

Fluid systems and fracture development during syn-depositional 1 fold growth: an example from the Pico del Aguila anticline, Sierras Exteriores, Southern Pyrenees, Spain.

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Nicolas Beaudoin, Damien Huyghe, Nicolas Bellahsen, Olivier Lacombe, Laurent Emmanuel, et al.. Fluid systems and fracture development during syn-depositional 1 fold growth: an example from the Pico del Aguila anticline, Sierras Exteriores, Southern Pyrenees, Spain.. Journal of Structural Geology, 2015, 70, pp.23-38. 10.1016/j.jsg.2014.11.003 . hal-01083732

HAL Id: hal-01083732 https://hal.sorbonne-universite.fr/hal-01083732

Submitted on 18 Nov 2014 $\,$

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Fluid systems and fracture development during syn-depositional fold growth: an 1 2 example from the Pico del Aguila anticline, Sierras Exteriores, Southern Pyrenees, 3 Spain. 4 5 Nicolas Beaudoin^{1,2,3,*}, Damien Huvghe⁴, Nicolas Bellahsen^{1,2}, Olivier Lacombe^{1,2}, Laurent Emmanuel^{1,2}, Frédéric Mouthereau^{1,2}, Laure Ouanhnon^{1,2}. 6 ¹ Sorbonne Universités, UPMC Univ Paris 06, UMR 7193, ISTEP, F-75005, Paris, France. 7 ² CNRS, UMR 7193, F-75005, Paris, France. 8 ³ School of Geographical and Earth Sciences, University of Glasgow, Gregory Building, 9 10 G12800 Glasgow, United Kingdom. ⁴ UMR 5563 - Geosciences Environnement Toulouse, Université Paul Sabatier Toulouse 11 12 III, Toulouse, France. 13 14 Keywords: Fluid system; Syn-sedimentary fold development; Pyrenean foreland; Fracture 15 population; Sierras Exteriores; Pico del Aguila anticline. 16 17 18 19 Abstract 20 21 This paper reports an integrated, spatio-temporal analysis of the fracture-controlled 22 paleo-fluid system in the Pico del Aguila anticline, a N-S trending fold located in the Sierras Exteriores, the southern front of the Spanish Pyrenees. Eight fracture sets (joints 23 or faults) are recognized throughout the fold and are separated into a fracture sequence 24 25 that is defined using field relationships and the remarkable temporal constraints offered by the syn-tectonic sedimentary deposits. This fracture sequence records a complex 26 27 Paleocene to Early Oligocene structural evolution, including map-view, clockwise 28 rotation and tilting of the fold axis. The geochemical analysis of calcite cements from the 29 different mineralized fracture/vein sets reveals a compartmentalized fluid system 30 during most of fold development. This initial paleofluid system was later perturbed 31 when bending-related fractures associated with foreland flexure and outer arc extension 32 triggered small-scale, vertical fluid migration. Fractures developed in shallow strata facilitated downward migration of surficial fluids that controlled the paleo-fluid system 33 in the Late Priabonian/Stampian continental deposits. The study of the Pico del Aguila 34 35 anticline depicts for the first time the evolution of a fluid system in a shallow, syn-36 depositional compressional setting, and results further strengthen the statement that

fluids migrate vertically across stratigraphic boundaries take place during fold hinge-related deformation.

39

40 1. Introduction

41 Fluid-rock interactions during folding control diagenesis and deformation, hydrocarbon migration, and heat transport (Qing and Mountjoy, 1992; Roure et al., 2005; Katz et al., 42 2006; Lacombe et al., 2014). A recent review of factors governing the temporal and 43 44 spatial distribution of fluids in folds (Evans and Fischer, 2012) highlights that the 45 development of a sub-seismic fracture network is essential in fluid migration. In 46 particular, the vertical persistence and lateral connectivity of joints usually promotes 47 alternating vertical and lateral fluid migrations at local and large-scale (e.g. Evans and 48 Battles, 1999; Van Geet et al., 2002; Fischer et al., 2009; Barbier et al., 2012a; Beaudoin 49 et al., 2011, 2014). Although no simple rule arises since each natural case of fold-50 fracture-fluid interactions differs, a common characteristics occurs (Evans and Fischer, 51 2012): the development of curvature-related fracture sets promotes vertical fluid 52 migration and mixing of various preexisting hydrologic reservoirs delimited by 53 stratigraphic seals. This kind of evolution can be deciphered when both the large faults 54 and the fracture network are studied. Indeed, sub-seismic fracture patterns experience a 55 succession of deformation steps at fold-scale (Stearns and Friedman, 1972; Fischer and Wilkerson, 2000; Bergbauer and Pollard, 2004; Bellahsen et al., 2006a, b; Cooper et al., 56 2006; Tavani et al., 2006; Beaudoin et al., 2012, 2013). However, most studies were 57 58 performed in settings where deformation substantially postdates strata compaction 59 (Evans and Battles, 1999; Van Geet et al., 2002; Fischer et al., 2009; Beaudoin et al., 60 2011, 2014; Barbier et al., 2012a). Consequently, the evolution of fluid-rock interactions 61 in strata folded at shallow depth during sediment deposition remains incompletely 62 documented.

Here, we study the case of the Pico del Aguila anticline, one of the N-S trending folds of the Sierras Exteriores positioned at the southern structural front of the Pyrenees (Fig. 1). The Pico del Aguila anticline is interpreted as a detachment fold with a décollement level located within Triassic evaporitic rocks (Millán, 1996). The growth of the structure is recorded by the syn-tectonic deposition of deep marine to continental sediments from the Late Lutetian to Priabonian times (Millán et al., 1994; Hogan and Burbank, 1996; Castelltort et al., 2003). The kinematics and mechanics of the Sierras

Exteriores and especially the Pico del Aguila anticline have been extensively documented (Millán et al., 1994; Poblet and Hardy, 1995; Poblet et al., 1998; Novoa et al., 2000; Anastasio and Holl, 2001; Castelltort et al., 2003; Nalpas et al., 2003; Huyghe et al., 2009; Vidal-Royo et al., 2012, 2013). Our investigation of the fluid system in the southernmost fold structures of the Pyrenean foreland allows a comparison with recent studies of fluid systems in the northern hinterlandward fault-related folds (Travé et al., 2000, 2007; Lacroix et al., 2011, 2014).

This contribution aims to describe the fracture network in the Pico del Aguila anticline, and then using the remarkable record of fold evolution granted by the growth strata to decipher the history of fracturing. Geochemical analyses of calcite vein cements as well as fault-coating calcite are used to identify and interpret the sources of fluids that flowed in fractures, their pathways, and their interactions with surrounding rocks. Beyond regional implications, results shed light on the evolution of the paleo-fluid system during growth of syn-depositional detachment folds.

