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Climate of the Past
Discussions

Extreme warming rates affecting alpine areas in SW Europe deduced from algal lipids

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Abstract. Alpine ecosystems of the southern Iberian Peninsula are among the most vulnerable and the first to respond to modern climate change in southwestern Europe. While major environmental shifts have occurred over the last ~1500 years in these alpine environments, only changes in the recent centuries have led to extreme responses, but factors imposing the strongest stress have been unclear until now. To understand these environmental responses, here, for the first time, we calibrated algal lipids (long-chain diols) to instrumental data extending alpine temperatures backward 1500 years. These novel results highlight the enhanced effect of greenhouse gases on alpine temperatures during the last ~200 years and the long-term modulating role of solar forcing. This study also shows that warming rates during the 20th century (~0.18ºC/decade) increased ~2.5 times with respect to the last stage of the Little Ice Age (~0.07ºC/decade), even exceeding temperature trends of the high-altitude Alps during the 20th century. As a consequence, temperature exceeded the pre-industrial threshold in the 1950s, being one of the major forcings of the enhanced recent change in the alpine ecosystems from southern Iberia. Nevertheless, other factors reducing the snow and ice albedo (i.e. atmospheric deposition) may have influenced local glacier loss, since steady climate conditions predominated from middle 19th century to the first decades of the 20th century.

1. Introduction

Global mean annual surface temperatures have risen by ~0.85ºC from 1880 to 2012 and the recent decades have been the warmest ones in the Northern Hemisphere during the Common Era (IPCC, 2013). This trend is alarming, since over the last decade temperature records have been broken yearly. For example, in Spain the highest temperatures ever recorded in September and July occurred in 2016 (45.5ºC) and 2017

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(46.9ºC-47.3ºC), respectively (Spanish National Weather Agency - AEMet Open Data, 2019). Increasing global temperatures are contributing not only directly to land and ocean surface warming, but are also changing the global hydrological cycle through the disturbance of atmospheric circulation patterns and moisture (Easterling et al., 2000; IPCC, 2013). As a result, the term "global warming" is migrating towards recent "global change" in order to express the variety of modern climate extremes witnessed across the world. The effects of modern global warming and associated climate change events may be causing extreme environmental impacts, beyond what is recorded in the recent geologic record (Waters et al., 2016). Hence, it is crucial to identify warming thresholds, rates, and forcing mechanisms from past high-resolution temperature records to understand modern climate change. It is especially important in fragile regions such as high elevation ecosystems of southern European mountains. This is the case for the Mediterranean alpine realm, an environmentally vulnerable biodiversity "Hot-Spot" (Schröter et al., 2005;Giorgi, 2006) where recent climate change is affecting species richness and distribution (Pauli et al., 2012;Médail and Quézel, 1999). Therefore, alpine wetlands in the Mediterranean region, such as the ones from the Sierra Nevada in the southern Iberian Peninsula, are sensitive recorders of changing climate and their sedimentological records archive the ecological and biogeochemical responses to different environmental forcings (Catalan et al., 2013).

In order to contribute to a better understanding of recent climate change events in these vulnerable areas, here, for the first time, we calibrate a recently developed algal-lipid temperature proxy in an alpine lacustrine record that overlaps with instrumental temperature time-series. This calibration allows the reconstruction of temperatures in alpine areas of the southern Iberian Peninsula during the Common Era when instrumental

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records are discontinuous or non-existent. Temperature-dependent biomarkers, such as those produced by algae (alkenones) or bacteria/archaea (glycerol dialkyl glycerol tetraether: GDGTs) had been commonly used in a wide range of marine records as quantitative paleothermometers, and their further application in lake environments has widely increased in the last decade (i.e., Castañeda and Schouten, 2011;Theroux et al., 2010;Longo et al., 2018;Colcord et al., 2015;Foster et al., 2016). A novel algal-lipid biomarker based on long-chain diols (LCDs), has also been applied as a temperature proxy in marine environments (Rodrigo-Gámiz et al., 2015;Rampen et al., 2012;Rampen et al., 2014b;Rodrigo-Gámiz et al., 2014); however, it has only been used tentatively in freshwater records (Rampen et al., 2014a). In this regard, studies using LCDs as different (paleo)environmental proxies in marine environments (not just only for temperature reconstructions) have increased in the last years, showing the potential of LCDs as upwelling proxies (Versteegh et al., 1997;de Bar et al., 2016), riverine inputs in marine settings (de Bar et al., 2016;Lattaud et al., 2017a), or nutrient proxies (Gal et al., 2018). Nevertheless, only a few studies have applied them for paleoenvironmental reconstructions, such as for paleoproductivity (Shimokawara et al., 2010), past rainfall anomalies (Romero-Viana et al., 2012), or paleotemperatures (Rampen et al., 2014a), among others. In any case, despite the great potential of LCDs for paleoenvironmental reconstructions, there are still a lot of ongoing questions about the applicability of diols at high latitude areas (Rodrigo-Gámiz et al., 2015), in freshwaters records (Rampen et al., 2014a), or even about their biological producers (Villanueva et al., 2014; Balzano et al., 2018).

The LCD distribution in marine environments shows significant correlations with mean annual sea surface temperature through the ratio of the fractional abundances of C_{28}

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1,13-diol, C_{30} 1,13-diol, and C_{30} 1,15-diol that are used in the Long Chain Diol Index (LDI, Eq. (1)) (Rampen et al., 2012). The use of this index is novel in freshwater environments and only a preliminary calibration with other indirect temperature proxies (i.e., GDGTs) is available (Rampen et al., 2014a). Here, we improve the biomarker paleothermometry by establishing the first temperature calibration for freshwater LCDs using a comparison with historical temperature records for the last ~100 years. These new data support and reinforce the use LCDs as a paleotemperature proxy in freshwater environments.

Equation (1) LDI= (F_{C30} 1,15-diol)/(F_{C28} 1,13-diol + F_{C30} 1,13-diol + F_{C30} 1,15-diol) (Rampen et al., 2012)

1.1 Regional settings

This paper focuses on the LCD record of two adjacent cores from Laguna de Río Seco (LdRS), a small alpine lake (~0.42 ha and less than 3m of water depth) at 3020 masl in the protected Sierra Nevada National Park, southern Spain (Fig. 1). Alpine Sierra Nevada wetlands, including LdRS, are low primary production (oligo-mesotrophic) systems and their biogeochemical cycles partially depends on aeolian nutrient supplies (i.e. Saharan aerosol deposition), since catchment basins are small and barren in nutrients (Pulido-Villena et al., 2005;Morales-Baquero et al., 2006;Reche et al., 2009).

Sierra Nevada is the southwestern-most mountain range in Europe, where latest Pleistocene cirque glaciers carved the metamorphic (mica schist) bedrock in the highest peaks (Castillo Martín, 2009). Massive glacier melting at the latest Pleistocene-Holocene

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transition transformed the former glacial depressions into wetlands (Castillo Martín, 2009) that evolved gradually into either shallow lakes or peatlands around the middle-to-late Holocene transition (Jiménez-Espejo et al., 2014;Garcia-Alix et al., 2017). Small glaciers re-appeared at the highest peaks of the Sierra Nevada in the 15th century, during the Little Ice Age, and remained until the 20th century (Oliva et al., 2018). The presence of these glaciers is observed in the sedimentary record in some alpine lakes in the Sierra Nevada (deposit of coarse sediments), like in Laguna de la Mosca (Oliva and Gomez-Ortiz, 2012). However, these kinds of deposits have not been registered in Laguna de Río Seco, where the last 1500 years are characterised by continuous laminated clays and bryophyte layers (Anderson et al., 2011). In any case, glacial effects have not caused any disturbance on wetland sedimentation (i.e., erosion), since local alpine sedimentary records show continuous sedimentation patterns (Anderson et al., 2011;Ramos-Román et al., 2016;García-Alix et al., 2012;Oliva and Gomez-Ortiz, 2012;Mesa-Fernández et al., 2018). During the 20th century this sensitive alpine region of southern Iberia has experienced significant impacts from modern climate change as evidenced, for example, by the first permanent European glacier loss there in the ~1920s (Grunewald and Scheithauer, 2010) and the extreme permafrost reduction during recent decades (Oliva and Gomez-Ortiz, 2012). As a consequence, this glacier and permafrost melting supplied a great amount of water (water availability) (Jiménez et al., 2018a) that boosted the occasional development of local aquatic environments, contrasting with the general environmental aridification trends observed throughout the 20th century (Garcia-Alix et al., 2017;Ramos-Román et al., 2016;Jiménez et al., 2018a).

