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4 **Ophicalcites from the Northern Pyrenean Belt: a field, petrographic and**
5 **stable isotope study.**

6

7 by:

8 Camille Clerc (1), Boulvais Philippe (2), Yves Lagabrielle (3) and Michel de Saint Blanquat
9 (4)

10

11 (1) Laboratoire de Géologie, CNRS-UMR 8538, Ecole Normale Supérieure, 24 rue Lhomond,
12 75231 Paris Cedex 5, France.

13 (2) Géosciences Rennes, CNRS-UMR 6118, Université de Rennes 1, Campus de Beaulieu,
14 35042, Rennes Cedex, France

15 (3) Géosciences Montpellier, CNRS-UMR 5243, Université de Montpellier 2, Place Eugène
16 Bataillon, 34095 Montpellier, France

17 (4) GET, CNRS-UMR 5563, Université Paul Sabatier, 14 avenue Edouard Belin, 31400
18 Toulouse, France.

19

20 Corresponding author:

21 Camille Clerc

22 clerc@geologie.ens.fr

23 **Abstract**

24

25 Brecciated and fractured peridotites with a carbonate matrix, referred to as
26 *ophicalcites*, are common features of mantle rocks exhumed in passive margins and mid-
27 oceanic ridges. Ophicalcites have been found in close association with massive peridotites,
28 which form the numerous ultramafic bodies scattered along the North Pyrenean Zone (NPZ),
29 on the northern flank of the Pyrenean belt. We present the first field, textural and stable
30 isotope characterization of these rocks. Our observations show that Pyrenean ophicalcites
31 belong to three main types: (1) a wide variety of breccias composed of sorted or unsorted
32 millimeter-to meter-sized clasts of fresh or oxidized ultramafic material, in a fine-grained
33 calcitic matrix; (2) calcitic veins penetrating into fractured serpentine and fresh peridotite; and
34 (3) pervasive substitution of serpentine minerals by calcite. Stable isotope analyses (O, C)
35 have been conducted on the carbonate matrix, veins and clasts of samples from 12 Pyrenean
36 ultramafic bodies. We show that the Pyrenean ophicalcites are the product of three distinct
37 genetic processes: i) pervasive ophicalcite resulting from relatively deep and hot hydrothermal
38 activity; ii) ophicalcites in veins resulting from tectonic fracturing and cooler hydrothermal
39 activity; and iii) polymictic breccias resulting from sedimentary processes occurring after the
40 exposure of subcontinental mantle as portions of the floor of basins which opened during the
41 mid-Cretaceous. We highlight a major difference between the Eastern and Western Pyrenean
42 ophicalcites belonging respectively to the sedimentary and to the hydrothermal types. Our
43 data set points to a possible origin of the sedimentary ophicalcites in continental endorheic
44 basins, but a post-depositional evolution by circulation of metamorphic fluids or an origin
45 from relatively warm marine waters cannot be ruled out. Finally, we discuss the significance
46 of such discrepancy in the characteristics of the NPZ ophicalcites in the frame of the variable
47 exhumation history of the peridotites all along the Pyrenean realm.

48

49

50

51 **Key words:** ophicalcite, ophicarbonates, stable isotopes, oxygen, carbon, veins, matrices,

52 Pyrenees, Lherz, Urdach, mantle exhumation, tectonic fracturation, hydrothermalism,

53 sedimentary deposits.

54 I. Introduction

55 During uplift and exhumation of the sub-continental mantle, the peridotites are commonly
56 serpentinized through interactions with fluids, with direct consequences on their bulk density
57 and their rheological, seismic, gravimetric and magnetic properties (Brun & Beslier 1996;
58 Boschi et al. 2006). In oceanic and passive margin environments, besides serpentinization, the
59 peridotites show evidence of carbonation expressed through the occurrence of a bimodal,
60 ultramafic and carbonate association known as ophicalcites or ophicarbonates (Spooner &
61 Fyfe 1973; Bonatti et al. 1974; Dietrich et al. 1974; Gianelli & Principi 1977; Ohnenstetter
62 1979; Lemoine 1980; Cortesogno et al. 1981; Lagabrielle & Cannat 1990). More recently, the
63 carbonation of the exhumed mantle rocks has been clearly described as the last event affecting
64 faulted rocks uprising along oceanic detachment faults at the axis of slow-spreading ridges
65 (Picazo et al. 2012).