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85 2. Geological Setting

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The Pico del Aguila anticline is a 160°E trending anticline located in the Sierras 87 88 Exteriores, a range comprising a set of NW-SE to N-S trending folds markedly oblique to 89 the south-Pyrenean thrust front (Fig. 1). The Sierras Exteriores are located at the 90 southern front of the Jaca piggyback basin, which borders the southwestern part of the 91 Central Pyrenees. The Pico del Aguila anticline plunges 30° toward the North, because it 92 is linked to the late thrusting of the Jaca Basin over the Ebro basin during the activation 93 of the Guarga basement thrust (Fig. 1B, Teixell, 1996; Jolivet et al., 2007). The Pico del 94 Aguila anticline is a detachment fold with a décollement level located within the Triassic 95 evaporite strata (Fig. 2). The growth of this anticline is well-constrained by the wealth of 96 biostratigraphic data (Canudo et al., 1988; Molina et al., 1988; Sztràkos and Castelltort, 97 2001; Huyghe et al., 2012a) and paleomagnetic studies (Hogan and Burbank, 1996) 98 obtained on Middle Eocene to Oligocene growth strata. The sedimentary succession (Fig. 99 1C) comprises dolostones and gypsiferous clays composing the Triassic Muschelkalk 100 and Keuper facies, respectively (Millán et al., 1994). The overlying formations are the 101 upper Cretaceous platform limestones of the Adraén-Bona Formation (Fm.), the 102 Paleocene continental and fluvial sandstones and mudstones of the Tremp Fm., and the

103 Lutetian shallow marine limestones of the Guara Fm. The sedimentary record indicates 104 that folding began during the deposition of the upper part of the Guara Fm. (upper 105 Lutetian, Millán et al., 1994; Huyghe et al., 2012a) and lasted during deposition of the 106 Bartonian-early Priabonian prodeltaic marls of the Arguis Fm. and the Middle-107 Priabonian marine shallow-deltaic sandstone of the Belsué-Atarès Fm. This 108 interpretation is in accordance with the growth model of Hogan and Burbank (1996) 109 that indicates that folding began during the late Lutetian (42 Ma) and ended during early 110 Priabonian (35 Ma). On top of the latter formation, the Late Priabonian-Stampian 111 continental sandstones and claystones of the Campodarbe Fm. were deposited after 112 folding (Millán et al., 1994). Subsequent thrusting of the Jaca basin over the Ebro basin is 113 due to the development of the southern Pyrenean frontal thrust during Late Oligoceneearly Miocene (Millán, 1996; Jolivet et al., 2007; Huyghe et al., 2009). 114

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116 **3. Methodology to decipher fluid-fracture-fold evolution**

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This work focuses on fracture generations observed in pre-, syn- and post-folding strata of the Pico del Aguila anticline. Fracture generations are mainly composed of joints and veins opened in mode I, as well as faults along which movement is recorded by slickensides. Pressure-solution seams (stylolites) related to both compaction and tectonic loading also occur. Mode I-opening of joints and veins was checked in the field and in thin-sections by the offset of pre-existing elements in the matrix or by the lack of grain crushing in the matrix near the borders of the fractures (Fig. 3a).

Nearly 1500 joint/vein orientation data were collected along with 120 fault-slip data at the outcrop-scale (Fig. 4-7, Table S2 as supplementary material) in different formations (pre-, syn-, and post-folding strata extending from Triassic to Priabonian) and structural positions (hinge and limbs). About 50 sites were defined by a common structural position and bedding dip, half of them being located in the pre-folding strata (25 sites). The post-folding strata of the Campodarbe Fm. have fewer samples in our dataset (6 sites).

132

133 3. 1. Identification of fracture sets

134Identification of fracture sets is now a well-established and powerful tool to135unravel the deformation history of folded strata (eg, Bergbauer and Pollard, 2004;

136 Bellahsen et al., 2006; Lacombe et al., 2012). Fracture sets can be defined as fracture 137 populations that share a common deformation mode, a common orientation regarding 138 the bedding dip and with statistically consistent chronological relationships compared 139 to other fracture sets. For our study, we compute the mean orientation of measured 140 fractures for each site by means of a Kernel statistical analysis (software developed at 141 IFPEN for the definition of fracture sets, see Bellahsen et al., 2006; Ahmadhadi et al., 142 2008). This data processing is first done for the present position of strata, then after the 143 correction of the fold axis plunge (by removing the tilt of 30° due to the frontal thrust 144 activity), and in a third step after the removal of the local bedding dip. Results are 145 presented on stereonets for each measurement site at each step (Figs. 3 to 6). Diagrams 146 are not weighted by abundance, as we believe that this modification can be biased by 147 outcrop conditions. However, vertical persistence, spacing or relative abundance of 148 fractures all were considered for the interpretations. Indeed, we believe that a fracture 149 set is relevant to constrain the tectonic evolution of strata only if it is observed 150 everywhere in the fold, or at least in numerous sites from a single structural or 151 stratigraphic position. Therefore, data processing results in the recognition of different 152 fracture sets that are each related to a deformation event. We assume that the 153 development of a fracture set will not overlap the development of another fracture set 154 except if the stress conditions required are similar for these two sets and if 155 chronological relationships suggest synchronism. Consequently, we use chronological 156 relationships to constrain the development of fracture sets through time so to build the 157 fracture sequence.

158 Four approaches are used to determine the relative age of the different fracture 159 sets: (a) the relative age of fractures based on abutting or offset relationships at field 160 sites; (b) the restriction of fractures to particular units in the pre-, syn- and/or post-161 folding stratigraphic sequence; (c) the assumption that mode I fractures formed 162 vertically with a horizontal least compressive principal stress (Anderson, 1951); and (d) 163 the assumption that bed-perpendicular fractures striking parallel to the fold axis and 164 located near the hinge of the fold are related to local extension due to strata bending. We 165 carefully observed abutments and crosscutting relationships on pavements at the Pico 166 del Aguila, using a rule that a set composed of fractures that terminate at fractures of 167 another set are inferred to have developed later (Fig. 8). These relationships observed at 168 site-scale are checked on thin sections, and the consistency of the chronology is checked

at the fold-scale. Finally, the sequence is checked with the record provided by growthstrata.

- 171
- 172 3.2. Inversion of fault-slip data for paleostress
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174 Some fracture sets comprise only faults, which slips were inverted to reconstruct 175 the related stress tensor using the inversion methods described in Angelier (1984)(see 176 Lacombe, 2012 for a recent review of fault-slip data inversion for paleostresses). The 177 identification and separation of successive generations of faults and related stress 178 regimes are based on both mechanical incompatibility between individual fault slips 179 (with the computed stress tensor) and relative chronology observations (e.g., 180 superimposed striations on fault surfaces, crosscutting relationships between faults). As 181 with the fracture sets, we provide stereonets to show the results of fault-slip data 182 inversion (1) in the current strata attitude (post-thrusting); (2) after removing the 183 regional tilt of the fold axis due to the activation of the frontal thrust (pre-thrusting and 184 post-folding); (3) and after removing the local bedding dip (pre-folding). If one assumes 185 that a principal stress axis remains generally vertical without local stress rotations, 186 which could be due to stress channelization within shallow-dipping strata separated by 187 low friction interlayers along which bedding-parallel slip occurs (Tavani et al., 2006), (1) 188 inversion of a fault set formed before folding (or thrusting) and measured in a fold limb 189 would have one of the computed stress axes perpendicular to bedding, with the other 190 two lying in the bedding plane; (2) inversion of a post-folding or post-thrusting fault set 191 yields stress tensors with compression horizontal irrespective of bedding dip in the 192 current or pre-thrusting attitude (e.g., Lacombe, 2012). Note that pre-folding stress 193 tensors are presented on Schmidt's stereonets only in cases where stress axes are 194 consistent with these Andersonian conditions once corrected from plunge and/or 195 bedding dip.

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197 **3.3.** Petrographic and geochemical analyses of veins and host-rock cements

Samples of vein calcite cements and fault-coating calcite within their surrounding matrix were collected in a variety of stratigraphic and structural positions. We use standard and cathodoluminescence petrography, as well as stable isotopes of carbon and oxygen to constrain the geochemistry of the fluids from which calcite precipitated.

Although we observe fluid inclusions in the samples, microthermometric work proved fruitless, and yielded only the observation that the inclusion population was dominated by monophase (liquid), aqueous inclusions (Fig 3 e-f) where the attempts to nucleate vapor bubble by freezing failed. Lack of vapor bubble in inclusions suggests a precipitation of fluids about 80 ± 20°C (Roedder, 1984).

