Late Neogene to Early Quaternary climate evolution in southwestern Europe from a continental perspective

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14 Abstract

15 Paleoclimate reconstructions are mostly based on continuous oceanic records, but 16 continental records, controlled by global and regional conditions, are paramount in 17 identifying long- and short-term climatic variability between regions and investigating 18 forcing mechanisms. Here we present a high-resolution lacustrine log from a western 19 Mediterranean intramountain basin; it is based on calcite oxygen isotope composition 20 $(\delta^{18}O_c)$ and records detailed paleoclimatic information from the Late Miocene to the Early 21 Pleistocene (9.8-1.8 Ma). Evidence is found for orbital forcing in the regional 22 paleoclimate, with minimum and maximum eccentricity related to drier and wetter 23 conditions respectively. Superimposed onto this variability, the long-term trend reflects 24 the influence of global paleogeographic and climate change. Variations inferred in 25 precipitation-evaporation (P-E) are related to SST in the North Atlantic, which evidences 26 a connection between marine dynamics and continental climate in areas far from the coast 27 in southwestern Europe and a relation between dry periods and high SST inland. It is 28 proposed that the regional climate was impacted by the effect of the closure of the Central 29 Atlantic Seaway and changes in the Atlantic Meridional Overturning Circulation 30 (AMOC). Warmer/drier conditions were related to a more permanent, stable, high-31 pressure centre over the mid-Atlantic in a situation of strengthened AMOC, which would 32 have blocked westerly winds, increasing aridity in southwestern Europe. The inferred 33 warm/dry connection differs from other western Mediterranean records, supporting 34 previous interpretations of a regional climate gradient in western Europe. As occurs at

35 present, isolation from the influence of the humidity of the Mediterranean Sea during 36 warm periods as a result of to the local orography could well have been the cause of 37 regional differences.

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39 Keywords

40 Paleoclimate, Lacustrine record, AMOC, Stable isotopes, Late Neogene, SW Europe

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42 **1. Introduction**

43 After the Miocene Climatic Optimum (17-14.5 Ma), the global climate underwent 44 a general cooling trend during the Late Miocene - Early Pleistocene, which resulted in a 45 global temperature decrease of the order of 5-6°C (e.g. Utescher et al., 2000; Zachos et 46 al., 2001; Lear et al., 2003; Lisiecki & Raymo, 2005; Mosbrugger et al., 2005; Bruch et 47 al., 2011; Quan et al., 2014; Holbourn et al., 2018). Deep geological and paleogeographic 48 changes during the Late Neogene reconfigured the climate system (e.g. Utescher et al., 49 2000; Zachos et al., 2001), and the climate zones came close to reaching the current setup. 50 The main changes include the rise of mountain ranges (the Himalayas, Carpathians, Alps, 51 Pyrenees, etc.), a new configuration of seaways and ocean circulation system, the 52 appearance of the Sahara Desert, and the establishment of a permanent ice cap in the 53 northern hemisphere (e.g. Ehleringer & Monson, 1993; Haug et al., 2001; Griffin, 2002; 54 Tuenter, 2004; Wang et al., 2006; Potter & Szatmari, 2009; Dowsett et al., 2009; 55 Haywood et al., 2009, 2016; Athanasiou et al., 2015; Jiménez-Moreno et al., 2018). 56 Overlapping the long-term cooling, shorter-scale changes also occurred, whose local 57 effects are less well known, especially in continental areas in which long and well-dated 58 climate records are still lacking (e.g. Ditlevsen & Ashwin, 2018). At mid-latitudes, heat 59 and moisture transport by ocean winds impacted on the continental rainfall and 60 temperature (e.g. Bruch et al., 2011; Quan et al., 2014), but regional climates did not 61 respond in a similar way to global changes, and climate variability and gradients between 62 regions have been brought to light (e.g. Suc, 1978; Fauquette et al., 1999; Hernández-63 Fernández et al., 2007; Matson & Fox, 2010; Prista et al., 2015). The response of 64 continental ecosystems differs depending on their latitude, continentality, altitude, and 65 landscape (e.g. Fauquette et al., 2006; Bohme et al., 2011; Ribera d'Alcalà, 2019). In 66 light of the foregoing, detailed continental paleoclimate reconstructions in diverse

settings are key to understanding the variability and gradients within continents (e.g.
Utescher *et al.*, 2012; Grygar *et al.*, 2017), as well as the factors forcing the changes.

69 Global circulation models of the Earth's changing climate agree that, with 70 increasing temperatures in the future, a decline in precipitation is expected in 71 Mediterranean areas as a result of the decline in the winter precipitation, in contrast with 72 other regions in the world, where increasing precipitation is projected (Tuel & Eltair, 73 2020). The Western Mediterranean reached its present paleogeographic configuration 74 during the Miocene, so deciphering Miocene-Pleistocene climate changes on multiple 75 time scales and reconstructing the regional climate forcing, internal variability, and 76 system feedback is paramount to understanding both past and future scenarios (Meyers et 77 al., 2010). Although some studies have provided information on how terrestrial 78 environments have reacted to long- and short-term climatic changes in the Western 79 Mediterranean area (Suc, 1984; Bertini, 2001; Combourieu-Nebout et al., 2004; Jiménez-80 Moreno et al., 2010, 2013; De Miguel et al., 2018), more studies are still needed, 81 including well-dated and calibrated proxy records in different geological/geographical 82 settings (Ambar & Scarascia-Mugnozza, 2012).

83 Many continental proxies based on fauna, flora, or sedimentary features have 84 proven very useful for paleoclimate studies (e.g. Fauquette et al., 1998, 1999; van Dam 85 & Weltje, 1999; Utescher et al., 2000; Teranes & McKenzie, 2001; Leng & Marshall, 86 2004; Mosbrugger et al., 2005; van Dam, 2006; van Dam et al., 2006; Domingo et al., 87 2009; Barrón et al., 2010; Domingo et al., 2013; Suc et al., 2021). Paleontological data 88 provide outstanding information but are sometimes lacking in resolution due to the 89 inherent characteristics of the fossil record. Sedimentological, physical, and chemical 90 proxies provide more continuous records (e.g. Quade et al., 2007; Abels et al., 2009a,b; 91 Pla-Pueyo et al., 2015; Grygar et al., 2017; Oliva-Urcía & Moreno, 2019). Among these, 92 oxygen isotope values (δ^{18} O) in lacustrine carbonates are widely used (e.g. Andrews *et* 93 al., 2000; Teranes & McKenzie, 2001; Leng & Marshall, 2004; Luzón et al., 2009). 94 Moreover, lacustrine basins potentially contain the most continuous and highest-95 resolution terrestrial records (see Fortelius et al., 2006).

The Teruel Basin (NE Iberian Peninsula), situated in a region of transition between Atlantic and Mediterranean influence, is a key area for the study of the Neogene continental climate in southwestern Europe. It records continuous deposition in a closed lake dating from 9.8 to 1.8 Ma and houses a huge amount of paleontological and

100 magnetostratigraphic information, being a reference area for European Neogene mammal 101 paleontology (e.g. Lindsay et al., 1989). This basin is located in an intramountainous area 102 with a significant orographic barrier to the east (Fig 1), and its current paleogeographic 103 configuration was attained in the Late Miocene. As occurs at present, this situation would 104 have isolated the study area from the influence of high Mediterranean Sea humidity. This 105 work presents a new lacustrine calcite oxygen isotope record ($\delta^{18}O_c$) from this basin. As 106 far as we know, it is the longest and highest-resolution continental climate curve of the 107 Late Neogene in the Iberian Peninsula. Our main objectives are to decipher the climate 108 signal recorded in the isotope data at different time-scales and interpret the principal 109 triggers of the relevant changes, in order to provide new insights into the evolution of the 110 climate and the variability of the western Mediterranean during the Late Miocene-Early 111 Pleistocene. The $\delta^{18}O_c$ series records the most relevant climate changes described on a 112 global scale and, from the Late Tortonian, shows a reasonable correlation with the sea 113 surface temperatures (SST) in the North Atlantic. A warm/dry connection is inferred that 114 differs from other western Mediterranean records, supporting previous interpretations of 115 a regional climate gradient in western Europe (Suc, 1978; Fauquette et al., 1999; Matson 116 & Fox, 2010; Prista et al., 2015; Jiménez-Moreno et al., 2009). Changes in the Atlantic 117 Meridional Overturning Circulation (AMOC), superimposed orbital cyclicity (with 118 eccentricity minima being related to drier conditions), and landscape barriers are 119 suggested as triggers for the recorded paleoclimate changes.

