1 **Early Pliocene climatic optimum, cooling and early glaciation deduced**

2 **by terrestrial and marine environmental changes in SW Spain**

3

4

5 Gonzalo Jiménez-Moreno[1,*], José N. Pérez-Asensio[2], Juan C. Larrasoaña[3,4], Francisco J.

6 Sierro[5], Daniel Garcia-Castellanos[4], Ángel Salazar[6], Josep Maria Salvany[7], Santiago

7 Ledesma[8], M. Pilar Mata[6] and Carlos Mediavilla[9]

8

9 [1]*Departamento de Estratigrafía y Paleontología, Universidad de Granada, 18002*

10 *Granada, Spain*

11 [2]*GRC Geociències Marines, Departament de Dinàmica de la Terra i de l'Oceà,*

12 *Universitat de Barcelona, 08028 Barcelona, Spain*

13 [3]*Instituto Geológico y Minero de España, Unidad de Zaragoza, 50006 Zaragoza, Spain*

14 [4]*Institute of Earth Sciences Jaume Almera, Consejo Superior de Investigaciones*

15 *Científicas, 08028 Barcelona, Spain*

16 [5]*Department of Geology, University of Salamanca, 37008 Salamanca, Spain*

17 [6]*Instituto Geológico y Minero de España, 28760 Tres Cantos, Madrid, Spain*

18 [7]*Departament d'Enginyeria Civil i Ambiental, Universitat Politècnica de Catalunya,*

19 *08034 Barcelona, Spain*

20 [8] *Naturgy Energy Group, S.A., 28033 Madrid, Spain*

21 [9]*Instituto Geológico y Minero de España, 41013 Sevilla, Spain*

22

23

1

24

25   *Corresponding author: Tel: +34 958 248727.

26   E-mail address: gonzaloj@ugr.es (G. Jiménez-Moreno).

27

30

Abstract

31

32   The Pliocene is a key period in Earth's climate evolution, as it records the transition from

33   warm and stable conditions to the colder and more variable glaciated climate of the

34   Pleistocene. Simultaneously, climate became more seasonal in the Mediterranean area,

35   and Mediterranean-type seasonal precipitation rhythm with summer drought established.

36   These climatic changes presumably had significant impacts on terrestrial environments.

37   However, the response of terrestrial environments to such climate changes is still not fully

38   understood due to the lack of detailed studies dealing with this period of time. In this

39   study, multiproxy analyses of continuous core sampling from La Matilla (SW Spain)

40   shows detailed and continuous record of pollen, sand content and abundance of benthic

41   foraminifer *Bolivina spathulata* to describe paleoenvironmental and paleoclimate trends

42   during the early Pliocene. This record shows warmest, most humid climate conditions

43   and highest riverine nutrient supply at ~ 4.35 Ma, coinciding with the Pliocene climatic

44   optimum and high global sea level. A climate cooling and aridity trend occurred

45   subsequently and a significant glaciation occurred at ~ 4.1-4.0 Ma, during a period known

46   by very little terrestrial evidence of glaciation. Our multiproxy data thus indicate that

47   terrestrial and marine environments were significantly variable during the early Pliocene

48   and that major glaciation-like cooling occurred before the intensification of northern

49   hemisphere glaciation at the beginning of the Pleistocene (~2.7 Ma). This major climate

50   cooling and aridity maxima between 4.1-4.0 Ma is independently validated by a coeval

51   sea-level drop (third order Za2 sequence boundary). This sea level drawdown is supported

52   by enhanced coarse sedimentation and minima in riverine nutrient supply, showing paired

53   vegetation and sea-level changes and thus a strong land-ocean relationship. This study

54   also shows that long-term climatic trends were interrupted by orbital-scale cyclic climatic

55   variability, with eccentricity, obliquity and precession acting as the main triggers

56   controlling climate and environmental change in the area.

57

58   **1. Introduction**

59

60   The early Pliocene (Zanclean stage; between 5.3 and 3.6 Ma) is a very interesting

61   period because it encompasses the so-called Pliocene climatic optimum (PCO) from 4.4

62   to 4 Ma, probably the warmest interval in this epoch with global average temperature ~

63   4 °C higher than today (Brierley and Fedorov, 2010; Fedorov et al., 2013). Carbon

64   dioxide reconstructions show higher-than-present values around 410 μatm (Bartoli et al.,

65   2011) and a global sea-level about 25 m higher than present (Miller et al., 2005).

66   Therefore, a great effort has been put into obtaining palaeoclimate records for the

67   Pliocene, as it is recognized as the most recent example of prolonged global warmth in a

68   higher-than-today CO_2 context in the geological past (Fedorov et al., 2013) and a very

69   good analog for future climate change (Robinson et al., 2008).

70   Marine isotopic records suggest that a climate cooling took place from the warmer

71   and more stable early Pliocene to the colder and more variable Pleistocene (Lisiecki and

72   Raymo, 2005). This gradual cooling was probably due to a decrease in atmospheric

73   carbon dioxide of about 100 parts per million (Bartoli et al., 2011). Even though the

3

74 Pliocene is thought to be a warmer period than today, a recent review by De Schepper et

75 al. (2014) shows evidences of significant glaciation events in both northern and southern

76 Hemispheres during this epoch. A progressive aridification in the Mediterranean area also

77 happened during the Pliocene (Fauquette et al., 1998; Popescu et al., 2010; Jiménez-

78 Moreno et al., 2013a), as well as the establishment of the Mediterranean-type seasonal

79 precipitation rhythm (summer drought) at ~ 3.4 Ma (Suc, 1984; Suc and Popescu, 2005;

80 Jiménez-Moreno et al., 2010). All these changes greatly affected past terrestrial

81 environments in the Mediterranean area and vegetation experienced a decrease and

82 disappearance of many thermophilous and hygrophilous species and the increase in

83 xerophytes and Mediterranean adapted taxa (Suc, 1984; Popescu et al., 2010). Very few

84 studies provide with information about how terrestrial western Mediterranean

85 environments reacted to such long- and short-term climatic changes (Suc, 1984; Bertini,

86 2001; Combourieu-Nebout et al., 2004; Jiménez-Moreno et al., 2010; 2013a). Therefore,

87 detailed pollen studies are needed to improve our knowledge about vegetation changes

88 and the main forcings triggering cyclical vegetation changes during the early Pliocene in

89 the Mediterranean area (i.e., Gauthier and Muñoz, 2009).

90   In the present study, we show a detailed and continuous record of pollen,

91 percentage of sand and abundance of the benthic foraminifer *Bolivina spathulata* from a

92 marine sedimentary record from the Guadalquivir Basin (SW Spain). This was done with

93 the main goal of describing long-term paleoenvironmental and paleoclimate trends during

94 the early Pliocene climatic optimum in the study area. This record shows that besides

95 long-term trends, climate was also characterized by cyclical variability (i.e., orbital

96 changes) that forced both terrestrial (e.g., vegetation changes of forested vs. open

97 vegetation) and marine (sedimentation, organic matter fluxes, sea-level) environmental

98 changes. The possible triggers of the observed orbital-scale variability are also discussed.

99

2. Geological setting

101

The study area is the western sector of the Guadalquivir Basin in SW Spain (Fig. 1A). This ENE-WSW-elongated foreland basin is limited to the north by the passive Iberian Massif and to the south by the active Betic Cordillera (Braga et al., 2002; González-Delgado et al., 2004). The Guadalquivir Basin has been linked to the isostatic subsidence produced by the loading of the Betic units on the south Iberian margin during the Neogene. By the earliest Tortonian, the Guadalquivir Basin was the Atlantic side of the North Betic Strait, connecting the Atlantic Ocean and the Mediterranean Sea (Martín et al., 2014; Braga et al., 2010). Afterwards, its paleogeography became an open and wide Atlantic bay (Martín el al., 2009), mostly in absence of fault tectonics (Fernàndez et al., 1998; Garcia-Castellanos et al., 2002), although subject to slow geodynamic vertical motions related to the mantle dynamics under the Betic cordillera (Garcia-Castellanos et al., 2002; Garcia-Castellanos & Villaseñor, 2011; Pérez-Asensio et al., 2018). During the Pliocene, the coastline steadily migrated westwards (Sierro et al., 1996) due to the axial sediment input together with the flexural isostatic response of the Betic-Guadalquivir-Hercynian massif system. Flexural basculation towards the south, in combination with the main sediment source being in the Betic cordillera, determined an axial (along-strike) fluvial drainage running along the northern boundary of the sedimentary basin (Garcia-Castellanos, 2002) as the Guadalquivir River does today. Sediments filling the Guadalquivir Basin are organized in several marine and continental units ranging from the Tortonian to the Holocene and the olistostromic deposits from the Betic Cordillera (Riaza and Martínez del Olmo, 1996; Sierro et al., 1990, 1996; González-Delgado et al.,

123   2004; Salvany et al., 2011; Larrasoaña et al., 2014; Aguirre et al., 2015; Rodríguez-

124   Ramírez et al., 2016).

125   Neogene sediments from the study area have been ascribed to eight marine and

126   continental lithostratigraphic units (Civis et al., 1987; Salvany et al., 2011) (Fig. 1). The

127   lowermost unit over the Iberian Massif basement is the Niebla formation (late Tortonian),

128   which is composed of coastal carbonate-siliciclastic deposits (Civis et al., 1987; Baceta