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208 Petrographic and cathodoluminescence observations were conducted on 209 oriented thin-sections of 35 selected samples that are representative of fracture sets 210 observed at the Pico del Aguila anticline. We use observations of vein crystal textures 211 and offset wall-rock markers to constrain the mode of fracture wall displacement (Fig. 212 3). Microscopy also allowed checking and refining the chronological relationships that have been initially defined from field observations. We use a Cathodyne Opea cold 213 214 cathode system to examine the cathodoluminescence of the samples (Fig. 9). These observations constrain the number of precipitation events, the conditions of 215 216 precipitation, and the diagenesis of the veins and host rocks. Operating conditions are in 217 the range of 200–400 IA and 13–18 kV gun current with a constant 60 mTorr vacuum.

218

 δ^{18} O and δ^{13} C have been measured in calcite collected from 70 veins and related 219 220 host-rocks, covering various structural and stratigraphic positions in the Pico del Aguila 221 anticline (Fig. 10, Table S1). Measurements are performed using an automated 222 preparation device coupled to an Isoprime gas-ratio mass spectrometer. Between 40 223 and 100 µg of calcite powder is collected from each veins, using either hand-drill or 224 scalpel to avoid mixture with host-rocks. Samples are placed in glass vials and reacted 225 with dehydrated phosphoric acid under vacuum at 90°C, before being measured 10 226 times each. A correction for dolomite samples was conducted (Rosenbaum and 227 Sheppard, 1986). Hereinafter, all values for both veins and host-rocks are reported in 228 per mil (‰) relative to the Vienna Pee Dee Belemnite (VPDB or PDB) for carbon and for 229 oxygen with an accuracy of 0.05‰ and 0.1‰, respectively (Table S1).

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231 **4. Fracture system: observations and interpretations**

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We present a new geological map, based on recent field observations and highresolution aerial photographs that slightly differs from the previous maps (Fig. 2; e.g.

Puigdefabregas, 1975; Millán, 1996; Vidal-Royo et al., 2012). The main difference is that
Triassic rocks are not observed in the northern part of the fold where sub-vertical
limestone strata of the Cretaceous Adraen-Bona Fm. were observed.

- Eight fracture sets are defined by field observations, statistical analyses, and stress
 inversion processes, including 5 joint/vein sets (Figs. 4-6) and 3 striated fault sets (Fig.
 7). For the sake of simplicity, the fracture sequence is presented with a nomenclature
 defined by relative timing with respect to folding (Fig. 8).
- 242 Sets J1 and J2 appear mainly in the oldest pre-tectonic formation (Table 1, figs. 4-5-243 6), making them the oldest fractures to have developed. Both are bed-perpendicular. 244 Once the plunge and local dip are removed, set [1 strikes 120° while set [2 strikes 090°. 245 Cross-cutting relationships show younger J2's abutting against older J1's (Fig. 8-b). 246 Other sets are observed in all stratigraphic units. Set J3 joints and veins strike 070° and 247 are bed-perpendicular, and abut against set J2 fractures (Fig. 8-c). Set J4 joints and veins 248 strike 40° and are bed-perpendicular, reopen J3 fractures and abut against J2 fractures 249 (Fig. 8-c). Sets J3 and J4 are observed in every structural position, whereas set J5 is only 250 observed near the anticline hinge and in the syncline. Set J5 joints and veins are roughly 251 normal to bedding and strike mainly 170°. Chronological relationships from outcrops 252 and thin sections indicate that set J5 is younger than sets J1, J2, J3, and J4 (Fig 8-a,d), 253 whereas chronological relationships with set J4 are ambiguous.
- 254 Sets F1, F2 and F3 comprise faults that are defined by a common causative 255 paleostress reconstructed using a stress inversion process (Fig. 7). Because the lack of 256 crosscutting relationships, the chronology of these fault sets is poorly constrained by 257 field and petrographic observations. Age of the fault sets with respect to folding and 258 thrusting events can however be assessed by assuming that they developed when the 259 stress tensor has a vertical principal stress, as predicted by Anderson (1951). Set F1 260 comprises steeply dipping, N-S striking normal faults that were only observed at the fold 261 hinge, and developed under an E-W extensional stress regime during folding (Fig. 7). Set F2 comprises newly-formed N-S reverse faults (ex: site 434, Fig. 7) with strike-slip 262 263 reactivation of fractures oriented 045°E/060°E (ex: site 433-2, Fig. 7) and 160° (ex: 474, 264 Fig. 7), and are compatible with a nearly E-W compression which respect to Anderson's 265 theory in the current attitude of strata or just plunge-corrected, meaning they developed 266 since post-folding. Lastly, set F3 comprise a group of conjugate E-W reverse faults that 267 formed in a predominantly N-S compressional stress regime (ex: site 136-1, Fig. 7).

These faults developed after folding, some of them postdate thrust activation (ex: site 136-1, Fig. 7) while the observed orientation, motion and steep angle of some other reverse faults (ex: site 34, Fig. 7) suggests that they developed at a lower angle during the thrusting.

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273 Prior work has shown that the area of the southern Pyrenean thrust front 274 experienced rotation around a vertical axis. This rotation is interpreted as being related 275 to the southwestward propagation of the deformation in the south Pyrenees and is 276 believed to partially explain the N-S striking of the folds in the Sierras Exteriores. 277 Rotations of 15° to 50° have been proposed from paleomagnetic studies (Puevo et al., 278 2002; Oliva-Urcia and Pueyo 2007) and on displacement field reconstruction (Huyghe et 279 al., 2009). Timing of such a rotation for the Pico del Aguila area is inferred to have 280 started as soon as Bartonian and terminated during Oligocene times (Huyghe et al., 281 2009).

282 Consequently, this rotation history is of primary importance for interpreting the 283 fracture sequence with respect to anticline development (Fig.11). Drawing on timing of 284 sedimentation with respect to folding (Fig. 1B), and on the stratigraphic distribution of 285 the fracture sets (Figs. 3 to 8, Table 1), we propose that the progressive development of 286 fractures from a 090° strike (set J2 only in oldest strata) to a 040° strike (set J4 in all 287 strata) reflects a progressive clockwise rotation (Fig. 11). Joint sets J2 to J4 are inferred 288 to have formed sequentially parallel to the direction of maximum contraction as part of 289 the structural suite recording regional layer-parallel shortening (LPS) that we here 290 documented as striking NE-SW (Fig. 11-a-b-c (2)). This interpretation is based on (1) the 291 established existence of a rotation in the area, (2) the fact that set [2 is a bed-292 perpendicular, fold-axis perpendicular-striking set of joints which can be related to LPS 293 (e.g. Bellahsen et al., 2006a), and (3) the stratigraphic distribution of fracture sets, 294 where [2 is observed only in prefolding and prerotation Guara Fm. (Huyghe et al., 2009) 295 while J3 and J4 developed also in syn-rotation strata (Table 1). The angular difference in 296 present-day strikes for sets J2 to J4 is 50°, which we infer to reflect the maximum 297 magnitude of the rotation around a vertical axis (Fig.11-a-b-c). Our interpretation differs 298 from previous work that predicts a rotation of only 20° at the Pico del Aguila (Huyghe et 299 al., 2009). Considering our data, because of possible local heterogeneities and bed-scale 300 stress perturbations, the strikes of fracture sets are only given within 10° of accuracy

301 (Table 1). Given this limitation and considering mean strikes for each fracture set, the
302 minimal value for the vertical rotation is the difference between J2's (80°) and J4's (50°)
303 strikes. Therefore, our dataset suggests a vertical rotation of about 30° and is more
304 likely considering results from other studies (Pueyo et al., 2002; Oliva-Urcia and Pueyo
305 2007, Huyghe et al., 2009).

