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1 **TO BE SUBMITTED TO QUATERNARY RESEARCH (2.583)**

2 **Climatic implications of the Quaternary fluvial tufa record in the NE Iberian Peninsula**
3 **over the last 500 ka**

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16 **Abstract**

17 Drainage of the Iberian Ranges (NE Spain) houses one of the most extensive
18 Quaternary fluvial tufaceous record in Europe. Specifically tufa deposits in the
19 Añamaza, Mesa, Piedra and Ebrón river valleys were mapped and stratigraphically
20 described. In addition they were chronologically referenced from a large dataset of
21 U/Th disequilibrium series, Aminoacid Racemization and Radiocarbon dates to infer
22 long-term climatic fluctuations in NE Iberia over the past 500 ka. High consistency
23 among the dating methods, also supported by geomorphic and stratigraphic relations
24 was observed. Tufa deposits accumulated in cascades, barrage-cascades and related
25 damming areas developed in stepped fluvial systems of variable slope, that display
26 sequences of cut-and-fill and nested-fill terraces. Tufa accumulation occurred during
27 discrete multi-millennial time periods along the Middle and Upper Pleistocene and

28 Holocene. Maximum frequency of tufa deposition was identified at 120 ka (MIS-5e),
29 105 ka (MIS-5c), 85 ka (~ MIS-5a) and 7 ka (MIS-1), probably under warmer and wetter
30 conditions than today. Additional phases of tufa deposition appear at ~ 345 ka (~ MIS-
31 10), 284 ka (MIS-9a), 206 ka (MIS-7) and 154 ka (MIS-6). Although most tufa growth
32 episodes are clearly correlated to interstadial periods, it is remarkable the occurrence
33 of tufa activity during the Penultimate Glaciation (MIS-6), indicating that the onset of
34 this stage was climatically favorable in the Iberian Peninsula. Biostatic conditions and
35 dynamics of karstic systems regulating tufa deposition seem to be very sensitive to 1)
36 precipitation regime, controlled by shifts in the position of North Atlantic atmospheric
37 belts and 2) summer insolation, regulated by orbital forcing.

38 **Key words:** fluvial tufas, dating techniques, Quaternary climate, Iberian Ranges.

39 **1 Introduction**

40 Tufas are terrestrial freshwater deposits of calcium carbonate (Viles and Goudie, 1990)
41 in karstic terrains, containing remains of macro- and microphytes, invertebrates and
42 bacteria (Ford and Pedley, 1996). They could be considered as the external
43 sedimentary response to karstic systems dynamics. A close relationship between
44 karstic activity, including tufa formation, and environmental conditions is established
45 in terms of suitable temperature, water availability, vegetation expansion, soil
46 development, carbonate dissolution after infiltration and water discharges saturated in
47 calcite (Henning et al., 1983; Magnin et al., 1991; Baker et al., 1993; Martín-Algarra et
48 al., 2003; Domínguez-Villar et al., 2011; among others). As a consequence, tufa records
49 become a useful tool to decipher long-term/low-frequency Quaternary climatic
50 changes because they can be accurately placed in time by using several dating
51 techniques.

52 Palaeoclimate interpretation of tufa archives (Ford and Pedley, 1996; Pedley et al.
53 1996) seems to indicate that temperature is the limiting factor in temperate areas of
54 high latitudes (Pazdur et al., 1988; Pedley, 1993; Frank et al., 2000), while precipitation
55 could limit tufa formation in low latitude regions (Livnat and Kronfeld, 1985; Kronfeld
56 et al., 1988; Smith et al., 2004; Viles et al., 2007; Cremaschi et al., 2010). In fact,

57 Henning et al. (1983) proposed a different pattern in the frequency of tufa formation
58 under both temperate and Mediterranean environments. This simple pattern becomes
59 more complex in the Iberian Peninsula due to its intermediate latitudinal location as
60 well as because of its highly sensitivity to shifts in the location of the North Atlantic
61 atmospheric belts (Calvo et al., 2001; Cacho et al., 2002; Moreno et al., 2005; Eynaud
62 et al., 2009). In this way, paleoclimate information derived from the fluvial tufa record
63 in the northeastern Iberian Peninsula can significantly contribute to improve the
64 available palaeoenvironmental framework at Mediterranean scale during Quaternary
65 times.

66 Tufas are found in a wide range of karst environments around the world and are very
67 common in Alpine Ranges of the Mediterranean region reaching great relevance in the
68 Iberian Peninsula (Durán, 1989; Pentecost, 1995; Ford and Pedley, 1996; Pedley, 2009).
69 Specifically the Iberian Ranges (NE Spain) house an exceptional tufa record through time
70 and space mainly associated with low and medium order reaches of the fluvial
71 network. Although the palaeoenvironmental studies of Iberian Ranges tufas have
72 recently experienced a significant impulse (e.g. Ortiz et al., 2009; Domínguez-Villar et
73 al., 2011; Vázquez-Urbez et al., 2011a), these advances can be supplemented with new
74 data. This study contributes to improve the palaeoclimatic information in northeastern
75 Iberian Peninsula by using a compilation of chronological data from Quaternary fluvial
76 tufa records in the Añamaza, Mesa, Piedra and Ebrón rivers valleys.

77 Palaeoenvironmental implications derived from tufaceous archives, supported by a
78 new chronological database by using U/Th series dating, Aminoacid Racemization and
79 Radiocarbon, have been inserted in the Middle-Upper Pleistocene and Holocene
80 regional paleoclimatic context. At the moment, only glacial and fluvial archives has
81 been used to infer cold signatures within a biased palaeoenvironmental scenario
82 (Fuller et al., 1998; Lewis et al., 2009; Benito et al., 2010; Calle et al., 2013; García-Ruiz
83 et al., 2013). Besides, the deduced long-term (multi-millennial) climatic changes have
84 been correlated with the Quaternary climate pattern at both Mediterranean and
85 global scale. Preliminary chronological data and paleoclimatic interpretation of local

86 tufa records from the studied sector of the Iberian Ranges has been supplied by Arenas
87 et al. (2010), Sancho et al. (2010) and Lozano et al. (2012).

88 **2 Studied areas in the Iberian Ranges**

89 The Iberian Ranges are a NW-SE trending alpine intraplate mountain chain located in
90 north-eastern Iberian Peninsula (Fig. 1). This mountain range is about 500 km in length
91 reaching the Mediterranean coast. The highest peaks exceed 2300 m in altitude.

92 The landscape is dominated by extensive high altitude platforms and planation
93 surfaces at 1600-1000 m a.s.l., partitioned by tectonic grabens. The most extensive
94 planation surface is named *Superficie de Erosión Fundamental* of the Iberian Ranges
95 (Peña et al., 1984). Its development could be finished in Pliocene times (Gutiérrez and
96 Peña, 1994). It erodes the alpine compressional structures affecting thick and
97 extensive marine carbonate formations, Jurassic and Upper Cretaceous in age. An
98 intensive karstic period leading large fields of dolines and poljes occurred during the
99 final stage of the planation surface development. The resulting erosive surface was
100 partitioned by extensional tectonics during Pliocene and Quaternary times (Gutiérrez
101 and Peña, 1994). In addition, the resulting morphotopographic framework constitutes
102 the starting point for the subsequent fluvial downcutting during Quaternary (Gutiérrez
103 et al., 2008; Scotti et al., 2014).