The sedimentary archive of LdRS has been selected for this study in order to 1) improve the freshwater LCDs paleothermometry by proposing a new temperature

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calibration for freshwater LCDs , 2) reconstruct temperatures beyond the instrumental record in a site at the leading edge of changing climate, 3) assess the role of different radiative forcing (i.e., solar radiation or greenhouse gas concentrations) on temperature change in alpine wetlands of the southwestern Europe during the Common Era, and 4) understand the responses to recent climate change in this highly sensitive environment.

2. Materials and methods

2.1. Sediment sampling

Two sediment cores were taken at the deepest part of LdRS, an alpine lake at 3020 masl in the Sierra Nevada (southern Spain) (Fig. 1c). A long sediment core (150 cm) was retrieved in 2006 (LdRS lgc). A short sediment core of 16 cm was collected in 2008 (LdRS shc) using a slide-hammer gravity corer (Aquatic Research Instruments, Hope, Idaho, USA). Independent age models were performed in each sediment core to avoid potential correlation problems caused by changes in the sedimentation rates between both coring sites (Fig. 1c) and different sampling dates (2006-LdRS lgc and 2008-LdRS shc). The age model of LdRS lgc is based on ^{210}Pb and ^{137}Cs in the uppermost part (first 15 cm), and ^{14}C analyses in older sediments (Anderson et al., 2011). The age model of the LdRS shc is based on gamma spectroscopy by measuring the ^{210}Pb, ^{137}Cs, and ^{226}Ra radionuclides in the first ~14 cm, and afterwards the age was extrapolated to the core bottom (16 cm) (Jiménez et al., 2018b;Jiménez et al., 2018a). Both records show that the sediment accumulation rate for the uppermost 15-16 cm ranges between 0.13 and 0.9 cm/yr (Anderson et al., 2011;Jiménez et al., 2018b), and lower sedimentation rates below this depth (~0.008 cm/yr) (Anderson et al., 2011). Ages models show that the LdRS shc

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extends back to ~200 years with a sample resolution ranging from 5 to 7yr (high-resolution) (Jiménez et al., 2018b;Jiménez et al., 2018a). In the case of the LdRS lgc, the section studied in this paper covers the last ~1500 years with a lower sample resolution. In this case, the sample resolution is around 6-7 years in the first 10 cm, and from 24 to 150 years in older samples (Anderson et al., 2011).

2.2. Geochemical analyses

Thirty-two sediment samples were collected consecutively every 0.5 cm along LdRS shc and twenty samples in the first 22 cm of LdRS lgc. The samples were freeze-dried and homogenized. The total lipid content was extracted from the sediment samples using a Thermo Scientific™ Dionex™ ASE™ 350 Accelerated Solvent Extractor system at 100 ºC and 7 × 10^6 Pa using a mixture of dichloromethane (DCM) and methanol (9:1, v:v). Afterwards, the neutral fraction was separated by means of aminopropyl-silica gel chromatography using DCM:isopropanol (1:1, v:v). This neutral fraction was subsequently eluted with hexane, DCM, ethyl acetate:hexane (25:75, v:v), and methanol through a 230-400 mesh/35-70 micron silica-gel chromatographic column, in order to obtain four neutral sub-fractions (N1-N4). Long chain diols were obtained in the third neutral fraction (N3, alcohol fraction), which was derivatised by bis-(trimethylsilyl) trifluoroacetamide (BSTFA) before running the analyses. 30μL of BSTFA and 40μL of pyridine were added to each N3 fraction and heated at 80 °C for 2 hours. When vials were at room temperature, a volume between 140 μl and 220 μl of DCM was added to each sample. Firstly, the derivatised N3 fractions were analysed with a Gas Chromatography with Flame-Ionization Detector (GC-FID Shimadzu 2010) in order to obtain the proper concentration. Subsequently, they were measured in a Shimadzu QP2010-Plus Mass

Spectrometer interfaced with a Shimadzu 2010 GC using a scan mode between *m/z* 50 – 650, in order to obtain a general picture of the mass spectrum of the samples, and on the basis of a Selected-Ion Monitoring mode (SIM), selecting the characteristic fragment ions of the most important long chain diols, i.e. *m/z* 299, 313, 327, and 341(Rampen et al., 2012;Versteegh et al., 1997). Fractional abundances of the C_{28} 1,13-diol, C_{30} 1,13-diol, C_{30} 1,15-diol are used in Eq. (1) to calculate the Long chain Diol Index (LDI) (Rampen et al., 2012). Fractional abundances of C_{28} 1,13-diol, C_{30} 1,15-diol, and C_{32} 1,15-diol are used to characterise the potential diol source (i.e. marine, lacustrine, or specific algae groups) (Rampen et al., 2014a;Lattaud et al., 2018).

2.3 Reference temperature time-series for LDI temperature calibration

Generating an accurate diol-temperature calibration in alpine Sierra Nevada wetlands is challenging, because there is a lack of long and continuous temperature time-series at such high elevations. The meteorological observatories at the Sierra Nevada ski resort (ranging in elevation from 2500 to 3020 masl) only provided discontinuous temperature records from 1965 to 2011(Spanish National Weather Agency - AEMet Open Data, 2019;Observatorio del cambio global de Sierra Nevada, 2016) that show a significant correlation ($r>0.95$; $p<0.0001$) with low elevation temperature time-series (Table S1, S2). This lack of reference temperature time-series in Sierra Nevada alpine areas does not allow the development of a direct LDI-temperature calibration at high-elevation. Therefore, one potential way to obtain a LDI-temperature calibration is by means of the correlation of LDI data with long and reliable historical temperature time-series at nearby lower elevation areas, followed by a correction of the altitudinal effect on temperatures. After testing the correlations between LDI and different low elevation

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observatories at different scales (mean warm season and mean annual temperatures) (Table S3), we decided to develop the LDI temperature calibrations against the mean annual temperatures from Sevilla-Tablada and Madrid-Retiro observatories, since these are the longest temperature time-series in the region showing the highest correlation with LDI data (see Table S3 for further explanations and Fig. 1a for the location of these low-elevation observatories).

Two different groups of reference temperature time-series at 3020 masl have been estimated in order to overcome the scarceness of high-elevation temperature time-series in the Sierra Nevada and obtain a reliable mean LDI-temperature calibration: 1) based on the elevational gradient between low and high elevation observatories and 2) based on the direct correlation between temperature time-series from Madrid and Sevilla observatories and that at 3020 masl (Cetursa 5 observatory) in the Sierra Nevada, which is near LdRS and at the same elevation (Table S1).

Reference temperature time-series 1) The elevational gradient among the reference temperature time-series from Madrid, Sevilla and LdRS is high (there is a difference of more than 2200 m: Table S1); therefore, they cannot be used directly to calibrate the LDI data of LdRS. In order to solve this problem, the environmental lapse rate ($\Delta_{temperature}/\Delta_{elevation}$ in ºC/m) has been estimated between lower elevation observatories with long temperature time-series (Granada, Sevilla, and Madrid) and those from Sierra Nevada at higher elevation (with shorter temperature time-series: Albergue, and Cetursa 1, 3 and 5) (Table S1). This comparison between low and high elevation observatories show significant correlations ($r>0.95$; $p<0.0001$) (Table S2; Fig. S1). Due to very few annual data points from high elevation sites, monthly and annual (twelve continuous months) environmental lapse rates were calculated to compare both datasets. The calculated temperature shifts between the reference low elevation observatories and LdRS site at 3020 masl, worked out from Fig. S1 equations (Table S4), was applied to the temperature time series from Madrid and Sevilla for the last ~100 years in order to obtain a temperature reconstruction (from 1908 to 2008 CE) at 3020 masl.

Reference temperature time-series 2) The direct comparison between Madrid and Sevilla temperatures and those from the observatory Cetursa 5 (3020 masl) by means of Ordinary Least Square regressions has given rise to two equations (Fig. S2a and b) that allow the reconstruction of temperature time-series at 3020 masl from 1908 to 2008.