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121 2. Study area

122 The Neogene Teruel Basin is an elongated intramountain graben with an NNE-123 SSW trend (mean altitude: 989 m a.s.l.), located in the central-eastern Iberian Range, NE 124 Iberian Peninsula (Álvaro et al., 1979) (Fig. 1). The Iberian Range was formed as a result 125 of the oblique collision between the Iberian microplate and the European plate from the 126 Late Cretaceous to the Early Miocene (Álvaro et al., 1979; Capote et al., 2002). From the 127 Middle Miocene, the central-eastern part of the mountain range was affected by 128 extensional tectonics associated with the evolution of the Valencia Trough 129 (Mediterranean Sea). As a result, the Iberian Range was cut obliquely by a series of 130 extensional basins such as the Teruel Basin, which made it possible to accommodate a 131 fairly continuous sedimentary infill (Simón, 1986; Capote et al., 2002).

The Neogene Teruel Basin covers an area of 735 km² and is divided into two large 132 133 sectors, the northern and southern sectors, which show differences in the structure and 134 age of formation. The northern sector is the focus of this work (Fig. 2a), corresponding 135 to a half-graben basin with an active N-S segmented margin at the eastern boundary 136 (Ezquerro et al., 2020). It hosts an endorheic sedimentary succession spanning from the 137 Late Miocene (~11.2 Ma) to the Early Pleistocene (~1.8 Ma) (Moissenet, 1983; Simón et 138 al., 2012; Ezquerro et al., 2016, 2020). The estimated age is based on a large number of 139 mammalian sites (e.g. Mein et al., 1990; Alcalá et al., 2000), as well as on 140 magnetostratigraphic data (e.g. Krijgsman et al., 1996; Opdyke et al., 1997; Ezquerro et 141 al., 2016) that yield a good chronological control related to Neogene continental mammal 142 zones (MN zones; Mein, 1975), the European Land Mammal Ages (ELMA, e.g. Lindsay 143 et al., 1989), and the Geomagnetic Polarity Time Scale (GPTS; Ogg, 2012).

144 The vertical throw associated with the faults is 700 m, but the endorheic infill is 145 less than 500 m thick, since the basin was captured by an external fluvial drainage around 146 1.8 Ma, when it evolved to exorheic conditions (Weerd, 1976; Moissenet, 1980; Godoy 147 et al., 1983; Ezquerro et al., 2012, 2016; Ezquerro, 2017). During the Mio-Pleistocene, 148 large and small alluvial fans, located at the passive and active basin margins respectively, 149 graded to palustrine and lacustrine environments towards the central areas (Godoy *et al.*, 150 1983a,b; Moissenet, 1983; Alonso-Zarza et al., 2000; Ezquerro et al., 2014, 2019; 151 Ezquerro, 2017). Thus, the endorheic infill comprises coarse-grained terrigenous facies 152 close to the boundaries, corresponding to proximal sectors of alluvial fans, passing 153 basinwards to medium and fine-grained detrital facies with local developments of 154 calcretes, related to middle and distal areas of the alluvial fans. The distal alluvial facies 155 grade to carbonates and minor gypsum, generated in shallow lacustrine-palustrine 156 environments in the central part of the basin (Fig. 2b). The sedimentological features 157 indicate that a closed, shallow lake existed in the Teruel Basin during the Mio-Pliocene, 158 covering approximately 360 km² during stages of maximum expansion (Ezquerro et al., 159 2014; Ezquerro, 2017). The overall lacustrine succession is represented by 209 m in 160 thickness of carbonate deposits in the central areas of the basin, becoming thinner and 161 disappearing towards the passive margin.

162 The sedimentary evolution was controlled by extensional tectonics (Ezquerro *et al.*, 2014; Ezquerro, 2017; Liesa *et al.*, 2019). The propagation of faults at the basin

164 margin controlled changes in sediment supply, as well as the distribution and evolution 165 of the sedimentary systems. Angular unconformities, growth strata, and other evidence 166 of active tectonism (e.g. seismites) have been found in relation to episodes of alluvial fan 167 progradation and lake retraction (Ezquerro, 2017; Ezquerro et al., 2019, 2020). In stages 168 of decreasing tectonism, the alluvial fans retrograded, and the lake expanded, especially 169 towards the passive margin. Climate also exerted notable control over sedimentation at 170 different scales, and several climate-induced lake expansions/retractions have been 171 inferred (Alonso-Zarza et al., 2012; Ezquerro, 2017; Ezquerro et al., 2014, 2019).

172 **3. Methods**

173 *3.1. Sampling methods*

174 The study record corresponds to a 212-m-thick composite lacustrine section that 175 represents carbonate sedimentation in the Neogene lacustrine system of the Teruel Basin 176 (Fig. 3). The sampled succession is based on the correlation of three stratigraphic profiles 177 (selected for the isotope study due to their high carbonate content), in turn based on well-178 dated guide levels or packages, identifiable by particular lithological or sedimentological 179 characteristics. The sampling protocol focused on obtaining nearly constantly spaced (one 180 sample per metre), unaltered samples of the lacustrine succession, resulting in a total of 181 202 samples. The sampling density allowed an accurate correlation between different 182 outcrops, avoiding large data gaps. In each profile, all the samples were located in their 183 precise stratigraphic position at decimetre precision. Rock blocks were collected using 184 hammers and chisels in the field to obtain a significant volume of sample (cm-scale) for 185 performing thin sections as well as mineralogical and geochemical analyses. The 186 subsequent laboratory procedures were based on extracting small cores from the blocks, 187 which avoids alterations or disturbances from the surrounding materials. The cores were 188 extracted with an electric drill and a hollow drill bit made of stainless steel with a diamond 189 head or, when this was not possible, with rotary saws.

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191 *3.2. XRD mineralogical analyses*

The mineralogy of the lacustrine carbonates was determined using RIGAKU D/max2500 and Philips PW 1729 diffractometers from the X-ray Diffraction and Fluorescence Analysis Service (Research Support Services) and the Earth Sciences Department, respectively, in the University of Zaragoza. The configuration of the diffractometers was 40Kv and 80 mA with Cu anodes and a graphite monochromator to 197 select the Cu K-alpha radiation. The analyses were carried out on powdered samples, 198 crushed manually by an agate mill, and sieved at 53 μ m. The measurement conditions 199 were established at room temperature with 20 angles from 3° to 60° and steps of 0.04° and 200 0.4 s/step. The resulting diffractograms were interpreted by the XPowder 12 software 201 using the fundamentals of reference intensity (RIR; Chung, 1974) and the Rietveld 202 method (Rietveld, 1969).

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3.3. Microtextural analyses: FESEM and optical microscopy

205 Selected samples were analysed by field emission scanning electron microscopy 206 (FESEM) using secondary (SE) and backscattered electron (BSE) images and energy-207 dispersive X-ray (EDS) analysis. The observations were performed using a Carl Zeiss 208 MERLIN FESEM equipped with an Oxford Instruments INCA 350 EDS detector of the 209 Research Support Services of the University of Zaragoza. Elemental analyses were made 210 with an accelerating voltage of 133 eV to 5.9 KeV and a beam current of 80 nA. Samples 211 were gold-coated. Photomicrographs were taken in imaging mode with secondary 212 electrons. Petrographic observations were made by optical microscopy using 213 conventional thin sections made at the Research Support Services of the University of 214 Zaragoza.

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216 3.4. Stable isotope analyses ($\delta^{18}O$ and $\delta^{13}C$)

217 The selection of samples was based on mineralogical X-ray and petrographic results 218 in order to avoid non-primary carbonates. Only samples with authigenic calcite and 219 without dolomite or other carbonate minerals were considered. With a view to avoiding 220 any influence from non-authigenic calcite sources, samples with appreciable fossil 221 content or carbonate extraclasts were not selected. Likewise, samples with low 222 percentages of calcite were discarded. Isotope analyses were performed on calcite 223 samples acquired on whole sample by grinding an agate mill and sieving at 53 µm. In 224 each sample, 10 to 15 mg of rock powder was used and treated with 1 ml of 103% H₃PO₄. 225 The CO₂ was extracted following the techniques described by Walters et al. (1972), and 226 each of the fractionated extractions was treated with Ag₃PO₄ to eliminate the possible 227 SO₂ released. Sampling and analyses were carried out at the Stable Isotope Laboratory of 228 the University of Salamanca, using a dual inlet SIRA-II mass spectrometer. The 229 measurements were made under vacuum conditions (McCrea, 1950), with a temperature 230 of 25°C in the carbonate line and 90°C with the IsoCaRB system. The precision of the

method is 0.2‰ for δ^{18} O and 0.1‰ for δ^{13} C. The obtained results are presented in relation to the international VPDB standard (Craig, 1957; Gonfiantini, 1984).