66 Ophicalcites were first discovered in the Ligurian Alps (Bonney 1879), and then described in
67 many ophiolite sequences (see Artemyev & Zaykov 2010 and Bogoch 1987 for a
68 comprehensive historical literature on ophicalcites). Ophicalcites commonly display large
69 variations in the proportions of ultramafic and carbonate material. They range from in-situ
70 fractured peridotites with carbonate infill, through clast-supported breccias with multiple
71 generations of carbonate infilling and internal sediments, to matrix-supported breccias.
72 Lemoine et al. (1987) distinguish two main types of ophicalcite. Ophicalcite type 1 (OC1) is
73 represented by massive serpentinites exhibiting a dense mesh of calcite-infilled fractures.
74 Ophicalcite type 2 (OC2) refers to sedimentary breccias having a calcitic matrix, often
75 deposited above ophicalcite of type 1. The clasts comprise sorted or unsorted millimeter- to
76 meter-sized fresh or oxidized ultramafic fragments. In some cases, exotic clasts of gabbroic or
77 basaltic composition can be observed. The matrix of OC2 is either a fine-grained calcitic

78 sediment or a cement consisting of sparry calcite. It varies in color from red to pink and gray
79 to green, depending on the chlorite or hematite content; sparry calcite is generally white.
80 Ophicalcites often record a polyphase history, revealed by different generations of cements
81 and sediment infill, which can be highlighted by color changes, and bimodal grain distribution
82 (Abbate et al. 1970; Bonatti et al. 1974; Bernoulli & Weissert 1985; Lemoine et al. 1987;
83 Früh-Green et al. 1990).

84 Because of the wide variety of ophicalcites and their occurrence in different oceanic or
85 continental margin settings, it is important to recall that the term ophicalcite does not refer to
86 a genetic process, but to a generic rock-type. Among the processes invoked for their
87 formation one group refers to endogenic evolution, involving various deep seated phenomena
88 such as: (1) mantle originated gas seeps (Bonatti et al. 1974; Haggerty 1991; Kelemen et al.
89 2004); (2) magmatic intrusions (Cornelius 1912; Bailey & McCallien 1960); (3) contact and
90 regional metamorphism (Peters 1965; Trommsdorff et al. 1980) and (4) hydrothermal fluid
91 interactions (Cornelius 1912; Lavoie & Cousineau 1995; Artemyev & Zaykov 2010). Another
92 group refers to surficial processes involving mechanical mixing of carbonates and ultramafic
93 rocks through tectonic crushing, sedimentary reworking and gravity-driven infilling of veins
94 and fractures (Bortolotti & Passerini 1970; Knipper 1978; Bernoulli & Weissert 1985; Früh-
95 Green et al. 1990; Treves & Harper 1994; Treves et al. 1995; Knipper & Sharas'kin 1998).

96 In the French Pyrenees, ophicalcites have been reported in some ultramafic bodies associated
97 with mid-Cretaceous basins and recently re-interpreted as resulting from mantle exhumation
98 during Mid-Cretaceous rifting (Lagabrielle & Bodinier 2008; Lagabrielle et al. 2010; Jammes
99 et al. 2009; Clerc et al. 2012). In order to document and to characterize the variable typology
100 of ophicalcites and to decipher their origin with respect to exhumation processes, we have
101 performed a comprehensive field, petrological and geochemical study of samples taken
102 throughout the Pyrenees (fig. 1). We identify three types of ophicalcites related to three

103 distinct genetic processes: i) pervasive ophicalcite resulting from replacement of ultramafic
104 minerals due to relatively deep and hot hydrothermal activity, ii) ophicalcites as veins
105 resulting from tectonic fracturing and cooler hydrothermal activity and iii) polymictic breccias
106 resulting from syn-sedimentary processes. The first two types record the activity associated
107 with the final emplacement of the peridotites, whereas the last one is associated with the final
108 exhumation of mantle rocks as portions of the basement of newly formed basins.