129   and Pendón, 1999). The second unit is the Gibraleón Formation (latest Tortonian-early

130   Pliocene) and includes greenish-bluish clays with glauconitic silts at its base (Civis et al.,

131   1987; Flores, 1987; Sierro et al., 1993; Pérez-Asensio et al., 2018). A Transitional Unit

132   (late Messinian-early Pliocene) comprising transitional facies with alternating silts and

133   sands is found over the Gibraleón Formation in central areas of the basin (De Torres,

134   1975; Mayoral and González, 1986–1987; Muñiz and Mayoral, 1996; Pérez-Asensio et

135   al., 2018). The third unit, the Huelva Formation (early Pliocene), consists of silts and

136   sands with a glauconitic layer at the base (Civis et al., 1987). The fourth unit is the sandy

137   Bonares Formation (early Pliocene) (Mayoral and Pendón, 1986-87). Four continental

138   units have been described above the marine units (Salvany et al., 2011). The Almonte

139   Formation (Upper Pliocene-Lower Pleistocene) is the first continental unit, and is

140   comprised of gravels and sands from proximal-alluvial deposits. Proximal sediments

141   from this unit are the so-called High Alluvial Level (Pendón and Rodríguez-Vidal, 1986–

142   1987; Salvany et al., 2011). The second continental unit is the Lebrija Formation (late

143   Pliocene-late Pleistocene), which includes sands, gravels and clays from distal-alluvial

144   deposits. The two last continental units (latest Pleistocene-Holocene) are the continental

145   Abalario Formation and the continental-estuarine Marismas Formation. They consist of

146   eolian sands and alluvial-estuarine clays, respectively.

147

3. Material and methods

The 276-m-long La Matilla (LM) core was drilled in the vicinity of Mazagón (Huelva) in the western Guadalquivir Basin (37º10'26.77''N, 6º43'27.14''W; 47 m altitude; Fig. 1). The LM core, currently curated at the Geological and Mining Institute of Spain (IGME) core repository, was drilled by IGME in 2006 using a rotary drilling rig with continuous core sampling. The core description, including lithostratigraphy, facies analysis and assignation of different units to the different formations outcropping in the lower Guadalquivir Basin area, was previously done in Pérez-Asensio et al. (2018).

The LM core chronology was developed from a combination of biostratigraphy and magnetostratigraphy (for a detailed explanation, see Pérez-Asensio et al., 2018; Fig. 2). Following the age model produced by these authors, the LM core contains a continuous early Pliocene sedimentary record comprised between 4.50 and 3.95 Ma (Pérez-Asensio et al., 2018; Fig. 2). The basinward migration of depositional facies in the Guadalquivir Basin explains why stratigraphic formations classically attributed to the Late Miocene in land sections (e.g., Gibraleón Formation) have an early Pliocene age at the location of LM core (Pérez-Asensio et al., 2018). Age estimates for the studied samples from the La Matilla core were assigned by assuming linear accumulation rates between the age tie points provided by the paleomagnetic boundaries (the tops of chrons C3n.2n, C3n.1r, and C3n.1n). For the lowermost part of the core (N1 polarity interval), the sedimentation rate of the R1 polarity interval (42.9 cm/ka) was extrapolated downward (Fig. 2). In the upper part of the core (R2 and an interval of uncertain polarity), the sedimentation rate of the R2 polarity interval (49.9 cm/ka) was extrapolated upward (Fig. 2). This resulted in an age of 3.95 Ma for the end of marine sedimentation (Fig. 2).

171   The percentage of sand content in the different facies along the 3 studied marine

172   units (Fig. 2) was calculated every 3 m (83 samples) dividing the weight of the sand

173   fraction (>63 μm) by the total sample weight (50 g) and multiplying by 100.

174   Samples (2 cm^3) for palynological analyses were taken roughly every 5 m

175   throughout the core, with a total of 52 samples analyzed. The palynomorph extraction

176   method followed a modified Faegri and Iversen (1989) methodology. Counting was

177   performed at x400 and x1000 magnifications to a minimum pollen sum of 300 terrestrial

178   pollen grains. Fossil pollen was compared with present-day relatives using published keys

179   (e.g., Beug, 1961) and a modern pollen reference collection at the University of Granada

180   (Spain). The raw counts were transformed to pollen percentages based on the terrestrial

181   sum excluding *Pinus* - usually overrepresented in marine environments because of the

182   advantage of bisaccate pollen for long-distance transport (Heusser, 1988; Jiménez-

183   Moreno et al., 2005). The pollen zonation of the detailed pollen diagram was

184   accomplished using CONISS (Grimm, 1987; Fig. 3). Arboreal Pollen (AP) abundance

185   was calculated based on the total pollen sum with and without *Pinus*. Furthermore, pollen

186   taxa were grouped, according to present-day ecological bases, into thermophilous

187   (subtropical) trees. The thermophilous tree group includes Arecaeae, Taxodioideae,

188   Rutaceae, Euphorbiaceae, *Cissus*, *Prosopis*, *Engelhardia* and Sapotaceae. An AP/arid

189   ratio was calculated in order to discriminate between warm/humid (i.e.,

190   interglacial/interstadial phases) and cold/arid (i.e., glacial/stadial phases). As arid taxa,

191   we included *Artemisia*, *Ephedra, Lygeum* and Asteraceae total (Cichorioideae and

192   Asteroideae). Dinoflagellate cysts (dinocysts) occur with the pollen in the studied

193   samples but at very low occurrences, which precluded their statistical study.

194   Principal Components Analysis (PCA) using PAST version 3.24 (Hammer et al.,

195   2001) was run on the most abundant pollen taxa time series (Fig. 4). This was done to

find hypothetical variables (components; i.e., environmental or climate parameters) accounting for as much as possible of the variance in the pollen data. A PCA correlation scatter diagram is shown in Fig. 4. This diagram shows to what degree the different taxa correlate with the principal component 1 (PC1) and 2 (PC2). PC1 data time series is represented in Figures 4B and 5.

A cyclostratigraphic analysis was performed on the *Quercus* and AP raw pollen percentages and PC1 pollen time series from the LM sedimentary sequence (Fig. 6). We used the software PAST (Hammer et al., 2001) with the program REDFIT (Schulz and Mudelsee, 2002) with the objective of characterizing the different periodicities present in the unevenly spaced raw pollen data.

A total of 83 samples of 50 g were analyzed for benthic foraminifera. At least 300 benthic foraminifera from the size fraction >125 μm were counted and identified. The relative abundances (%) of the benthic foraminifer *Bolivina spathulata* (Fig. 7) were obtained from transforming the raw counts of this species into percentages. Higher abundances of this shallow infaunal species were used as proxy for higher organic matter fluxes linked to riverine discharge (Duchemin et al., 2008; Schmiedl et al., 2010; Pérez-Asensio et al., 2014).

4. Results

4.1. Sand content

Sand content of the marine units of the LM sediment core shows an overall increasing trend towards the top of the core and varies from 0.2 (255 m; 4.44 Ma) to 93.4 % (48 m; 4.01 Ma) (Fig. 2). Very low sand % values (lower than 10%) characterize the core bottom, from 275 (4.49 Ma) to 150 m (4.21 Ma). Sand % increases in three cyclical increasing-

221 decreasing cycles since then, with peaks of 14, 20 and 33% at 147 (4.21 Ma), 132 (4.18

222 Ma) and 114 m (4.14 Ma), respectively. Minimum values (<5%) are reached again

223 between 102-96 m (4.12-4.10 Ma). An increase in sand % is observed later on, with a

224 maximum of 68% at 81 m (4.07 Ma). Sand values decreased after this peak, and another

225 minimum of ~20% is recorded at 78-75 m (4.07-4.06 Ma). A substantial increase is

226 observed then with a peak of ~80% recorded at 60 m (4.03 Ma). A relative minimum of

227 58% is recorded at 57 m (4.02 Ma). Another increase-decrease cyclical oscillation is

228 observed between 57 and 39 m (4.02-3.99 Ma), with a maximum in sand of 93% recorded

229 at 48 m (4.01 Ma) and another drop in the sand values at 39 m (3.99 Ma). An increase in

230 sand % with values around 70% characterize the record between 36-33 m (3.987-3.985

231 Ma).

232

233 *4.2. Pollen analysis*

234 Seventy-three different pollen taxa have been identified in the LM sedimentary record

235 pollen spectra. The record shows diverse pollen taxa. However, many of the identified

236 taxa occur in percentages lower than 1% and have not been plotted in Figure 3. These

237 rare species include thermophilous (subtropical) plants such as Arecaceae, Taxodioideae,

238 Rutaceae, Euphorbiaceae, *Cissus*, *Prosopis*, *Engelhardia* and Sapotaceae. Temperate tree

239 species are also diversified, but many of them occur in very low percentages (i.e., *Acer*,

240 *Castanea-Castanopsis* type, *Rhus*, *Pterocarya*, *Carya*, *Juglans*, *Corylus*, *Liquidambar*,

241 *Distylium*, *Lonicera*, *Hedera* and *Ulmus*), except for *Quercus* (see below).