104 Thus, the formation of tufas in the Iberian Ranges was triggered by the occurrence of
105 high altitude, extensive limestone flattened areas, partitioned by neotectonics and
106 dissected by a fluvial network with non-equilibrium longitudinal stretches, steep
107 gradients and marked knick points. Under this morphostructural framework, tufa
108 accumulation was increased during favourable intervals with high water discharges
109 saturated in calcite from karstic aquifers and warm environmental conditions.

110 In the Iberian Ranges, the most important tufa build-ups are associated with the
111 drainage network flowing radially to the N and NE (into the Ebro depression), to the
112 SW (into the Tajo and Guadiana depressions) and to the SE (into the Mijares, Turia and
113 Júcar depressions). The Quaternary fluvial tufa records studied are located in the
114 valleys of the Añamaza, Mesa and Piedra rivers, tributaries of the Ebro River, and the

115 Ebrón River, a tributary of the Turia River (Fig. 1) distributed from the northwestern
116 sector to the central sector of the Iberian Ranges.

117 The Añamaza valley is located in the northwestern sector of the Iberian Ranges (Fig. 1).
118 The Añamaza River is a 37 km-long tributary of the Alhama River, which in turn flows
119 into the Ebro River. It drains an area of around 210 km² with an altitude ranging
120 between 1040 and 460 m. The mean annual temperature is 11.2 °C and the mean
121 annual precipitation is 520 mm. The geological bedrock (Fig. 2a) is composed of Middle
122 Jurassic marine limestones and marls and Upper Jurassic-Cretaceous continental and
123 transitional rocks (Weald facies), affected by folds and faults with NW-SE trends.
124 Continental Tertiary detrital and limestone deposits overlie the sequence. The River
125 Añamaza is fed mainly from a karstic aquifer hosted in Dogger limestones of 200 m
126 maximum thickness. Springs are mainly found between Añavieja and Débanos. A mean
127 annual discharge around 0.16 m³/s is recorded at Débanos. Current tufa sedimentation
128 has been reported by Auqué et al. (2014).

129 The Mesa and Piedra valleys are located in the northern side of the central sector of
130 the Iberian Ranges (Fig. 1). Both rivers join in the La Tranquera Reservoir and are
131 tributaries of the Jalón River, which in turn flows into the Ebro River. The Mesa River
132 flows along 50 km with a general northeast trend. The surface of the drainage basin is
133 622 km² and the altitude ranges between 1520 and 690 m. Mean annual temperature
134 is 13 °C, with strong seasonal contrasts. Mean annual precipitation is 410 mm,
135 irregularly distributed. The river flows across Triassic sandstones, dolostones and
136 gypsum, Jurassic and Cretaceous limestones and dolostones and Miocene
137 conglomerates, sandstones and mudstones (Fig. 3a). Jurassic and Cretaceous rocks
138 hold the regional aquifer that feeds the drainage network in the area. Mean discharge
139 of the River Mesa reaches 1.58 m³/s including low-thermal groundwater inputs in the
140 lower valley stretch. Present day tufa sedimentation and hydrological conditions in the
141 Mesa River has been analyzed by Auqué et al. (2013).

142 The River Piedra flows along 41 km with a north trend. Its drainage area occupies some
143 1545 km² and its altitude ranges from 1010 to 600 m. The mean annual air
144 temperature recorded in the area is 13.7 °C and the mean annual precipitation reaches

145 370 mm. The bedrock (Fig. 3a) is composed of Jurassic limestones, Lower Cretaceous
146 sands and sandstones, and a thick Upper Cretaceous sequence of limestones and
147 dolostones. All these units are slightly deformed by NW–SE trending folds and faults.
148 Continental Tertiary detrital deposits overlie the sequence. The River Piedra is fed
149 mainly by groundwater from an aquifer hosted in Lower Jurassic and Upper Cretaceous
150 limestones and dolostones some 500 m thick. The mean discharge of the River Piedra
151 is around 1.22 m³/s. Current tufa dynamics in the Piedra has been intensively studied
152 over last 13 years (Vázquez-Urbez et al., 2010; Osácar et al., 2013; Arenas et al.,
153 2014a).

154 The Ebrón valley is located in the southeastern sector of the Iberian Ranges (Fig. 1).
155 The Ebrón River, 30 km long, is a tributary of the Turia River on the Mediterranean side
156 of the Iberian Peninsula. Its drainage area is around 246 km² and the altitude range
157 between 1720 and 720 m. Mean annual temperature in the area is around 10.8 °C,
158 with strong seasonal contrasts, and a mean annual precipitation reaches 450 mm,
159 irregularly distributed. Geological bedrock (Fig. 4a) is made of Triassic dolostones,
160 mudstones and gypsum rocks, Jurassic limestones, Lower Cretaceous detrital rocks and
161 Upper Cretaceous dolostones. These materials are affected by NE-SW and NW-SE
162 trending folds and faults. Tertiary conglomerates, sandstones and mudstones overlie
163 the Mesozoic sequence. Mean discharge of the Ebrón River, around of 1.20 m³/s, is
164 mostly derived from groundwater contributions in the middle reach of the valley, near
165 El Cuervo village.

166 **3 Material and methods**

167 The chronological study of Quaternary tufa records requires a geological mapping
168 approach, as well as a detailed geomorphic and stratigraphic control of the identified
169 tufa build-ups, prior to sample and to perform dating techniques. In a first stage,
170 geological mapping of tufa outcrops along the Añamaza, Mesa, Piedra and Ebrón
171 valleys was undertaken on an aerial photographic base (1:18,000 scale). Later,
172 intensive field work was carried out to check the mapped tufa build-ups, to distinguish
173 morphosedimentary units, to describe selected stratigraphic sections and, finally, to
174 sample for dating. A multi-technique strategy, including U/Th disequilibrium series

175 (U/Th), Aminoacid Racemization (AR) and Radiocarbon (^{14}C) methods was developed
176 according the tufa facies characteristics and the expected age of the deposits. A total
177 of 93 dates are presented. As all of them do not have the same level of reliability and
178 uncertainty two techniques have been applied simultaneously on the same
179 stratigraphic section, when possible. Sometimes replicated ages have been obtained.
180 Although discrete ages of individual tufa samples may be subject to selection biases,
181 the set of samples from a particular build-up allows to constrain the time elapsed for
182 its accumulation. As a consequence, the whole population of data appears to be
183 representative of the major stages of tufa formation in the Iberian Ranges.

184 **3.1 U/Th disequilibrium series**

185 Uranium-Thorium disequilibrium dating was performed using parallel ion-counting
186 multi-collector ICP-MS (Hellstrom, 2003; Cheng et al., 2013). A total of 35 samples
187 from tufa build-ups outcropping in the valleys of the four rivers were developed (Table
188 1). Samples correspond to various facies: stromatolites, boundsones of mosses, calcite
189 coatings on phytoclasts and bioclastic mudstones and wackestones, in all cases
190 avoiding cements. Quality of data is variable, according the initial amount of detrital
191 Th, the post-depositional locking of the geochemical system and the recrystallization
192 processes. Many samples show contamination with external Th. In these cases,
193 corrected ages for detrital Th were calculated according Hellstrom (2006). In open
194 systems the post-deposition uranium mobility usually leads to U loss, which makes
195 samples appear older than they really are (as the Th/U ratio is increased). Taking
196 account the closeness to closed systems, the reliability of results is variable. In some
197 cases, replications indicate high quality data. Despite the inconveniences observed in
198 the application of U/Th on tufa deposits, the obtained ages contribute to establish a
199 reasonably accurate chronological framework.