109

110 **II. Geological setting of the ultramafic Pyrenean bodies:**

111 The Pyrenees are a narrow, 400 km long continental fold and thrust belt resulting from the
112 collision of the northern edge of the Iberian Plate and the southern edge of the European Plate
113 during the late Cretaceous to Tertiary (Choukroune & ECORS Team 1989; Muñoz 1992;
114 Deramond et al. 1993; Roure & Combes 1998; Teixell 1998). Triassic and Jurassic aborted
115 rifting events predated the development of a major Cretaceous crustal thinning event, which
116 culminated in displacement between the Iberian and European plates (Puigdefàbregas & P.
117 Souquet 1986; Vergés & Garcia-Senz 2001). Continental rifting in the Pyrenean domain
118 occurred in response to the counterclockwise rotation of Iberia relative to Europe, coeval with
119 the onset of oceanic spreading in the Bay of Biscay between Chron M0 and A33o
120 (approximately 125-83 Ma) (Le Pichon et al. 1970; Choukroune & Mattauer 1978; Olivet
121 1996; Gong et al. 2008; Jammes et al. 2009). About forty metric- to kilometer-sized
122 fragments of subcontinental mantle rocks are found along the northern flank of the Pyrenees,
123 in the North Pyrenean Zone (NPZ). They reside within or next to numerous lozenge-shaped
124 basins flanking the North Pyrenean Fault (NPF). These basins are interpreted as the remnants
125 of isolated, pull-apart or transtensive half graben basins formed in response to the eastward
126 drift of Iberia along the NPF and later inverted during the Late Cretaceous-Early Cenozoic

127 Pyrenean orogeny (Le Pichon et al. 1970; Choukroune & Mattauer 1978). A typical flysch
128 sedimentation started during the mid-Albian within these basins (black flysch), which later
129 enlarged during the Late Albian and connected into one single, wider basin trough during the
130 Cenomanian (Debroas 1976; P. Souquet et al. 1985; Debroas 1990).

131 Various scenarios have been proposed for the emplacement of the ultramafic bodies, ranging
132 from purely tectonic mechanisms, such as solid intrusion of hot or cold mantle rocks into
133 sediments during strike-slip events (Avé-Lallemand 1967; Minnigh et al. 1980; Vielzeuf &
134 Kornprobst 1984), to tectono-sedimentary processes in which mantle rocks were exhumed
135 during Variscan time (Mattauer & Choukroune 1974; Fortane et al. 1986) and reworked in a
136 mid-Cretaceous wild flysch (Fortane et al. 1986). In recent re-examinations, various authors
137 propose that some of these bodies are fragments of sub-continental mantle basement partially
138 exhumed during Albian-Cenomanian times (Lagabrielle & Bodinier, 2008; Jammes et al.
139 2009; Lagabrielle et al. 2010; Debroas et al. 2010; Clerc et al. 2012). Within the NPZ, the
140 metasediments are locally strongly deformed and underwent a High Temperature - Low
141 Pressure (HT-LP) mid-Cretaceous metamorphic event, which lasted nearly 30 Ma from 110
142 Ma to 80 Ma (Azambre & Rossy 1976; Albarède & Michard-Vitrac 1978b; Montigny et al.
143 1986; Golberg et al. 1986; Goldberg & Maluski 1988; Thiébaud et al. 1988; Thiébaud et al.
144 1992). This metamorphism is considered as a consequence of crustal thinning (Golberg &
145 Leyreloup 1990) and developed in relation with hydrothermal circulations (Dauteuil & Ricou
146 1989). Hydrothermal circulations are also responsible for extensive albitization of some of the
147 North Pyrenean Massifs (Demange et al. 1999; Boulvais et al. 2007; Poujol et al. 2010), and
148 formation of massive talc deposits (Moine et al. 1989; Schärer et al. 1999; Boulvais et al.
149 2006) probably in relation to the activity of major ductile extensive shear-zones (Passchier
150 1984; St Blanquat et al. 1986; Costa & Maluski 1988; St Blanquat et al. 1990; St Blanquat
151 1993).