242 Overall, the LM core pollen record shows high abundances of non-arboreal pollen

243 (NAP) (i.e., herbs and grasses), with average abundances around 75%. NAP pollen is

244 dominated by Asteraceae Cichorioideae, Asteraceae Asteroideae, Poaceae and

245 Amaranthaceae, and in smaller abundances, *Plantago*, Apiaceae, Cistaceae, *Ephedra*,

246 *Artemisia* and Liliaceae. With respect to the AP, taxa are dominated by *Pinus* varying

247 around average values of 26%. Less abundant trees are mostly dominated by evergreen

248 and deciduous *Quercus*, *Olea*, Cupressaceae, *Cedrus* and *Cathaya*. Aquatics such as

249 Cyperaceae also occur in this record but in low average abundances around 1%. We used

250 variations in pollen species to objectively zone the pollen data using the program CONISS

251 (Grimm, 1987), producing three pollen zones for the LM record (Fig. 3). The pollen

252 results in these zones are described below:

253 Pollen zone LM-1 (267 – 250 m, 4.47 – 4.43 Ma) is characterized by high NAP

254 percentage, with high abundance of Asteraceae Cichorioideae of around 40%, Poaceae

255 around 22% and Asteraceae Asteroideae around 10%.

256 Pollen zone LM-2 (250 – 105 m, 4.43 – 4.12 Ma) features a significant decrease in

257 NAP and the highest percentages of AP, which shows many cyclical oscillations and

258 several peaks above 45%. This zone is then characterized by the highest occurrences in

259 *Quercus* total (evergreen and deciduous) of 23% at the beginning of this zone at 216 m

260 (~4.35 Ma). *Pinus* and *Quercus* total stay relatively abundant but show a decreasing trend

261 throughout this zone. *Olea*, *Cathaya* and *Cedrus* occur in this zone with discrete

262 occurrences.

263 Pollen zone LM-3 (105 – 30 m, 4.12 – 3.97 Ma) is characterized by the decrease in

264 AP (notably in *Pinus* and *Quercus* but also in *Cathaya* and *Cedrus*) and the highest values

265 of NAP of the LM record, with two maxima centered at ~100 and 50 m (4.1 and 4.0 Ma).

266 *Olea* and Cupressaceae also increased and show maxima in this zone, featuring an

267 increasing trend interrupted by two cyclical oscillations.

268 PCA analysis on a selection of most abundant pollen taxa from the LM record shows

269 two main groups of distinctive taxa (Fig. 4A). PC1+ group, characterized by positive

270 correlation to PC1, is made up of the most abundant tree taxa such as *Pinus*, *Quercus*

271 (both evergreen and deciduous), *Cathaya*, *Cedrus* and the halophytic herb

272 Amaranthaceae. PC1- group is characterized by negative correlation to PC1 and is

273 dominated by non-arboreal species, such as Asteraceae Cichorioideae, Asteraceae

274 Asteroideae, Poaceae, *Ephedra*, and Mediterranean-adapted tree species *Olea* and

275 Cupressaceae. PCA analysis indicates that PC1 (probably a combination of temperature

276 and precipitation, see below) is strong, explaining the 44.12% of the variance. Pollen data

277 and CONISS cluster analysis agree with the PC1 (correlation of PC1 with AP: r=0.92;

278 p<0.01; PC1 with *Quercus* total r=0.48; p<0.01) and pollen zones LM-1 and LM-3 are

279 characterized by low abundances in the PC1+ pollen group and zone LM-2 is

280 characterized by the highest values in the PC1+ (Fig. 4B).

281

282 *4.3. Periodicity of pollen changes*

283 Pollen data show a very clear cyclical pattern in the relative abundance of many taxa

284 such as *Quercus* (Fig. 3). The PC1 is also characterized by cyclical oscillations, which

285 covary with those observed in the raw *Quercus*, AP, thermophilous and AP/arid ratios

286 (Figs. 4B, 5; see discussion below). Spectral analysis on the *Quercus* and AP pollen data

287 and PC1 shows statistically significant (above the 80 and 90% confidence level) spectral

288 peaks with periodicities between 80 - 67, 51 - 44, 37, 30 and 22-21 ka (Fig. 6).

289

290 *4.4. Benthic foraminifer* Bolivina spathulata

291 Relative abundance (%) of *Bolivina spathulata* changes considerably throughout the

292 studied sediment core and varies from 0 to 50% (Fig. 7). The bottom part of the record

293 between 273 – 240 m (4.49 – 4.41 Ma) is characterized by low values around 10%. An

294 increase is observed then, reaching the maximum value of the record (~48%) at 213 m

295 (4.35 Ma). A decreasing trend is observed since then, interrupted by cyclical variability,

296   reaching several minima and the occasional absence of this species between 100 and the

297   top of the record (since 4.12 Ma).

298

299   **5. Discussion**

300

301   *5.1. Vegetation, benthic foraminifera and sedimentation as paleoenvironmental and*

302   *paleoclimate proxies*

303   Pollen analysis on Miocene and Pliocene marine sedimentary sequences from the

304   Iberian Peninsula area have shown the highly sensitive response of pollen records to

305   climate change, recording vegetation changes related to long-term and short-term orbital-

306   scale climate variability (Jiménez-Moreno et al., 2007; 2010; 2013b). These previous

307   studies show that the abundance of thermophilous forest species such as subtropical and

308   temperate taxa can be used to track temperature changes through time. Hygrophilous

309   species can also be used as proxies for precipitation, although sometimes it is difficult to

310   separate the temperature vs. precipitation signals, for certain species require both high

311   temperature and precipitation (or the opposite) to thrive. In this study we also used

312   present-day thermophilous (i.e., subtropical) and temperate adapted species as proxies for

313   temperature (Fig. 5). In this respect, *Quercus* (both evergreen and deciduous species)

314   abundance has been shown to be an excellent proxy recording warm-humid vs. cold-arid

315   (e.g., interglacials vs. glacials) paleoclimate phases in the southern Iberian Peninsula

316   (Combourieu-Nebout et al 2002; Jiménez-Moreno et al., 2013b; Camuera et al., 2018,

317   2019). AP abundance can be used as a proxy for precipitation, for forested species require

318   more water availability to grow than NAP taxa (Faegri and Iversen, 1987; Herzschuh,

319   2007). Here we added the arid indicators (*Artemisia*, *Ephedra* and Asteraceae total) to the

320   forested humid/non-forested arid equation, calculating an AP/arid ratio as a better proxy

321   for precipitation. The observed covariation between the PC1 with the above mentioned

322   proxies for temperature and precipitation also indicate that increases in PC1 are related

323   to warm/humid periods (Fig. 5). We are unsure about how to interpret the climatic

324   inference of the herb Amarathaceae in this pollen record. Statistically it seems to be

325   associated with the tree-dominated PC1+ group, however, in zone LM-02 it also shows a

326   timid and progressive increasing trend, covarying at that time with the aridity proxies so

327   it looks like it might be a mixed signal.

328   *Bolivina spathulata* abundance can be used as a proxy for organic matter fluxes

329   related with high freshwater riverine input (Duchemin et al., 2008; Schmiedl et al., 2010).

330   This species can tolerate low-oxygen conditions and prefer environments with supply of

331   terrestrial degraded organic matter related to river runoff (Barmawidjaja et al., 1992;

332   Stefanelli, 2004; Duchemin et al., 2008; Schmiedl et al., 2010) (Fig. 7). Previous studies

333   showed high abundance of *B. spathulata* during interglacial-like periods, characterized

334   by high riverine discharge bringing nutrients into the ocean (Pérez-Asensio et al., 2014).

335   Lithological changes (i.e., sand content) of the LM sedimentary core can be used as a

336   proxy for relative sea-level oscillations controlling the proximity to coast of the studied

337   site and thus the amount of coarse detritic input from the continent (Fig. 7).

338

339   *5.2. Climate change recorded in the LM core during the early Pliocene*

340

341   High abundances of NAP in the pollen spectra from LM indicate overall open

342   vegetation environments and arid conditions during the early Pliocene in the studied area.

343   The occurrence of herbaceous subdesertic taxa such as *Lygeum*, supports this

344   interpretation. Occurrences of subtropical species also point to a warm subtropical

345   climate. Climate was warmer than today, which is indicated by the higher occurrence of

346   thermophilous taxa currently living in warmer areas of North Africa (Suc et al., 1995).

347   This agrees with previous studies from the area for the early Pliocene, which show similar

348   vegetation and climate estimations of mean annual temperatures around 21ºC and mean

349   annual precipitation of ~400-600 mm (Andalucía G1 site; Jiménez-Moreno et al., 2010).

350   A mosaic of different plant associations inherited from the Miocene and organized in

351   altitudinal belts characterized the vegetation at that time. Subtropical evergreen-

352   deciduous species lived at low elevations, an open mixed deciduous forest (mainly made

353   up of conifers like *Pinus*, and several deciduous trees such as *Quercus*), occurred at higher

354   altitude, and conifers such as *Cathaya* and *Cedrus* grew at higher elevations on the

355   surrounding mountains.

356   Significant long-term changes in climate are observed during the early Pliocene

357   between 4.5 and 3.9 Ma in the SW Guadalquivir Basin region. This is deduced by changes

358   in the terrestrial vegetation that parallel marine benthic fauna and sedimentary variations

359   in the LM core (Fig. 7). The beginning of the studied record (including LM-1 pollen zone)

360   at around 4.45 Ma begins with overall cold and arid climate conditions. Relatively low

361   organic matter fluxes (low *B. spathulata* abundances) is also recorded at that time,

362   indicating low riverine runoff, which further support aridity at that time. This

363   interpretation agrees with climate inferences obtained from previous palynological

364   studies showing a cooling event at ~4.5 Ma, in the marine pollen record from the

365   Andalucía G1 site (Suc et al., 1995), located offshore of S Iberia and 300 km SE of LM,

366   and in the marine pollen record from the Garraf 1 site in NE Iberian Peninsula (Suc and

367   Cravatte, 1982). Globally, this time period corresponds with relatively low sea level, right

368   after the Za1 minimum sea-level sequence boundary (Handerbol et al., 1998), which

369   would agree with cold conditions during a period of eccentricity minima (Laskar et al.,

370   2004; Fig. 8).