306 We infer that set [1 predates the rotation and its final orientation fully records 307 the rotation. Given a present day strike of 120°, the regional trend of J1 before the 308 rotation would be 090° (Fig. 11-a (1)). Thus, the joints could be interpreted as having 309 developed during N-S extension related to foreland flexure and/or forebulge in the area 310 (Hervouët et al., 2005). Set [5 is inferred to have developed at the anticline hinge during 311 outer-arc bending that occurred during vertical axis rotation. As these fractures reflect 312 hinge-related deformation, their orientation remained parallel with the anticlinal hinge 313 as it rotated (Fig. 11 a-b-c). Similarly, during fold growth, hinge extensional strain was 314 accommodated by development of F1 faults that were also rotated with the anticline as it grew and spun (Fig. 11-c (3)). Crosscutting relationships between J4 and J5 are 315 316 ambiguous, so the development of J4 fractures parallel to regional contraction could 317 have been before or coeval with local extension at the hinge.

After rotation around a vertical axis was completed (Fig. 11-d (4)), fold tightening locally perturbed tectonic stress that became perpendicular to fold axis (e.g., Amrouch et al., 2010), in response to which set F2 formed. Later, the Pyrenean-related N-S contraction triggered E-W-trending thrusts ramps (Teixell, 1996; Jolivet et al., 2007), tilted the folds axis during its overthrusting above the 30°-dipping frontal ramp and caused E-W small reverse F3 faults (Fig. 11-d (5)).

324 The proposed fracture sequence reflects a tectonic history starting from foreland 325 flexure and/or forebulge until the late activation of regional thrusts due to Pyrenean N-S 326 orogenic contraction. Similar relationships between regional-scale foreland flexure and 327 the development of systematic sets of parallel joints/veins have been proposed in other 328 foreland basins (Billi and Salvini, 2003; Beaudoin et al., 2012; Quintà and Tavani, 2012), 329 and supports a growing body of evidence that many fractures observed in folded strata 330 may in fact predate folding history (e.g., Bergbauer and Pollard, 2004; Bellahsen et al., 331 2006a; Ahmadhadi et al., 2008; Lacombe et al., 2011; Quinta and Tavani, 2012).

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333 **5. Fluid system: observations and interpretations**

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5.1. Sample selection according to microstructural observations

Using observations of veins and surrounding host-rocks from optical and cathodoluminescence microscopy (Fig. 3, 9), four different textures in veins can be described, following the classifications of Durney and Ramsay (1973) and Machel (2000).

340 The textures are (1) blocky calcite with single-phase, bright orange luminescence with 341 brighter fringes at the external rim of crystals (Fig. 9-a); (2) blocky calcite with grain-342 scale luminescence zonation from bright to dull orange luminescence (Fig. 9-b); (3) 343 Elongated blocky veins with orange luminescence variation in the vein while each 344 fibrous crystal has homogeneous luminescence (Fig. 9-c); and (4) Crack-seal textures 345 characterized by fringes of fine stretched grains on the outer bound of the vein 346 recording one single (Fig. 3-d) or multiple (Fig. 3-a) events. The four textures are 347 observed in all veins sets, so correlation does not exist between texture and either with 348 stratigraphic position or structure or vein set, suggesting precipitation mechanisms 349 independent from these parameters.

350 Textures in veins can be used as indicators of mode I opening and for a single 351 event of fluid precipitation. Blocky calcite texture showing growth competition (Fig. 3-d) 352 is symptomatic of a single fluid precipitation event (Bons et al., 2012). Most of veins 353 from our samples displays such a texture, and cathodoluminescence observed in case 354 (1) (Fig. 9-a) is related to dynamic recrystallization due to growth competition (Machel, 355 2000), while small-scale zonation of case (2) reflects slower precipitation under variable 356 redox condition or simply variation in precipitation kinetics (Machel, 2000). Elongated 357 blocky veins described as texture (3) (Figs. 3-b, 9-c) can be interpreted to reflect 358 precipitation kinetics of fluid equivalent to opening kinematics of the fracture (Bons et 359 al., 2000). Also, the elongation direction relates to the direction of opening of veins, and 360 is useful to distinguish mode I veins fractures (Fig. 3-b) from oblique opening veins 361 fractures (Fig. 3-a), where the latter were discarded from our geochemical study. Finally, 362 multiple crack-seal events were discarded for geochemistry (Fig. 3-a) as they reflect 363 multiple or discontinuous fluid precipitation events, possibly involving different sources 364 (Bons et al. 2000).

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5.2. Fluid sources from isotopic measurements

367 To interpret the geochemical dataset in terms of fluid system evolution, we 368 divided the data into pre-, syn-, and post-folding groups based on host stratigraphic 369 units to determine if isotopic data from veins and host rock show correlations with 370 stratigraphy and/or fold timing (Fig. 10, Table 2). Considering the data this way, we 371 identify four patterns: (1) Veins from Triassic Fms. and some veins from pre-folding 372 Guara Fm. exhibit δ^{18} O and δ^{13} C values lower than those of their host-rocks; (2) all other 373 veins belonging to pre-folding formations exhibit δ^{18} O values lower than those of their 374 host-rocks, while simultaneously exhibiting δ^{13} C values that are broadly similar to those 375 of their host-rocks. (3) Veins in the syn-folding Arguis Fm. have δ^{18} O and δ^{13} C similar to 376 their host-rock values. (4) Veins from the post-folding Campodarbe Fm. exhibit δ^{18} O 377 values that are significantly greater than their host-rock values, but δ^{13} C values similar 378 to their host-rock values. These four patterns in the data suggest that fluids from 379 different sources interacted with the host-rocks before, during and after folding, such 380 that fluid flow and precipitation were a function of stratigraphic interval and 381 deformation timing.

Isotopic disequilibrium in vein cements that belong to pattern (1) suggests an 382 383 opening of each reservoir to an external source of fluids, characterized by negative δ^{13} C 384 values. As shown in Fig. 10-a, the low δ^{13} C values in some Guara Fm. and Triassic veins 385 seem to match those of host rocks in the Paleocene Tremp Fm, and we note that the 386 negative δ^{13} C values are consistent with sedimentary rocks inferred to represent 387 paleosoils and lakes (Pujalte et al., 2009) that contain organic matter. The shallow burial 388 depth attained in this area makes it unlikely that the lower δ^{13} C values in veins reflect 389 the influence of hydrocarbons. Instead, we infer paleohydrological connection and 390 mixing between fluids from these Paleocene and Triassic units. Interestingly, this 391 connection is recorded broadly in veins of sets J1 and J5, meaning that most of the 392 vertical fluid migration between units was triggered by curvature-related fractures, 393 either due to folding or to regional foreland flexure (Fig. 12-a).

Isotopic patterns of cases (2) and (3) reflect a closed stratified fluid system that experienced a different evolution regarding the timing of deposit of host-rock regarding evolution of folding (Fig. 10-a,-b):

397- Veins in pre-folding formations that are not case (1) can be defined by isotopic pattern398(2). The lower δ^{18} 0 of veins relative to host rocks can be interpreted resulting from399precipitation of local fluids after a burial (e.g. Ferket et al., 2000; Travé et al., 2007; Fitz-

Diaz et al., 2011; Evans et al., 2012; Vandeginste et al., 2012). According to the isotopic difference between veins and host-rocks (Δ on Fig. 10-b) and considering temperaturedependent fractionation between H₂O and CaCO₃ after Kim and O'Neil (1997), we estimate that pore fluids precipitated 30°C higher than host-rock precipitation temperature. Considering "normal" geothermal gradient, this interpretation implies a burial of 1km, consistent with the sedimentary history (Vidal-Royo et al., 2012).