152

153 **III. Geology of the sampling sites**

154 The ophicalcites and related ultramafic breccias selected for the oxygen and carbon isotope
155 study were sampled from peridotite bodies exposed all along the NPZ (fig. 1). We selected
156 nine localities which are representative of the variety of the Pyrenean peridotites, and
157 presumably of the different geological processes involved in their exhumation history. Two
158 sampling sites are located in the western Pyrenees (Urdach and Tos de la Coustette), one in
159 the central Pyrenees (Moncaup) and nine in the Eastern Pyrenees (Ercé-Angladure, Lherz,
160 Fontête Rouge, Freychinède, Berqué, Vicdessos, Urs, Bestiac, Caussou).

161

162 **III.1. Western peridotites**

163 Several peridotite bodies outcrop in the Chaînons Béarnais, within the fold and thrust belt
164 Mesozoic sequence of the NPZ, at a longitude corresponding to the western termination of the
165 Paleozoic Pyrenean axial zone. The base of the stratigraphic sequence, exposed along the
166 Mail Arrouy, Sarrance and Layens post-Cenomanian thrusts (Casteras 1970), is composed of
167 Late Triassic evaporites, breccias and ophites overlain by Mesozoic platform carbonates
168 forming the original cover of the northern Iberian margin (Canérot et al. 1978; Canérot &
169 Delavaux 1986). In contrast to the Eastern NPZ, evidence of HT-LP metamorphism is
170 restricted here to some scarce and narrow regions bordering fault contacts with Triassic and
171 mantle rocks (Fortane et al. 1986; Thiébaud et al. 1992; Lagabrielle et al. 2010) and the
172 temperatures during peak metamorphism barely exceeded 400°C (Clerc 2012). Petrological
173 and geothermobarometric studies of the western ultramafic bodies show that they underwent a
174 two step exhumation with a first rise from 60 km to 25 km depth (1050-950°C), probably
175 during late Hercynian times, followed by a further step from 25 km to a shallower and cooler

176 (600°C) level (Fabriès et al., 1998). This second step is marked by the development of a
177 mylonitic fabric, from 117 Ma to 109 Ma (Vissers et al. 1997; Fabriès et al. 1998).

178 The Urdach body is a 1.5 km wide peridotite slice exposed at the western termination
179 of the Mail Arrouy thrust (fig. 2A). It is overlain by Paleozoic basement slices and surrounded
180 on its western and southern sides by a large volume of unsorted sedimentary breccias and
181 olistoliths composed of peridotite fragments associated with Paleozoic basement clasts from
182 upper, middle and lower crustal levels. These debris are intermingled in the Cenomanian
183 black flysch (Casteras 1970; Vielzeuf 1984; Souquet et al. 1985; Jammes et al. 2009; Debroas
184 et al. 2010). Some authors considered that the Urdach body itself might be an olistolith settled
185 in Cenomanian sediments (Duée et al. 1984; Fortane et al. 1986). Peridotite hydrothermal
186 alteration led to pervasive serpentinization reaching 80%. This is a dominant character of the
187 Urdach body (Fabriès et al. 1998).