371   Rapid warming and enhanced humid conditions occurred after ~4.37 Ma, and

372   warmest and wettest conditions in the vicinity of the LM record were reached at around

373   4.35 Ma. (Figs. 7 and 8). This is deduced by the highest occurrences in *Quercus* total,

374   thermophilous and AP/arid ratios of the LM record (Fig. 5). Conifers requiring relatively

375   high humidity such as *Cedrus* and *Cathaya* reached their maxima around that time (Fig.

376   3), supporting highest humidity. Highest *B. spathulata* abundances are also recorded at

377   4.35 Ma in the LM record (Fig. 7), indicating highest fluvial runoff supplying continental

378   degraded organic matter. Warmest conditions at that time are supported by low global ice

379   volume deduced by global isotopic composition of benthic foraminifera (perhaps the CN3

380   isotopic event; Lisiecki and Raymo, 2005), and high insolation and eccentricity,

381   coinciding with the early Pliocene climatic optimum and high global sea level (Fig. 8).

382   Warmest conditions at this time are also recorded regionally in other pollen records from

383   the Mediterranean area such as the marine sedimentary sequences of Cap d'Adge 1 and

384   Pichegu (Suc et al., 2018).

385   Overall, warm and humid conditions prevailed after this warmest maximum during

386   pollen zone LM-02 until 4.13 Ma. However, a progressive climate cooling and aridity

387   trend interrupted by cyclic climatic variability also characterized the LM record (Fig. 8).

388   This is deduced by the progressive decrease in abundance of *Quercus*, thermophilous,

389   high AP/arid ratios and PC1 (Fig. 5). Riverine nutrient supply deduced from the LM

390   record covaried with the vegetation also featuring a significant decreasing trend at that

391   time (Fig. 7). This indicates a progressive decrease in riverine runoff with a decaying

392   supply in degraded organic matter from the continent into the marine environment, which

393   agrees with the increasing trend in aridity. This environmental change observed in the

394   LM record could be related with the cooling trend and increase in global ice volume

395   deduced by the increase in the global isotopic $\delta^{18}O$ stack record (Lisiecki and Raymo,

396   2005), perhaps related with a decrease in eccentricity and its modulating effect in summer

397   insolation (Laskar et al., 2004) (Fig. 8). A progressive increase in sand content occurred

398   in the LM record between 4.21 and 4.14 Ma (Fig. 7). This could be due to a sea-level

399   glacio-eustatic lowering due to the progressive cooling, generating enhanced influence of

400   coarse terrestrial sedimentation due to closeness to coast. West-southwestward

401   progradation of terrigenous depositional systems along the axial part of the Guadalquivir

402   Basin and also from North-east (passive border), filling up the basin, would account for

403   the important increase in coarser detritics and sedimentary rates in a context of sea-level

404   lowering in this area, as deduced by previous studies (Sierro et al., 1996; Pérez-Asensio

405   et al., 2018; Jiménez-Moreno et al., 2013b). This gradual filling could have been

406   accelerated in a context of eustatic sea level lowering with the subsequent decrease in

407   accommodation space in the Guadalquivir basin.

408   This cooling trend ended with significant glaciation-like cold and arid events that

409   affected the terrestrial environments in the study area at ~ 4.1 and 4.0 Ma (LM-03 pollen

410   zone) (Fig. 8). Coldest/driest conditions at this time are deduced by lowest *Quercus*,

411   AP/arid and PC1 values (Fig. 5). Similar very low percentages (<5%) in *Quercus* forest

412   were reached during the last two glaciations (ca. 20 and 140 ka) in the southern Iberian

413   Peninsula (Camuera et al., 2019), supporting our climatic interpretations. Related with

414   this major climate cooling and aridity maxima, was enhanced coarse sedimentation and

415   minima in organic matter fluxes to the seafloor due to low riverine discharge, showing

416   paired vegetation and sea-level changes and a strong land-ocean relationship (Fig. 7).

417   Coldest/driest conditions of the LM record could be related to significant glaciation

418   events recorded in the marine isotopic records of benthic $\delta^{18}O$ Gi24-20 (Lisiecki and

419   Raymo, 2005) and minima in eccentricity (Fig. 8). The glacio-eustatic sea-level drop

420   associated with this especially cold glacial period was recorded globally as a third order

421 (Za2) sequence boundary (Fig. 8; Handerbol et al., 1998), affecting our study site with a

422 significant regression, increasing its proximity to the coast and enhancing coarse

423 sedimentation (Fig. 7). The evidences of glacial-like conditions between 4.1-4.0 Ma in

424 the LM area are supported by a recent review about climate variability during the Pliocene

425 by De Schepper et al. (2014), who show evidences of a significant glaciation event in

426 both northern and southern Hemispheres at around 4.0 Ma. This glacial-like climatic

427 event occurred during the early Pliocene climatic optimum, a period known by very little

428 terrestrial evidence of glaciation, and indicates that major glaciation-like cooling occurred

429 before the intensification of northern hemisphere glaciation at the beginning of the

430 Pleistocene (~2.7 Ma; Lisiecki and Raymo, 2005). This cooling trend and associated

431 glacio-eustatic sea-level lowering might have contributed to the final continentalization

432 of the study area that occurred at about 30 m core depth in the LM sediment core (Almonte

433 and Abalario Fm.; Fig. 1). A previously mentioned cause that could have triggered

434 enhanced cooling and glaciation at this time in the Northern Hemisphere is the

435 constriction of the Indonesian Seaway between 4-3 Ma, generating a weakening of the

436 Atlantic Meridional Overturning Circulation, which regulates climate in the area (Karas

437 et al., 2017).

438 An increase in climate seasonality (i.e., summer drought) seems to have affected the

439 terrestrial environments in the study area since 4.12 Ma and related with the above

440 mentioned significant glacial event. This is deduced by the increasing trend in *Olea*, a

441 typical Mediterranean and summer-drought-adapted sclerophyllous taxon, from 4.12 Ma

442 towards the top of the LM record at 3.97 Ma (Fig. 7). Previous studies noticed a major

443 reduction in subtropical taxa and an increase in sclerophyllous vegetation during the late

444 Pliocene at around 3.4-3.2 Ma, which they interpreted as the effect on the vegetation of

445 the onset of the Mediterranean-type seasonal precipitation rhythm in the Mediterranean

446   area (Suc, 1984; Suc and Popescu, 2005). Our pollen data from the LM record indicate

447   that this change towards a more seasonal Mediterranean-like climate could have started

448   earlier, at around 4 Ma ago. However, more data from additional sites in the area are

449   necessary to confirm this, as the *Olea* pollen change is small (from 0 to 4%; Fig. 7).

450   Anyway, the climate change to a colder, drier and more seasonal climate at this time had

451   considerable effects on Pliocene environments and vegetation, reducing tropical species

452   worldwide, and triggering the disappearance of forests and the spread of more open and

453   xeric landscapes (see regional syntheses in Jiménez-Moreno et al., 2010; Biltekin et al.,

454   2015; Fauquette et al., 2018).

455

456

457   *5.3. Climate variability during the early Pliocene*

458

459   Previous paleoclimatic studies concluded that initial assumption of persistent global

460   warmth and stable Pliocene climate conditions were not substantiated (Prescott et al.,

461   2014), and temperatures were as variable as those during the late Pleistocene (Lawrence

462   et al., 2009). In this study, we show that long-term scale trends in climate in the early

463   Pliocene LM record are interrupted by shorter-orbital-scale cyclical climatic changes,

464   deduced by the alternation between thermophilous-temperate forest (i.e., AP) and herbs

465   (i.e., NAP), most-likely representing warm/humid-cold/arid climatic cycles, respectively

466   (see explanation above). Different scale cyclicities can be observed visually in the pollen

467   abundances (Fig. 5) and statistically through spectral analysis of the *Quercus*, AP and

468   PC1 data time-series (Fig. 6). Two main and statistically significant vegetation and

469   climate cyclicities can be noticed in the three studied proxies, with periodicities around