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234 *3.5. Spectral and cyclicity analyses (depth domain)*

235 The spectral and cyclostratigraphic analyses were developed in a continuous 236 quantitative time series of 118 data corresponding to the δ^{18} O values of the lacustrine calcite ($\delta^{18}O_c$). Before performing the spectral analysis, taking into account the different 237 238 sampling intervals, an interpolation of the time series (every 1 m) was performed using 239 the software Acycle v2.1 (Li et al., 2019). The methodology also includes trend 240 elimination (with the LOESS model; Cleveland, 1979) and spectral analysis (depth 241 domain) using the Multi-Taper method (MTM; Thomson, 1982). With the aim of 242 characterizing the lower-frequency cycles, the time series was smoothed with a Gaussian 243 filter. The anchoring between the cycles resulting from the spectral analysis and the 244 Milankovitch cycles was performed using the Correlation Coefficient tool (COCO and 245 eCOCO) of the Acycle v2.1 software, which provides the most probable value for the 246 sedimentation rate; the anchor allows thickness cycles (depth domain) to pass into time 247 cycles (time domain).

248

4. Results

250 4.1. Sedimentological features

251 The sedimentary succession is mainly formed by limestones, marls, carbonate silts, 252 and nodular carbonates, with some intercalations of mudstones, gypsums, and sandstones 253 towards the upper part (Fig. 3). Greyish to whitish limestones, in tabular or irregular 254 strata, predominate in the lower and middle part of the succession, exhibiting two main 255 facies: massive mudstone-wackestone and bioturbated wackestone-packstone (Fig. 4); 256 laminated facies are recognized only occasionally. Bioturbated facies show vertical root 257 traces and brecciation. Charophytes and gastropods are the most common fossils in both 258 kinds of facies, which rarely show intraclasts. Massive limestones represent lacustrine 259 inner or sublittoral facies, whereas bioturbated limestones indicate littoral to palustrine 260 conditions. Greyish to greenish marls show a massive, rarely laminated texture and 261 contain charophytes, gastropods, and plant debris. They usually appear as tabular or 262 irregular layers between limestones and carbonate silts. Whitish carbonate silts are more 263 common in the middle-upper part of the succession and form massive or laminated tabular 264 beds with occasional gastropods and plant remains (Fig. 3). Marls and silts represent the entry of water and terrigenous supplies into the lake. On the basis of these features,
sedimentation in a wide, shallow, low-energy carbonate lake has been inferred (AlonsoZarza *et al.*, 2000, 2012; Ezquerro *et al.*, 2014; Ezquerro, 2017).

268 According to the optical microscopy and scanning electron microscopy (FESEM) 269 observations, the mineralogical data indicate an authigenic origin for the calcite (Cc), 270 since no textural evidence of diagenetic calcite replacement is observed (Fig. 5). Calcite 271 forms small euhedral-subhedral crystals with knobby surfaces. Calcite and quartz (Qz) + 272 phyllosilicates (Phy) show a negative correlation (R = -0.58; 202 samples) (Fig. 6a), 273 indicating that they were favoured under different sedimentary conditions. On the other 274 hand, quartz and phyllosilicates show a positive correlation, which is indicative of a 275 common origin related to detrital input (Fig. 6b). A very high negative correlation 276 emerges when (Qz + Phy)/(Cc + Gy) are compared (R = -1), indicating a non-genetic 277 relation between the two sets (Fig. 6c). Moreover, gypsum (Gy) and calcite (Cc) show a 278 strong negative correlation (R = -0.76), which suggests non-simultaneous precipitation 279 (Fig. 6d), with gypsum precipitation being favoured in more concentrated waters.

280 4.2. The $\delta^{18}O_c$ lacustrine record

The calcite $\delta^{18}O_c$ curve is based on detailed sampling and analysis of 202 281 282 lacustrine carbonate samples (approximately one per metre). In the end, however, 182 283 samples were included in the composite record due to the overlapping of 20 samples 284 between correlated profiles. The isotopic signal of the overlapping samples is 285 reproducible and replicable in the sense of Mangini et al. (2007) or Dorale & Liu (2009), 286 which reinforces the hypothesis of a precipitation of carbonates in equilibrium with lake 287 waters. The different $\delta^{18}O_c$ values of the Mesozoic rocks in the source area (Luzón *et al.*, 288 2009; Colás et al., 2010; Val et al., 2017) rule out a significant influence of detrital carbonates on the isotopic signal (Fig. 7). The positive correlation between the $\delta^{18}O_c$ and 289 $\delta^{13}C_c$ values indicates closed lacustrine hydrological conditions (Fig. 7), the influence of 290 291 the air-water exchange being reflected in the isotopic signal (Talbot, 1990; Li & Ku, 1997; 292 Valero-Garcés et al., 1997; Ezquerro et al., 2014).

293 The $\delta^{18}O_c$ values are always negative and range between approximately -2% and 294 -10%, with an average (χ) of -5.8% and a standard deviation (σ) of 1.3 % (Fig. 3). 295 Despite the significant variability of the $\delta^{18}O_c$ values, three main periods in the order of $10^{6} \text{ years can be recognized by comparing the } \delta^{18}O_{c} \text{ trend with the average value (Fig. 3).}$ The oxygen isotope data also reveal high climate variability, especially for Periods 2 and
3.

299 Period 1 comprises 40 data with values between -5.0 ‰ and -7.4 ‰ and covers 300 from the Middle to the Upper Tortonian (~9.8 to ~8.6 Ma). This part of the succession is 301 characterized by lower $\delta^{18}O_c$ values than the average, with $\chi = -6.3$ ‰ and $\sigma = 0.5$ ‰ 302 (Fig. 3). Low $\delta^{18}O_c$ intra-variation exists, with a slight increase towards the upper part of 303 the period (Fig. 3). Previous studies have shown that during this period the lake system 304 showed a high width/depth ratio in the initial phases of basin development (Ezquerro *et* 305 *al.*, 2014, 2020; Ezquerro, 2017).

306 Period 2 includes 67 samples that represent the Late Tortonian-Late Zanclean 307 interval (~8.6 to ~3.6 Ma). The data range from -6.8 ‰ to -2.3 ‰, with $\chi = -4.8$ ‰ and $\sigma = 1.0$ ‰, and with a higher $\delta^{18}O_c$ mean value in this period than in the succession as a 308 whole and notably higher than the average for Period 1 (Fig. 3). The $\delta^{18}O_c$ variability 309 310 increases with respect to Period 1, and most values exceed the average. Higher values are 311 found in three intervals over the course of this period: from ~8.5 to ~8.1 Ma, ~6.0 to ~5.7 312 Ma, and ~ 5.1 to ~ 4.2 Ma; the latter interval shows the highest values for the whole time 313 span under study. Some intervals (~7.1 to ~6.6 Ma; ~6.1 to ~6.0 Ma, ~5.3 to ~5.1 Ma) 314 and several excursions (e.g. taking place between ~4.8 and ~4.0 Ma) with values below the basin average can also be recognized. As a consequence, the series of $\delta^{18}O_c$ values is 315 316 highly oscillatory, especially from the middle-upper part of Period 2, with variations 317 sometimes of up to ~ 4.0 %. In Period 2, two main lake expansions were recorded, with 318 maxima around 6.9 Ma and 4.4 Ma. These were separated by a phase of lacustrine 319 retraction around 6.0 Ma (Ezquerro et al., 2014, 2020; Ezquerro, 2017). Facies analysis 320 indicates that shallow lacustrine conditions always prevailed.