200 **3.2 Aminoacid Racemization**

201 A total of 48 samples from tufa build-ups located in the four fluvial valleys were
202 analyzed by AR techniques (Table 2). Samples correspond to carbonate silts and sands.
203 Ostracode specimens from various species were recovered, although we selected

204 mostly *Herpetocypris reptans* for the analysis because it is the most common in tufa
205 deposits of the Iberian Peninsula. In some few cases, where *H. reptans* was absent or
206 scarce, *Cyprideis torosa*, *Candona neglecta*, *Candona marchica* and *Ilyocypris gibba*
207 shells were picked. Moreover, the age calculation algorithms were calculated for these
208 species (Ortiz et al., 2004). However, due to certain genera show different amino acid
209 racemization rates (Ortiz et al., 2013), a conversion factor had to be applied to D/L
210 ratios of these three species in order to be directly comparable with *H. reptans*
211 aspartic and glutamic DL values. When possible, we performed 7 analyses of the same
212 sample. Ostracode valves were carefully cleaned sonically in distilled deionized water
213 and rinsed in the same water to remove sediment. In order to remove secondary
214 organic molecules adsorbed to the shells, the valves were then submerged in 3%
215 hydrogen peroxide for 2 hours following Kaufman (2000) and Hearty et al. (2004).

216 Aminoacid concentrations and ratios were quantified using HPLC (high-performance
217 liquid chromatography), following the sample preparation protocol described by
218 Kaufman and Manley (1998) and Kaufman (2000).

219 We measured D/L ratios of aspartic acid and glutamic acid because in most ostracode
220 valves they account for over ca. 50% of the amino acid content (Kaufman, 2000; Bright
221 and Kaufman, 2011). Likewise, they have high racemization rates, thus making them
222 suitable to date relatively young samples. The numerical age of each sample was
223 determined by introducing the aspartic acid and glutamic acid D/L ratios obtained in
224 ostracodes into the age calculation algorithms established by Ortiz et al. (2004). The
225 age of a sample is the average of the numerical dates. The age uncertainty is one
226 standard deviation of all the numerical ages calculated from the amino acid D/L ratios
227 of each sample.

228 The use of the age calculation algorithms obtained in central and southern Spain
229 ostracodes by Ortiz et al. (2004) for the dating of these deposits is justified because a
230 similar thermal history can be inferred for these areas, which are located in the
231 Mediterranean climatic zone of the Iberian Peninsula, with a similar CMAT (current
232 mean annual temperature). Likewise, the age calculation algorithms were established
233 for the ostracode species analyzed here (*C. torosa* and *H. reptans*), which show similar

234 racemization rates (Ortiz et al., 2013) and, therefore, DL ratios are directly comparable
235 without any conversion factor.

236 **3.3 Radiocarbon**

237 Radiocarbon dating was performed on 10 samples taken from different tufa build-ups
238 outcropping in the four valleys (Table 3). Samples correspond to gray peaty intervals
239 and charcoal remnants. Occasionally gastropods and eggshells were also dated. The
240 dating analyses were carried out by the Radiocarbon Laboratory of the Department of
241 Geography at the University of Zurich. The AMS (accelerator mass spectrometer) used
242 was the tandem accelerator of the Institute of Particle Physics at the Swiss Federal
243 Institute of Technology Zurich. Radiocarbon dates were calibrated using the Calib6
244 program and the IntCal09 calibration curve (Stuiver and Reimer, 1993; Reimer et al.,
245 2009).

246 **4 Results**

247 **4.1 General stratigraphic and sedimentological features of fluvial tufas**

248 Fluvial tufa deposits appear as successive build-ups that crop out along the main, and
249 some secondary, valleys as stepped terraces in longitudinal profiles as well as
250 transverse sections. In all cases, tufa morfosedimentary units lie unconformably on the
251 bedrock conforming sequences of cut and fill and nested fill terraces. Commonly, these
252 build-ups are composed of lenticular and wedge-shaped bodies that open
253 downstream, with lenticular (channel-like) transverse sections. The thickness of single
254 build-ups ranges from a few meters up to 90 m, and their extent can reach
255 approximately 1.5 km long and several hundred meters wide.

256 A wide variety of sedimentary facies make these deposits (Figs. 5a, 6a, 7a and 8a):
257 stromatolites (Ls), boundstones of bryophytes (Lbr) and of other macrophyte stems
258 (up-growing plants, Lst 1; down-growing plants, Lst 2), rudstones of phytoclasts (Lph)
259 and of oncoids (Lo), mudstones to packstones of bioclasts and intraclasts (Lb), marls
260 (M) and carbonate, mainly bioclastic, sands and silts (Sb). The latter two facies may
261 contain variable amounts of microscopic and macroscopic organic matter, in some

262 cases making up to peaty intervals. Gravels, conglomerates (G), sands, sandstones (S)
263 and mudstones (F) are minor components; typically, coarse facies appear related to
264 erosional surfaces.

265 Facies Ls, Lbr and Lst 2 constitute highly inclined to vertical strata, which, in most
266 cases, make up hemi-domed bodies up to several metres high. Lenticular bodies of
267 phytoclastic facies (Lph) are commonly interbedded. All these facies formed in typically
268 fast flowing water conditions: cascades, barrage-cascades and moderate-sloped river
269 stretches. In contrast, facies Lb, Sb, Lph, M and minor Ls and Lo mostly compose
270 horizontal and slightly inclined strata. Facies Lb, Sb, Lph, Lo and M formed in slow-
271 flowing and standing-water areas, dammed by barrage-cascades. Oncolites can also be
272 structured as shallow, low-sinuosity bars. Stromatolites developed also in moderate-
273 sloped river stretches between cascades. Boundstones of up-growing plants (Lst 1)
274 formed in moderate- to slow-flowing water conditions both along free-flowing river
275 stretches and standing-water areas (e.g., palustrine conditions) (Arenas-Abad et al.,
276 2010; Vázquez-Urbez et al., 2012). Coarse detrital facies (G, S and Lph) appear related
277 to erosional surfaces at the base of the build-ups, but also within them giving rise to
278 composite build-ups (i.e., made of several lenticular and wedged bodies) (Vázquez-
279 Urbez et al., 2012; Arenas et al., 2014).