- Syn-folding formation is characterized by an isotopic equilibrium between veins and
host-rocks (case (3)) that reflects local pore-fluids precipitation without significant
change in temperature since host-rock underwent diagenesis. This is consistent with the
limited burial experienced by the Arguis Fm. after it deposited in the area of the Pico del
Aguila (Millán, 1996).

The isotopic pattern of case (4) in the post-folding Campodarbe Formation is inferred to represent an opening to external source of fluids characterized by a higher δ^{18} O values. According to the continental paleo-environmental conditions at that time (Millán, 1996), such a source could be either river-derived fluid or meteoric fluids. As river-derived fluids isotopic range cannot be used to explain the measured signatures (-8‰ to -5.5‰PDB, Zamarreno et al., 1997, Huyghe et al., 2012b), we propose that isotopic signatures of case (4) record precipitation from meteoric fluids.

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419 **6.** Discussion : fluid-rock evolution during syn-depositional folding

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In their recent review focusing on fold-related fluid systems, Evans and Fischer (2012) stressed the fact that these fluid systems have some common characteristics before and during folding. Analysis of paleo-fluid systems during growth of both detachment and basement-cored folds show that fluid systems are compartmentalized by stratigraphy and exhibit little vertical fluid migration. During subsequent folding, syn-folding joints and faults rupture stratigraphic seals and trigger vertical fluid migration and mixing.

The fluid system evolution of the Pico del Aguila (Fig. 12-a) is accordingly interpreted as a stratified fluid system during most of the geological history, with a strong control of lithology on the fluid isotopic signatures (e.g. Fischer et al., 2009; Evans and Fischer, 2012). However, inter-formational fluid flow is documented for sets J1 and J5, which are related to flexural forebulge and local extension due to folding,

respectively. Once the paleo-environment switches from marine to continental during
Priabonian (Millán, 1996), the source for formational fluid switched from marinederived pore fluids to surficial, likely meteoric-derived fluids.

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436 The likely common opening of fluid systems to vertical migration during folding 437 (e.g., Evans and Fischer, 2012, Fig.12) is therefore once more supported by the Pico del 438 Aguila case study. Opening during flexural forebulge has also been documented in the 439 Bighorn Basin (Beaudoin et al., 2014). Being the first syn-depositional fold developed at 440 shallow depth for which the fluid system has been studied, the Pico del Aguila 441 additionally illustrates, beyond the strong lithological control on the fluid system, the 442 progressive switch from marine to continental environment as documented by δ^{18} O 443 values of calcite-cemented veins. This interpretation is consistent with observations of 444 current fluid flow in anticlines developed at shallow or significant water depth as in the 445 Central Basin in Iran or in Brunei (Morley et al., 2014). The difference in burial depth at 446 the time of deformation also impacts on the scale of the vertical migration triggered by 447 effective tension-related fracture sets (Fig. 12). For example in a deep buried basement-448 cored fold, such as the Sheep Mountain anticline (Fig. 12-a, Beaudoin et al., 2011), 449 curvature-related fractures developed enough vertical permeability to allow fluid from 450 depth to invade all the strata, while such big-scale migration is not recorded for the Pico 451 del Aguila (Fig. 12-b). This difference could be directly related to the limited burial and 452 related mechanical compaction of the reservoir, and could illustrate the influence of 453 mechanical properties of strata on hydraulic behavior of curvature related fractures, as 454 highlighted in numerous natural cases (Cooke, 1997; Fischer and Jackson, 1999; 455 Laubach et al, 2009; Savage et al., 2010; Barbier et al., 2012a; b; Morley et al., 2014).

456 More generally, studies of regional fluid flows in the southern Pyrenean foreland 457 depict large-scale flows of hydrothermal fluids in structures closer to the Pyrenean range (Travé et al., 2000; 2007), and in the Gavarnie thrust, structurally above the 458 459 Guarga thrust (McCaig et al. 1995; Henderson and McCaig, 1996; McCaig et al., 2000). In 460 the Pico del Aguila, no hydrothermal fluid flow overprinted the system during activation 461 of the underlying Guarga thrust that developed set F3 faults, in which syn-kinematic 462 calcite coating precipitated from local fluids (Table S1). The lack of evidence of deep 463 fluid flow has also been documented along the thrust system of the Monte Perdido, 464 South of Gavarnie (Lacroix et al., 2011) and can be related to the large distance from the 465 range (Figure 1, Lacroix et al., 2014). Alternatively, this lack could be related to limited

faults and joints development after folding. The related vertical permeability creation
was too limited and prevented fluids from the basement to flow through the nonpermeable evaporites underlying the limestone. Such a case is opposed to what can
happen in a basement-cored fold (Fig. 12-a).

470

471 **7. Conclusions**

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473 1. The sub-seismic fracture pattern recognized in folded strata of the Pico del 474 Aguila anticline comprises 8 sets of joints/veins and faults. The oldest fracture set is 475 likely related to the regional-scale foreland flexure that affected strata during Lutetian 476 and therefore clearly predates folding history. Three fracture sets (J2 to J4) then 477 developed in progressively youngering strata, recording a clockwise vertical-axis 478 rotation of the area. Their E-W to NE-SW trends indicate that they developed under a 479 far-field, relatively static NE-SW shortening during the 30-40° rotation around a vertical 480 axis. Among the four remaining sets, two are related to local outer-arc extension during 481 folding (sets [5 and F1), one is related to E-W compression during late-stage fold 482 tightening (set F2), and the last is a set of post-thrusting faults (set F3) that formed in 483 the same N-S compressional stress regime that activated the Guarga basement thrust.

2. The paleo-fluid system related to the fracture pattern is stratified and controlled by depositional environments during most of the history of vein mineral precipitation. The development of regional-scale foreland flexure and local-scale strata curvature-related vein sets triggered small-scale, interformational, vertical fluid migrations between Triassic and Paleocene reservoirs. The progressive switch from marine to continental paleo-environment occurring during Priabonian is recorded by a change of fluid source from local marine fluids to terrestrial surficial fluids.

491 3. Our interpretation of the fluid system in the Pico del Aguila anticline supports 492 the hypothesis that fluid systems exhibit a common behavior during folding, wherein 493 curvature-related facilitate of joints vertical migration fluids from one 494 hydrostratigraphic reservoir to another. It also illustrates that the extent of such a 495 vertical migration may be strongly reduced when folding affects poorly compacted 496 sediments. Other similar case studies are needed to confirm if the fluid system evolution 497 deciphered in the Pico del Aguila anticline is archetypal of fold-related fluid systems in 498 shallow, syn-tectonic sedimentary settings.

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500 Acknowledgments

- 501 Authors thank N. Labourdette for analytical support and F. Delbas for thin-sections.
- 502 Authors are grateful to M. Fischer, M. Evans, and Editor W. Dunne for their inspiring and
- 503 highly helpful reviews, which have significantly improved this manuscript. S. Castelltort
- 504 is thanked for help in the field and fruitful discussions. This work was supported by
- 505 ISTeP and material support of LFC-R laboratory.
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729 Captions

730 Figure 1: A- Regional geological map of the Southern Central Pyrenees with location of 731 the balanced cross-sections that cross the Pico del Aguila anticline (Huyghe et al., 2009); 732 B- Stratigraphic column for the Pico del Aguila area (after Castelltort et al., 2003). 733 Lithological key of patterns from bottom to top: carets - evaporate, dashs - clay, 734 parallelograms – dolostone, rectangles – limestone, big dots in irregular shapes – river 735 conglomerates, plain black – marls, fine dots - sandstone; C- Transverse and longitudinal 736 regional cross-sections in the Sierras Exteriores, focusing on the Pico del Aguila Area 737 (Huyghe et al., 2009).