Period 3 is defined by 75 data covering Piacenzian and Early Gelasian stages (from ~3.6 to ~1.8 Ma). In contrast to Period 2, $\delta^{18}O_c$ values are lower than the basin average, especially in the lower part (Fig. 3), ranging from -9.8 ‰ to -3.5 ‰ ($\chi = -6.5$ ‰ and $\sigma = 1.3$ ‰). The intra-period variation is high, and values oscillate very quickly (changes of up to ~4.5 ‰) but are only higher than the basin average around 3.1-3.2 Ma and between ~2.9 and ~2.5 Ma. From 2.2 Ma to more modern times, the values are close to the basin average, and very few excursions are recognized. During this period, the lake 328 system became more restricted and shallower due to the segmentation of the basin and
329 isolation of several depocentres in consequence of fault activity (Ezquerro *et al.*, 2014,
330 2020; Ezquerro, 2017).

331 4.3. Spectral analysis of the $\delta^{18}O_c$ time series

332 In order to recognize an external forcing superimposed upon the tectonics, the cyclicity analysis was based on the $\delta^{18}O_c$ values and their correlation with the thickness 333 334 of the sedimentary succession. Previous works (Krijgsman et al., 1996; Garcés et al., 335 1999) have suggested the existence of a hiatus around the Middle Turolian (Tortonian -336 Messinian boundary), between chrons C4n.1r and C3Ar, but the absolute dating has 337 remained unresolved. Recently, Ezquerro (2017) and Ezquerro et al. (submitted), 338 integrating information from the stratigraphic framework, mammal sites, and the 339 magnetostratigraphic context, have shown that the proposed hiatus coincides with an 340 interval with a very low sedimentation rate between ~7.9 and ~6.6 Ma. In our stratigraphic 341 succession, this interval occurs in the middle part and is poorly defined, with seven 342 samples representing a period of around one million years. Taking this into consideration, 343 and in order to avoid any disturbance related to the existence of a hiatus or a period of 344 very low sedimentation, the spectral analysis focused on the 6.15 to 1.8 Ma interval (66-345 209 m).

346 The spectral analysis was performed over 143 metres that correspond to 118 data, 347 with an average sampling interval of 1.2 m (interpolated to 1 m for the spectral analysis). 348 Bearing in mind that to define a cycle at least two data are needed (Nyquist frequency), 349 the thinnest cycle that could be recognized would be 2 m thick. As according to the 350 spectral analysis results (depth domain, Fig. 8), the thickness of the obliquity and 351 precession cycles should be 1.9 and 0.95 m, respectively, it was not possible to 352 discriminate them with the available sampling density. Two cycles (13.33 m and 4.77 m) 353 exceed the 99.9% confidence band (Fig. 8b). Given the resulting average sedimentation rate of 3.61 cm/ka for the interval between 66 and 209 m of the $\delta^{18}O_c$ time series (Fig. 9), 354 355 these cycles lasted 369 ka and 132 ka, respectively. However, the sedimentation rate 356 analysis undertaken with the Acycle-eCOCO software (Li et al., 2019) indicates 357 noticeable changes throughout the series, although the rate remained very constant 358 between 3.1 and 1.8 Ma (140 and 209 m, respectively; Fig. 9). The spectral analysis of 359 this part of the succession suggests that the two maxima above the 99.9% confidence band

360 correspond to cycles of 14.58 m and 4.32 m (Fig. 8c). Based on the inferred average
361 sedimentation rate (3.61 cm/ka), the cycles span ca. 404 ka and 120 ka, respectively,
362 which is very close to the two eccentricity modes (< 10% error).

363 After applying a Gaussian filter (interpolated every metre and removing the trend with a LOESS model), with the filter centre at 405 ka and 2.4 Ma, the anchoring of the 364 filtered $\delta^{18}O_c$ time series to the La2010a solution (Laskar *et al.*, 2011) was performed 365 366 using the paleontological and magnetostratigraphic data for the Teruel Basin (e.g. Mein 367 et al., 1990; Krijgsman et al., 1996; Opdyke et al., 1997; Ezquerro, 2017) in order to 368 introduce the time coordinate (Fig. 8d). The $\delta^{18}O_c$ oscillations of the filtered curve match 369 the eccentricity component of the orbital forcing, with the maximum and minimum eccentricity values being correlated, respectively, with the lowest and highest $\delta^{18}O_c$ 370 371 values.

372

373 **5. Discussion**

374 5.1. On the origin of the $\delta^{18}O_c$ signal

375 The isotopic composition of lacustrine authigenic carbonates is directly related to the temperature and δ^{18} O of the lake waters (δ^{18} O_w) (e.g. Leng & Marshall, 2004; Hoefs, 376 2009). In open lakes with a short residence time, the $\delta^{18}O_c$ values reflect seasonality, 377 378 temperature, and the rainfall water δ^{18} O composition. By contrast, closed shallow lakes 379 are more sensitive to long-term changes in the P-E rate due to the long residence time of 380 the water (Li & Ku, 1997; Andrews et al., 2000; Leng & Marshall, 2004). In closed and 381 shallow lacustrine systems, the P-E ratio, controlled by temperature and humidity 382 conditions, thus has a greater impact on the isotopic composition of the lake water 383 compared to other factors such as changes in the source of moisture. However, more 384 complex relationships are possible, since the factors that control basin hydrology can 385 interact with each other (e.g. Jones & Roberts, 2008; Benavente et al., 2019). In order to shed light on the climate signal recorded in the $\delta^{18}O_c$ in the Neogene Teruel Basin, the 386 387 climate factors that controlled the isotopic composition of the lacustrine calcite are here 388 discussed.

As explained above, the sedimentological data support a large palustrine-shallow lacustrine area dominated by closed hydrological conditions during the Late Neogene-Early Pleistocene. The $\delta^{18}O_c$ values generally lie within the range of freshwater lakes, and

392 the positive $\delta^{18}O_c / \delta^{13}C_c$ covariance agrees with the closed hydrological conditions 393 inferred (Talbot, 1990; Li & Ku, 1997; Leng & Marshall, 2004). By the same token, the 394 high $\delta^{18}O_c$ variability is not consistent with an open lake in which the water composition 395 is mainly homogeneous (e.g. Valero-Garcés et al., 1997; Jones & Roberts, 2008), leading 396 to low isotope variation (Quade et al., 1995; Dunagan & Turner, 2004; Luzón et al., 397 2017). Only the low variation in $\delta^{18}O_c$ recorded in Period 1 could indicate damping 398 processes, which Ezquerro et al. (2014) relate to fault activity. Whatever the case, as 399 suggested by Talbot (1990), these isotopic features are typical of lakes with a low 400 width/depth ratio, which is consistent with the sedimentological interpretation for Period 401 1. The $\delta^{18}O_c / \delta^{13}C_c$ covariance, the generally high $\delta^{18}O_c$ variability, and the amplitude of the range values point to the P-E ratio as the direct control factor for the calcite isotope 402 403 values. Several studies point out that, in palustrine areas and shallow lakes, the influence 404 of temperature through surface evaporation on isotope composition is greater than 405 groundwater (e.g. Jones & Roberts, 2008; Benavente et al., 2019).

406 In summary, it is proposed that the calcite isotope values in the Late Neogene Teruel 407 Basin would have been controlled by changes in humidity but also in temperature. With higher temperatures, relatively δ^{18} O-enriched precipitation and increasing lake water 408 evaporation (with preferential ¹⁶O loss) would have led to a rise in the δ^{18} O of the lake 409 water and in the precipitated carbonates. Therefore, higher $\delta^{18}O_c$ values would record 410 drier (a lower P–E ratio) and warmer periods. Conversely, lower $\delta^{18}O_c$ values may be 411 412 related to lower temperatures, which would have favoured isotopically depleted rain 413 (Craig, 1961) and reduced evaporation (a higher P–E ratio) in the lake.

414 Comparisons between our data and previous results providing mean annual temperatures (MAT) and mean annual precipitation (MAP) based on the δ^{18} O signature 415 416 of mammal dentition (van Dam & Reichart, 2009; Matson & Fox, 2010; Domingo et al., 2009, 2013) or micromammal associations (van Dam & Weltje, 1999; van Dam, 2006; 417 418 van Dam et al., 2006) reinforce this interpretation (Fig. 10), since the curves proposed for these authors show very similar trends to that of our $\delta^{18}O_c$. The $\delta^{18}O$ isotope data from 419 420 large-mammal tooth enamel (van Dam & Reichart, 2009; Matson & Fox, 2010; Domingo 421 et al., 2009, 2013) present lower resolution than the small-mammal records but are also 422 key to relating temperature and humidity conditions. The range and position of the $\delta^{18}O_c$ 423 variations in the Teruel Basin are comparable to the macromammal data, which also support the influence of temperature. The δ^{18} O variations recorded in the dentition were 424

interpreted as changes in temperature between 11.1 °C and 23.8 °C (van Dam & Reichart,
2009; Matson & Fox, 2010), and, with minor variations, they show a general decrease in
temperature (5 to 6°C) over the course of the study period (e.g. van Dam & Reichart,
2009; Domingo *et al.*, 2013; De Miguel *et al.*, 2019).