280 The geometry of deposits, facies associations and lateral relations among facies within
281 build-ups conform the barrage-cascade model (Pedley, 1990; 2009) or stepped fluvial
282 system (Arenas-Abad et al., 2010), consisting of cascade and barrage-cascade
283 structures laterally related to dammed areas. However, distinct attributes characterize
284 each of the studied fluvial systems, mostly as a function of variations in slope along the
285 valley and as the result of existing diffidence processes. The Añamaza deposits
286 correspond to both moderate and very high-sloped fluvial systems, which determined,
287 respectively, the presence of large pooled areas (e.g., in the upper part of the valley)
288 and of steep river stretches dominated by facies Ls and Lbr (e.g., in the middle part of
289 the valley) (Arenas et al., 2014). In the Mesa valley, the moderate slope conditioned
290 the development of large dammed areas, with wide palustrine fringes, separated by
291 barrage-cascades. In contrast, the outstanding feature of the River Piedra tufas is the

292 abundance of thick barrage-cascade deposits in the middle-low stretch, as a result of
293 increased slope. At some moments, the height of the barrages surpassed the water
294 divide and caused the upstream dammed areas to spill over a secondary course (fluvial
295 diffidence mechanism), which also recorded tufa deposits (Vázquez-Urbez et al., 2011,
296 2012). In the Ebrón valley, with moderate to high slope, thick deposits of facies Sb
297 formed in dammed areas upstream of barrage-cascades. In addition, extensive
298 development of stromatolites occurred in some gently sloped stretches between
299 barrages (Lozano et al., 2012).

300 In brief, the studied tufa deposits formed in stepped fluvial systems of variable slope,
301 as a result of alternating stages of valley filling and erosional processes, which
302 produced successive build-ups along the main and secondary valleys.

303 **4. 2 Tufas of the Añamaza River valley**

304 Tufa deposits crop out along the middle stretch of the Añamaza valley (northeast of
305 Dévanos) (Figs. 2a). Quaternary tufa build-ups are distributed in two main
306 morphosedimentary units forming a staircase terrace system. The thickness is variable
307 reaching 70 m in the Salto del Cajo build-up (Fig. 2b). Most common tufa facies are
308 macrophyte boundstones and rudstones, moss boundstones, stromatolites, carbonate
309 sands and silts (Fig. 2c), and bioclastic mudstones, wakestones, packstones and
310 floatstones.

311 Some selected build-ups were sampled for dating (A-1, A-2, A-3, A-4, A-5, A-6, A-7, A-8,
312 A-9 and A-10) (Fig. 2a). Data from U/Th show variable reliability because some of them
313 represent open geochemical systems. In addition, corrected ages considering the
314 occurrence of detrital Th introduce small changes to the final proposed age (Table 1).
315 Respect to AR age data, only some samples show certain recent contamination (Table
316 2).

317 The Salto del Cajo build-up (Fig. 2b) (A-4 to 8 sections; Fig. 5a and b) is a wedge-shaped
318 body that opens northwards. It is made of five depositional bodies separated by
319 erosional surfaces composing a complex filling of a steep stretch (Arenas et al., 2014).
320 A broad agreement between chronological data (U/Th and AR) and the stratigraphic

321 correlation among stratigraphic sections was observed. Thus, considering the reliable
322 data, the first depositional stages are dated as 282.5 ± 7 ka (U/Th sample SC1-2) and
323 204.3 ± 6.7 ka (U/Th sample SC4-2r), and the rest of the depositional stages are dated
324 between 120.8 ± 3.8 ka (U/Th sample SC2-11) and 80.5 ± 16.6 ka (AR sample SC2-16).
325 In addition, a cascade tufa unit that overlies previous Pleistocene deposits yielded an
326 age of 1.5 ± 0.2 ka (U/Th sample SC5-1r).

327 In the Las Parideras sector (A-3 section) a wedge-shaped tufa build-up, approximately
328 17 m thick, has been dated. The approximate age of this outcrop could be between
329 83.5 ± 18.9 ka (AR sample AÑA13-1) and 105.8 ± 3.9 ka (U/Th sample AÑA 13-2). Two
330 other build-ups (A-10 and A-9) (Fig. 5a) located in the northern part of the studied
331 Añamaza valley stretch provide reliable ages of 115.4 ± 33.0 ka (AR sample AÑA 7N-8)
332 to 99.6 ± 1.8 ka (U/Th sample AÑA 7N-15) for A-10 section, and 129.1 ± 16.8 ka (AR
333 sample AÑA 8-4), 121.7 ± 27.0 ka (AR sample AÑA 8-3) and 119.1 ± 2.5 ka (U/Th
334 sample AÑA 8-5) for A-9 section (Fig. 2c). A sample (AÑA 8-1) of this build-up gave a
335 U/Th age with a great uncertainty of 337.0 ± 110.1 ka.

336 Other tufa build-ups encased in Pleistocene tufas are those of El Molino (A-2 section)
337 (Fig. 5a) and Dévanos (A-1 section). Several samples yielded AR ages of 10.6 ± 3.3 and
338 10.5 ± 4.4 ka in Dévanos, and between 7.7 ± 3.8 ka and 3.3 ± 0.9 in El Molino (Table 2).
339 Besides, an age of 1816-1509 ka (sample AÑA-M-H) has been obtained by calibrated
340 radiocarbon in El Molino (Table 3). Correlative fluviolacustrine sediments studied by
341 Luzón et al. (2011) from drilling cores in the upper stretch of the valley span from 10.6
342 ka to 0.6 ka.

343 In summary according to the obtained ages several episodes of tufa deposition
344 correlated to marine isotope stages MIS 9a, 7, 5 and 1 respectively can be inferred. The
345 presence of MIS 10 is uncertain. Most tufas in the River Añamaza valley formed during
346 MIS 5 (130-80 ka) (Figs. 5a and 9a).

347 **4.3 Tufas of the Mesa River valley**

348 Tufa build-ups crop out scattered along 30 km in the River Mesa valley from Mochales
349 to Ibdes villages (Fig. 3a). The thickness of single build-ups ranges from a few metres to

350 25 m. The build-ups can be grouped in two generations developing staircase terraces.
351 Several build-ups belonging to the older stage have been chronologically characterized
352 (Valdetechada, M-1 section; Jesus Nazareno, M-3 section; Los Villarejos, M-4 and M-5
353 sections; and La Rinconada, M-6 section). In addition, two other younger build-ups
354 have also been dated near Villel de Mesa village (M-2 section). Most common tufa
355 facies are macrophyte boundstones and rudstones, moss boundstones, stromatolites,
356 carbonate sands and silts, and bioclastic mudstones, wakestones, packstones and
357 floatstones.

358 The Valdetechada build-up (M-1 section) (Fig. 6a), located upstream of Mochales, is
359 15-20 m thick. Three samples were dated by AR (Table 2), yielding ages 110.4 ± 22.4 ka
360 (sample BVT-A), 215.5 ± 31.6 ka (sample BVT-C) and 194.5 ± 24.1 ka (sample BVT-D).
361 The former sample (BVT-A) is contaminated with recent aminonacids and, therefore,
362 the age should be older. In the Los Villarejos Norte outcrop (M-4 section) (Figs. 3b and
363 6a), the AR ages obtained for two laterally related samples (LVN-A and LVN-B), located
364 at the top of the build-up, are 74.1 ± 12.6 ka and 80.9 ± 11.2 ka respectively. These
365 dates are consistent with two ages obtained by U/Th in the Casas Villarejos profile (M-
366 5 section) (Fig. 6a) from samples located in very close stratigraphic position. This
367 method supplied two accurate and precise ages, 80.9 ± 1.5 ka and 81.1 ± 2.0 ka (Table
368 1). In addition, another sample (JNE1.1) from the Jesús Nazareno stratigraphic section
369 (M-3 section) (Fig. 6a and b) gave an age of 98.7 ± 0.6 ka, close to the above datings.
370 Finally, the lower interval of the La Rinconada stratigraphic section (M-6 section) (Fig.
371 6a), near Ibdes village, was dated by AR (Table 2). The obtained ages are 129.1 ± 20.2
372 ka and 102.8 ± 21.7 ka (samples LR-A and LR-B). Nevertheless, these ages should be
373 older because samples are contaminated with recent aminoacids.