3. Results

3.1 Long chain diols in alpine lakes from southern Iberia

The distribution of the dominant long-chain diol isomers in LdRS (C_{28}, C_{30} and C_{32} 1,13- and 1,15-diols) is different from that of marine and lake sediments as well as of algal cultures and particulate organic matter published up until now (Rampen et al., 2014a;Lattaud et al., 2018) (Fig. 2a). This suggests that diol producers in LdRS are likely different from those identified so far in lacustrine environments and algal culture studies, making the diol puzzle even more complex than originally thought. Eustigmatophyceae algae have been commonly proposed as diol producers in freshwater environments (e.g. *Vischeria* sp., *Eustigmatos* sp.) (Volkman et al., 1999;Rampen et al., 2014a;Villanueva et al., 2014). Nevertheless, in the case of Sierra Nevada wetlands, where planktonic algae communities are very simple (Sánchez-Castillo, 1988), Eustigmatophyceae algae have

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not been identified so far (Barea-Arco et al., 2001;Sánchez-Castillo, 1988). Conversely, preliminary evidence would point instead to *Chromulina* spp. and/or other planktonic chrysophytes as potential diol producers in this area (Fig. S3). These observations suggest that further work is needed to unravel the real biological source for diols in these alpine wetlands.

The LDI record obtained from the C_{28}, C_{30} diols in LdRS depicts important fluctuations during the last ~1500 years, in agreement with the temperature trends during the Common Era. More specifically, LDI values in the long record, LdRS lgc, range from ~0.23 to 0.05 from ~400 to 1900 Common Era years (CE), with the maximum and minimum values at ~930 and ~1690 CE, respectively (Fig. 2b). These changes are coeval with the maximum temperatures of the Medieval Climate Anomaly (MCA) and with the minimum temperatures of the Little Ice Age (LIA). LDI fluctuations were extremely abrupt during the 20th century: from 0.10 to 0.31, according to the lowest resolution record (LdRS lgc) and from 0.13 to 0.32, following the highest resolution record (LdRS shc). These minimum and maximum values were reached in both cases during the first and last decades of the 20th century, respectively (Fig. 2b). As it can be observed, a significant Pearson correlation ($r>0.7$; $p<0.004$) exists among the LDI from LdRS shc and global (Hansen et al., 2010) and regional annual instrumental temperatures (Spanish National Weather Agency - AEMet Open Data, 2019) (Table S3, and S5).

3.2 LDI temperature calibration

An Ordinary Least Square Regression was run between the two groups of reference temperature time-series at 3020 masl and the LDI record from LdRS shc (Fig.

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3a). The obtained equation represents the relationship between mean annual temperatures (MAT) and LDI, providing a calibration equation (Eq. (2); Fig. 3a). The residual errors of the obtained temperature calibration, according to both the LDI-reconstructed temperatures and the reference temperature time-series, are lower than 0.8ºC, being the standard error 0.26ºC. The histogram showing the frequency of the residuals reveals that ~80% of the residuals range from 0.3 to -0.3. Apparently, data from 1973 slightly outlie (residuals ≦-0.60). This means that the residual errors would be lower than 0.6ºC and the standard error ~0.2ºC if these outliers were omitted (Fig. 3b).

Equation (2) MAT (ºC) =9.7028xLDI - 0.37501 (n=76; R^2= 0.81)

The application of the obtained calibration to the LDI values of LdRS provides the first temperature reconstruction for the Common Era in this alpine area (Fig. 2c). In order to estimate the real magnitude of temperature variations during this period, the Mean Annual Temperature Anomaly (MATA ºC) has been calculated with reference to the mean annual temperatures of the last 30 years of the record (2008-1979). The lowest temperatures were recorded between ~1600 and ~1780 CE, with a temperature anomaly ranging from ~ -2.0 to ~ -2.3 ºC. These temperature anomalies only reached positive values after 1998 (Fig. 2c).

4. Discussion

4.1 LdRS record in the environmental context of the Iberian Peninsula during the Common Era

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Abrupt changes in temperature and precipitation have been depicted during the last 2000 years in the Iberian Peninsula and surrounding marine areas (Sánchez-López et al., 2016;Moreno et al., 2012). Precipitation was highly variable, showing arid conditions during the MCA, especially in southern Iberia, overall humid conditions throughout the LIA (with a complex internal structure showing large variability in humidity and extreme events), and arid conditions for the Industrial Period (Moreno et al., 2012;Sánchez-López et al., 2016;Oliva et al., 2018;Rodrigo et al., 1999), especially in high elevation wetlands from southern Iberia (Anderson et al., 2011;Jiménez-Espejo et al., 2014;Garcia-Alix et al., 2017).

As far as reconstructed temperature variations are concerned, although the Early Middle Ages displayed a great temperature variability in the Iberian Peninsula and surrounding marine sites (Sánchez-López et al., 2016;Moreno et al., 2012), three main stages have been identified for the last millennium deduced from different proxies: a warm period throughout the MCA followed by cold temperatures during the LIA, ending in an abrupt warming in the second half of the 20th century (Moreno et al., 2012;Sánchez-López et al., 2016;Oliva et al., 2018). One of the proxies used to reconstruct such temperature variations in continental areas of the Iberian Peninsula were tree ring data. Long tree ring temperature archives of the Iberian Peninsula showed the same overall variations as the ones registered in LdRS, such as high temperatures before 1250 CE (Büntgen et al., 2017), some temperature drops coeval with solar minima during the LIA (i.e. the end of Spörer or Maunder Minima), as well as a period of moderate-low temperatures from ~1850 to ~1940, followed by a temperature increasing trend in the second half of the 20th century with several temperature drops between ~1960 and ~1990 (Büntgen et al., 2017; Tejedor et al., 2017). The same trends have been observed in the

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European summer temperatures deduced from tree ring records (Luterbacher et al., 2016) (Fig. 4b and 5b). Surprisingly, tree ring data from the Pyrenees and Iberian range show minor temperature variations, and even a slight temperature decrease from ~2000 to 2008, which is supported by the LdRS diol record (Fig. 5c). This temperature cooling/stabilization at the beginning of the 21st century is coeval with globally reduced warming rates over the 2001–2014 period (Fyfe et al., 2016).

Contrasting with these continental temperature reconstructions, high-resolution sea surface temperature (SST) estimations from marine sites surrounding the Iberian Peninsula, such as those derived from alkenones in the Tagus Delta (Iberian Atlantic Margin) or in the Balearic Islands (western Mediterranean Sea), showed a general decreasing trend for the last ~2000 years, with a warm MCA, a cold LIA, and cold/moderate temperatures for the Industrial Period that do not appear to mirror the modern global warming observed throughout the 20th century (Abrantes et al., 2005;Moreno et al., 2012). Only high-resolution alkenone and TEX_{86} (from GDGTs) derived-SST records of the cores 384B and 436B from the Alboran Sea (Nieto-Moreno et al., 2013) and the alkenone-SST record of core Gol-Ho1B from the Gulf of Lions (Sicre et al., 2016) have shown a clear temperature increase during the 20th century, similar to the LdRS LDI temperature record (Fig. 4a,c, 5a,c). The observed heterogeneity in the SST reconstructions based on biomarkers such as alkenones (Moreno et al., 2012;Abrantes et al., 2005;Rodrigo-Gámiz et al., 2014), GDGTs (Nieto-Moreno et al., 2013) or long chain diols (Rodrigo-Gámiz et al., 2014) could be explained by each record belonging to a different biogeographical area with specific temporal and dynamic oceanographic regimes, and potential dissimilar primary productivity patterns of each algal source (i.e. seasonality or bloom length) (Sicre et al., 2016).

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The previously described climate variability in precipitation and temperature during the last ~1500 years in the Iberian Peninsula have been explained by different forcing mechanisms such as the effect of the westerlies-North Atlantic climate dynamics, internal climate variability, solar irradiance, volcanism, or anthropogenic forcing (Gómez-Navarro et al., 2011;Gómez-Navarro et al., 2012;Moreno et al., 2012;Sánchez-López et al., 2016). Their potential effect on the LDI record from LdRS is discussed in the following section.