738

739 Figure 2: Geological map of the Pico del Aguila Anticline based on previously published 740 maps (Puigdefabregas, 1975; Millán, 1996; Vidal-Royo et al., 2012), aerial photographs 741 and new field observation and measurements. A balanced cross-section following line C-742 C' is presented, with a length balance of layers for strata younger than Triassic and an 743 area balance for the Triassic rocks of the décollement. Dotted frames on the maps are 744 areas of maps presented in figures 4-6. White-labeled black dots locate fracture 745 measurement sites or sample sites for geochemical analyses. Please refer to 746 supplementary material 2 for GPS values. In the stratigraphy caption box, "cont." refers 747 to continental environment.

748

749 Figure 3: Photomicrographs of thin- and thick-sections using polarized and cross-750 polaryzed microscopy. a – Multiple crack-seals and fibrous vein where calcite crystals 751 exhibit the growth direction, recording a mixed mode I-mode II 'transtensional' opening 752 mode of the vein (sample A14); b – Vein with elongated blocky calcite texture where the 753 grain growth direction is perpendicular to the border of the vein, indicating a mode I 754 opening (sample A44). c – Photomicrograph of fluid inclusions in a thick-section. Biggest 755 primary or pseudo-secondary fluid inclusions are circled in red and the stained parts in 756 crystals corresponds to an increase in density of secondary fluid inclusion trails (sample 757 A45). d – Calcite vein with blocky texture fringed by microsparitic crystals, recording a 758 two-stage opening of the vein (sample A86), analogous to the crack-seal model (Ramsay, 759 1980).

760

Figure 4: Results of fracture analysis in the Triassic to Lutetian prefolding strata in the
anticline. Results are presented on 3 diagrams (Schmidt' lower-hemisphere, equal-area
stereonets) displaying raw data in current strata attitude (left), then corrected for fold
axial plunge related to the Gavarnie thrust activation (middle), and corrected for local
bedding dip (right). Location of the measurement area is given in Fig. 2 and in
supplementary material 2. Abbreviations on the map are for the formations: Ar – Arguis
Fm., Gu – Guara Fm., Tp – Tremp Fm., A-B – Adraen Bona Fm., Tr – Triassic Fms.

768

Figure 5: Results of fracture analysis in the western syncline located in the Pico del
Aguila area. Same key as in Fig. 4. Abbreviations on the map are for formations: Ar –
Arguis Fm., Gu – Guara Fm., Tp – Tremp Fm., A-B – Adraen Bona Fm.

772

Figure 6: Results of fracture analysis in the syn-folding and post-folding strata of
Bartonian to Priabonian age, respectively. Same key as in Fig. 4. Abbreviations on the
map are for formations: Cp – Campodarbe Fm., B-A – Beslusé Atares Fm., Ar – Arguis
Fm., Gu – Guara Fm.

777

Figure 7: Results of fault-slip inversion (Schmidt' stereonets lower hemisphere). 778 779 Computed stress axes are reported as stars with three branches (σ_1), four branches (σ_2) 780 and five branches (σ_3). Convergent/divergent black arrows indicate the direction of 781 compression/tension. Results are represented in the current attitude of strata, and 782 diagrams labeled as "corrected" correspond to the same computed tensor and fault-slip 783 data but corrected for removal of fold axial plunge related to the activation of Gavarnie 784 thrust. Diagrams labeled as "unbasc" are corrected for fold axis plunge then for bed tilt. 785 Diagrams are gathered according to consistency of reconstructed paleostress tensors. 786 Abbreviations on the map are for formations: Cp – Campodarbe Fm., B-A – Belsué Atares 787 Fm., Ar – Arguis Fm., Gu – Guara Fm., Tp – Tremp Fm., A-B – Adraen Bona Fm., Tr – 788 Triassic Fms.

789

Figure 8: a-d – Field photographs with chronological interpretations of fracture
networks. Sites for photographs: site 39(a), site 497 (b), site 476 (c), and site 447(d). e-f
Photographs of faults and fractures showing the spacing and vertical persistence of
fractures observed at site 433 (refer to Figs. 4 to 7 for location).

794

Figure 9: Photomicrographs of thin sections under cathodoluminescence microscopy: a
- Heterogeneous blocky-type calcite in vein, with brightness variation related to crystal
boundaries or to atomic-scale defects (sample A77); b - Heterogeneous blocky-type
calcite in vein exhibiting bright to dull orange zonation related to crystal growth (sample
A37); c - Elongated blocky-type calcite in vein exhibiting brightness variation related to
crystals (sample A44, Fig. 3-b). Please refer to the electronic version for colors.

801

Figure 10: Results of δ^{18} O and δ^{13} C analysis of veins and host-rocks displayed in context of fracture age with respect to folding. a - δ^{18} O vs δ^{13} C plot, b - δ^{18} O of veins vs δ^{18} O of related host-rocks. On all charts, solid symbols are for vein cements and fault coatings, whereas empty symbol are for related host-rocks samples. Please refer to the text for the explanation of labels (1) to (4). All values are expressed in ‰PDB. Labels XX/YY refers to the number of analyzes performed / number of samples collected. See Table 2 and Supplementary Table S1 for detailed isotopic data.

809

Figure 11: Schematic block diagrams of structural, sedimentary and mesostructural evolutionary scenario of the Pico del Aguila anticline. Fracture sets and related contractional/extensional trends are illustrated. Stratigraphic timing and timing with respect to folding stated for each block diagram. Fractures are not represented according to abundance. Labels (1) to (5) are related to local and regional stress orientations (see text).

816

Figure 12: a - Schematic cross-section illustrating evolution of the paleo-fluid system in
the Pico del Aiguila Anticline. b - Comparison with the Sheep Mountain anticline,
Wyoming, USA (after Beaudoin et al., 2011; Evans and Fischer, 2012).

820

Table 1: Results of the statistical interpretation of fracture set orientation, stratigraphicdistribution and indicators for relative chronology.

823

Table 2: Number of samples used for isotopic analysis in each formation, along with therelated range of isotopic values measured.