374 The younger stage of tufa development has been studied in two outcrops located
375 upstream and downstream of Villel de Mesa village (Fig. 3) and dated by AR. Upstream
376 of Villel de Mesa the dated outcrop has an age of 8.4 ± 2 ka (sample RM1-A, Table 2).
377 In the outcrop downstream of Villel de Mesa (M-2 section) (Fig. 6a), the obtained ages
378 are 46.8 ± 8.2 ka (sample CMI-A), 6.7 ± 2.3 ka (sample CMI-B) and 13.7 ± 5.9 ka (sample
379 CMI-C) (Table 2). Another sample (CMI-D), stratigraphically equivalent to CMI-A,

380 provided a calibrated ^{14}C age of 6400-6278 years BP (Table 3). Thus, the AR results
381 indicate that this technique does not offer precise ages for Holocene times.

382 Briefly, considering the proposed ages for the studied tufa build-ups in the River Mesa
383 valley, several stages of tufa development could be differentiated (Figs. 6a and 9b).
384 The oldest phase (215-194 ka) correlates to MIS-7. The more important stage (130-75
385 ka) coincides with the whole MIS-5. It may be that the end of the MIS-6 is also
386 recorded in this tufa deposition period. The occurrence of MIS 3 has been, locally,
387 evidenced. Finally, the youngest phase is Lower Holocene in age (6-7 ka) (MIS-1).

388 **4.4 Tufas of the Piedra River valley**

389 Fluvial tufa deposits are very extensive and thick in the lower stretch of the Piedra
390 River valley. They are basically located in two sectors: near the Monasterio de Piedra
391 Natural Park and around Nuévalos village (Fig. 3a). From a morphosedimentary point
392 of view several episodes of tufa formation arranged in cut-and-fill and nested-fill
393 terrace patterns were distinguished.

394 Several tufa build-ups were studied and sampled. According to geomorphic and
395 stratigraphic criteria they were grouped in an older period (La Requiada, P-3, P-4, P-5
396 sections; Barranco de Los Arcos, P-6 section; Los Bancales, P-2 section; Ermita de la
397 Blanca, P-7 section; Monasterio, P-9 section; and Nuévalos Viejo, P-10 section) and a
398 younger period (Arco de la Yedra, P-1 section; Cola de Caballo and Piscifactoría, P-8
399 sections). A great variety of facies is present.

400 The obtained U/Th ages are of variable quality. Some samples suffer badly from initial
401 thorium contamination as well as from open system behavior (Table 1). Nevertheless,
402 as a rule, the proposed ages, once corrected for initial thorium contamination, are
403 reliable. A few cases from the AR analysis show signs of recent aminoacid
404 contamination (Table 2).

405 Several tufa build-ups were studied near the Monasterio de Piedra, related to the River
406 Piedra but also to the tributary network. The U/Th age obtained for the Monasterio
407 build-up (P-9 section) (Fig. 7a) is 340.7 ± 15.6 ka (sample MON Top). The La Requiada

408 tufa build-up (P-3, P-4 and P-5 sections) (Figs. 3c and 7a) is some 75 m thick and in
409 which two main overlapped stages of tufa aggradation, separated by an important
410 erosional surface, were identified. Deposits corresponding to the older stage were
411 dated by AR (sample RS-1-C) and an age of 237.9 ± 27.4 ka was obtained. Tufas of the
412 younger stage were also dated and a U/Th age of 174.1 ± 22.6 ka was yielded by
413 sample RS-2.2b. The AR ages in this stage are 143.2 ± 28.8 ka and 194.5 ± 27.8 ka
414 (samples RS 1-B and RS 1-A), even both samples two show evidence of recent
415 aminoacid contamination (Table 2). The Barranco de los Arcos deposits (P-6 section)
416 crop out as several isolated build-ups (Fig. 7a) that reach 20 m in thickness. Two
417 replicated U/Th analysis of a sample (BLA-1.1 and BLA-1.2) have provided ages of 254.7
418 ± 48.5 ka and 229.3 ± 35.0 ka respectively. These deposits where, thus, correlated with
419 the lower section of the La Requijada build-up. This result, along with mapping data,
420 allowed reporting a fluvial difffluence episode (Vázquez-Urbez et al., 2011). On the
421 other hand, the Los Bancales build-up (P-2 section) is an 18-20 m thick deposit that
422 extends between the La Requijada and Barranco de los Arcos. The three ages obtained
423 by AR are 233 ± 49 ka (sample MP-A-02), 192.0 ± 26 ka (sample MP-A-01) ka and $148 \pm$
424 17 (sample MP-A-05) respectively. Taking into account these ages, the mapping data
425 and the spatial relationships between tufa deposits, it was possible to assert that the
426 Los Bancales tufa deposit and the upper section of the La Requijada build-up were
427 genetically related. The Los Bancales deposit represents other fluvial difffluence
428 episode in this area (Vázquez-Urbez et al., 2011). Finally, downstream of La Requijada,
429 the Ermita de la Blanca build-up (P-7 section) (Fig. 7a) is some 70-80 m thick. The top
430 of this deposit (sample ER-15) yielded a U/Th age of 89.7 ± 0.9 ka.

431 Other important tufa accumulations have been identified 2-3 km downstream the
432 Monasterio de Piedra, around Nuévalos village (Fig. 3a). In fact the old village is settled
433 on a ~25 m thick tufa build-up (P-10 section). The lower part of this builp-up (Nuévalos
434 Viejo section; Fig. 7a and c) is 254.0 ± 28.8 ka (sample NV-B) and the top could be older
435 than 65.2 ± 24.9 ka (sample NV-A) according AR analysis. A laterally related sample
436 (NG-A) provided a probable AR age older than 130.9 ± 29.0 ka.

437 In addition to the Pleistocene tufa build-ups several Holocene build-ups, confirmed by
438 ^{14}C ages, have been recognized along the Piedra River valley (Table 3). In the Arco de la
439 Yedra outcrop (P-1 section) (Fig. 7a and b), the sample MP-3, from a peaty level,
440 provided a calibrated age of 2878-2735 years BP. Gastropod shells embedded in moss
441 boundstones (sample MP-2) from the Cola de Caballo (Fig. 7a) deposit provided an age
442 of 933-761 years BP. Downstream in the infilled valley of the Monasterio de Piedra
443 tufa (Piscifactoría; Fig. 7a) deposits have an age of 772-653 years BP (charcoal sample
444 MP-1).

445 In brief, according to the distribution of the reliable obtained ages, tufa deposits in the
446 River Piedra corresponds to several periods correlated to MIS-9 (340 ka), MIS-7-6,
447 which shows two important pulses (255-230 ka and 145-195 ka) separated by an
448 important erosional event, MIS-5 (80 ka) and, finally, MIS-1 (2.7-0.8 ka BP) (Figs. 7a
449 and 9c).