429 The connection between the warmer/drier stages is also in line with climate 430 interpretations for the Teruel Basin and other Iberian records (Daams et al., 1988; van 431 Dam and Weltje, 1999; Hernández-Fernández et al., 2007; Domingo et al., 2009, 2013; 432 van Dam & Reichart, 2009; Matson & Fox, 2010; De Miguel et al., 2018). Nevertheless, 433 in Mediterranean coastal zones, a connection between warming and more humid stages 434 has been proposed, as well as a latitudinal temperature and precipitation gradient between 435 northern and southern areas since the Middle Miocene (Suc, 1978; Fauquette et al., 1999; 436 García Alix et al., 2008; Matson & Fox, 2010; Suc et al., 2018; Jiménez Moreno et al., 437 2019). Accordingly, in the Iberian area when the climate was warmer and more humid in 438 the northern Mediterranean, warmer and drier conditions prevailed in the southern 439 Mediterranean. At present, the climate of the Iberian Peninsula features marked regional 440 and seasonal variability, which is the result of the interaction of different air masses with 441 a complicated orography and is strongly influenced by the surrounding water masses 442 (Cabos et al., 2020). Regional differences in the Late Neogene would have been 443 associated, as at present, with the orographic barriers in the S-SE (in this case the Iberian 444 Range), preventing a high Mediterranean oceanic influence. Currently, vegetation models 445 for the Mediterranean based on pollen records show a zonal distribution through the 446 Iberian Peninsula (Favre et al., 2007; Jiménez-Moreno et al., 2010), with a clear limit 447 (xerophytic vegetation/subtropical forests) near the Teruel Basin. The presence of such a 448 limit agrees with the occurrence of the particular environmental conditions for this 449 intramountainous basin during the Late Miocene (which more general climate models fail 450 to establish).

451

452 5.2. Climate changes inferred from $\delta^{18}O_c$ and correlation with other climate records

453 The isotope curve of the Teruel Basin records short and long-term climate changes 454 and three periods with very distinct climate conditions. The general cooling trend 455 documented worldwide after the MCO (Miller *et al.*, 1991; Zachos *et al.*, 2001; van Dam 456 & Reichart, 2009; Super *et al.*, 2020) is recorded by a general decrease in $\delta^{18}O_c$ values 457 from Period 1 to 3. The range of the $\delta^{18}O_c$ changes (in average values) recorded in the 458 Teruel lacustrine carbonates could represent an extreme increase of ~6°C between Periods 459 1 and 2, and a decrease of ~7°C between Periods 2 and 3 (Fig. 10). It should be noted that 460 in comparison with previous records, our $\delta^{18}O_c$ curve additionally shows high-frequency 461 climate variability in the Teruel Basin, considerably improving the resolution of climate 462 information for this Iberian area.

463 Broadly, Period 1 (~9.8 to ~8.6 Ma) was relatively humid (high P–E) and cool, as inferred from the lower $\delta^{18}O_c$ values (average -6.4‰) with respect to the mean. 464 Furthermore, a low standard deviation for $\delta^{18}O_c$ suggests low climate variability. Such 465 466 conditions in the Teruel Basin correlate well with a stage of relative global cooling, which 467 has been associated with lower levels of atmospheric pCO_2 (see Fig. 11; Pearson & 468 Palmer, 2000; Pagani et al., 2010; Seki et al., 2010; Stap et al., 2016) and the arrival of 469 ice-rafted debris at low latitudes (Thiede et al., 1998; Winkler et al., 2002) during this 470 time. The models of mean annual precipitation (MAP) for the Teruel Basin, based on 471 changes in the arboreal and invertivore mammal communities, also indicate wetter-cooler 472 conditions until ~8.6 Ma. Other pollen and micromammal proxies point to an increase in 473 precipitation in Europe, with (sub)humid climatic conditions prevailing during this period 474 (van Dam, 2006; Donders et al., 2009; Utescher et al., 2012; Quan et al., 2014). 475 Nevertheless, some authors propose warm and humid conditions in southwestern and 476 central Europe (Böhme et al., 2008). Low climate variability during the Late Tortonian 477 has also been evidenced in ocean records (Herbert et al., 2016; Super et al., 2020), as well 478 as in other continental proxies such as pollen records from the south of the Iberian 479 Peninsula (Postigo-Mijarra et al., 2009). This could be related to the fact that a 480 Mediterranean-type climate, dry in summer, had not yet been established (Quan et al., 481 2014).

A noticeable increase in $\delta^{18}O_c$ values (average ~2‰) at ~8.6 Ma characterizes the 482 483 transition to Period 2 (~8.6 to ~3.6 Ma) and testifies to a net decrease in the P-E ratio and 484 increasing temperatures in the Teruel Basin (Figs. 10 & 11). Such a change coincides 485 with a general increase in pCO_2 (e.g. Pagani *et al.*, 2005) and an increase in SST (~5°C) 486 at mid-latitudes (Herbert et al., 2016 and references therein), as well as with the expansion 487 of C4 plants (Cerling et al., 1997; Edwards et al., 2010) on a global scale. A wet-dry 488 seasonal climate has been interpreted for the Teruel Basin until ~8 Ma (van Dam & 489 Weltje, 1999), although a faster decrease in the winter rainfall than in the MAP suggests 490 increasing aridification around 8.5 Ma, when the establishment of a dry season was

491 definitive and higher mean temperatures were reached during the colder months (van 492 Dam, 2006). The disappearance of many paleotropical taxa and aquatic plants in the 493 Iberian Peninsula (Böhme et al., 2008; Postigo-Mijarra et al., 2009) and faunal changes 494 (e.g. Blanco et al., 2018; De Miguel et al., 2019) also indicate warmer conditions, which 495 in turn suggest more effective evaporation than precipitation (Fig. 10). However, in the 496 Granada and Crevillente basins (SE Spain), the climate in the Latest Tortonian was 497 temperate and more humid conditions than for the Teruel Basin have been deduced, evidencing regional differences in Iberia. At a global scale, the European Temperate Wet 498 499 Zone (ETWZ) contracted from ~9 Ma onward, and the Subtropical High-Pressure Zone 500 (SHPZ) expanded, as southern and western Europe and eastern Asia evolved towards 501 drier conditions (Haug et al., 2001; Griffin, 2002; Barrón et al., 2010; Böhme et al., 2008, 502 2011).

503 After the increase in SST at ~9 Ma, a general cooling trend characterized the Late 504 Miocene (Miller et al., 1991; Zachos et al., 2001; Herbert et al., 2016; Super et al., 2020). 505 By contrast, warm conditions, low P–E rates, and increasing climate variability are 506 deduced from our isotope curve for the Teruel Basin between the Late Tortonian and the 507 Zanclean (ca. 4.3 Ma). This trend changed towards the Piacenzian to a more humid and 508 cooler climate (Period 3), with the record showing a clear shift towards lower $\delta^{18}O_c$ 509 values; the lowest values are recorded from 3.6 to 3 Ma. The models of mean annual 510 precipitation (MAP) for the Teruel Basin, based on changes in the arboreal and 511 invertivore mammal communities, also indicate warmer/drier conditions from the Late 512 Tortonian to the Zanclean, which correspond to increases in MAT of up to 7°C and 513 evaporative enrichment during the summer (van Dam & Reichart, 2009; Matson & Fox, 514 2010). Although a generally warm climate is demonstrated for the Late Miocene-Early 515 Pliocene, wetter-cooler conditions prevailed during some periods in the region, as indicated by several gentle excursions to lower $\delta^{18}O_c$ values (e.g. ~7.1 to 6.6 Ma, 6.1 to 6 516 517 Ma, ~5.3 to 5.1 Ma, and some other episodes in the Zanclean; Fig. 11). This agrees with 518 previous interpretations (van Dam & Weltje, 1999; van Dam, 2006; van Dam et al., 2006, 519 2009) proposing drops in the MAT due to the higher influence of the low winter 520 temperatures and cooler summers (van Dam & Reichart, 2009; Matson & Fox, 2010).