188 The 400 m long Tos de la Coustette ultramafic body is located 3 km west of the
189 Sarailié summit, at the western termination of the Sarrance anticline (fig 2a). Apart from the
190 peridotites, the faulted heart of the Sarrance anticline includes, Paleozoic basement rocks and
191 ophite lenses embedded within cataclastic Triassic sediments. It is thrust over the
192 verticalized Urgonian limestones and Albian flysch of the Lourdios Syncline (Casteras 1970;
193 Lagabrielle et al., 2010). The Tos de la Coustette body itself is in tectonic contact with small
194 lenses of Paleozoic crustal rocks and Triassic metaevaporites outcropping both above and
195 beneath the peridotites. Like the Sarailié peridotites, the environment of the Tos de la
196 Coustette body is devoid of sedimentary breccias; instead these bodies are entirely surrounded
197 by cataclastic breccias limited by tectonic contacts and are thought never to have been
198 exposed to the seafloor (Canérot & Delavaux 1986; Lagabrielle et al. 2010).

199

200 **III.2. Central peridotites**

201 The Moncaup ultramafic body is part of a group of peridotites exposures lying around the
202 Milhas massif, in the central Pyrenees (fig. 1). They are associated with basement rocks,
203 variably brecciated Triassic sediments, ophites and Albian mafic intrusions. They are overlain
204 in tectonic contact by highly metamorphosed Mesozoic marbles (Debeaux & Thiébaud 1958;
205 Hervouët et al. 1987; Barrère et al. 1984). Although the peridotites have risen to near surface
206 levels, there is no evidence for sedimentary reworking indicating their exhumation on the
207 basin floor. Indeed, based on their geological setting, it can be deduced that the mantle rocks
208 have remained capped by the Mesozoic marbles together with small slices of continental
209 crust, during their uplift along the detachment fault (Lagabrielle et al. 2010).

210

211 **III.3. Eastern peridotites:**

212 The eastern Pyrenean peridotites are found within narrow belts of Mesozoic sediments of the
213 NPZ, mainly limestones, pinched between the Axial Zone to the south and blocks of
214 Paleozoic crust to the North representing the continental basement of the NPZ (North
215 Pyrenean massifs) (fig. 1). Although the eastern Pyrenean mantle outcrops are often small and
216 disconnected from an original substratum, one can still observe, on a decametric scale, a
217 progressive transition from carbonate-free massive peridotites to high carbonate content
218 breccias with all intermediates. On the rim of the ultramafic bodies, the massive peridotites
219 are often crosscut by millimetric to centimetric calcite veins over the first few meters. In
220 addition, in many localities, the peridotites are reworked within sedimentary polymictic
221 breccias together with highly variable proportions of carbonate clasts. The amount of matrix
222 of these breccias increases toward the carbonated end-member.

223 Most of the ultramafic bodies sampled for this study, from West to East, namely Ercé-
224 Angladure, Lherz, Fontête Rouge, Freychinède, Berqué, Videssos, Urs, Bestiac and Caussou,
225 display geological settings consistent with an origin as olistoliths surrounded by polymictic
226 detritic formation (fig. 2B, 3). They are interpreted as sedimentary records of the exhumation
227 of the peridotites on the floor of the Cretaceous basins (Lagabrielle & Bodinier 2008; Clerc et
228 al. 2012). The ultramafic-bearing breccias show sedimentary features such as grain-sorting
229 and crossbeddings and can be found away from the main bodies, indicating that they have
230 been transported by sedimentary processes. Lagabrielle & Bodinier (2008) showed that the
231 polymictic ultramafic-carbonate clastic sediments have been emplaced into fissures opened
232 within the exhumed massive peridotites, in a way similar to OC2 sedimentary ophicalcites of
233 Lemoine et al. (1987). Similar features are reported in more detail, together with the presence
234 of ultramafic-rich debris flows and evidence of ultramafic rock-fall in the vicinity of the
235 Lherz body by Clerc et al. (2012). The peridotites show little serpentinization, developed
236 mainly along discrete, localized joints and fissures. The carbonates reworked in the detritic
237 formations surrounding the peridotites are strongly deformed and underwent HT-LP
238 metamorphism with peak temperatures commonly as high as 600°C (Golberg & Leyreloup,
239 1990; Clerc 2012). By contrast to the western peridotites, the eastern ones underwent a single
240 and rapid uplift event, which probably limited hydrothermal alteration and serpentinization
241 (Albarède & Michard-Vitrac 1978a; Fabriès et al. 1991; Henry et al. 1998).