450 **4.5 Tufas of the river Ebrón valley**

451 In the valley of the River Ebrón, around Castielfabib village, mapping and field work
452 allowed to differentiate tufa deposits corresponding to two encased terraces (Fig. 4a
453 and b). There is a noticeable rupture in the longitudinal profile of the River Ebrón near
454 Castielfabib. Several tufa build-ups were sampled (Fig. 8): Mirador (E-4 section) and
455 Cascada (E-2 section) stratigraphic sections in the upper-older unit, and Convento (E-1
456 section) and Central (E-3 section) stratigraphic sections, in the lower-younger unit. In
457 the older unit, most common tufa facies are moss boundstones and stromatolites,
458 phytoclastic rudstones and lime mud and carbonate sands. In the younger unit the
459 dominant facies are carbonate sands and silts, but there is also stromatolites, moss
460 and macrophyte boundstones and phytoclastic rudstones.

461 The Mirador stratigraphic section (M-4 section) is around 77 m thick (Fig. 8a and b).
462 Seven samples were dated by AR (Table 2). The age (152.9 ± 21.3 ka) of the sample
463 MIR-5 has been corrected because the racemization rate of *Candona* genus is slower
464 than *Herpetocypris* genus. Sample MIR-8 shows significant amounts of L-sirina
465 indicating a possible present-day aminoacids contamination. As a consequence the real

466 age would be older than the measured age. Samples MIR-2A and MIR-7 show a similar
467 age (~ 490 ka), older than the whole set of samples. These samples contain *Cyprideis*
468 *torosa* which indicate waters with high contents in sulphate and chlorine with a wide
469 range of water salinity. That means that sampled tufa deposits are likely inherited from
470 older tufa deposits located upstream and fed by water running across the Upper
471 Triassic evaporites. In addition, two samples correlated with the upper part of the
472 stratigraphic section were dated by U/Th (Table 1). Replicated analyses indicate
473 consistent and reliable ages ranging between 98.3 ka and 100.5 ka (samples MIR B5
474 and MIR B6). Summarizing, the more reasonable ages point to 186 ka at the bottom,
475 152 ka in the intermediate stretch and 100 ka in the top of the stratigraphic section.

476 The Cascada stratigraphic section (E-2 section) (Figs. 4c and 8a) is 20 m in thickness.
477 Four samples from the stratigraphic section (LC-4.4 y LC-B) and from a laterally
478 equivalent deposit (LC-A y LC-B) were dated by AR (Table 2). Samples from the section
479 were corrected in the same way as the sample MIR-5 above. Therefore, the age of this
480 tufa build-up ranges between 150 ka and 98 ka. These ages are supported by U/Th
481 data from a replicate sample (LC-3 A and B) supplying an age of 117.4 ka (Table 1).

482 The thickness of younger tufa unit exceeds 50 m. The Convento stratigraphic section
483 (E-1 section) (Fig. 8a) (25 m thick) corresponds to the upper section of this unit. Dates
484 obtained by AR in two samples (CON-2 and CON-8) are 10.3 ± 3.1 ka and 16.8 ± 1.8 ka
485 respectively (Table 2). These ages appear reversed respect to the stratigraphic location
486 of samples, however, calibrated ^{14}C ages of three successive samples (CON-1, CON-7
487 and CON-9) are 6129-5982, 2489 ± 2335 and 2065-1896 years BP respectively (Table
488 3), which are consistent with their stratigraphic position.

489 Stratigraphic section Central (E-3 section) (Fig. 8a), located around 1 km downstream
490 of El Convento section, is 25 m thick. In this section AR yielded two ages 19.1 ± 2.5 ka
491 (sample CN-1) and 14.6 ± 1.3 ka (sample CN-5) (Table 2). Nevertheless, younger
492 reliable calibrated ages of 3475-3359 year BP (sample CN-2) and 2961-2787 year BP
493 (CN-4) have been obtained by ^{14}C (Table 3). Comparison of ages by AR and ^{14}C
494 indicates that AR technique does not supply reliable and accurate ages for Holocene
495 times.

496 In summary, distribution of the reliable ages obtained for the River Ebrón valley
497 evidenced the occurrence of tufa deposition during MIS 6 and 5 (180-100 ka) and MIS
498 1 (6-2 ka) (Figs. 8a and 9d). Evidence of older tufaceous deposits during MIS 13 (490
499 ka) should be also taken into consideration.

500 **5. Discussion**

501 Quaternary tufa deposits are widespread along the fluvial network draining the Iberian
502 Ranges (NE Spain). Tufas have been studied in four selected valleys (Añamaza, Mesa,
503 Piedra and Ebrón rivers) arranged in a latitudinal and longitudinal slight gradient. As a
504 rule, tufa build-ups crop out as cut-and-fill and nested terraces. Different fluvial
505 patterns (ramp, ramp wedges, barrage-cascade and associated dammed areas) related
506 to stepped fluvial systems (Arenas-Abad et al., 2010) are recognized. Several
507 techniques (U/Th series dating, Aminoacid Racemization and Radiocarbon) were
508 applied on tufa records to produce a reliable chronological approach well framed in
509 both regional and global paleoclimatological contexts. A general agreement among the
510 different techniques is observed, although U/Th dating provides ages of variable
511 reliability according the features of the geochemistry of tufa systems, and Aminoacid
512 Racemization does not yield accurate ages for Holocene tufa deposits. Anyhow, the
513 ages obtained through the several techniques can be considered as consistent with the
514 geomorphic position of build-ups and the location of samples in stratigraphic sections.

515 ***5. 1 Chronological framework of tufa record and paleoclimatic implications***

516 Ages for the Quaternary Iberian Ranges tufa record derived from a multi-technique
517 dating strategy provide a base to decipher favorable conditions for tufa deposition
518 and, therefore, to infer Quaternary palaeoclimatic implications in western
519 Mediterranean areas.

520 The cumulative probability density function (CPDF), obtained from summed individual
521 probability distributions of the tufa deposits reliable datings (replicates were not
522 considered), reveals several multi-millennial periods of tufa deposition in the studied
523 sector of the Iberian Ranges (Fig. 9e). Firstly, some tufa ages to be confirmed evidence
524 an old period of tufa deposition during MIS-13 (around 490 ka) in the Ebrón River

525 valley. On the other hand, the frequency distribution plot indicates that noticeable
526 periods of tufa accumulation occurred during MIS-10 (345 ka) (with high uncertainty),
527 MIS-9a (284 ka), MIS-7 (206 ka) and MIS-6 (154 ka). Nevertheless, the most active
528 periods of tufa deposition coincide with MIS-5 (85 ka, 105 ka and 120 ka) and MIS-1 (7
529 ka) respectively (Fig. 9g and h). MIS-3 is poorly represented and the occurrence of MIS-
530 3 tufas in this area remains uncertain. Correspondence between increase in tufa
531 development and interstadials MIS is widely recognized under temperate climates
532 (Henning et al., 1983) but also in the Iberian Peninsula (Martínez-Tudela et al., 1986;
533 Martín-Algarra et al., 2003; Ordoñez et al., 2005; Ortiz et al., 2009; Domínguez-Villar et
534 al., 2011). Some authors accentuate the relevance of humidity during interglacial
535 periods in increasing tufa deposition rates in the Mediterranean region (Martínez-
536 Tudela et al., 1986; Pedley, 2009). First data from speleothem growing stages in NE
537 Iberian Peninsula indicate they are clearly connected to warm climates, with positive
538 hydrological balance and high insolation, during interglacial periods (preferably MIS7, 5
539 and 1) (Moreno et al., 2013). On the other hand, speleothem growth rates distribution
540 in the NW Iberian Peninsula shows increasing intervals at 200 ka (MIS7), 125 ka, 105 ka
541 and 85 ka (MIS5 e, c and a respectively) and 9-6 ka (MIS1) approximately. In the Iberian
542 Ranges the agreement between these speleothemic stages and the tufa deposition
543 intervals is highly remarkable. However, more surprising is the extensive occurrence of
544 tufa deposition during stadial stages. In fact, tufa formation particularly during MIS-6
545 in the Iberian Ranges is not uncommon.