470   51-37 and 22-21 ka, which could be related with obliquity (~41 ka) and precession (~21

ka) orbital cycles, respectively (Laskar et al., 2004). We are unsure about the significance of the later 22-21 ka cycle, since the average sample resolution in the study record is 9.4 ka, perhaps insufficient to statistically support this cycle. However, the occurrence of this cycle is supported by previous studies suggesting that precession induced climatic oscillations in rainfall in this area generated cyclical variations in the input of freshwater and sedimentation into the Guadalquivir Basin and Gulf of Cadiz during the early Pliocene (Ledesma, 2000; Sierro et al., 2000). Two other obtained statistically significant cyclicities are ~80-67 and 30 ka, which are more difficult to interpret but could be related to a mixed signal of different orbital-scale cycles (eccentricity, obliquity and precession; Berger, 1977). Lisiecki and Raymo (2005) also obtained a similar spectral peak at 29 ka on the last 860 ka of the LR04 benthic $\delta^{18}O$ stack, which they interpreted as a nonlinear climate response. We are unassertive about why the 100-ka eccentricity cycle is not showing up in the spectral analyses, as visually it seems to be characterizing the main variability of the environmental and vegetation changes (see *Quercus* data filtered to 100-ka and correlations to short-term eccentricity in Fig. 8). Long-term eccentricity (~400 ka) cycles are not obtained in the spectral analysis either as the record is too short to register several of these cycles but seems to be the one controlling long-term environmental and climatic evolution in this area (long-term minima in *Quercus* or AP/arid ratios in pollen zones LM-01 and LM-03, at 4.5-4.4 and 4.1-4.0 Ma; Fig. 8). The 400-ka cycle has previously been reported to be one of the main controls of cyclical marine sedimentation in the Guadalquivir and Gulf of Cadiz area (Ledesma, 2000; Sierro et al. 2000). The sea-level drop recorded at ~4.0 Ma, coinciding with the 3rd order sequence boundary Za2 (Hardenbol et al., 1998), is simultaneous with an eccentricity minimum related to the long-term eccentricity cycle and thus is interpreted here to have a glacio-eustatic origin. This orbital-scale variability seems to be reciprocated in the proxy for continental organic

496   matter fluxes to the seafloor (*B. spathulata* abundance), showing minima during cold

497   events (Fig. 7) and pointing to simultaneous variations and fast responses in the terrestrial

498   and marine environments due to same orbital-scale climate oscillations. Overall, the

499   presence of orbital changes in the studied LM record support the age model of Pérez-

500   Asensio et al. (2018) even for its uppermost part, were magnetobiostratigraphic tie points

501   could not be retrieved due to a change in paleoenvironmental conditions and a loss in

502   paleomagnetic quality.

503   This study reveals the strong influence of the eccentricity and obliquity orbital-

504   scale climatic changes on the environment in the study area. Long- and short-term

505   eccentricity are one of the most significant orbital-scale cycles controlling climate and

506   environmental changes recorded in late Miocene and Pliocene sedimentary sequences

507   (Braga and Martin, 1996; Shackleton and Crowhurst, 1997; Ledesma, 2000; Vidal et al.,

508   2002; Pérez-Asensio et al., 2012, 2013; Jiménez-Moreno et al., 2013a, b; van den Berg

509   et al., 2015, 2018). In the mid-latitude Mediterranean area, cyclical changes in the

510   vegetation are mostly forced by precession (Kloosterboer-van Hoeve, 2006; Tzedakis,

511   2007). Nevertheless, obliquity seems to have also played an important role in shaping the

512   vegetation during the Pliocene and Pleistocene period (Popescu, 2001; Popescu et al.,

513   2010; Suc et al., 2010; Joannin et al., 2007, 2008; Jiménez-Moreno et al., 2013a; this

514   study). This indicates a high-latitude impact on the climate from the Mediterranean area

515   through variations in seasonal contrast, mainly controlling temperature changes (Tuenter

516   et al., 2003; Suc et al., 2010).

517

518   **Conclusions**

519

520   The detailed multiproxy study on an early Pliocene marine sedimentary record

521   from SW Spain shows:

522   1.   Warmest and wettest conditions during the 4.5-3.9 Ma period occurred in this

523   area at ~4.35 Ma. Highest organic matter fluxes to the seafloor related to

524   riverine discharges are also recorded at this time, supporting high fluvial

525   runoff supplying continental degraded organic matter. This agrees with

526   warmest conditions shown by low global ice volume deduced by global

527   isotopic composition of benthic foraminifera, and high insolation and

528   eccentricity, coinciding with the early Pliocene climatic optimum and high

529   global sea level.

530   2.   Vegetation and riverine nutrient supply proxies show a subsequent cooling

531   and drying trend after the warmest maximum until ~4.13 Ma. This

532   environmental change could be related with the cooling trend and increase in

533   global ice volume, perhaps caused by a decrease in eccentricity and its

534   modulating effect on summer insolation. A progressive increase in sand

535   content in the LM record between 4.21 and 4.14 Ma could also indicate a

536   glacio-eustatic lowering of the sea-level. West-southwestward progradation

537   of terrigenous depositional systems along the axial part of the Guadalquivir

538   Basin and important detrital inputs from the Iberian Massif prograding

539   towards the center, filling up the basin, would account for the important

540   increase in coarser detritics in a context of sea-level lowering in this area.

541   3.   This cooling trend ended with significant glaciation-like cold and arid events

542   affecting the study area between ~4.1 and 4.0 Ma. Related with this major

543   climate cooling and aridity maxima was enhanced coarse sedimentation and

544   minima in riverine nutrient supply to the ocean, showing paired vegetation

545   and sea-level changes and a strong land-ocean relationship. Coldest/driest

546   conditions of the LM record could be related to significant glaciation events

547   (Gi24-20) recorded in the marine isotopic records and minima in eccentricity.

548   The glacio-eustatic sea-level drop associated with this glaciation affected our

549   study site with a significant regression, increasing its proximity to the coast

550   and enhancing coarse sedimentation and was recorded globally as a third order

551   (Za2) sequence boundary.

552   4. Our study shows that terrestrial and marine environments were forced by long-

553   and short-term eccentricity, obliquity and perhaps precession orbital cycles,

554   shaping vegetation, fluvial organic matter fluxes to the seafloor and

555   sedimentary changes in SW Iberia.

556

557

566 **References**

567 Aguirre, J., Braga, J.C., Martín, J.M., Puga-Bernabéu, Á., Pérez-Asensio, J.N., Sánchez-

568   Almazo, I.M., and Génio, L., 2015. An enigmatic kilometer-scale concentration

569   of small mytilids (late Miocene, Guadalquivir Basin, S Spain). Palaeogeography,

570   Palaeoclimatology, Palaeoecology 436, 199–213.

571   Baceta, J.I., Pendón, J.G., 1999. Estratigrafía y arquitectura de facies de la Formación

572   Niebla, Neógeno superior, sector occidental de la Cuenca del Guadalquivir.

573   Revista de la Sociedad Geológica de España 12, 419–438.

574   Barmawidjaja, D.M., Jorissen, F.J., Puskaric, S., van der Zwaan, G.J., 1992. Microhabitat

575   selection by benthic foraminifera in the northern Adriatic Sea. Journal of

576   Foraminiferal Research 22, 297–317.

577   Bartoli, G., Hönisch, B., Zeebe, R.E., 2011. Atmospheric CO_2 decline during the Pliocene

578   intensification of Northern Hemisphere glaciations, Paleoceanography, 26,

579   PA4213.

580   Berger, A.L., 1977. Support for the astronomical theory of climatic change. Nature 269,

581   44–45.

582   Bertini, A., 2001. Pliocene climatic cycles and altitudinal forest development from 2.7

583   Ma in the Northern Apennines (Italy): evidence from the pollen record of the

584   Stirone section (−5.1 to −2.2 Ma). Geobios 34, 253–265.

585   Beug, H.-J., 1961. Leitfaden der Pollenbestimmung für Mitteleuropa und angrenzende

586   Gebiete, Fisch. Stuttg., 61.

587   Bitelkin, D., Popescu, S.-M., Suc, J.-P., Quézel, P., Yavuz, N., Jiménez-Moreno, G.,

588   Safra, A., Namik Cagatay, M., 2015. Anatolia: a plant refuge area during the last

589   23 Million years according to pollen records from Turkey. Review of

590   Palaeobotany and Palynology 215, 1-22.

591   Braga, J.C., Martín, J.M., 1996. Geometries of reef advance in response to relative sea-

592   level changes in a Messinian (uppermost Miocene) fringing reef (Cariatiz reef,

593   Sorbas Basin, SE Spain). Sedimentary Geology 107, 61–81.

594   Braga, J.C., Martín, J.M., Aguirre, J., 2002. Tertiary. Southern Spain. In: Gibbons, W.,

595   Moreno, T. (Eds.), The Geology of Spain. The Geological Society, London, pp.

596   320–327.

597   Braga, J.C., Martín, J.M., Aguirre, J., Baird, C.D., Grunnaleite, I., Jensen, N.B., Puga-

598   Bernabéu, A., Saelen, G., Talbot, M.R., 2010. Middle-Miocene (Serravallian)

599   temperate carbonates in a seaway connecting the Atlantic Ocean and the

600   Mediterranean Sea (North Betic Strait, S Spain). Sedimentary Geology 225, 19–

601   33.

602   Brierley, C., Fedorov, A., 2010. Relative importance of meridional and zonal sea surface

603   temperature gradients for the onset of the ice ages and Pliocene-Pleistocene

604   climate evolution. Paleoceanography 25, PA2214.

605   Camuera, J., Jiménez-Moreno, G., Ramos-Román, M.J., García-Alix, A., Toney, J.L.,

606   Anderson, R.S., Jiménez-Espejo, F.J., Kaufman, D., Bright, J., Webster, C.,

607   Yanes, Y., Carrión, J.S., Ohkouchi, N., Suga, H., Yamame, M., Yokoyama, Y.,

608   Martínez-Ruiz, F., 2018. Orbital-scale environmental and climatic changes

609   recorded in a new ~ 200,000-year-long multiproxy sedimentary record from

610   Padul, southern Iberian Peninsula. Quaternary Science Reviews 198, 91-114.

611   Camuera, J., Jiménez-Moreno, G., Ramos-Román, M.J., García-Alix, A., Toney, J.L.,

612   Anderson, R.S., Jiménez-Espejo, F., Bright, J., Webster, C., Yanes, Y., Carrión,

613   J.S., 2019. Vegetation and climate changes during the last two glacial-interglacial

614   cycles in the western Mediterranean: A new long pollen record from Padul

615   (southern Iberian Peninsula). Quaternary Science Reviews 205, 86-105.