826

- 827 Table S1: Results of isotopic analyses.
- 828 Table S2: Geographical location of samples and measurement sites

829

Set name	Set strike*	Set inclination	Stratigraphic units	Evidence for relative age
J1	120	bed-normal	Guara - lower Arguis	RD, CC, A
J2	90	bed-normal	Guara	RD, CC, A
J3	70	bed-normal	All	СС, А
J4	40	bed-normal	All	CC, A
J5	170	sub bed-normal	All	CC, A

*: mean strike within 10° computed statistically

RD: Restricted to Stratigraphic units CC: cross-cutting relationships; A: respect andersonian criterion

<u>s; A: respe</u>

Age relative	Forma	tion	number of analyses/number	Range of isotopic v	alues (>80% samples)
tororang			or samples	δ ¹⁸ O (‰PDB)	δ ¹³ C (‰PDB)
		veins	7/8	-2.20 to -0.02	-1.77 to -0.77
Post-folding	Campodarbe	host-rock	7/8	-6.05 to -5.4	-1.2 to 0.05
	Belsue	veins	2/3	-4.65 to -2.91	-2.61 to -1.38
Sup fal-line	Atares	host-rock	3/3	-5.81 to -4.81	-1.53 to 0.11
syn-tolding	Arguis	veins	25/25	-4.35 to -0.66	-1.64 to 0.48
		host-rock	20/25	-4.47 to -2.55	-1.7 to 0.43
	Guara	veins	29/29	-10.4 to -1.42	-0.05 to 2.25
		host-rock	20/29	-6.7 to -1.42	-1.5 to 2.15
Dro folding	Tremp	veins	5/5 1/2	-7.20 10 -0.71	-7.77 10 -0.54
Pre-Iolaing		NOSL-FOCK	1/3	-5.43	-0.56
	Adraen Bona	host-rock	1/1	-3.9	1.49
		veins	3/4	-7 65 to -6 73	-5 43 to -3 51
	Triassic	host-rock	4/4	-5.78 to -3.28	-1.37 to 2.65









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Highlights:

- . Fluid system deciphered during syn-sedimentary folding
- . Role of tectonic and sedimentary environment on fluid system evolution
- . Role of burial on vertical permeability of fractures
- . Fracture development witnesses fold rotation around a vertical axis

Deposition	Sample	Formation	Age	Fracture Set	Vein isotop	pic ratios	Host rock is	otopic ratio	Site Number
regarding folding					$\partial^{18}0$	$\partial^{13}C$	$\partial^{18}0$	$\partial^{13}C$	(Figure for location)
	A040	Campodarbe	Priabonian	Set J4			-5.36	-0.13	447 (Fig. 6)
	A041	Campodarbe	Priabonian	Set J5	-0.3	-1.77	-5.35	-1.06	449 (Fig. 6)
	A042	Campodarbe	Priabonian	Set 15	-0.68	-1.62	-6.03	-0.24	450 (Fig. 6)
1dine	A044	Campodarbe	Priabonian	Set J4	-0.13	-1.56	-5.28	-0.36	452 (Fig. 6)
at for	A045	Campodarbe	Priabonian	Set J4	-0.5	-1.43	-5.64	-0.34	453 (Fig. 6)
802	A048	Campodarbe	Priabonian	Set 13	-0.02	-0.77			455 (Fig. 6)
	A050	Campodarbe	Priabonian	Set J4	-2.2	-1.08	-3.52	-0.02	458 (Fig. 6)
	A050	Campodarbe	Priabonian	Set J2	-4.79	-1.02	-3.52	-0.02	458 (Fig. 6)
	A103	Belsue-Atarès	Bartonian	Set 13			-4.81	0.11	507 (Fig. 4)
	A054	Belsue-Atarès	Bartonian	Set J3	-2.91	-1.38	-5.81	-0.05	465 (Fig. 4)
	A104	Belsue-Atarès	Bartonian	Set J2	-4.65	-2.61	-5.11	-1.53	511 (Fig. 4)
	A001	Arguis	Bartonian	Set 12	-3.48	-1 41	-3 19	-0.9	433 (Fig. 4)
	A003	Arguis	Bartonian	Set F1	-3 53	-8 19	-3.91	-0.97	433 (Fig. 7)
	A004	Arguis	Bartonian	Set F1	-3.4	-1 47	5.51	0.57	433 (Fig. 7)
	A005	Arguis	Bartonian	Set F1	-3.55	-1 /9	-3 77	-1 55	433 (Fig. 7)
	A003	Arguis	Bartonian	Set 13	-4.35	-1.45	-4.16	-1.55	433 (Fig. 7)
	A000	Arguis	Bartonian	Sot E1	2.66	1.40	4.10	1.14	433 (Fig. 4)
	A009	Arguis	Bartonian	Set F1	2.00	-1.04	2	0.07	433 (Fig. 7)
	A010	Arguis	Bartonian	Set F1	-3.20	-0.51	-3	0.02	433 (Fig. 7)
	A011	Arguis	Bartonian	Set JZ	-2.31	-0.76	-3.87	0.12	433 (Fig. 4)
	A012	Arguis	Bartonian	Set F1	-2.54	-0.78	-3.35	-0.72	433 (Fig. 7)
	A013	Arguis	Bartonian	Set J2	-2.51	-0.78	-4.72	-1./	433 (Fig. 4)
foldi	AU13	Arguis	Bartonian	Set J2	-2.51	-0.78	-4.67	-1.37	433 (Fig. 4)
SAUL	A015	Arguis	Bartonian	Set J1	-2.94	-0.29	C		434 (Fig. 4)
	A020	Arguis	Bartonian	Set J4	-3.79	-0.23	-3.99	-0.46	436 (Fig. 4)
	A021	Arguis	Bartonian	Set J3	-2.86	-0.77	-3.69	-0.29	436 (Fig. 4)
	A022	Arguis	Bartonian	Set F2	-2.96	-0.9	-3.35	-0.72	439 (Fig. 7)
	A071	Arguis	Bartonian	Set J2	-3.56	-1.46	-3.61	-0.23	476 (Fig. 4)
	A072	Arguis	Bartonian	Set J2	-2.43	-1.19	-2.55	-0.26	476 (Fig. 4)
	A073	Arguis	Bartonian	Set J3	-2.85	-0.82	-3.09	-0.28	477 (Fig. 4)
	A077	Arguis	Bartonian	Set J2	-2.7	-1.04	-4.47	-0.75	482 (Fig. 4)
	A077	Arguis	Bartonian	Set J3	-2.43	-1.23	-4.47	-0.75	482 (Fig. 4)
	A078	Arguis	Bartonian	Set J3	-2.37	-1.64			482 (Fig. 4)
	A107	Arguis	Bartonian	Set J3	-2.93	-1.13			513 (Fig. 4)
	A121	Arguis	Bartonian	Set F3	-0.66	0.48	-3.03	0.43	522 (Fig. 7)
	A124	Arguis	Bartonian	Set F2	-3.16	-0.64	-4.45	0.28	523 (Fig. 7)
	A128	Arguis	Bartonian	Set J2	-3.08	-1.00	-3.4	-1.44	527 (Fig. 4)
	A031	Guara	Lutetian	Set F2	-5.8	1.88	-1.66	2.55	444 (Fig. 7)
	A032	Guara	Lutetian	Set F3	-8.45	-0.34	-1.42	2.38	445 (Fig. 7)
	A033	Guara	Lutetian	Set F3	-3.99	2.25	-2.73	0.69	445 (Fig. 7)
	A037	Guara	Lutetian	Set 12	0.38	2 34	-1.62	1.83	445 (Fig. 4)
	A056	Guara	Lutetian	Set 13	-8.99	0.57	-5.94	-0.74	443 (Fig. 4)
	A057	Guara	Lutetian	Set 15	-4.19	0.66	-5.04	-0.41	467 (Fig. 4)
	A060	Guara	Lutotian	Sot IS	4.15	0.00	5.45	0.41	467 (Fig. 4)
	A000	Guara	Lutotian	Set IE	3.75	0.5	4.07	0.35	407 (Fig. 4)
	A060	Guara	Lutetian	Set JS	-5.75	0.9	-4.97	-0.27	467 (Fig. 4)
	A061	Guara	Lutetian	Set 13	-4.74	0.46	-5.58	-0.82	409 (Fig. 4)
	A064	Guara	Lutetian	Set J2	-2.52	1.07	-2.08	0.23	470 (Fig. 4)
	A064	Guara	Lutetian	Set J2	-2.66	1.12			470 (Fig. 4)
	A064	Guara	Lutetian	Set J5	-8.29	-3.24			470 (Fig. 4)
	A066	Guara	Lutetian	Set J3	-2.42	1.01			472 (Fig. 4)
	A066	Guara	Lutetian	Set J3	-1.94	-0.31			472 (Fig. 4)
	A067	Guara	Lutetian	Set J1	-10.4	-3.71	-3.52	-0.02	473 (Fig. 4)
	A068	Guara	Lutetian	Set J3	-1.03	1.62	-2.36	1.36	474 (Fig. 4)
	A070	Guara	Lutetian	Set J3	-2.19	1.26	-2.51	0.72	475 (Fig. 4)
Aine	A087	Guara	Lutetian	Set J5	-6.14	-0.41	-5.1	1.05	497 (Fig. 4)
a foll	A088	Guara	Lutetian	Set J1	-8.49	-5.17			497 (Fig. 4)
Pre	A089	Guara	Lutetian	Set J3	-7.91	1.81			498 (Fig. 4)
	A091	Guara	Lutetian	Set J2	-7.67	0.92	-6.12	0.38	503 (Fig. 4)
	A094	Guara	Lutetian	Set J1	-8.8	-4.56	-6.7	-1.27	499 (Fig. 4)
	A097	Guara	Lutetian	???	-3.13	-3.28	-2.41	0.22	503 (Fig. 4)
	A100	Guara	Lutetian	???	-5.39	0.8	-1.85	0.67	503 (Fig. 4)
	A100	Guara	Lutetian	Set J2	2.1	0.98	-1.85	0.67	503 (Fig. 4)
	A109	Guara	Lutetian	Set J3	-0.71	2.38	-3.12	1.83	515 (Fig. 4)
	A115	Guara	Lutetian	Set J3	-8.74	2.27			519 (Fig. 4)
	A118	Guara	Lutetian	Set J5	-1.96	2.38			520 (Fig. 4)
	A119	Guara	Lutetian	222	-2.55	2 14			520 (Fig 4)
	A086	Adraen-Bona	Cretaceous	Set 12	-5.43	0.56	-3.9	1 49	493 (Fig 4)
	Δ082	Tremn	Cretaceous	Set IS	-7 08	-6 2/	5.5	1.40	489 (Fig 1)
	A002	Tromp	Crotacoous	Sct 15	-7.00	.34			180 (Fig. 4)
	AU83	Tromp	Crotococci	Set J3	-7.20	-1.2	2 77	6 20	405 (FIB. 4)
	AU84	Triossia		Set 14	-0./1	-7.77	-3.//	-0.29	450 (FIB. 4)
	AISI		Tail	Set J1	-7.23	-3.51	-3.28	2.65	553 (FIg. 4)
	A131	Triassic	Triassic	Set J4	-7.65	-4.09	-3.28	2.65	553 (Fig. 4)
	A132	Triassic	Triassic	111	-6.73	-5.43	-4.11	-1.37	555 (Fig. 4)
<u> </u>	A133	Triassic	Triassic				-5.78	0.21	555 (Fig. 4)
Accuracy of the me	asurment	s reported in this table is ba	ased on the s	tandard deviatio	n of the values	obtained from t	he standards,	and is of 0.05	5‰ for carbon and
0.1‰ for oxygen									