4.2. Control mechanisms on alpine temperatures in SW Europe during the Common Era

Solar, volcanic, and anthropogenic (i.e. CO_2 and CH_4) radiative changes, along with the internal variability are usually attributed as the leading factors controlling temperatures during the Common Era (IPCC, 2013;Ammann et al., 2007). In addition, North Atlantic climate dynamics such as the North Atlantic Oscillation (NAO) or the Atlantic Multidecadal Oscillation (AMO) are other potential drivers of natural climate variability in the Iberian Peninsula (O'Reilly et al., 2017;Moreno et al., 2012;Sánchez-López et al., 2016;López-Moreno et al., 2011). The control of the North Atlantic climate dynamics in the studied alpine wetlands is evident, at least for precipitation and humidity fluctuations, since the NAO and solar forcing have been described as the main controls on the paleoenvironmental evolution recorded in this area (Ramos-Román et al., 2016;Garcia-Alix et al., 2017). Conversely, other studies have shown that the NAO climate mode had little effects on temperatures in this alpine area from 1950 to 2006 CE (López-Moreno et al., 2011). LdRS data agree with this observation, since no correlation

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(Tables S5, S6) has been detected between the NAO reconstruction (Trouet et al., 2009) and the obtained LDI record for the last millennium. In the case of the AMO, it has an impact on the North Atlantic atmospheric blocking mechanisms (Häkkinen et al., 2011) and on the European and Mediterranean temperatures, especially during the AMO warm phases (O'Reilly et al., 2017). In the study area, the AMO shows a moderate long-term correlation (Figs. 4c,d and 5c,d $r>0.60$; $p<0.01$) with both long and short core derived-LDI records, but the correlation decreases when long-term trends are removed ($r<0.32$; $p>0.1$) (Tables S5, S6). Since the nature of the AMO and its specific drivers are still a matter of debate, i.e., internal ocean variability control (multidecadal fluctuations in the Atlantic Meridional Overturning Circulation) versus solar or volcanic forcing for the last centuries (Knudsen et al., 2014), we cannot conclude whether the observed correlations represent the sole effect of the AMO or the influence of its underlying forcing mechanisms.

The significant correlation at long and short terms ($r>0.61$; $p<0.005$) between LDI-records from LdRS and greenhouse gases (Schmidt et al., 2011) (Fig. 4c,g; Table S6), especially since the beginning of the 20th century (Industrial Period) (Fig. 5c,f; Table S5), suggests that greenhouse gases might have an important effect on temperatures at this high elevation site.

The potential impact of solar radiation and volcanic eruptions on climate over both short- and long-timescales is a topic of controversy in the literature (Ammann et al., 2007). In this regard, volcanic forcing, which should give rise to negative radiative forcing in the climate system (Ammann et al., 2007;Sigl et al., 2015), do not show a significant correlation with LDI-derived temperatures from LdRS over the last 1500 years

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(Fig. 4c,g and 5c,h; Table S5, S6). We suggest that this lack of influence at LdRS record is a function of its high-altitude location, at 3020 masl, in the free troposphere, which reduces the environmental impact of small volcanic tropospheric eruptions that likely have greater effects on lower elevation sites (Mather et al., 2013). In addition, the relatively short residence time of volcanic aerosols in the atmosphere mainly causes, at most, decadal-timescale effects (Sigl et al., 2015) that can be difficult to identify in most sedimentary records due to the age resolution, as in the case of older sediments than 200 years in LdRS. Nevertheless, large explosive volcanic eruptions delivering large amounts of stratospheric aerosols (Marotzke and Forster, 2015;Sigl et al., 2015), such as that for Agung Volcano in Bali, Indonesia (1963-1964 CE), may be associated with a small depression in LDI-derived temperatures observed in LdRS record (Fig. 5c,g). Although reconstructed-LDI cold temperatures occasionally seem to occur coevally with volcanic eruptions, for example, 560-510 and 320 years ago (~1450-1500 and 1690 CE) (Sigl et al., 2015), there is not a consistent relationship between the intensity and number of large eruptions and the reconstructed coolings in LdRS records, especially over the last ~200 years where the age sample resolution would be enough to detect them (LdRS shc).

Most of the above mentioned cooling events recorded in LdRS, such as those during the LIA, correspond to low solar activity periods like the Spörer Minimum (from ~1450 to 1550 CE) or the Maunder Minimum (from ~1645 to 1715 CE) (Stuiver and Quay, 1980) (Fig. 4 a,b,c,e,f). Thus, long-term correlations between LDI-derived temperatures and solar activity, based on reconstructions of the solar irradiance and cosmogenic isotopes (such as ^{14}C), are evident during the last ~1500 years in LdRS record ($r>0.69$; $p<0.002$) (Fig. 4c,e,f; Table S5). This correlation drops ($0.37< r <0.56$ and $0.04< p <0.14$) when long-term trends are removed (Table S6). The long-term solar influence

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agrees with previous observations in other alpine records of this area (Ramos-Román et al., 2016;Garcia-Alix et al., 2017). Solar activity slightly decreases its long-term influence in LdRS record during the last ~200 years ($r>0.56$; $p<0.001$) and disappears when long-term trends are removed (Table S5). Only some occasional temperature decreases or slower rates of warming such as during the 19th to 20th century transition, from ~1930 to 1940, from ~1960 to 1975, and around 1988 CE, correspond with slight declines in the total solar activity (Fig. 5c,e blue arrows).

In the same way, LdRS shc registered a small decrease in LDI-derived temperature (or stabilization) at the beginning of the 21st century (Fig. 5c), also recorded in the Madrid and Sevilla temperature time-series (Spanish National Weather Agency - AEMet Open Data, 2019), tree ring records of the the Pyrenees and Iberian Range (Tejedor et al., 2017;Büntgen et al., 2017), in marine platforms of the western Mediterranean (Fig. 5a) (Sicre et al., 2016) or globally (Fyfe et al., 2016). Although this slowdown agrees with a decreasing trend in solar activity and a slight stabilization of atmospheric methane concentrations, the causes are more complex, and probably related to a combination of internal variability and radiative forcing (i.e. volcanic and solar activity, or decadal timescale changes in anthropogenic aerosols) (Fyfe et al., 2016).

4.3. Exceeding natural thresholds in alpine areas

The LDI-derived temperature reconstructions from LdRS show that in the early 1950s temperature exceeded for the first time the highest temperature values reached in the record during the MCA (Fig. 4c). Although this means that the local temperature natural threshold was surpassed, the low sample density during the MCA precludes

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setting a robust turning point. In any case, temperature values increased by ~1.2 ºC in this alpine area after ~1950 CE, under full anthropogenic influence. The comparison between pre-industrial and post-industrial scenarios in the study site highlights the human impact on natural trends. The temperature increase during the last stages of the LIA (from ~1690 to ~1850 CE), an analogue for a non-anthropogenic temperature-increase scenario, was ~1.2 ºC (~0.07 ºC/decade; Fig. 6), whereas the mean temperature rise throughout the 20th century was ~1.8 ºC (~0.18 ºC/decade; Fig. 6). Although this means that on average, the warming rate was 2.5 times faster throughout the 20th century than at the end of the LIA (Fig. 6), these observations are based on a low sample density for the LIA, which might slightly increase the uncertainty for this period. By comparison, average global temperatures rose by ~0.85 ºC from 1880 to 2012 CE, corresponding to 0.06 ºC/decade (IPCC, 2013), which highlight the high-elevation amplification effect of temperatures on this vulnerable area.

An even more alarming result is that other European alpine areas in the Mediterranean region, such as those from the Alps, experienced a slower warming rate during the 20th century (~0.11 ºC/decade) (Fig. 6) (Auer et al., 2007;Böhm et al., 2010). This is ~1.6 times slower than the warming rate recorded in the Sierra Nevada. This evidence, along with the generally smaller amount of precipitation in the alpine areas of the western Mediterranean region (Auer et al., 2007;Rodrigo et al., 1999), allows us to conclude that the 20th century environmental stress in this area was greater than in the Alps.

Future scenarios are not optimistic for Sierra Nevada alpine areas either as projected temperature may rise at least ~1.4 ºC by the end of the 21st century (Fig. 7c).