616   Civis, J., Sierro, F.J., González-Delgado, J.A., Flores, J.A., Andrés, I., de Porta, J., Valle,

617   M.F., 1987. El Neógeno marino de la provincia de Huelva: Antecedentes y

618   definición de las unidades litoestratigráficas. In: Civis, J. (Ed.), Paleontología del

619   Neógeno de Huelva. Ediciones Universidad de Salamanca, Salamanca, pp. 9–21.

620 Combourieu-Nebout, N., Turon, J.L., Zahn, R., Capotondi, L., Londeix, L., Pahnke, K.,

621   2002. Enhanced aridity and atmospheric high-pressure stability over the western

622   Mediterranean during the North Atlantic cold events of the past 50 k.y. Geology

623   30 (10), 863-866.

624

625

626 Combourieu-Nebout, N., Foucault, A., Mélières, F., 2004. Vegetation markers of

627   palaeoclimate cyclical changes in the Pliocene of Punta Piccola (Sicily, Italy).

628   Palaeogeography, Palaeoclimatology, Palaeoecology 214, 55–66.

629 De Schepper, S., Gibbard, P.L., Salzmann, U., Ehlers, J., 2014. A global synthesis of the

630   marine and terrestrial evidence for glaciation during the Pliocene Epoch. Earth-

631   Science Reviews 135, 83–102.

632 De Torres, T., 1975. Mapa Geológico Nacional: Instituto Geológico y Minero de España

633   Sheet Number 984, scale 1:50,000, 14 pp.

634 Duchemin, G., Jorissen, F.J., Le Loc'h, F., Andrieux-Loyer, F., Hily, C., Thouzeau, G.,

635   2008. Seasonal variability of living benthic foraminifera from the outer

636   continental shelf of the Bay of Biscay. Journal of Sea Research 59, 297–319.

637 Faegri, K., Iversen, J., 1989. Textbook of Pollen Analysis. New York: Wiley.

638 Fauquette, S., Guiot, J., Suc, J.-P., 1998. A method for climatic reconstruction of the

639   Mediterranean Pliocene using pollen data. Palaeogeography, Palaeoclimatology,

640   Palaeoecology 144, 183–201.

641 Fauquette, S., Suc, J.-P., Médail, F., Muller, S.D., Jiménez-Moreno, G., Bertini, A.,

642   Martinetto, E., Popescu, S.-M., Zheng, Z., de Beaulieu, J.-L., 2018. The Alps: A

643   geological, climatic, and human perspective on vegetation history and modern

644   plant diversity. In: Hoorn, C., Perrigo, A., Antonelli, A., (Eds.), Mountains,

645   Climate, and Biodiversity, First Edition. Wiley-Blackwell, pp. 413-428.

646 Fedorov, A.V., Brierley, C.M., Lawrence, K.T., Liu, Z., Dekens, P.S., Ravelo, A.C.,

647   2013. Patterns and mechanisms of early Pliocene warmth. Nature 496, 43-49.

648 Fernàndez, M., Beràstegui, X., Puig, C., García-Castellanos, D., Jurado, M.J., Torné, M.,

649   Banks, C., 1998. Geophysical and geological constraints on the evolution of the

650   Guadalquivir foreland basin, Spain. In: Mascle, A., Puigdefabregas, C.,

651   Luterbacher, H. & Fernandez, M. (eds.). Cenozoic Foreland Basins of Western

652   Europe. Geological Society of London Special Publications, 134, 29-48.

653

654 Flores, J.A., 1987. El nanoplancton calcáreo en la formación «Arcillas de Gibraleón»:

655   Síntesis bioestratigráfica y paleoecológica. In: Civis, J. (Ed.), Paleontología del

656   Neógeno de Huelva. Ediciones Universidad de Salamanca, Salamanca, Spain, pp.

657   65–68.

658 Garcia-Castellanos, D., 2002. Interplay between lithospheric flexure and river transport

659   in foreland basins. Basin Research 14(2), 89-104.

660 Garcia-Castellanos, D., Fernàndez, M., Torné, M., 2002. Modelling the evolution of the

661   Guadalquivir foreland basin (South Spain). Tectonics 21(3),

662 Garcia-Castellanos, D., Villaseñor, A., 2011. Messinian salinity crisis regulated by

663   competing tectonics and erosion at the Gibraltar Arc. Nature, 480, 359-363.

664 Gauthier, A., Munoz, A., 2009. Seasonal sedimentation in the Pliocene Villaroya Lake

665   (N Spain) inferred from pollen analysis. Sedimentary Geology 222 (1-2), 111-

666   123.

667 González-Delgado, J.A., Civis, J., Dabrio, C.J., Goy, J.L., Ledesma, S., Pais, J., Sierro,

668   F.J., Zazo, C., 2004. Cuenca del Guadalquivir. In: Vera, J.A. (Ed.), Geología de

669   España. SGE-IGME, Madrid, pp. 543–550.

670 Grimm, E.C., 1987. CONISS: a Fortran 77 program for stratigraphically constrained

671   cluster analysis by the method of incremental sum of squares. Computers and

672   Geosciences 13, 13–35.

673 Hammer, O., Harper, D.A.T., Ryan, P., 2001. Past: paleontological statistics software

674   package for education and data analysis. Palaeontologia Electronica 4 (1) (art. 4,

675   9 pp.).

676 Hardenbol, J., Thierry, J., Farley,M.B., Jaquin, T., de Graciansky, P., Vail, P.R., 1998.

677   Mesozoic and Cenozoic sequence chronostratigraphic framework of European

678   basins - chart 2: Cenozoic sequence chronostratigraphy. In: de Graciansky, P.,

679   Hardenbol, J., Jaquin, T., Vail, P.R. (Eds.), Mesozoic and cenozoic sequence

680   stratigraphy of european basins: SEPM Special Publication, 60.

681 Herzschuh, U., 2007. Reliability of pollen ratios for environmental reconstructions on the

682   Tibetan Plateau. Journal of Biogeography 34, 1265–1273.

683 Heusser, L., 1988. Pollen distribution in marine sediments on the continental margin of

684   northern California. Mar. Geol. 80, 131– 147.

685 Hilgen, F.J., Lourens, J.A., Van Dam, L.J., Beu, A.G., Boyes, A.F., Cooper, R.A.,

686   Krijgsman, W., Ogg, J.G., Piller, W.E., Wilson, D.S., 2012. The Neogene Period.

687   In: Gradstein, F.M., Ogg, J.G., Schmitz, M.D., Ogg, G.M., (Eds.), The Geologic

688   Time Scale. Boston Elsevier, pp. 923-978.

689 Jiménez-Moreno, G., Rodríguez-Tovar, F.-J., Pardo-Igúzquiza, E., Fauquette, S., Suc, J.-

690   P., Müller, P., 2005. High-resolution palynological analysis in late Early–Middle

691   Miocene core from the Pannonian Basin, Hungary: climatic changes,

692   astronomical forcing and eustatic fluctuations in the Central Paratethys.

693   Palaeogeography, Palaeoclimatology, Palaeoecology 216 (1–2), 73–97.

694 Jiménez-Moreno, G., Abdul-Aziz, H., Rodríguez-Tovar, F. J., Pardo-Igúzquiza, E., Suc,

695   J.-P., 2007. Palynological evidence for astronomical forcing in Early-Middle

696   Miocene lacustrine deposits from Rubielos de Mora Basin (NE Spain).

697   Palaeogeography, Palaeoclimatology, Palaeoecology 252, 601-616.

698 Jiménez-Moreno, G., Suc, J.-P., Fauquette, S., 2010. Miocene to Pliocene vegetation

699   reconstruction and climate estimates in the Iberian Peninsula from pollen data.

700   Review of Palaeobotany and Palynology 162, 403–415.

701 Jiménez-Moreno, G., Burjachs, F., Expósito, I., Oms, O., Carrancho, A., Villalain, J.J.,

702   van der Made, J., Agustí, J., Campeny, G., Gómez de Soler, B., 2013a. Late

703   Pliocene vegetation and orbital-scale climate changes from the western

704   Mediterranean area. Global and Planetary Change 108, 15-28.

705 Jiménez-Moreno, G., Pérez-Asensio, J.N., Larrasoaña, J.C., Aguirre, J., Civis, J., Rivas-

706   Carballo, M.R., Valle-Hernández, M.F., González-Delgado, J.A., 2013b.

707   Vegetation, sea-level and climate changes during the Messinian salinity crisis.

708   Geological Society of America Bulletin 125, 432-444

709 Joannin, S., Quillévéré, F., Suc, J.-P., Lécuyer, C., Martineau, F., 2007. Early Pleistocene

710   climate changes in the central Mediterranean region as inferred from integrated

711   pollen and planktonic foraminiferal stable isotope analyses. Quaternary Research

712   67, 364–374.