242

243 **IV. Description of the analyzed samples**

244 **IV.1. Sampling strategy and collected samples**

245 The sampling strategy was to collect samples from the four main geological environments
246 identified and distinguished as follows: (1) poorly serpentinized peridotites surrounded by hot

247 metasediments (Eastern and central peridotites) either exhumed to the basin floor (Lherz,
248 Bestiac, Caussou, Vicdessos, Urs, Ercé-Angladure) or only unroofed but never exhumed
249 (Moncaup), and (2) highly serpentinized peridotites surrounded by cooler sediments (Western
250 peridotites) either exhumed to the basin floor (Urdach) or only unroofed but never exhumed
251 (Tos de la Coustette). The list of the 48 studied samples is given in Table 1. We focused this
252 study on the Lherz and Urdach bodies since they are among the largest mantle outcrops in the
253 Pyrenees and because they represent two well-studied end-members in terms of their
254 geological environment. Furthermore, these two localities offer better outcrop conditions
255 compared with the smaller bodies poorly exposed in areas presenting important vegetal
256 covering and rock alteration.

257 **IV.2. Field and macroscopic aspects:**

258 IV.2.a. Western Pyrenean Ophicalcites:

259 The western ophicalcites appear essentially as veins of calcite infilling fissures and fractures
260 opened within the ultramafic rocks. The fractures present relatively constant and repeated
261 orientations (fig. 4A). Particularly well observable in a quarry opened on the western side of
262 the Urdach lherzolite body, they were first described by Monchoux (1970) and later
263 interpreted as typical ophicalcite textures by Jammes et al. (2009). These authors highlighted
264 their similarities with structures observed within exhumed mantle in the Alps and drilled off
265 Iberia (Manatschal 2004). They consist mainly in millimetric to decimetric veins of clear
266 white calcite (fig. 4B). The thickest veins are actually constituted of an accumulation of
267 numerous veins and veinlets separated by thin fragments of peridotite strapped from the rims.

268 At Tos de la Coustette, the ophicalcites also appear as a dense mesh of very fine veinlets and
269 as a pervasive substitution of serpentine minerals by patches of carbonate, barely visible on a

270 macroscopic scale, invading highly serpentized peridotites (fig. 4C and D). Similar textures
271 have also been described further east in the Avezac-Moncaut peridotites (Fabriès et al. 1998).

272 IV.2.b. Central Pyrenean Ophicalcites:

273 At least two types of ophicalcites were identified in the Moncaup peridotites. The first one,
274 observed close to the damage zone of the tectonic contact between the peridotites and the
275 overlying marbles, is represented by millimetric veins of coarse translucent sparite (fig. 4E).
276 The veins crosscut and hence post-date a mylonitic fabric affecting the peridotites. The
277 second one, observed in the damage zone of the detachment fault, consists of light brown
278 micro-conglomerates and micrite infilling veins and cavities opened in the altered and
279 dislocated peridotites (fig. 4F). The cavities present contorted and rounded rims. The micro-
280 sediments show complex multi-generation evolutions with successive stages of deposition
281 indicated by several color shades and crosscutting sparitic veins. The micro-conglomerates
282 are laminated and present clear grain-sorting.

283 IV.2.c. Eastern Pyrenean Ophicalcites:

284 In the Eastern Pyrenees, ophicalcites and polymictic ultramafic-marble bearing breccias are
285 observed within, close to, and even far away from the main peridotite bodies. Most of the
286 clasts are composed either of ultra-fresh subcontinental peridotites or of marbles bearing
287 mineral assemblages typical of the HT-LP mid-Cretaceous metamorphism. They are
288 associated with a minor proportion of fragments deriving from gabbros, Triassic ophites
289 Mesozoic meta-pelites and meta-evaporites and Paleozoic basement rocks (Lagabrielle &
290 Bodinier 2008; Clerc et al. 2012).