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This means that MAT at ~3020 masl would increase to ~4 ºC, which is more than 1.5 times higher than present mean temperatures (ºC). This projection exceeds the ones from the CMIP5 models discussed in the IPCC-2013 report for the end of the 21st century under a low emission scenario (RCP2.6). It is closer to the average temperature increase under the medium-low emission scenario (RCP4.5), and in the low range of a medium-high emission scenario (RCP6.0) (IPCC, 2013). The projected temperature increase in Sierra Nevada would give rise to abrupt and foreseeable environmental consequences such as an evapotranspiration increase and a dramatic reduction in the snow cover. This would consequently amplify the observed decrease in water availability in this semiarid alpine area during the 20th century (Garcia-Alix et al., 2017;Jiménez et al., 2018b) and thus, affecting the water reservoirs for the human population at lower elevations. This scenario would impact on more than 500,000 people in the city of Granada and neighbouring towns, on more than 20,000 ha of irrigated agriculture in southwestern Spain, and would reduce the hydroelectricity production in this area. This would have a more significant impact locally in Sierra Nevada, where endemic and endangered species inhabit (Munguira and Martin, 1993;Blanca, 2001). The projected warming would amplify the gradual reduction in alpine wetland areas that has been observed during the last millennia as a consequence of long-term natural variations along with the superimposed effect of the human-induced environmental change (Garcia-Alix et al., 2017). Alpine wetlands in the southern Iberian Peninsula might disappear under this scenario, resulting in an environmental crisis.

4.4. Impact on the southwesternmost European alpine glaciers

The studied alpine area supported the southernmost glaciers in Europe during the LIA. Glaciers and permanent snow fields below ~3000 masl, such as those of Corral del Mulhacen (~2950 masl) whose last mention in the literature was between 1809 and 1849 CE (Oliva and Gomez-Ortiz, 2012), would have totally disappeared by the decrease in regional precipitation at the beginning of the 19th century (Rodrigo et al., 1999). The climatic features at the end of the 19th century and the beginning of the 20th century did not allow this glacier to re-establish itself (Fig. 7). Post-LIA climatic conditions have also been proposed as the trigger for the melting of the Corral del Veleta Glacier in Sierra Nevada (~3100 masl) at the beginning of the 20th century (Garcia-Alix et al., 2017;Oliva and Gomez-Ortiz, 2012;Oliva et al., 2018). However, our new temperature record show that temperatures did not exceed the levels of the 1850s until the late 1940s CE. Precipitation was low in the southern Iberian Peninsula during the first half of the 20th century, but similar, and even lower, precipitation values were registered before ~1850 CE (Rodrigo et al., 1999;Spanish National Weather Agency - AEMet Open Data, 2019) (Fig. 7). Therefore, how could the glacier have retreated under this almost steady-state scenario? A similar paradox has been described in the Alps (Painter et al., 2013), where glaciers began to sharply retreat after the mid-19th century, even though temperature and precipitation records would suggest that glacier expansion should have occurred at least until the first decades of the 20th century. In this case the trigger of the glacial retreat was the industrial black carbon deposition that amplified the solar radiation absorbed at the snow surface and caused its subsequent melting - not a temperature or precipitation change (Painter et al., 2013). Our data suggest that temperature and precipitation were not the only drivers of glacial retreat that led to the melting of permanent glaciers in the Sierra Nevada in the 1920s. Instead, mirroring the case of the Alps, it is plausible that other factors reducing the albedo, such as enhanced atmospheric deposition may have

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played a strong role. In this regard, important atmospheric depositional events have been recorded in the study alpine sites of southern Iberia from the mid-19th century to the first decades of the 20th century caused by both enhanced North African dust fluxes (Mulitza et al., 2010) (Jiménez et al., 2018b) as well as a spike in atmospheric pollution (as observed in anthropogenic Pb and Hg records in Sierra Nevada; Fig. 7) (Garcia-Alix et al., 2017;Garcia-Alix et al., 2013). Similarly, both phenomena have been demonstrated as triggers for glacier retreat (Painter et al., 2013) and snow melt in the Alps (Di Mauro et al., 2018).

Melting of the last glaciated area in the Sierra Nevada during the first decades of the 20st century (Grunewald and Scheithauer, 2010) represents an important turning point regarding recent environmental change in this alpine region (Garcia-Alix et al., 2017;Jiménez et al., 2018a). The rapid pace of environmental change in the area after this date is attributed to an amplified effect of warming and aridification (Fig. 7b,c) that increased stress on vulnerable ecosystems (Garcia-Alix et al., 2017;Jiménez et al., 2018b;Jiménez et al., 2018a) with little hope for return of local glaciers.

5. Concluding remarks

This study shows the vulnerability of alpine regions and the importance of their monitoring for a better understanding of climate variability and future rapid responses. In this regard, algal-derived biomarkers from LdRS records have given rise to the first long-chain diol temperature calibration in freshwater environments by means of the comparison with instrumental temperature data. The combination of a short and long sediment core has provided both a highly accurate LDI-temperature calibration for the

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instrumental period and a long-term historical perspective on the modern warming. This approach delivers a better time-integrated temperature model than discrete temperature measurements for the 20th century.

The low sample resolution in the longer core before~1500 CE does not allow us to totally constrain the main natural controls on temperatures in this high-elevation site for the whole Common Era. However, the general trends support that the presumed primary effect of greenhouse gases on temperatures reconstructed from algal-lipids in this alpine region of southern Iberia is likely modulated by long-term solar forcing. In recent times, greenhouse gases seem to be the major temperature driver in this high elevation site. Volcanic forcing appears to have little effect on reconstructed temperatures in this alpine area. The Atlantic Multidecadal Oscillation (AMO) have also shown to have a long-term effect in the study area; however, due its complex nature, the real effect of the AMO on reconstructed temperatures in LdRS cannot be fully constrained. In any case, the effect of the internal climate variability on local temperatures cannot be ruled out. LdRS record also highlights the potential impact that non-climatic environmental drivers such as atmospheric dust and pollution deposition can have exerted on these remote alpine environments (i.e. glacier melting).

Alpine temperatures of southern Iberia abruptly increased in the 1950s, exceeding the highest temperature scores reached in pre-industrial times. This means that the rate of warming throughout the 20th century increased ~2.5 times compared to the rate in the last stages of the LIA. Furthermore, this modern warming rate is even higher in the Sierra Nevada than in the Alps, pointing towards more environmental stress in these ecosystems. In addition to the amplified effect of warming and aridification, the local environmental pressure may have enhanced throughout the 20th century due to the disappearance of perennial snow fields and the gradual reduction of the seasonal snow cover affecting the local water availability. All these evidences in this fragile alpine region point towards an even worse scenario by the end of the 21st.

Data availability. Fractional abundances of the C_{28} 1,13-diol, C_{30} 1,13-diol, C_{30} 1,15-diol, and C_{32} 1,15-diol from both studied cores (LdRS shc and LdRS lgc) are available in Table S7.

Supplement. The supplement related to this article is available online at: *https://doi.org/*

Author contributions. The study was conceived by AG-A and JLT. CPM, LJ, GJM, and RSA recovered the sediment cores. AG-A analysed the samples and processed the data. All co-authors discussed the data and equally contributed to the preparation of the manuscript.

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

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FIGURES

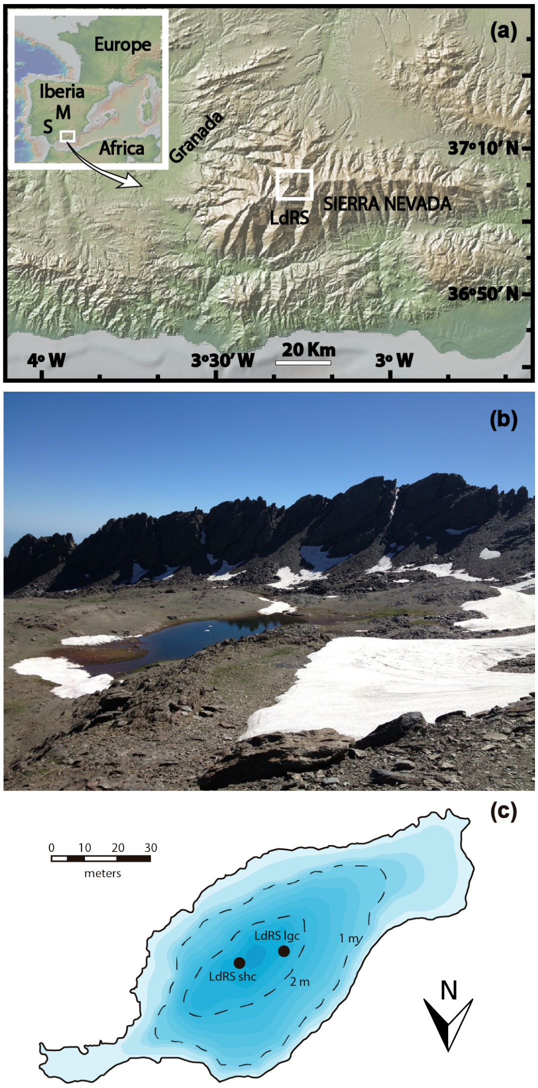

Figure 1. Geographical setting. (**a**) Location of the Sierra Nevada in the western Mediterranean region, Madrid (M), Sevilla (S) and Granada observatories, as well as the studied area: Laguna de Río Seco (LdRS), (**b**) LdRS catchment basin (0.42 ha) in spring 2013, **(c)** bathymetry map of LdRS along with the sampling points of both cores. Data source and software: (**a**) map created by A. García-Alix using GeoMapApp (3.6.6)

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(http://www.geomapapp.org), **(b)** picture from A. García-Alix, **(c)** digitalized map of a bathymetry report from Egmasa S.A.