546 At regional scale, preferential accumulation of Quaternary tufa deposits during
547 interglacial periods (Fig. 9g and h) in the Iberian Ranges broadly alternates with the
548 occurrence of glacial and non-carbonate bearing fluvial records in NE Spain (Pyrennes,
549 Ebro Basin and Iberian Ranges) during cold phases (Fig. 9; vertical grey shaded areas)
550 (Fuller et al., 1998; Sancho et al., 2008; Lewis et al., 2009; Benito et al., 2010; García-
551 Ruiz et al., 2013). Lewis et al. (2009) and Benito et al. (2010) recognize extensive fluvial
552 terraces correlated to periods of glacial stability in glaciated headwaters Pyrenean
553 valleys (basins of the Aragón, Gállego and Cinca rivers) at 263 ka, 178-140 ka, 110-97
554 ka, 68-61 ka, 45-47 ka, 17 ka and 11 ka. It has been established that the glacial system
555 activity and the related fluvial dynamics seem sensitive to insolation controlled by

556 orbital forcing (Lewis et al, 2009). In addition, Fuller et al. (1998) indicate that river
557 aggradation episodes in the Guadalupe River valley (Iberian Ranges) occurred at 183-
558 130 ka, 122 ka, 111 ka, 88 ka, 49 ka, 39-36 ka, 24-22 ka, 19-16 ka, 14-12 ka and short
559 Holocene stages. These alluviation phases were controlled by winter storm frequency
560 under stadial periods with prevailing periglacial conditions.

561 However this broad zipper-like environmental evolutionary model, composed of
562 alternative cold and warm long-term periods shows some inconsistencies. It is truly
563 noticeable the accumulation of tufa records during the Penultimate Glaciation (MIS-6
564 stage). In NE Iberian Peninsula MIS-6 period is characterized by high water availability
565 in fluvial systems that allows the development of extensive terraces controlled by
566 glacial outwash discharges and/or effective runoff from slopes (Benito et al., 2010)
567 derived from increased seasonal storm frequency (Fuller et al., 1998). At regional scale,
568 the penultimate glaciations (MIS-6) is characterized by an early phase, made of a
569 sequence of warming-cooling events, and a late part with prolonged glacial conditions
570 in western Mediterranean (Martrat et al., 2004) but also in the Portuguese margin
571 (Margari et al., 2014). Stalagmite records from the northern Iberian Peninsula indicate
572 periods of speleothem growth at 149-151 ka (Muñoz-García et al., 2007) and around
573 175 ka (Stoll et al., 2013), suggesting that growth persisted through the Penultimate
574 Glaciation. In the same way, Wainer et al. (2013) suggest that MIS-6e was relatively
575 warm and humid in south-western France from speleothem record. Wilson et al.
576 (2013) reported increased moisture availability during MIS-6.5 in Greece from
577 lacustrine records. In brief, it seems probed that at lower latitudes favorable
578 conditions (warmer and wetter) during the early MIS6 were enhanced because of the
579 intensification of Asian and African monsoons (Wainer et al., 2013) and related
580 northward deflected North Atlantic polar front (Calvo et al., 2001), in a context of
581 maxima summer insolation (Stoll et al., 2013) and orbital variability (Margari et al.,
582 2014). As a consequence, climate in the Iberian Ranges during the onset of this period
583 would not be as unfavourable as to preclude tufa formation, and the prevailing
584 environmental conditions allowed the formation of significant tufa deposits.

585 Therefore, it seems that tufa record in the Iberian Peninsula is sensitive to latitudinal
586 shifts of atmospheric fronts and jet streams, as well as perturbations in North Atlantic,
587 due to its location between high and low latitude regions. However, tufa deposition is
588 not sensitive enough to the slight latitudinal and longitudinal gradient at local scale.
589 Differences through time between different fluvial tufa systems located from NW to SE
590 in the Iberian Ranges are not been evidenced. Thus tufa systems response does not
591 record significant changes in environmental conditions (source area and amount of
592 rainfall and/or the vegetation cover) from more continental and Atlantic conditions in
593 the Añamaza River valley to more Mediterranean conditions in the Ebrón River valley.

594 Insolation is another environmental factor to be considered for driving tufa
595 accumulation. In fact, the cumulative probability density function evidences a close
596 relationship between summer insolation peaks and tufa growth episodes in the Iberian
597 Ranges (Fig. 9f). Under humid climates, summer insolation would favor the increase in
598 temperature, but also in CO₂ production by vegetation and, as a consequence, the CO₂
599 input in the karstic system dynamics. In endokarstic environments, Baldini et al.
600 (2005) show a clear relationship between speleothem growth rate and the degree of
601 forest and shrub vegetation coverage. In fact, Stoll et al. (2013) indicate that hiatuses
602 and reductions in stalagmite growth rate occur during extreme minima in summer
603 insolation. The influence of insolation on tufa formation has been also evidenced
604 during the Last Interglaciation in northern hemisphere (Southern Germany) by Frank et
605 al. (2000). The key role of insolation has been also evidenced from seasonal variations
606 of tufa thickness in modern fluvial systems (e.g. Piedra River) (Arenas et al., 2014).

607 **5. 2 Palaeoenvironmental setting**

608 Climate during phases of tufa accumulation in the Iberian Ranges was characterized by
609 prevailing humid conditions producing high groundwater discharges. In addition to
610 water availability, warm temperatures and insolation, seem to reinforce the rates of
611 tufa deposition. The resulting palaeoenvironmental context included the increase in
612 forest vegetation cover, soil CO₂ production, exo- and endokarstic activity and the
613 discharge of chemically active groundwater. This general biostatic context involves the
614 landscape stability, biologically induced (i.e. Goudie et al., 1993; García del Cura et al.,

615 1997). Favourable conditions are intensified particularly during MIS 5, leading to the
616 accumulation of the most important tufa deposits at regional scale, which suggest the
617 wetter and warmer conditions over the last 500 ka in NE Iberia. Similar
618 palaeoenvironmental conditions have been also interpreted at local, regional and
619 continental scales (Henning et al., 1983; Frank et al., 2000; Ordoñez et al., 2005;
620 Dominguez-Villar et al., 2011).

621 An increase of precipitation over the Mediterranean region under interstadial
622 conditions has been deduced from the Alborán sea record (Moreno et al., 2002) and
623 from growth rates of speleothem in northern Iberian Peninsula (Stoll et al., 2013). In
624 addition, a relationship between warm periods and increased global atmospheric CO₂
625 concentration has been also observed from ice cores in Antarctica (Indermühle et al.,
626 2000). The increase in atmospheric CO₂ favoured the increase in forest biomass
627 (Higgins and Scheiter, 2012). Thus, higher temperature, precipitation and atmospheric
628 CO₂ concentration favour the soil-vegetation productivity. Therefore, warm and humid
629 conditions could favour a well developed vegetation cover and a high production of
630 soil CO₂. Under these conditions, solution processes were accelerated and, thus, ionic
631 concentrations in groundwater discharges were increased.