521 Increasing temperature and warming conditions for the Late Miocene-Early 522 Pliocene have also been inferred from fauna and vegetation in western European and 523 Mediterranean regions (Fauquette *et al.*, 2007; Hernández-Fernández *et al.*, 2007), with 524 severe aridity occurring in many regions (Fauquette et al., 1999; Barrón et al., 2010; 525 Pellegrino et al., 2018; Suc et al., 2018; Jiménez Moreno et al., 2019), followed by a 526 general temperature decrease (Fauquette et al., 2006). Our isotope curve suggests that 527 although aridity intensified during the Messinian, no dramatic climate changes occurred 528 during the Messinian Salinity Crisis (~5.9 to 5.3 Ma), as previously proposed for the 529 Mediterranean coastal zone (Fauquette et al., 1999, 2006; Jiménez Moreno et al., 2009; 530 Barrón et al., 2010). Between 5 and 4 Ma, aridification became more severe in the Teruel 531 Basin, and wet-season precipitation decreased (van Dam & Weltje, 1999; van Dam, 2006; 532 van Dam *et al.*, 2006, 2009); this is recorded by the higher $\delta^{18}O_c$ values in our record, 533 which rarely fall as far as average levels.

534 The transition to Period 3 (~3.6 to ~1.8 Ma) is characterized by a sharp decrease in 535 the $\delta^{18}O_c$ values (average: -6.4%), the lowest values of the whole succession generally being recorded. The higher P-E ratio and lower temperatures in the Teruel Basin (Figs. 536 537 10 & 11) are consistent with the global climate cooling that occurred from the Mio-538 Pliocene boundary to the end of the Zanclean (Lisiecki & Raymo, 2005; Karas et al., 539 2017), which has been associated with the onset of northern hemisphere glaciation, with 540 a temperature decrease in Europe and the growth of the Arctic ice sheet (e.g. De Schepper 541 et al., 2013). CO₂ concentrations decreased during the Piacenzian-Gelasian (Pearson & 542 Palmer, 2000). Our curve also reflects a clear increase in climate variability, which 543 chimes with the SST records (e.g. Lawrence et al., 2010; Herbert et al., 2016). Many 544 shorter-term drier and warmer episodes (more positive $\delta^{18}O_c$) can be deduced in the 545 Teruel record, such as the mid-Pliocene Warm Period (mPWP) between 3.2 and 3.0 Ma, as well as many other positive $\delta^{18}O_c$ excursions, which we propose, on the basis of their 546 547 age, to be related to Marine Isotope Stages (Fig. 11). The identification of MIS effects in 548 continental records has also been suggested by Suc et al. (2018). The driest conditions 549 during this period were reached from 2.5 to 2.9 Ma, in accord with the results published 550 by van Dam & Reichart (2009) and Rodríguez-López et al. (2012). As in previous periods, 551 high $\delta^{18}O_c$ variability could correspond to an increase in MAP and related seasonal 552 oscillations. The MAP estimates for the Teruel Basin are less well defined for the Late 553 Pliocene (from ~2.6 Ma on), but a general increase in precipitation rates has been inferred, 554 with high variations, as a consequence of glaciations (Fauquette et al., 1998).

555

556 5.3. Triggers of the climate intra-variability: orbital forcing and the dynamics of the north 557 Atlantic Ocean and Mediterranean Sea

558 The three long-term climate periods (1 to 3), as well as the shorter-term climate 559 variations defined on the basis of $\delta^{18}O_c$ values, suggest that several superimposed factors 560 controlled the environmental conditions in the eastern Iberian Peninsula. Here we discuss 561 proposals, apart from orbital forcing, for how the north Atlantic Ocean and the 562 Mediterranean Sea impacted on climate in the Neogene Teruel Basin.

563 As previously stated, the influence of astronomical forcing has been demonstrated 564 by correlating the lowest isotopic values with maxima in eccentricity, and vice versa (Fig. 8). In addition, lower $\delta^{18}O_c$ values are associated with deeper sedimentary facies, 565 566 indicating a relatively high water table. In light of the foregoing, wetter and cooler (low 567 $\delta^{18}O_c$) episodes are correlated with eccentricity maxima, whereas warming episodes with enhanced evaporation (high $\delta^{18}O_c$) are correlated with eccentricity minima (Fig. 12). This 568 569 interpretation is in line with that proposed by Abels et al. (2009a,b) based on the study of 570 palustrine-lacustrine facies in the southern part of the Teruel Basin. These authors 571 suggested a modulation of summer/winter insolation and winter precipitation through the 572 influence of obliquity and precession. It is highly probable that lower-order orbital cycles 573 influenced climate variability and shorter-term changes in our record, but the current 574 sampling resolution precludes confirmation of this hypothesis.

Comparison of the Teruel $\delta^{18}O_c$ curve with oceanic records (Fig. 11) suggests that 575 576 variations in the AMOC might have had a deep impact on changes in humidity and 577 temperature in the inland areas of the eastern Iberian Peninsula; the magnitude and timing of these changes can be inferred from our $\delta^{18}O_c$ curve (Fig. 11). It is known that although 578 579 orographic barriers may influence the circulation of air masses, ocean dynamics exerts 580 direct control over mid-latitude continental areas, since the winds that transport heat and 581 moisture landwards influence rainfall rates and temperature (Bruch et al., 2011; Herold 582 et al., 2012; Quan et al., 2014; Sherriff-Tadano et al., 2018). The correlation between 583 Mio-Pliocene terrestrial and marine climate records in Europe has been documented in 584 many works. Mid to Late Miocene pollen records in northwestern Europe show a strong 585 coupling between marine and terrestrial temperatures, although the cooling trend seems 586 to have been more gradual in inland areas (Donders et al., 2008, 2009). During the Middle 587 Miocene, central Europe was under oceanic influence, displaying a warm and wet climate 588 very consistent with increases in global warming intervals and seasonality during cooling

periods (Methner *et al.*, 2020). Around the Mediterranean Sea, vegetation and climate changes that occurred during the Pliocene are in good agreement with pollen records from NW Europe and δ^{18} O curves from Mediterranean and Atlantic deep-sea cores (Fauquette *et al.*, 1999). The δ^{18} O_c record for the Teruel Basin also shows a similarity with North Atlantic marine records, especially from 8.6 Ma, and at a lower scale marine isotope stages (MIS) are reflected in the curve (Fig. 11).

595 Today, the Atlantic Ocean is the primary source for Iberian precipitation. Recent 596 modelling work indicates that the precipitation gradient is controlled by the stronger 597 Atlantic influence in the northern and eastern areas, especially during the winter season, 598 with precipitation and temperatures being controlled by the air-ocean coupling (Cabos et 599 al., 2020). Modern temperatures and rainfall δ^{18} O values in eastern Iberia also show a 600 predominance of the Atlantic fronts, whereas the Mediterranean convective rainfalls have 601 less impact and play a significant role especially in summer, when the influence of the 602 large-scale Atlantic circulation is reduced (Moreno et al., 2014, 2021; Pérez-Mejías et al., 603 2018; Cabos et al., 2020). The present-day records in eastern Iberia also indicate a close 604 relationship between Atlantic Ocean temperatures and relative humidity as the main drivers of the P–E rate, suggesting that δ^{18} O values in the continental realm are highly 605 606 sensitive to these changes (e.g. Pérez-Mejías et al., 2018). Inter-annual rainfall 607 oscillations are controlled by the seasonal migration of the Subtropical High-Pressure 608 Zone (SHPZ) and changes in the North Atlantic Oscillation (NAO) (Moreno et al., 2014; 609 Hernández et al., 2015). As the main orographic barriers have not changed since the Late 610 Miocene, it is thought that a similar situation must have prevailed in the Iberian Peninsula 611 since then and that changes in temperature and moisture in the Atlantic have had a strong 612 influence on inland areas.

