has also revealed that high-mountain lakes are very sensitive to human

activities (Anderson et al., 2011).

#### 5.3 Significance of the eolian record from Laguna Hondera

505 Saharan dust influence over current alpine lake ecosystems is widely known (Morales-Baquero et al., 506 2006a, 2006b; Pulido-Villena et al., 2008b; Mladenov et al., 2011), nevertheless, none of the previous 507 record preserved the relationship between elements found in present-day Saharan dust. The most 508 representative elements of Saharan dust in LH record are Fe, Zr and Ca as shown by the PC2 loading 509 (Fig. 5), where Ca and Fe directly affect the alpine lake biogeochemistry in this region (Pulido-Villena et 510 al., 2006, 2008b). Zirconium is transported in heavy minerals in eolian dust (Govin et al., 2012) and has 511 largely been used in the Iberian Peninsula and the western Mediterranean as an indicator of eolian 512 Saharan input (Moreno et al., 2005; Nieto-Moreno et al., 2011; Rodrigo-Gámiz et al., 2011; Jiménez-513 Espejo et al., 2014; Martínez-Ruiz et al., 2015, and references therein). High Zr content has also been 514 identified in present aerosols at high elevations in Sierra Nevada (García-Alix et al., 2017). Considering 515 the low weatherable base cation reserves in LH bedrock catchment area, calcium is suggested to be 516 carried by atmospheric input of Saharan dust into alpine lakes in Sierra Nevada (Pulido-Villena et al., 517 2006, see discussion; Morales-Baquero et al., 2013). This is the first time that the Ca signal is properly 518 recorded in a long record from Sierra Nevada. This could be explained by higher evaporation rates at this 519 site promoting annual lake desiccation that could prevent Ca water column dissolution and 520 using/recycling by organism, preserving better the original eolian signal. These elements have an essential 521 role as nutrients becoming winnowed and recycled rapidly in the oligotrophic alpine lake ecosystem 522 (Morales-Baquero et al., 2006b). This phenomenon has also been observed in other high-elevation lakes 523 where the phytoplankton is supported by a small and continually recycled nutrient pool (e.g., Sawatzky et 524 al., 2006). 525

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

Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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537 North Africa, which culminated with the AHP demise and the massive Saharan dust input recorded in all 538 records in Sierra Nevada at ~3500 cal yr BP (Fig. 7).

#### 6. Conclusions

539 540 The multiproxy paleoclimate analysis from LH has allowed the reconstruction of the vegetation and 541 climate evolution in Sierra Nevada and southern Iberia during the Holocene, and the possible factors that 542 have triggered paleoenvironmental changes. Climate during the Early Holocene was predominantly 543 humid, with two relatively arid periods between 10000-9000 and ~ 8200 cal yr BP, resulting in less 544 detrital inputs and a change to more peaty lithology. The onset of an arid trend took place around 7000 545 cal yr BP, decreasing the runoff input in the area. A significant increase in eolian-derived elements 546 occurred between 6300-5500 cal yr BP, coinciding with the AHP demise. An arid interval is recorded 547 between 4000-2500 cal yr BP, with a vegetation assemblage dominated by xerophytes. 548 Relative humid conditions occurred in the area between 2500-1450 cal yr BP, interrupting the Late 549 Holocene aridification trend. This humid interval was characterized by expansion of forest vegetation, 550 high runoff input, and a more clayey lithology. But during the DA and the MCA (1450-650 cal yr BP) 551 there was enhanced eolian input and an expansion of xerophytes, indicating increased arid conditions. In 552 contrast, the LIA (650-150 cal yr BP) was characterized by predominant humid conditions as pointed out 553 high runoff and low eolian input. 554 The first human impact signals in LH is recorded at ~2800 cal yr BP with a rise of Pb/Al ratio, coinciding 555 with the onset of mining in the Iberian Peninsula. The IP (150 cal yr BP-Present) is characterized in the 556 LH record by the highest values of the Pb/Al ratio, indicating fossil fuel burning by metallurgy industry, 557 enhanced of mining and other human activities. 558 Importantly, the LH record shows a unique and exceptional Ca signal derived from eolian input (high Ca-559 Zr correlation) during the past ~6300 years in Sierra Nevada. The good preservation of the Ca record 560 might have been favoured by the high evaporation and the low lake depth that could have prevented Ca 561 column water dissolution and its re-use by organisms. Our record indicate that present-day inorganic 562 nutrient input from Sahara was established 6300 yrs ago and lasted until the present, with variations

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depending on the prevailing climate.

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Discussion started: 3 May 2018

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Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018





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Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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931 List of tables

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 Clam program (Blaauw, 2010; version 2.2).

Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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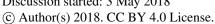


	Simulation								
Correlation		A		В		С		D	
Ca/Ca	0.63	p<0.01	0.50	p<0.01	0.57	p<0.01	0.54	p<0.01	
(XRF)									
K/K (XRF)	0.53	p<0.01	0.64	p<0.01	0.56	p<0.01	0.65	p<0.01	

**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.

Clim. Past Discuss., https://doi.org/10.5194/cp-2018-35 Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018







#### 984 List of figures







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Figure 1. (a) Location of the Laguna Hondera (LH) in Sierra Nevada, southern Iberian Peninsula, Mediterranean region, along with other nearby records mentioned in the text. (1) El Refugio Cave stalagmite record; (2) ODP 976 pollen record (Combourieu-Nebout et al., 2009); (3) MD95-2043 pollen record (Fletcher and Sánchez-Goñi, 2008); (4) CC26, Corchia Cave stalagmite record (Zanchetta et al., 2007; Regattieri et al., 2014). Sierra Nevada north-facing sites are encircled in red, south-facing sites are encircled in blue (LH: Laguna Hondera, the current study, is shown in bold). LdLM: Laguna de la Mula (Jiménez-Moreno et al., 2013); BdLV: Borreguil de la Virgen (García-Alix et al., 2012; Jiménez-Moreno and Anderson, 2012); LdRS: Laguna de Río Seco (Anderson et al., 2011; García-Alix et al., 2013; Jiménez-Espejo et al., 2014); BdlC: Borreguil de la Caldera (Ramos-Román et al., 2016; García-Alix et al., 2017) (b) Regional satellite photo of LH. The catchment area is indicated by the white line. (c) Photo of Laguna Hondera in September 2012, when the core was taken. Photo taken by Gonzalo Jiménez-Moreno

Clim. Past Discuss., https://doi.org/10.5194/cp-2018-35 Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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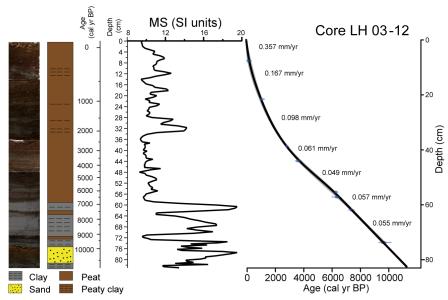


Figure 2. Photo of core LH 12-03, along with the lithology, magnetic susceptibility (MS, in SI units) 999 profile and age-depth model. Sediment accumulation rates (SAR in mm yr <sup>-1</sup>) are shown between 1000 1001 individual radiocarbon ages (see details in text for method of construction).

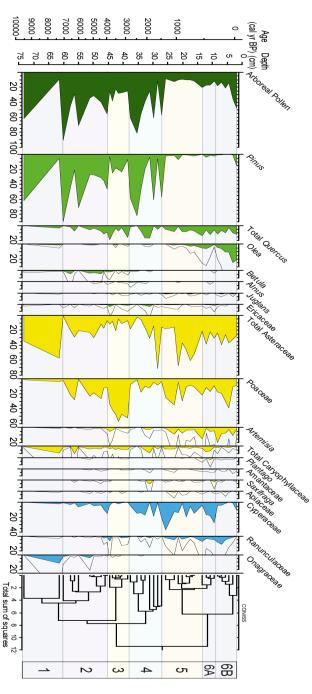
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Clim. Past Discuss., https://doi.org/10.5194/cp-2018-35 Manuscript under review for journal Clim. Past Discussion started: 3 May 2018

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**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).

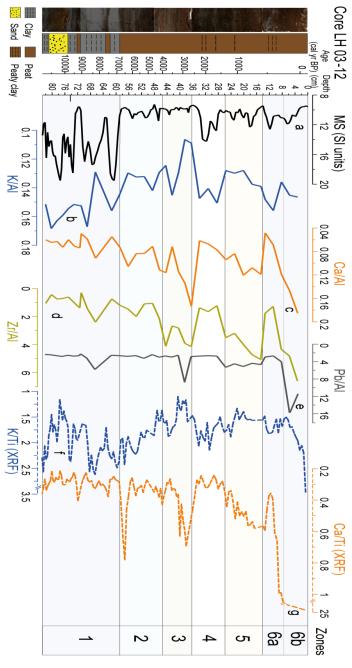
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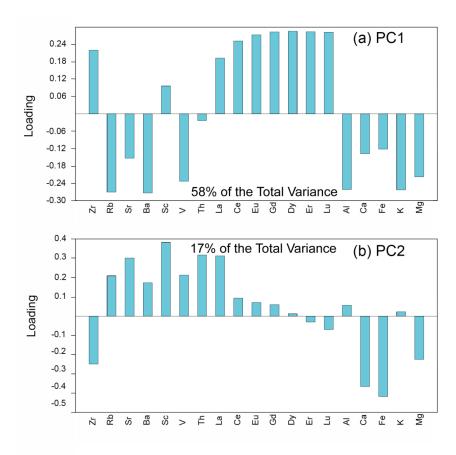
**Figure 4.** Detailed geochemical diagram of the LH 12-03 record showing the selected proxies: (a) MS; (b) K/Al; (c) Ca/Al; (d) Zr/Al; (e) Pb/Al; (f) K/Al (XRF); (g) Ca/Al (XRF) (MS in SI units, Zr/Al and Pb/Al scale x 10<sup>-4</sup> and XRF in counts). Pollen zonation described in section 4.3 was used.