713 Joannin, S., Ciaranfi, N., Stefanelli, S., 2008. Vegetation changes during the late early

714   Pleistocene at Montalbano Jonico (Province of Matera, southern Italy) based on

715   pollen analysis. Palaeogeography, Palaeoclimatology, Palaeoecology 270, 92–

716   101.

717   Karas, C., Nürnberg, D., Bahr, A., Groeneveld, J., Herrle, J.O., Tiedemann, R.,

718   deMenocal, P.B., 2017. Pliocene oceanic seaways and global climate. Scientific

719   Reports 7:39842.

720   Kloosterboer-van Hoeve, M.L., Steenbrink, J., Visscher, H., Brinkhuis, H., 2006.

721   Millennial-scale climatic cycles in the Early Pliocene pollen record of Ptolemais,

722   northern Greece. Palaeogeography, Palaeoclimatology, Palaeoecology 229, 321–

723   334.

724   Larrasoaña, J.C., Liu, Q., Hu, P., Roberts, A.P., Mata, P., Civis, J., Sierro, F.J., Pérez-

725   Asensio, J.N., 2014. Paleomagnetic and paleoenvironmental implications of

726   magnetofossil occurrences in late Miocene marine sediments from the

727   Guadalquivir Basin, SW Spain. Frontiers in Microbiology, 5, Article 71.

728   Laskar, J., Robutel, P., Joutel, F., Gastineau, M., Correia, A.C.M., Levrard, B., 2004. A

729   long-term numerical solution for the insolation quantities of the Earth. Astronomy

730   & Astrophysics 428, 261–285.

731   Lawrence, K. T., Herbert, T.D., Brown, C.M., Raymo, M.E., Haywood, A.M., 2009.

732   High-amplitude variations in North Atlantic sea surface temperature during the

733   early Pliocene warm period, Paleoceanography, 24, PA2218.

734   Ledesma, S., 2000. Astrobiocronología y estratigrafía de alta resolución del Neógeno de

735   la Cuenca del Guadalquivir-Golfo de Cádiz. PhD dissertation, University of

736   Salamanca, Spain.

737   Lisiecki, L. E., Raymo, M.E., 2005. A Pliocene-Pleistocene stack of 57 globally

738   distributed benthic $\delta^{18}O$ records. Paleoceanography 20, PA1003.

739   Martín, J.M., Braga, J.C., Aguirre, J., Puga-Bernabéu, A., 2009. History and evolution of

740   the North-Betic Strait (Prebetic Zone, Betic Cordillera): a narrow, early

741   Tortonian, tidal-dominated, Atlantic–Mediterranean marine passage.
742   Sedimentary Geology 216, 80–90.

743 Martín, J.M., Puga-Bernabéu, A., Aguirre, J., and Braga, J.C., 2014. Miocene Atlantic-
744   Mediterranean seaways in the Betic Cordillera (southern Spain). Revista de la
745   Sociedad Geológica de España 27, 175–186.

746 Mayoral, E., González, I., 1986–1987. Contribución al estudio de las facies de tránsito
747   Mioceno-Plioceno en el sector noroccidental de la Cuenca del Guadalquivir
748   (Valencina de la Concepción, Sevilla). Acta Geológica Hispánica 21–22, 177–
749   184.

750 Mayoral, E., Pendón, J.G. 1986-87. Icnofacies y sedimentación en zona costera. Plioceno
751   superior (?), litoral de Huelva. Acta Geologica Hispanica 21-22: 507-513.

752 Miller, K.G., Kominz, M.A., Browning, J.V., Wright, J.D., Mountain, G.S., Katz, M.E.,
753   Sugarman, P.J., Cramer, B.S., Christie-Blick, N., Pekar, S.F., 2005. The
754   Phanerozoic record of global sea-level change. Science 310, 1293–1298.

755 Muñiz, F., Mayoral, E., 1996. Características paleoambientales de las <<facies de
756   tránsito>> (Plioceno inferior) en el sector N.O. de la Cuenca del Gualdalquivir
757   (Albaida del Aljarafe, Sevilla, España). Geogaceta 20, 207–208.

758 Pendón, J.G., and Rodríguez-Vidal, J., 1986–1987. Caracteres sedimentológicos y
759   geomorfológicos del Alto Nivel Aluvial cuaternario en el litoral de Huelva. Acta
760   Geológica Hispánica 21–22, 107–111.

761 Pérez-Asensio, J.N., Aguirre, J., Schmiedl, G., Civis, J., 2012. Impact of restriction of the
762   Atlantic–Mediterranean gateway on the Mediterranean Outflow Water and
763   eastern Atlantic circulation during the Messinian. Paleoceanography 27, PA3222.
764   http://dx.doi.org/10.1029/2012PA002309.

765 Pérez-Asensio, J.N., Aguirre, J., Jimenez-Moreno, G., Schmiedl, G., and Civis, J., 2013.

766   Glacioeustatic control on the origin and cessation of the Messinian salinity crisis.

767   Global and Planetary Change 111, 1–8.

768 Pérez-Asensio, J.N., Aguirre, J., Schmiedl, G., Civis, J., 2014. Messinian productivity

769   changes in the northeastern Atlantic and their relationship to the closure of the

770   Atlantic–Mediterranean gateway: Implications for Neogene palaeoclimate and

771   palaeoceanography. Journal of the Geological Society [London] 171, 389–400.

772 Pérez-Asensio, J.N., Larrasoaña, J.C., Samankassou, E., Sierro, F.J., Garcia-Castellanos,

773   D., Jiménez-Moreno, G., Salazar, A., Salvany, J.M., Ledesma, S., Mata, M.P.,

774   Civis, J., Mediavilla, C., 2018. Magnetobiochronology of Lower Pliocene marine

775   sediments from the lower Guadalquivir Basin: Insights into the tectonic evolution

776   of the Strait of Gibraltar área. *GSA Bulletin* 130 (11-12), 1791-1808.

777 Popescu, S.-M., 2001. Repetitive changes in Early Pliocene vegetation revealed by high-

778   resolution pollen analysis: revised cyclostratigraphy of southwestern Romania.

779   Review of Palaeobotany and Palynology 120 (3–4), 181–202.

780 Popescu, S.M., Biltekin, D., Winter, H., Suc, J.-P., Melinte-Dobrinescu, M.C., Klotz, S.,

781   Rabineau, M., Combourieu-Nebout, N., Clauzon, G., Deaconu, F., 2010. Pliocene

782   and Lower Pleistocene vegetation and climate changes at the European scale: long

783   pollen records and climatostratigraphy. Quaternary International 219, 152–167.

784 Prescott, C.L., Haywood, A.M., Dolan, A.M., Hunter, S.J., Pope, J.O., Pickering, S.J.,

785   2014. Assessing orbitally-forced interglacial climate variability during the mid-

786   Pliocene Warm Period. Earth and Planetary Science Letters 400, 261-271.

787 Riaza, C., Martínez del Olmo, W., 1996. Depositional model of the Guadalquivir-Gulf of

788   Cádiz Tertiary basin. In: Friend, P., Dabrio, C.J. (Eds.), Tertiary basins of Spain.

789   Cambridge University Press, Cambridge, pp. 330–338.

790   Robinson, M., Dowsett, H.J., Chandler, M.A., 2008. Pliocene role in assessing future

791   climate impacts. Eos Transactions American Geophysical Union 89, 501-502.

792   Rodríguez-Ramírez, A., Villarías-Robles, J.J.R., Pérez-Asensio, J.N., Santos, A.,

793   Morales, J.A., Celestino-Pérez, S., León, Á., Santos-Arévalo, F.J., 2016.

794   Geomorphological record of extreme wave events during Roman times in the

795   Guadalquivir estuary (Gulf of Cadiz, SW Spain): an archaeological and

796   paleogeographical approach. Geomorphology 261, 103–118.

797   Salvany, J.M., Larrasoaña, J.C., Mediavilla, C., and Rebollo, A., 2011. Chronology and

798   tectono-sedimentary evolution of the Upper Pliocene to Quaternary deposits of

799   the lower Guadalquivir Basin, SW Spain. Sedimentary Geology 241, 22–39.

800   Schmiedl, G., Kuhnt, T., Ehrmann, W., Emeis, K.-C., Hamann, Y., Kotthoff, U., Dulski,

801   P., Pross, J., 2010. Climatic forcing of eastern Mediterranean deep-water and

802   benthic ecosystems during the past 22 000 years. Quaternary Science Reviews 29,

803   3006–3020.

804   Schulz, M., Mudelse, A., 2002. REDFIT: estimating red-noise spectra directly from

805   unevenly spaced paleoclimatic time series. Computers & Geosciences 28, 421–

806   426.

807   Shackleton, N.J., Crowhurst, S., 1997. Sediment fluxes based on orbitally tuned time

808   scale 5 Ma to 14 Ma, Site 926. In: Shackleton, N.J., Curry, W.B., Richter, C.,

809   Bralower, T.J., et al., (Eds.). Proceedings of the Ocean Drilling Program,

810   Scientific Results, Volume 154: College Station, Texas, Ocean Drilling Program,

811   pp. 69–82.