613 A remarkable, dramatic transition in the oceanic and atmospheric circulation was 614 associated with huge paleogeographic changes during the Late Neogene (Haug et al., 615 2001; Zachos et al., 2001; Quan et al., 2014). During this period, global climate 616 conditions changed until an Earth configuration similar to today's was reached. A shift in 617 the atmospheric circulation in the Tortonian (Quan et al., 2014), with a high-pressure 618 system over Atlantic mid-latitudes, imposed the ocean-water and atmospheric pattern that 619 currently prevails in western Europe, with dominant westerly winds in Europe (Quan et 620 al., 2014; Bell et al., 2015; Herold et al., 2012; Sherriff-Tadano et al., 2018). Although 621 the closure of the Central Atlantic Seaway (CAS) started ~15 Ma ago, the first emergence

622 of land and the early disconnection between Pacific and Atlantic deep-waters occurred 623 from ~10 to ~7 Ma, and the disconnection of surface waters took place between ~3.5 and 624 2.5 Ma (Molnar, 2008; Bell et al., 2015). The Pacific-Atlantic disconnection triggered the 625 intensification of the Atlantic Meridional Overturning Circulation (AMOC), and the 626 transport of heat and moisture in the Atlantic became more efficient (Karas et al., 2017), 627 impacting the global atmospheric circulation and the climate in western Europe 628 landwards (Böhme et al., 2008; Bell et al., 2015; Lohman et al., 2015; Karas et al., 2017). 629 In general, since the closure of the CAS (~7 Ma), water exchange between the Pacific and 630 Atlantic oceans has been severely restricted. Nevertheless, a temporary connection 631 between the two oceans has been reestablished during some episodes, triggering AMOC 632 weakness (Böhme et al., 2008; De Schepper et al., 2013). The strengthening or weakening 633 of the AMOC implies, respectively, an increase or decrease in the SST in the North 634 Atlantic, the changes at the mid-high latitudes of the northern hemisphere being more 635 marked since the closure of the CAS (Molnar, 2008; Bell et al., 2015). A general 636 synchrony between the oscillations of the SST in the different North Atlantic Ocean 637 Drilling Program (ODP) sites has been shown since the closure of the CAS (Molnar, 638 2008; Zhang et al., 2013; Fedorov et al., 2013; Bell et al., 2015) (Fig. 11). In addition to 639 the evolution of the CAS, changes in the Arctic gateways through the NADW (Bell et al., 640 2015) and the Mediterranean outflow into the Atlantic waters (Pérez Asensio et al., 2012; 641 Ivanovic et al., 2014) could have also impacted the strengthening or weakening of the 642 AMOC during some episodes (Karas et al., 2017).

As previously stated, until ~8.6 Ma our isotope record and the SST of the Atlantic 643 644 Ocean show low variability (Fig. 11), a situation that could be associated with the 645 incipient closure of the CAS and a possible water exchange between the Pacific and 646 Atlantic Oceans that drove a weak North Atlantic circulation (Molnar, 2008; De Schepper 647 et al., 2013; Bell et al., 2015). The still open oceanic connection could have induced 648 anomalies in surface winds (Quan et al., 2014; Herold et al., 2012; Sherriff-Tadano et al., 649 2018), resulting in low effective heat and moisture transfer to the continent at the eastern 650 Atlantic margin. Moreover, a Mediterranean-type climate, dry in summer, had not yet been established (Quan *et al.*, 2014). The agreement between the Teruel $\delta^{18}O_c$ curve and 651 652 SST and foraminiferal δ^{18} O datasets from ocean records at the same latitude and in the 653 same climate band (Herbert et al., 2016; Raymo et al., 2018) increases from the Late 654 Tortonian (~8.5 Ma), when the deep CAS waters disconnected (Molnar, 2008; Bell et al.,

655 2015). The AMOC system was then reinforced, and the current climatic dynamics and 656 temperature gradients were established, with climate oscillations in western Europe being 657 at least partially related to a situation of weaker or stronger AMOC (Fig. 11). At ~7.3 Ma, 658 the subtropical high at mid-latitudes and the westerly belt reached their current position 659 and from then on, the Atlantic conditions exerted a further influence on western Europe 660 (Quan *et al.*, 2014; Herold *et al.*, 2012; Sherriff-Tadano *et al.*, 2018). From this point on, 661 our data fit well with the SST variations in the Atlantic (Fig. 11).

662 A gradient is observed between Atlantic records from distinct latitudes (Fig. 11), 663 with the ODP 608 record in the south (42°N) showing higher and more constant 664 temperatures from the Late Tortonian to the Messinian, in line with the Teruel $\delta^{18}O_c$ data. 665 Moreover, with increasing latitude, a general cooling tendency from 8 Ma until the end 666 of the Late Messinian is more marked in the ocean records (Miller et al., 1991; Zachos et 667 al., 2001; van Dam & Reichart, 2009; Herbert et al., 2016; Super et al., 2020). Between 668 ~7 and 5.4 Ma (Late Miocene Cooling, LMC), the ocean water temperature dropped ~4°C 669 at a global scale at mid-latitudes (Turco et al., 2001; Herbert et al., 2016), and 670 temperatures in the northern hemisphere were frequently poised near, but not below, the 671 threshold for permanent continental ice formation (Herbert et al., 2016). Nevertheless, on 672 the basis of δ^{13} C data, some authors propose that during the LMC several shorter warming 673 events occurred related to temporary changes in the NADW (Bell et al., 2015; Otto-Bliesner *et al.*, 2017). As previously expounded, our $\delta^{18}O_c$ record, like other climate 674 675 datasets for the Iberian Peninsula (Fig. 10), suggests generally warm conditions in eastern 676 Iberia from the Late Tortonian to the Zanclean. Even so, gentle excursions to lower δ^{18} O 677 values (at ~7.1, ~6, and 5.3 to 5 Ma; Fig. 11) provide evidence of wetter-cooler periods in the region. These coincide with the decreases in temperature detected in the ODP 982, 678 679 ODP 907, and Monte dei Corvi records (Fig. 11), suggesting that profound changes in the 680 oceanic system could have had a clear effect on the SW Mediterranean area, even within 681 warm climate conditions.

The subsequent recovery of the AMOC (Fig. 11) induced northern hemisphere warming in the Early Pliocene (Karas *et al.*, 2017), and the later weakening of the AMOC from ~3.8 to 3 Ma is in line with an Early Piacenzian global cooling trend and SST variations on a global scale (Fedorov *et al.*, 2013; Tzanova *et al.*, 2015; Herbert *et al.*, 2016). Both changes are recognized in our isotope curve, with increasing $\delta^{18}O_c$ values from 5.3 to 4.2 Ma in the Zanclean and decreasing values thereafter (Figs. 10 & 11). From the Piacenzian on, it is also evident that our $\delta^{18}O_c$ record shows lower values during weak AMOC stages (3.8-3 and 2.5-2.2 Ma) and higher values during strong AMOC episodes (3-2.5 and 2.2-2 Ma), which clearly evidences the influence of Atlantic dynamics on the climate of continental Iberia (Fig. 11).

692 In light of the foregoing, it can be stated that changes in the AMOC influence the 693 climate of southwestern Europe mainly through modifications in the high-pressure centre 694 in the mid-Atlantic (Fig. 12). Under a weak AMOC scenario and with a weakening of the 695 Azores High, cold fronts from the Icelandic low-pressure system are allowed to sweep 696 across the moderate latitudes of continental Europe. The resulting climate is cold and dry 697 in northern Europe and wet in the Mediterranean area. It is proposed that under these conditions, an increasing P–E ratio and decreasing δ^{18} O precipitation values would induce 698 lower $\delta^{18}O_c$ in our lacustrine record. Conversely, a pronounced Azores High would block 699 westerly winds and storms, which would be diverted towards more northerly areas. As a 700 701 consequence, aridity would increase in southwestern Europe, the P-E ratio would fall, 702 and δ^{18} O precipitation values would increase; accordingly, higher δ^{18} O_c values were 703 recorded in the Teruel record under this situation.

704 Considering the influence of the AMOC and eccentricity-induced modulation, two 705 end-member scenarios can be envisaged (Fig. 12). During periods of strong AMOC and 706 minimal eccentricity, the driest conditions would be reached, which would induce higher values of $\delta^{18}O_c$ (Fig. 12a). In contrast, during periods with weak AMOC and maximum 707 eccentricity, the wettest conditions would be reached, which would imply lower values 708 of $\delta^{18}O_c$ (Fig. 12b). The intermediate scenarios would correspond to values of $\delta^{18}O_c$ 709 710 between the extremes. Moreover, the climate could have been modulated by the influence 711 of shorter-order orbital cyclicity.