291 In the Etang de Lherz area, Lagabrielle and Bodinier (1998) identified four main types of
292 breccias and ophicalcites that can be extended to the other peridotite outcrops of the Eastern
293 Pyrenees. (i) Type 1 is found in direct contact with or within the ultramafic body, it consists

294 of a carapace of monomictic breccias resulting from the cataclastic deformation of the
295 peridotites during exhumation. These breccias typically lack carbonate clasts and contain little
296 to no carbonate veins and cement. Therefore, they will not be considered further in this study.
297 (ii) Type 2 breccias, generally found in close contact with type 1 breccias, are ultramafic-
298 dominated polymictic breccias resulting from the sedimentary reworking of type 1. (iii) Type
299 3 breccias consist of thin layers of graded ultramafic litharenites bearing isolated cm-sized
300 clasts of peridotites and marbles and presenting slumps and syn-sedimentary normal faults
301 (fig. 5A and B). Within the type 3 breccias, the peridotite clasts display many different
302 lithologies (lherzolite, harzburgite, websterite, pyroxenite etc.), variable mantle textures
303 (equant coarse-granular to mylonitic) and variable degrees of serpentinization (totally fresh to
304 fully serpentinized, fig. 5C and D). These observations point to mixing and transport from a
305 relatively distant source by sedimentary processes. Furthermore, clasts of former monomictic
306 carbonate breccias and polymictic UM-marble breccia are also reworked in these formations,
307 pointing to their late deposition with regard to the exhumation history (Clerc et al., 2012). (iv)
308 Type 4 breccias correspond to clastic rocks closely resembling the OC2 sedimentary
309 ophicalcites of Lemoine et al. (1987). Clear white calcite veins penetrate fractured ultramafic
310 blocks in which they separate angular fragments. There is a striking association of these veins
311 with matrix-supported microbreccias similar to those forming the matrix of the type 2 breccias
312 (fig. 5E). The veins are smoothly rooted in the matrix and seem to be its extension in narrow
313 domains where the clasts were too big to fit. However, some veins crosscut pre-existing
314 matrices, pointing to a contemporaneous formation of matrices and veins during
315 sedimentation accompanied by multi-stage circulations of cementing fluids. In some outcrops,
316 the ultramafic clasts exhibit a centimeter thick orange-brown oxidation ring on their contact
317 with carbonate matrices and veins (fig. 5 E and F). This feature is also commonly observed in
318 oceanic ophicalcites (Boschi et al. 2006; Dick et al. 2008). This oxidation pattern does not

319 appear on the rims of the thinnest veins.

320 Ophicalcites at the Bestiac locality show similar features and offer exceptional
321 conditions to investigate the successive generations of serpentinization and carbonation. The
322 peridotite consists of tens of lens-shaped bodies less than a few hundred meters in size and
323 embedded in a metamorphic bimodal ultramafic-marble breccia. Angular to slightly rounded
324 centimeter-sized clasts of lherzolite are found more than 500 m away from the main blocks, a
325 pattern clearly not consistent with any fault-assisted mode of emplacement. A few outstanding
326 outcrops are visible in several abandoned caves and galleries dug for prospection of asbestos.
327 Fresh to dark green serpentinites are crosscut by networks of cm-thick light green fibrous
328 serpentine (likely chrysotile) delimitating metric lumps which mimict at a meter-scale the
329 classical microscopic mesh-texture of serpentized peridotites (Wicks & Whittaker 1977)
330 (fig. 5G). The serpentine veins commonly show oblique sigmoidal fibers indicating shearing
331 contemporaneous to brittle deformation responsible for vein development. Pluri-millimetric
332 veins of calcite crosscut through this latest generation of serpentine veins that acted as a weak
333 gateway for sediments and fluids during the ultimate stage of deformation and sedimentary
334 reworking (fig. 5H). Some of the calcitic veins have a reddish coloration and increased
335 concentration of oxides, suggesting possible syn- or post-diagenetic hydrothermal circulation.
336 As in the Lherz area, these remarkable ultramafic-bearing formations have a relatively
337 restricted extension and appear within large volumes of clastic formations devoid of any
338 ultramafic component.