812   Sierro, F.J., Gonzalez-Delgado, J.A., Dabrio, C., Flores, J.A., and Civis, J., 1990. The

813   Neogene of the Guadalquivir basin (SW of Spain). In: Agustí, J., Domènech, R.,

814   Julià, R., and Martinell, J., (eds.), Paleontologia i Evolució Memòria Especial 2:

815   Iberian Neogene Basins. Sabadell, Spain, Diputació de Barcelona, pp. 209–250.

816   Sierro, F.J., Flores, J.A., Civis, J., González-Delgado, J.A., Francés, G., 1993. Late

817   Miocene globorotaliid event-stratigraphy and biogeography in the NE-Atlantic

818   and Mediterranean. Marine Micropaleontology 21, 143–168.

819   Sierro, F.J., González Delgado, A., Dabrio, C.J., Flores, A., Civis, J., 1996. Late Neogene

820   depositional sequences in the foreland basin of Guadalquivir (SW Spain). In:

821   Friend, P.F., and Dabrio, C.J., (eds.), Tertiary Basins of Spain. Cambridge, UK,

822   Cambridge University Press, pp. 339–345.

823   Sierro, F.J., Ledesma, S., Flores, J.A., Torrescusa, S., Martínez del Olmo, W., 2000. Sonic

824   and gamma-ray astrochronology: Cycle to cycle calibration of Atlantic climatic

825   records to Mediterranean sapropels and astronomical oscillations. Geology 28,

826   695-698.

827   Stefanelli, S., 2004. Cyclic changes in oxygenation based on foraminiferal microhabitats:

828   Early-Middle Pleistocene, Lucania Basin (southern Italy). Journal of

829   Micropalaeontology 23, 81–95.

830   Suc, J.-P., Cravatte, J., 1982. Etude palynologique du Pliocène de Catalogne (nord-est de

831   l'Espagne). Paléobiologie Continentale 13 (1), 1–31.

832   Suc, J.-P., 1984. Origin and evolution of the Mediterranean vegetation and climate in

833   Europe. Nature 307, 429–432.

834   Suc, J.-P., Popescu, S.-M., 2005. Pollen records and climatic cycles in the Mediterranean

835   region since 2.7 Ma. In: Head, M.J., Gibbard, P.L. (Eds.), Early–Middle

836   Pleistocene Transitions, the Land–Ocean Evidence: Geological Society, London

837   Special Publications, vol. 247, pp. 147–158.

838   Suc, J.-P., Diniz, F., Leroy, S., Poumot, C., Bertini, A., Dupont, L., Clet, M., Bessais, E.,

839   Zheng, Z., Fauquette, S., Ferrier, J., 1995. Zanclean (Brunssumian) to early

840   Piacenzian (early–middle Reuverian) climate from 4 to 54° north latitude (West

841   Africa, West Europe and West Mediterranean areas). Mededelingen - Rijks

842   Geologische Dienst 52, 43–56.

843   Suc, J.-P., Combourieu-Nebout, N., Seret, G., Klotz, S., Popescu, S.-M., Gautier, F.,

844   Clauzon, G., Westgate, J., Sandhu, A.S., 2010. The Crotone series: a synthesis

845   and new data. Quaternary International 219, 121–133.

846   Suc, J.-P., Popescu, S.-M., Fauquette, S., Bessedik, M., Jiménez-Moreno, G., Bachiri

847   Taoufiq, N., Zheng, Z., Médail, F., Klotz, S., 2018. Reconstruction of

848   Mediterranean flora, vegetation and climate for the last 23 million years based on

849   an extensive pollen dataset. Ecologia Mediterranea 44 (2), 53-85.

850   Tuenter, E., Weber, S.L., Hilgen, F.J., Lourens, L.J., 2003. The response of the African

851   summer monsoon to remote and local forcing due to precession and obliquity.

852   Global and Planetary Change 36, 219–235.

853   Tzedakis, P.C., 2007. Seven ambiguities in the Mediterranean palaeoenvironmental

854   narrative. Quaternary Science Reviews 26, 2042–2066.

855   Van den Berg, B.C.J., Sierro, F.J., Hilgen, F.J., Flecker, R., Larrasoaña, J.C., Krijgsman,

856   W., Flores, J.A., Mata, M.P., Martín, E.B., Civis, J., González-Delgado, J.A.,

857   2015. Astronomical tuning for the upper Messinian Spanish Atlantic margin:

858   disentangling basin evolution, climate cyclicity and MOW. Global and Planetary

859   Change 135, 89–103.

860   Van den Berg, B.C.J., Sierro, F.J., Hilgen, F.J., Flecker, R., Larrasoaña, J.C., Krijgsman,

861   W., Flores, J.A., Mata, M.P., 2018. Imprint of Messinian Salinity Crisis events on

862   the Spanish Atlantic margin. Newsletters on Stratigratighraphy 51, 93–115.

863 Vidal, L., Bickert, T., Wefer, G., Röhl, U., 2002. Late Miocene stable isotope stratigraphy

864   of SE Atlantic ODP Site 1085: Relation to Messinian events. Marine Geology

865   180, 71–85.

866

867

868

869

870 **Figure captions**

871

872 Figure 1. (A) Simplified geological map of the study area and location of La Matilla (LM)

873 sedimentary core. The regional cross section in B is also indicated. (B) A NW-SE cross

874 section of the lower Guadalquivir Basin showing the main lithostratigraphic units and the

875 LM core location. Modified from Pérez-Asensio et al. (2018).

876

877 Figure 2. The LM core lithology, sand content, variations in inclination of the

878 characteristic remanent magnetization (ChRM) based on quality types 1 and 2 data and

879 the associated pattern of polarity intervals identified, and location of biostratigraphic

880 events recognized by planktonic foraminifera (modified from Pérez-Asensio et al., 2018).

881 On the bottom right is the age-depth model and sedimentary rates (cm/k.y.) for the LM

882 record (modified after Pérez-Asensio et al., 2018). Gray shadings and dashed lines

883 indicate the interpreted correlation to the GTS2012 time scale (Hilgen et al., 2012). FO—

884 first occurrence (lowest occurrence); LO—last occurrence (highest occurrence); LcO—

885 last common occurrence (highest common occurrence); G.—*Globorotalia*.

886

887   Figure 3. Detailed pollen diagram of the early Pliocene LM core in depth (m) and age

888   (Ma). Only species with percentages higher than 1% are shown. Arboreal pollen (AP)

889   percentages are also plotted. Green and yellow indicate the trees and herbs, respectively.

890   Shading in some pollen species is the exaggeration of their abundance x5. Aquatic

891   Cyperaceae is in blue. On the right is a cluster analysis (Grimm, 1987) of the pollen results

892   and pollen zones, Climatic inferences (with color shading, red indicating warm/humid,

893   blue cold/dry) are shown in the pollen zones.

894

895   Figure 4. Principal Component Analysis (PCA) from the LM pollen data. (A) PCA scatter

896   diagram with the most significant (abundant) pollen taxa. PCA (PC1+ and PC1-) groups

897   are shown. The analysis was carried out using PAST (Hammer et al., 2001). (B) Obtained

898   PC1 plot and AP (%) from LM record. Note the visual covariation with each other. Pollen

899   zones are shown on the right with climatic inferences (with color shading, red indicating

900   warm/humid, blue cold/dry).

901

902   Figure 5. Comparison of the pollen paleoclimatic proxies obtained from the early

903   Pliocene LM record. From bottom to top: percentages of thermophilous taxa, percentages

904   of *Quercus*, AP/arid ratios, and PC1. The pollen zones and climatic inferences (with color

905   shading, red indicating warm/humid, blue cold/dry) are shown. Note the covariation of

906   all proxies shown by dashed lines.

907

908   Figure 6. Spectral analysis of the most significant climatic pollen proxies from the early

909   Pliocene LM record: *Quercus*, AP and PC1. Confidence levels are shown in orange (80%

910   confidence level) and green (90% confidence level). Significant periodicities (above the

911   80 and 90% confidence level) are shown with numbers (all in ka). The spectral analysis

912   was done using the software PAST (Hammer et al., 2001). In pink shading are frequencies

913   of orbital-scale eccentricity, obliquity and precession cycles.

914

915   Figure 7. Comparison of paleoclimatic, riverine nutrient supply and sedimentation

916   (proximity to coast) proxies for the 4.5-3.9 Ma interval from LM record. From bottom to

917   top are abundances (%) of: *Bolivina spathulata*, *Quercus*, *Olea* and sand content. Blue

918   shading indicates cold/dry periods. Note the covariation of the proxies shown by dashed

919   lines. Blue dashed line highlight the significant cooling/drying at ~4.12 Ma (see text for

920   explanation).

921

922   Figure 8. Comparison of paleoclimatic pollen proxies from the LM record with global

923   paleoclimatic records of the early Pliocene 4.5-3.9 Ma interval. From bottom to top: raw

924   *Quercus* abundance (%) and filtered 100-ka component of the *Quercus* abundance from

925   LM, PC1 from LM, LR04 benthic $\delta^{18}O$ record showing major isotopic events (Lisiecki

926   and Raymo, 2005), orbital eccentricity (Laskar et al., 2004), summer insolation at 65ºN

927   (Laskar et al., 2004) and global sea-level cycles of Hardenbol et al. (1998) with sequence

928   boundaries Za1 and Za2. Blue shading indicates cold/dry periods. Dashed lines point to

929   correlations between the LM record and global paleoclimatic and glacio-eustatic records:

930   in blue are events corresponding to long-term eccentricity minima, in black are events

931   corresponding to 100-ka eccentricity cycles and in orange are correlations to obliquity

932   cycles (see text for explanation).

933

934

935

936

941

943

944

945