632 In this environmental context, tufa deposition is triggered in the fluvial network when
633 physical CO₂ outgassing is enhanced by the morphological discontinuities in the river
634 longitudinal profiles. High gradient and stepped fluvial systems (*sensu* Arenas et al.,
635 2010) showing different sedimentary architectures (barrage and dammed water areas,
636 prograding-aggrading wedges and high gradient ramps with jumps) resulted in tufa
637 build-ups, according to the local hydrological, geological and morphotopographic
638 context.

639 On the contrary, during phases of cold conditions (e.g., including wet MIS-4 and arid
640 MIS-2 among others periods; Lewis et al., 2009), deposition of tufa ceased and fluvial
641 incision destroyed partly the previous tufa deposits and favoured the development of
642 a stepped-terraces landscape. Erosion processes in fluvial tufa systems are strongly
643 related to degradation of the vegetation cover on the slopes (Ordoñez et al., 2005) and
644 to increase in detrital supply from mechanical weathering. In addition, the resulting

645 tufa build-ups sometimes include erosional episodes, when the previous tufa build-ups
646 were partially destroyed. These episodes are probably related to short stages of
647 environmental deterioration, including an increase of extreme hydrological events.
648 These erosive events, related to extreme water discharges, are common in present-day
649 fluvial tufa systems, controlling the development of tufa deposits (Vázquez-Urbez et
650 al., 2010).

651 **6 Conclusions**

652 Chronological advances on tufa record in a wide sector of the Iberian Ranges, based on
653 a multi-technique strategy, allow us to advance in the proposal of a regional
654 palaeoclimatic framework, composed of sequential warm and wet long-term stages
655 during the last 500 ka. The following conclusions can be highlighted:

656 - Chronological data were obtained from Aminoacid Racemization, U/Th disequilibrium
657 series and Radiocarbon. A high consistency was observed among the dating methods,
658 supported by geomorphic and stratigraphic field relations, even though Racemization
659 appears inaccurate for Holocene times.

660 - The regional tufaceous sequence includes periods of active tufa deposition at 345 ka
661 (MIS-10), 284 ka (MIS-9a), 206 ka (MIS-7), 154 ka (MIS-6), 85 ka (~MIS-5a), 105 ka
662 (MIS-5c), 120 ka (MIS-5e) and 7 ka (MIS-1). Major tufa growth stages occur during MIS-
663 5 and MIS-1 indicating warmer and wetter conditions than today.

664 - In addition to tufa deposition during interstadials stages, it is remarkable the
665 occurrence of high tufaceous dynamics during MIS-6, indicating that climate in Iberian
666 Peninsula for some time (probably at the onset) of the Penultimate Glacial stage was
667 not so unfavorable to tufa deposition.

668 - The tufa systems dynamics in NE Iberia over last 500 ka seems sensitive to summer
669 solar forcing, as well as to precipitation regime in the western Mediterranean area,
670 affected by latitudinal shifts of North Atlantic perturbations.

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927 **Figure captions**

928 Figure 1. Location of the studied areas (valleys of the Añamaza, Mesa, Piedra and
929 Ebrón rivers) in the Iberian Ranges (NE Iberian Peninsula).

930 Figure 2. Añamaza valley area. A) Geological map, with location of stratigraphic
931 sections and other studied sites. B) Field view of a wedge-shaped tufa deposit (Salto

932 del Cajo), with indication of the stratigraphic sections (A-5, A-7 and A-8) shown in
933 figure 5. C) Detail of section A-10 (metre 2-4), consisting of dominant carbonate sand
934 and silt.

935 Figure 3. Mesa and Piedra valley areas. A) Geological map, with location of
936 stratigraphic sections and other studied sites. B) Field view of tufa deposits (Los
937 Villarejos N; M-4 section) in the Mesa Valley. Thick carbonate sand and silt deposits that
938 grade eastward to moss and hanging-stem boundstones. C) Field view of tufa deposit
939 (La Requijada) in the Piedra valley, with indication of the stratigraphic sections (P-3, P-
940 4 and P-5) shown in figure 7. Note the angular unconformity of the Quaternary
941 deposits over Tertiary and Cretaceous rocks. The two main deposition stages are
942 depicted in the Quaternary.

943 Figure 4. Ebrón valley area. A) Geological map, with location of stratigraphic sections
944 and other studied sites. B) Field view of the valley, showing the two main Quaternary
945 deposition stages arranged in an encased terrace sequence. The locality is Castielfabib.
946 C) Detail of a moss boundstone from section E-2 (Cascada).

947 Figure 5. A) Stratigraphic sections of the Añamaza valley and their correlation. Note
948 position of samples used for dating and correspondence to MIS. Only samples that
949 yielded reliable dates are indicated. Location of sections in figure 2. B) Field view of the
950 lower half of stratigraphic section A-5 (Salto del Cajo).

951 Figure 6. A) Stratigraphic sections of the Mesa valley and their relative position. Note
952 position of samples used for dating and correspondence to MIS. Only samples that
953 yielded reliable dates are indicated. Location of sections in Figure 3. B) Field view of
954 the lower part of stratigraphic section M-3 (Jesús Nazareno Este), with abundant
955 stromatolite deposits.

956 Figure 7. A) Stratigraphic sections of the Piedra valley and their correlation. Note
957 position of samples used for dating and correspondence to MIS. Only samples that
958 yielded reliable dates are indicated. Location of sections in figure 3. B) Field view of
959 stratigraphic section P-1 (Arco de la Yedra). Note the presence of dark grey organic-

960 matter rich horizons between bioclastic and phytoclastic sands and limestones. C) Field
961 view of the middle part of stratigraphic section P-10 (Nuévalos).

962 Figure 8. A) Stratigraphic sections of the Ebrón valley and their relative position. Note
963 position of samples used for dating and correspondence to MIS. Only samples that
964 yielded reliable dates are indicated. Location of sections in figure 4. B) Field view of
965 stratigraphic section E-4 (Mirador) (approximately, metres 15 to 65).

966 Figure 9. Temporal distribution of tufa deposits in the Añamaza (diamonds) (a), Mesa
967 (triangles) (b), Piedra (squares) (c) and Ebrón (circles) (d) valleys. Blue, red and green
968 color symbols refer to U/Th series, racemization and radiocarbon data, respectively.
969 Cumulative probability density function plot of dates (e). Relationships to other
970 palaeoenvironmental records: summer insolation at 42 °N (Berger and Loutre, 1991)
971 (f), marine isotope stages (MIS) compilation (Bradley, 1999) (g) and SPECMAP isotope
972 curve (Martinson et al., 1987) (h). Vertical grey shaded areas show main stream
973 terrace regional aggradation periods.

974 **Table captions**

975 Table 1. U/Th disequilibrium series data and ages.

976 Table 2. Aspartic acid and glutamic acid racemization ratios data and ages. Tufa facies
977 samples are always carbonate sands and silts.

978 Table 3. Radiocarbon data and ages.