Supplementa	ry material 2: Site nur	nber lo cation		
Site number	Longitude (decimal)	Latitude (decimal)	Formation	Age
446	-0.398945	42.327804	Campodarbe	Priabonian Priabonian
447 449	-0.407133	42.329652	Campodarbe	Priabonian
450	-0.401991	42.329628	Campodarbe	Priabonian
452	-0.401818	42.329522	Campodarbe	Priabonian
453	-0.401759	42.329546	Campodarbe	Priabonian
455	-0.401005	42.329284	Campodarbe	Priabonian
458 528	-0.400551 -0.393591	42.329169 42.342126	Campodarbe	Priabonian
459	-0.393884	42.325935	Belsué-Atarès	Bartonian
464	-0.394159	42.325990	Belsué-Atarès	Bartonian
465	-0.394251	42.326038	Belsué-Atarès	Bartonian
507	-0.412320	42.328541	Belsué-Atarès	Bartonian
511	-0.412357	42.328458	Belsué-Atarès	Bartonian
43	-0.414050	42.326917	Arguis	Bartonian
44 122	-U.426783	42.325333	Arguis	Bartonian
433 434	-0.413300	42.326212	Arguis	Bartonian
436	-0.414191	42.326608	Arguis	Bartonian
439	-0.414035	42.326563	Arguis	Bartonian
476	-0.412080	42.326619	Arguis	Bartonian
477	-0.412371	42.326508	Arguis	Bartonian
482	-0.412419	42.327050	Arguis	Bartonian
513	-0.412389	42.326222	Arguis	Bartonian
522	-0.420804	42.327150	Arguis	Bartonian
523	-0.421153	42.32/128	Arguis	Bartonian
527	-0.424152	42.323338 42 305800	Aiguis Guara	bar (011181) Lutetian
26	-0.40723	42.314360	Guara	Lutetian
27	-0.407833	42.314367	Guara	Lutetian
28	-0.406483	42.313517	Guara	Lutetian
31	-0.404667	42.308367	Guara	Lutetian
32	-0.403767	42.307033	Guara	Lutetian
33	-0.403067	42.305917	Guara	Lutetian
34	-0.410050	42.323300	Guara	Lutetian
35	-0.414367	42.324433	Guara	Lutetian
30 27	-0.420330	42.23/339	Guara	Lutetian
38	-0.428286	42.301444	Guara	Lutetian
39	-0.411600	42.317750	Guara	Lutetian
40	-0.412467	42.321100	Guara	Lutetian
42	-0.414767	42.322667	Guara	Lutetian
45	-0.411300	42.325400	Guara	Lutetian
46	-0.422067	42.306017	Guara	Lutetian
51	-0.409400	42.309736	Guara	Lutetian
52	-0.402764	24.300739	Guara	Lutetian
136	-U.412183	42.326367	Guara	Lutetian
13/ 129	-0.411233	42.329083	Guara	Lutetian
138 120	-0.428007	42.303667	Guara	Lutetian
141	-0.446800	42.294267	Guara	Lutetian
142	-0.442767	42.310567	Guara	Lutetian
144	-0.393750	42.345333	Guara	Lutetian
145	-0.418917	42.324983	Guara	Lutetian
146	-0.402833	42.335867	Guara	Lutetian
444	-0.412102	42.325097	Guara	Lutetian
445	-0.412098	42.325157	Guara	Lutetian
467	-0.405834	42.312462	Guara	Lutetian
469	-0.405920	42.312555	Guara	Lutetian
4/U 177	-0.405842 -0 205998	42.312005	Guara	Lutetian
473	-0.406108	42.312823	Guara	Lutetian
474	-0.406552	42.313484	Guara	Lutetian
475	-0.406526	42.313602	Guara	Lutetian
497	-0.419564	42.306512	Guara	Lutetian
498	-0.420014	42.306393	Guara	Lutetian
499	-0.420358	42.306254	Guara	Lutetian
500	-0.429013	42.304032	Guara	Lutetian
503	-0.403275	42.306514	Guara	Lutetian
515	-0.416279	42.323905	Guara	Lutetian
519	-0.414092	42.322/08	Guara	Lutetian
493	-0.411017	42.309405	Adrean-Bona	Cretaceous
489	-0.407789	42.310467	Tremp	Cretaceous
490	-0.407872	42.310662	Tremp	Cretaceous
552	0 401047	42 205117	T - 1 1 -	Tringela
555	-0.401047	42.285117	Triassic	THASSIC