Clim. Past Discuss., https://doi.org/10.5194/cp-2018-35 Manuscript under review for journal Clim. Past Discussion started: 3 May 2018 © Author(s) 2018. CC BY 4.0 License.





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**Figure 5.** Principal Component Analysis (PCA) loadings from selected geochemical elements. (a) PC1, which describes 58% of total variance; (b) PC2, which describes 17% of total variance.

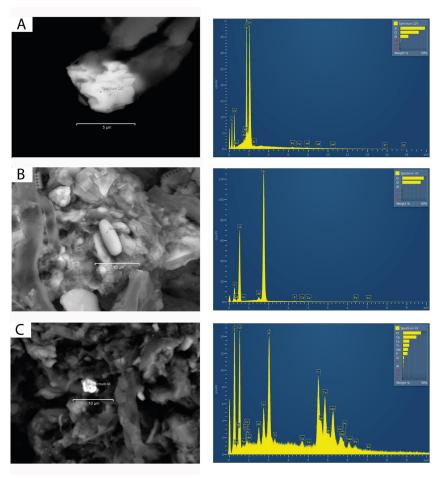
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**Figure 6.** Electron Backscatter Diffraction microphotographs of the LH record with clearer colours representing heavier minerals. (a) Zircon, with high Zr content (Dr. 01, 4-5 cm); (b) rounded quartz related with eolian transport (Dr. 01, 2-3 cm);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

Clim. Past Discuss., https://doi.org/10.5194/cp-2018-35 Manuscript under review for journal Clim. Past Discussion started: 3 May 2018

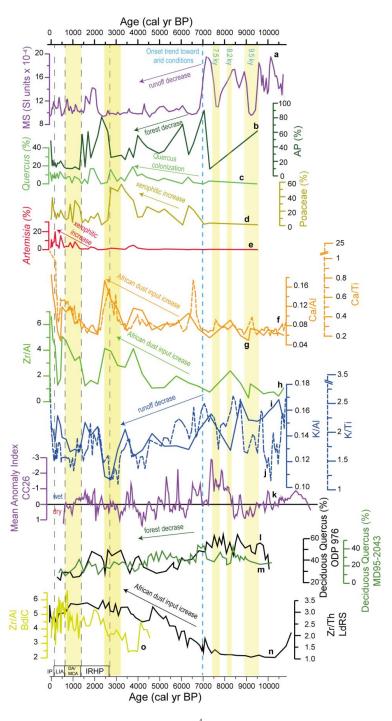
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**Figure 7.** Comparison of MS data (in SI units  $\times 10^{-4}$ ), the most important pollen taxa and geochemical proxies from LH 12-03 record, with nearby paleoclimate records. (a) LH Magnetic Susceptibility (MS) record; (b) Arboreal Pollen (AP) percentage from LH; (c) *Quercus* percentage from LH; (d) Poaceae

Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018





1023	percentage from LH; (e) Artemisia percentage from LH; (f) Ca/Ti (XRF) ratio from LH in dashed line;
1024	(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
1025	in dashed line; (k) Mean Anomaly Index from CC26 record (Corchia cave; Regattieri et al., 2014); (l)
1026	Deciduous Quercus ODP 976 (Alboran Sea; Combourieu-Nebout et al., 2009); (m) Deciduous Quercus
1027	MD95-2043 (Alboran Sea; Fletcher and Sanchez-Goñi, 2008); (n) Zr/Th ratio from Laguna de Río Seco
1028	$(LdRS); (o) \ Zr/Al \ ratio \ from \ Borreguil \ de \ la \ Caldera \ (BdlC). \ Yellow \ bands \ indicate \ more \ arid \ intervals.$
1029	Dark dashed lines are used for separating the different CE periods: IRHP: Iberian Roman Humid Period;
1030	DA: Dark Ages; MCA: Medieval Climate Anomaly; LIA: Little Ice Age; IP: Industrial Period. Blue
1031	dashed line indicates the onset of the trend toward arid conditions.
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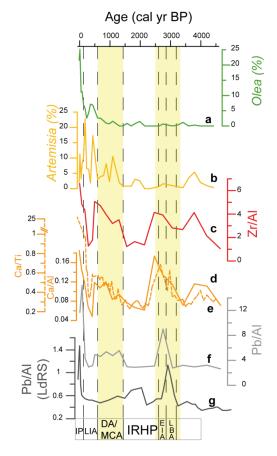
Clim. Past Discuss., https://doi.org/10.5194/cp-2018-35 Manuscript under review for journal Clim. Past

Discussion started: 3 May 2018

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Figure 8. Comparison of geochemical proxies with pollen taxa, related to anthropogenic impact for the last ~4500 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; (g) Pb/Al ratio from Laguna de Río Seco (LdRS). Yellow bands indicate more arid intervals. Dark dashed lines are used for separating the different CE and BCE periods: LBA: Late Broze Age; EIA: Early Iron Age; IRHP: Iberian Roman Humid Period; DA: Dark Ages; MCA: Medieval Climate Anomaly; LIA: Little Ice Age; IP: Industrial Period.