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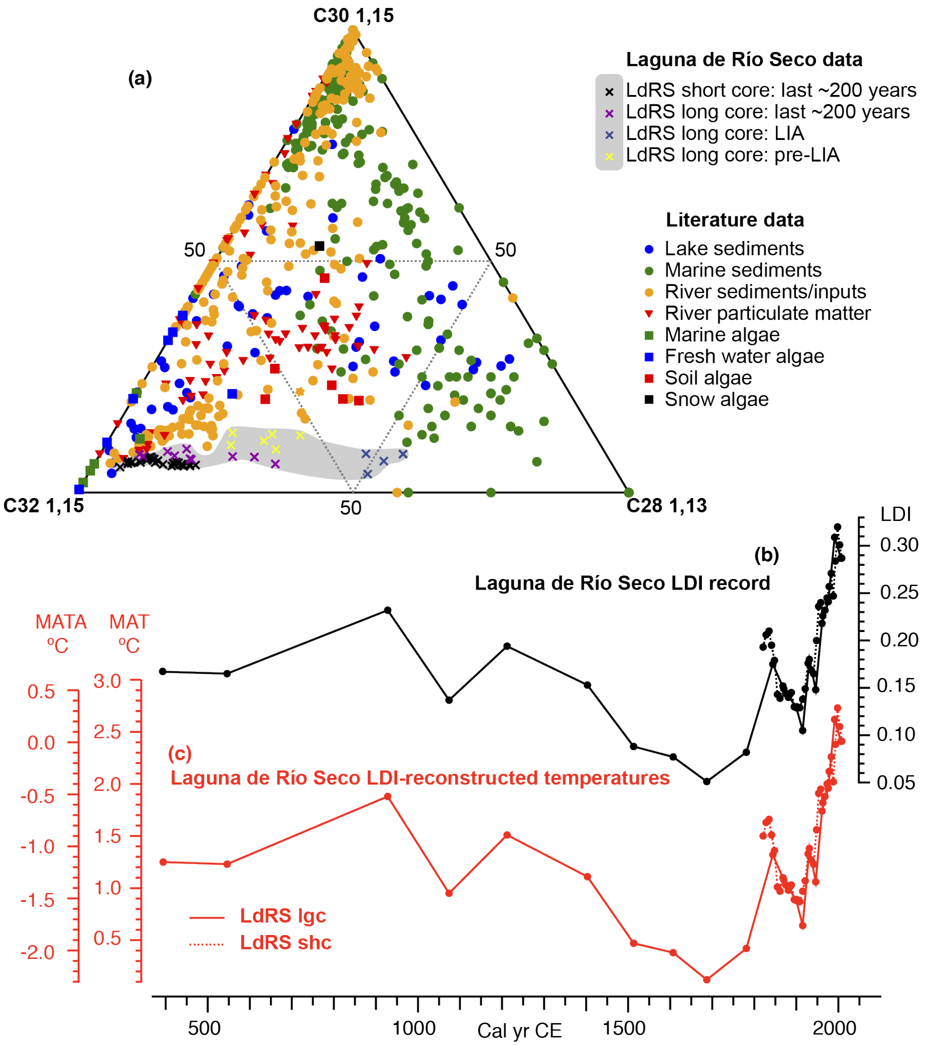

Figure 2. (a) Ternary diagram of the relative abundances of C_{28} 1,13-diol, C_{30} 1,15-diol and C_{32} 1,15-diol from LdRS short core (LdRS shc ~200 years) and LdRS long core (LdRS lgc ~1,500 years). Diol data compiled from the literature: lake sediments (Rampen et al., 2014a), algal cultures (Rampen et al., 2014a), marine sediments (Rampen et al., 2012;Rampen et al., 2014b;de Bar et al., 2016;Lattaud et al., 2017a), river sediments/inputs (de Bar et al., 2016;Lattaud et al., 2017b) (Lattaud et al., 2018), river particulate organic matter (Lattaud et al., 2018), **(b)** LDI record from LdRS, including

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both long core (line) and short core (dashed line), **(c)** Mean Annual Temperature (MAT ºC) reconstruction from LDI records of LdRS, as well as Mean Annual Temperature Anomaly reconstruction (MATA ºC) respect to the mean annual temperatures of the last 30 years (2008-1979). Long core (line), short core (dashed line).

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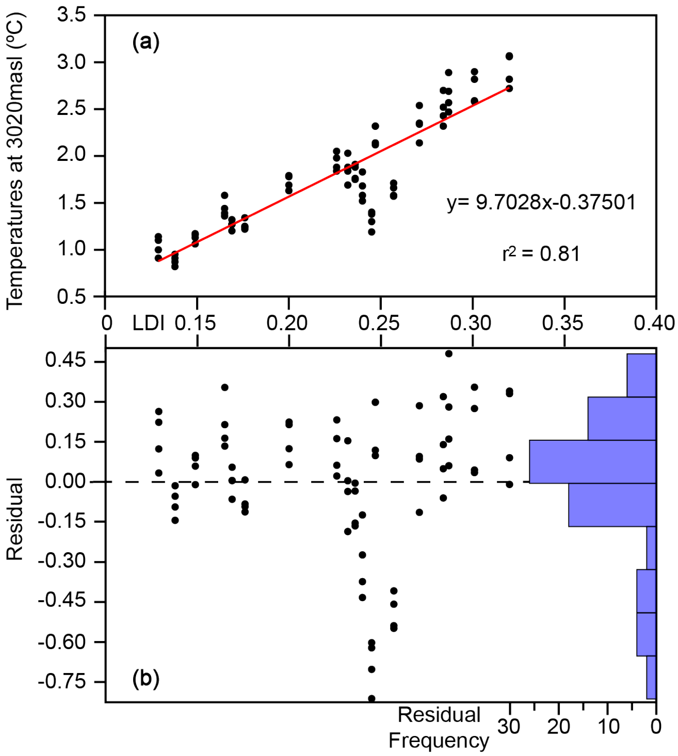

Figure 3. LDI temperature calibration. (a) Correlations by means of Ordinary Least Square regression between the LDI records from LdRS and the two groups of estimated temperature time-series at 3020 masl: Group 1) Temperatures obtained after correcting the instrumental temperature time-series from Madrid and Sevilla for the last ~100 years with the environmental lapse rate between low elevation and Sierra Nevada observatories (Spanish National Weather Agency - AEMet Open Data, 2019;Gonzalez-Hidalgo et al., 2015;Observatorio del cambio global de Sierra Nevada, 2016) (Table S4). Group 2) Temperatures obtained by means of the direct correlation among Madrid and Sevilla temperatures and those from the observatory Cetursa 5 at 3020 masl (Spanish National Weather Agency - AEMet Open Data, 2019;Gonzalez-Hidalgo et al., 2015;Observatorio del cambio global de Sierra Nevada, 2016) (Fig. S2), and **(b)** LDI values of LdRS vs.

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residual temperatures (calculated between the calibrated LDI temperatures vs. reference temperature time series at 3020 masl), as well as the histogram of the frequency of these residuals.