712

713 **6.** Conclusions

714

715 Climate changes impacted the continental systems of southwestern Europe at 716 different scales during the Late Miocene-Early Pleistocene. The isotope signature of 717 lacustrine carbonates in the Teruel Basin corroborates a relationship between $\delta^{18}O_c$ and 718 the climate signal through the P–E ratio and a connection between dry and warm periods 719 in this area of the western Mediterranean.

720 Orbital forcing controlled the climate conditions, and the relation between 721 eccentricity and $\delta^{18}O_c$ values shows a correlation of eccentricity minima and maxima with 722 dry and humid stages, respectively. The lacustrine record shows that the present-day 723 connection between Atlantic dynamics and climate far from the coast in eastern Iberia 724 already existed during the Late Miocene. The $\delta^{18}O_c$ curve agrees with North Atlantic sea-725 surface temperature curves, a connection existing between dry periods and high SST, 726 which supports the influence of marine climate conditions landwards through heat and 727 moisture transport from the Atlantic Ocean to the continent. A relation between climate 728 changes in the region and marine isotope stages is also hypothesized. The effect of the 729 closure of the Central Atlantic Seaway from ~8.6 Ma was to reinforce the warm/dry 730 synchrony related to a stable high-pressure centre in the mid-Atlantic during strong 731 AMOC stages. This diverted westerly winds northwards, increasing aridity in 732 southwestern Europe.

733 The inferred warm/dry connection in the Teruel Basin, which differs from other 734 western Mediterranean areas, supports previous interpretations proposing a regional 735 climate gradient in western Europe, where warming and higher moisture have run 736 parallel. The current paleogeographic configuration was established in the Late Miocene, 737 the basin being located in an intramountainous area with an orographic barrier to the E-738 SE. As occurs at present, this situation would have isolated the study area from the 739 influence of Mediterranean Sea humidity during warm periods, enhancing the Atlantic-740 continental connection.

The $\delta^{18}O_c$ curve presented here sheds substantial new light on the past climate in the western Mediterranean continental area and provides a powerful tool for other studies in eastern Iberia, especially of a paleontological or paleoecological nature. It is the longest paleoclimate record published and shows higher resolution than previous ones, helping to confirm past climate variations and fill information gaps between 9.8 and 1.8 Ma.

746

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- 756

757 Abbreviations

- 758 AMOC- Atlantic Meridional Overturning Circulation
- 759 CAS Central Atlantic Seaway
- 760 ELMA European Land Mammal Age
- 761 ETWZ European Temperate Wet Zone
- 762 GPTS Geomagnetic Polarity Time Scale
- 763 LMC Late Miocene Cooling
- 764 MAP Mean Annual Precipitation
- 765 MAT Mean Annual Temperature
- 766 MCO- Miocene Climate Optimum
- 767 mPWP mid-Pliocene Warm Period
- 768 NADW North Atlantic Deep Water
- 769 NAO North Atlantic Oscillation
- 770 ODP Ocean Drilling Program
- 771 P–E Precipitation–evaporation
- 772 SHPZ Subtropical High-Pressure Zone
- 773 SST Sea Surface Temperature
- 774

775 **Figure captions**

Fig. 1. Deep Sea Drilling Program Map with the location of the Teruel Basin (red star)
(inset shows the position of this intramountain basin in the eastern Iberian Range in
Iberia). Selected sites from the Ocean Drilling Program (ODP) and Integrated Ocean
Drilling Program (IODP), used in the discussion of the results (Figure 11) due to their
proximity to the studied region, are also included (marked as black dots). MC refers to
the Monte dei Corvi site.

Fig. 2. Geological context (a) and geological map (b) of the northern sector of the Teruel
Basin.

Fig. 3. Composite stratigraphic profile of the northern Teruel Basin showing the calcite $\delta^{18}O_c$, $\delta^{13}C_c$, and mineralogical results. Lithologies are distinguished by colours: limestones and marls (blue, grey, and green), carbonated silts (purple), and mudstones (yellow). The anchoring with the chronostratigraphic time scale was based on magnetostratigraphic data and paleontological sites. The three climate periods proposed in this work are indicated.

Fig. 4. Field images of the lacustrine beds. a. Massive limestones in tabular-irregular
strata (outcrop thickness ~20 m). b. Detail of the carbonate beds. c. Detail of the
bioturbated tabular limestones.

Fig. 5. Field emission scanning electron microscope images (FESEM) of the limestones.
Calcite crystals are in general small in size, with rhombohedral habits and common
knobby surfaces consistent with neoformation.

- **Fig. 6. a.** Qz + Phy and Cc correlation for the lacustrine samples from the Teruel Basin.
- **b.** Phy and Qz. **c.** Gy and Cc. **d.** Qz + Phy and Cc + Gy. Authigenic minerals such as
- calcite (Cc) and gypsum (Gy) show a negative linear relation with detrital minerals suchas quartz (Qz) and phyllosilicates (Phy).
- 800 **Fig. 7.** $\delta^{18}O_c$ and $\delta^{13}C_c$ plots of the lacustrine carbonates in the Teruel Basin (blue dots) 801 and the surrounding Mesozoic carbonate rocks (in black).
- **Fig. 8. a.** Stable oxygen isotope ($\delta^{18}O_c$) curve of the Teruel Basin, spanning from ca. 9.8 802 803 to 1.8 Ma, calculated from lacustrine calcites; three broad/main climate periods are 804 distinguished. **b.** Results from the spectral analysis (2σ MTM power spectrum with background AR(1) model and 95%, 99%, and 99.9% confidence levels) for the 66 to 209 805 m interval of the δ^{18} O time series and **c.** for the 140 to 209 m interval of the δ^{18} O time 806 807 series. **d.** Tuned cyclostratigraphy for the filtered δ^{18} Oc time series and eccentricity curve 808 calculated with the La2010a model (Gaussian filter passbands are 2.5 ± 0.5 and 0.4 ± 0.1). 809 Tuning is based on assigning the pervasive 13.33 m cycle to the 405-ka eccentricity cycle. It is found that the most negative $\delta^{18}O_c$ values correlate with the maximum eccentricity 810 811 of the Earth's orbit.

Fig. 9. COCO analysis and eCOCO sedimentation rate map of the δ^{18} Oc series in the Teruel profile (66-209 m). **a.** Composite stratigraphic profile for the northern Teruel Basin with calcite δ^{18} O_c and the Gaussian filter centred at 13.33 m (frequency = 0.075±0.015) for the section used in the spectral analysis. **b.** The correlation coefficient 816 (top) and evolutionary correlation coefficient (bottom, coloured area). **c.** Null hypothesis 817 test (top) and evolutionary null hypothesis (H₀) significance level (bottom). For both the 818 COCO and eCOCO analyses, the tested sedimentation rates range from 1 to 10 cm/ka 819 with a step of 0.1 cm/ka, and the number of Monte Carlo simulations is 2000. For the 820 eCOCO analysis, the sliding window size is 35.75 m; the sliding window step is 1 m.

Fig. 10. Correlation of the $\delta^{18}O_c$ curve for the Teruel Basin with published climatic 821 records for this basin and the east of the Iberian Peninsula. **a.** Our $\delta^{18}O_c$ curve with the 822 periods of high and low aridity interpreted. b. Temperature/aridity curves based on 823 824 micromammal associations in the Teruel Basin and other Neogene basins in Iberia. c. 825 Temperature/aridity curves interpreted from the dentition record of macromammals in 826 eastern Iberia. References are indicated in the figure. A comparison of the curves 827 reinforces our proposal that low values of $\delta^{18}O_c$ indicate wet/cool periods and high $\delta^{18}O_c$ 828 values indicate dry/warm periods.

Fig. 11. Correlation of the Teruel Basin $\delta^{18}O_c$ curve and other climate records. a. The 829 830 $\delta^{18}O_c$ record reconstructed in this work as explained in Figure 3; the temporal position of some geological milestones is indicated. **b.** The δ^{18} O curve of deep marine benthic 831 832 for a surface temperature (SST) and atmospheric pCO_2 833 changes inferred from North Atlantic Ocean and Mediterranean alkenone records. 834 Previously proposed changes in the AMOC have been included. The references from 835 which these data have been extracted are indicated. The MIS events as identified by 836 Shackleton et al. (1995) and Lisiecki & Raymo (2005). Mi-glacials as established Abreu 837 & Anderson (1998) and Wetershold et al. (2005).

Fig. 12. Synthesis of the possible scenarios involving eccentricity and AMOCoverlapping during the studied time period.

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