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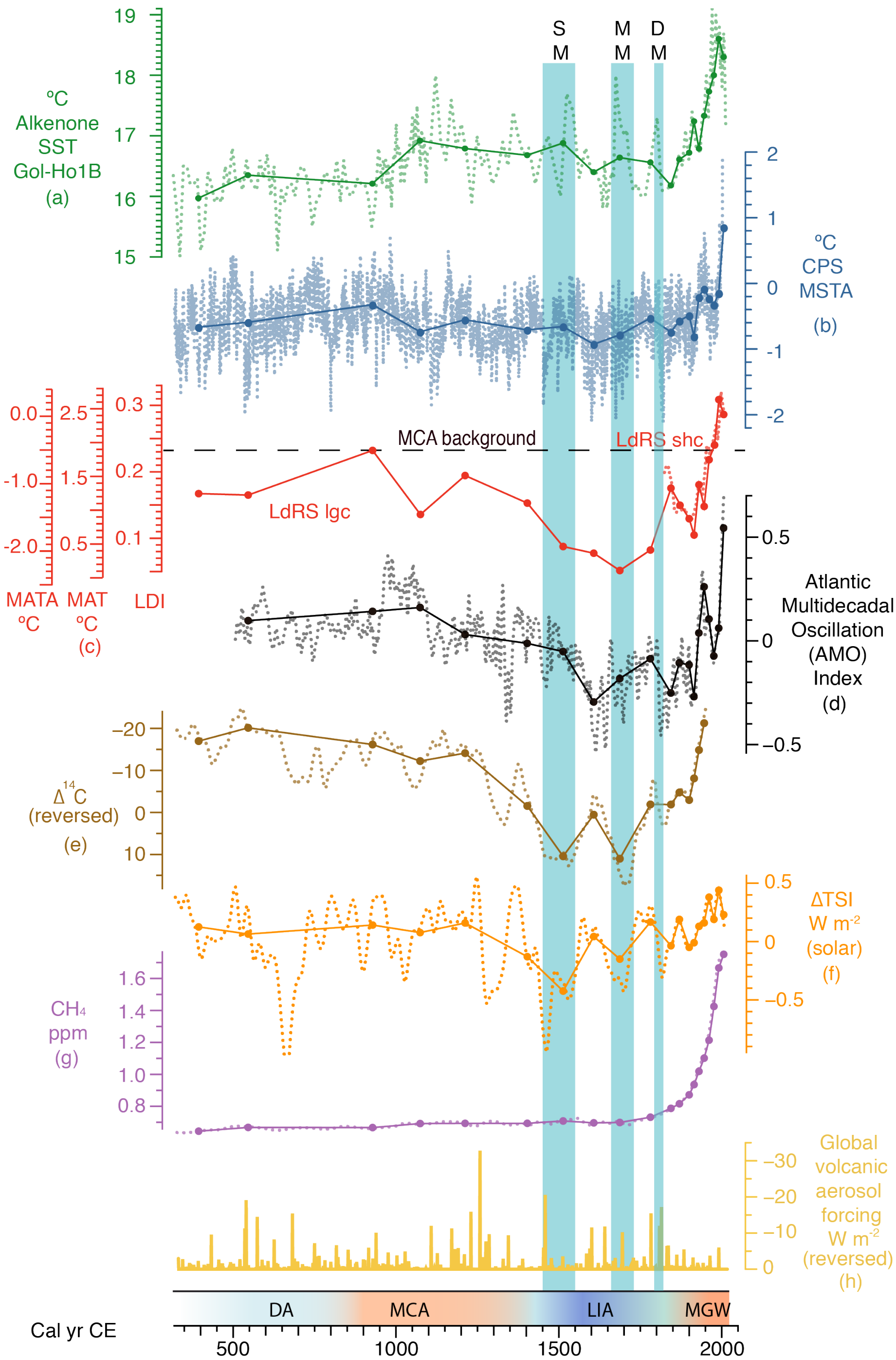

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Figure 4. Comparison of the LDI and the reconstructed temperature record for the last ~1500 years of LdRS with temperature records, greenhouse gases, solar radiation and volcanic eruption records. Original data are in dashed lines. Solid dots represent the same time averaging as the LDI data in LdRS lgc (data were linearly interpolated to the same resolution as the sampling points of LdRS lgc) to facilitate the Pearson correlation: **(a)** Alkenone-Sea Surface Temperatures (SST, ºC) of the core Gol-Ho1B_KSGC-31 (Gulf of Lion: NW Mediterranean Sea (Sicre et al., 2016)), **(b)** Composite-plus-scaling (CPS) mean summer temperature anomaly reconstruction from tree rings records in Europe with respect to 1974-2003 CE (MSTA ºC) (Luterbacher et al., 2016), **(c)** LDI record along with reconstructed mean annual temperatures (MAT ºC) and mean annual temperature anomalies with respect to 1979-2008 CE (MATA ºC) for the last 1500 years in LdRS (LdRS lgc, Laguna de Río Seco long core; LdRS shc, Laguna de Río Seco short core), **(d)** Atlantic Multidecadal Oscillation (AMO) reconstruction (Mann et al., 2009), **(e)** $\Delta^{14}C$ in the atmosphere (reversed) (Reimer et al., 2013), **(f)** reconstruction of the difference of the total solar irradiance from the value of the PMOD composite series during the solar cycle minimum of the year 1986 CE (1365.57 W m^{-2}) (ΔTSI) (Steinhilber et al., 2009), **(g)** reconstructed concentration of atmospheric CH_4 (ppm) (Schmidt et al., 2011), and **(h)** reconstruction of the global volcanic aerosol forcing (W m^{-2}) (reversed) (Sigl et al., 2015). Acronyms: DA, Dark Ages; MCA, Medieval Climate Anomaly; LIA, Little Ice Age; MGW, Modern Global Warming. Blue bars show three low solar activity periods, the Spörer Minimum (SM), the Maunder Minimum (MM), and the Dalton Minimum (DM).

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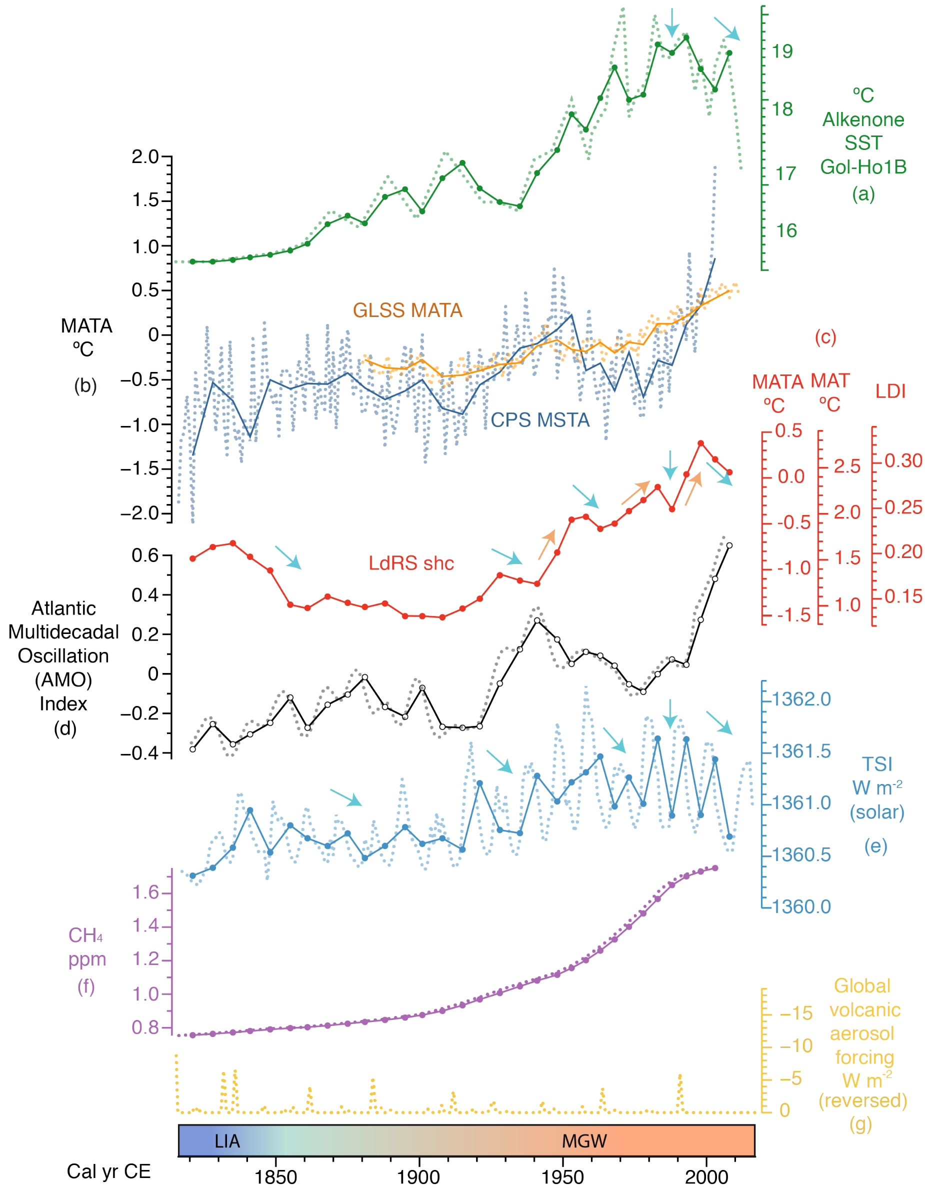

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1051 **Figure 5. Comparison of the LDI and the reconstructed temperatures record for the**
1052 **last ~200 years of LdRS with temperatures, greenhouse gases, solar radiation and**
1053 **volcanic eruption records.** Original data are in dashed lines. Solid lines represent the
1054 same time averaging as the LDI data in LdRS shc (data were linearly interpolated to the
1055 same resolution as the sampling points of LdRS shc) to facilitate the Pearson correlation.

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(a) Alkenone-Sea Surface Temperatures (SST, ºC) of the core Gol-Ho1B_KSGC-31 (Gulf of Lion: NW Mediterranean Sea (Sicre et al., 2016)), **(b)** Composite-plus-scaling (CPS) mean summer temperature anomaly reconstruction from tree rings records in Europe with respect to 1974-2003 (MSTA ºC) (Luterbacher et al., 2016) as well as global land and sea surface (GLSS) mean annual temperature anomalies with respect to 1979-2008 CE (MATA ºC) (Hansen et al., 2010), **(c)** LDI record along with reconstructed mean annual temperatures (MAT ºC) and mean annual temperature anomalies with respect to 1979-2008 CE (MATA ºC) for the last ~200 years in LdRS, **(d)** Atlantic Multidecadal Oscillation (AMO) reconstruction (Mann et al., 2009), **(e)** high resolution total solar irradiance reconstruction (TSI, W m^{-2}) (Coddington et al., 2016), **(f)** reconstructed concentration of atmospheric CH_4 (ppm) (Schmidt et al., 2011), and **(g)** reconstruction of the global volcanic aerosol forcing (W m^{-2}) (reversed) (Sigl et al., 2015). Acronyms: LIA, Little Ice Age; MGW, Modern Global Warming. Blue arrows: decrease; orange arrows: increase.

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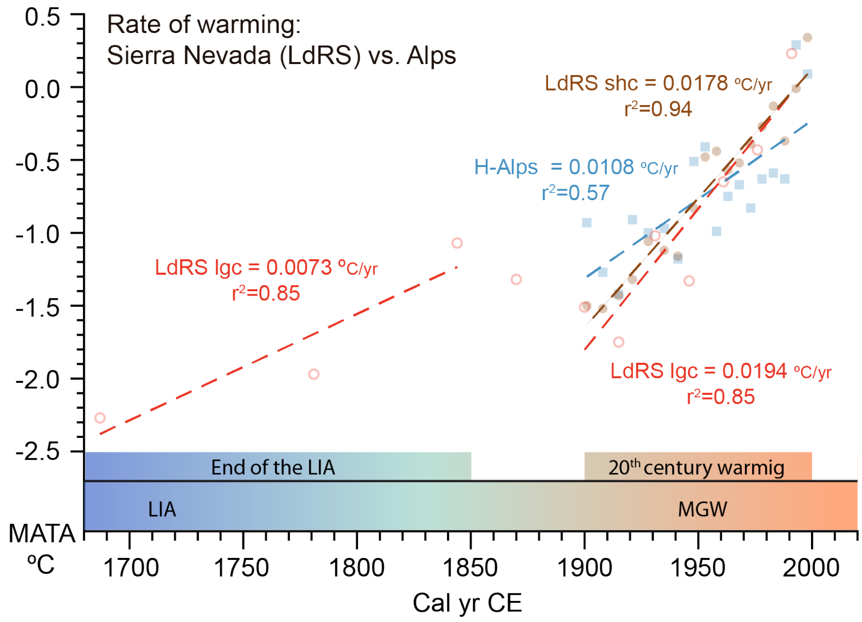

Figure 6. Comparison between the average temperature warming rates from LdRS and the alpine areas of the Alps by means of Ordinary Least Square Regressions. MATA (respect to the period 1979-2008 CE) from Laguna de Río Seco long core: LdRS lgc, red open circles, including the LIA and the 20th century; MATA for the 20th century (respect to the period 1979-2008 CE) from Laguna de Río Seco short core: LdRS shc, brown closed circles; and high-Alps historical (homogenised) temperature records from the Historical Instrumental Climatological Surface Time Series of the Greater Alpine Region (HISTALP) database (Auer et al., 2007;Böhm et al., 2010) at the same time averaging as LdRS shc (data were linearly interpolated to the same resolution as the sampling points of LdRS shc) to facilitate the comparison (blue closed squares).

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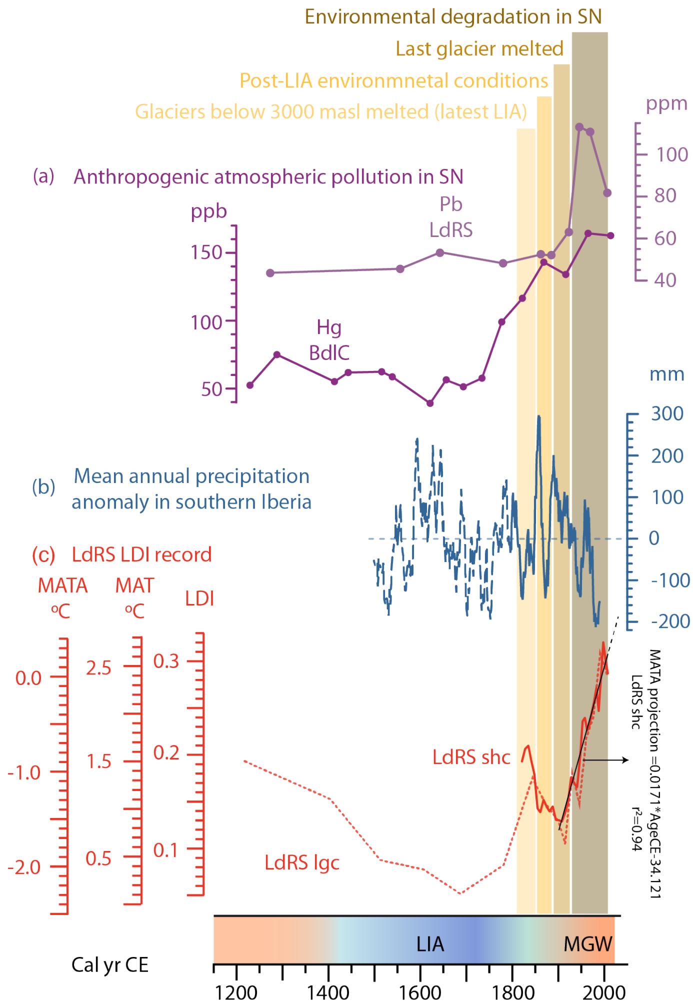

Figure 7. Comparison among different factors affecting the environmental evolution of alpine wetlands in Sierra Nevada. (a) records of anthropogenic heavy metal atmospheric pollution (Pb and Hg) in two alpine sites of Sierra Nevada: Laguna de Río Seco (LdRS) and Borreguil de la Caldera (BdlC) (Garcia-Alix et al., 2017;Garcia-Alix et al., 2013), **(b)** mean annual precipitation anomaly in southern Iberia from 1500 to 1990 CE with respect to the mean value of the instrumental period (1791-1990 CE): solid line- instrumental data from Gibraltar, Southern Iberia, dashed line- anomaly precipitation reconstruction (Rodrigo et al., 1999), **(c)** LDI and temperature reconstruction in LdRS, as

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well as the MATA projection from an Ordinary Least Square Regression including LdRS shc MATA for the last ~110 years (n=20). Colour bars indicate the four main environmental stages in the Sierra Nevada (SN) during the last 200 years. Acronyms: LIA, Little Ice Age; MGW, Modern Global Warming.