339

340 **IV.3. Microscopic description:**

341 Ophicalcite from the Urdach ultramafic body have millimetric veins of sparite. Several vein
342 generations crosscut each other with varying angles. The calcite veins commonly show crack-

343 seal aspects. Symmetric layers of calcite with varying intensity in transmitted light and
344 cathodoluminescence (CL), induced by minor variations of composition or inclusion
345 concentration, are separated by a central suture (fig. 6A). Some of the largest veins actually
346 consist of an accumulation of numerous sub-parallel veinlets, less than a micrometer wide,
347 invading the serpentinite (fig. 6B). Finally, some other veins show zoned botryoids of
348 fibrous radiating calcite (fig. 6C)

349 In the Tos de la Coustette opicalcites, the carbonates mainly appear as micrometric to
350 millimetric patches of calcite extensively dispersed within the serpentinite (fig. 6D). Intimate
351 repartition of calcite and serpentine indicate a pervasive calcification by replacement of some
352 serpentinous phases. The poorly elongated calcite aggregates develop following a general
353 foliation marked by magnetite alignments and thin yellow serpentine veinlets. Surface
354 estimate by digital image treatment indicates that the rock includes up to 55 % calcite, 40 %
355 serpentine and 5 % magnetite with minor phases.

356 The matrix of the eastern Pyrenean opicalcites and associated polymictic breccias consists of
357 a pale-orange litharenite composed of infra-millimeter-sized angular clasts of marbles mixed
358 with varyingly serpentinized ultramafic clasts and isolated minerals (pyroxenes, olivine, green
359 and brown spinels). This litharenite sometimes appears laminated and shows graded-bedding
360 due to sedimentary transport (Lagabrielle & Bodinier 2008; Clerc et al. 2012) (fig. 6E). In
361 contrast to the veins, the matrices of the breccias, show much less evidence for
362 recrystallization: there are, for example, a few newly formed metamorphic phyllosilicates and
363 amphiboles.

364 Most of the veins observed in the Eastern Pyrenean opicalcites consist of clear and equant
365 sparry calcites (fig. 6F). The veins cross cut alternatively the ultramafic clasts and the matrix
366 of the breccias in which they often seem to be rooted. In the simplest cases, a single

367 generation of calcite crystal nucleates from the rims of the veins and grows toward the center
368 where it joins in a central suture. In some cases, the growth resumed or the fracture was
369 reopened, leading to the formation of vugs. Most of the veins show multistep histories with
370 successive infillings of sparite particularly well highlighted by varying luminescence in CL
371 (fig. 6G). When pure enough, the veins are clearly recrystallized as evidenced by the
372 conservation of former zoned calcite dogteeth ghosts within bigger equant neoformed
373 crystals. The borders of the neoformed crystals are independent from those of the ghosts that
374 they overprint (fig. 6H). When thick enough, the veins are generally filled with detrital
375 material including micro-fragments of serpentine, oxides and calcite clasts mixed within a
376 micrite. Although they lack microfossils, such veins resemble the neptunian dykes and veins
377 formed on the subaquatic floor and consequently opened to sedimentary influx (Smart et al.
378 1987; Laznicka 1988; Winterer et al. 1991).

379 **V. Methods for determination of O and C isotope compositions**

380 Rock samples were sawed to select well-oriented and relevant planes. The sawed faces were
381 cleaned using water and pulsed with dry air before micro-drilling. A minimum of about 20 mg
382 of powder was collected for each sampling site.

383 The O and C isotope compositions were measured using a VG SIRA 10 triple collector mass
384 spectrometer at the University of Rennes 1, on the CO₂ released during reaction of calcite
385 with anhydrous H₃PO₄ in sealed vessels at 50°C (McCrea 1950). NBS 19 and internal-lab
386 standard references materials (Prolabo Rennes) were continuously measured during the course
387 of this work. NBS 19 measured values were $\delta^{18}\text{O} = 28.26 \pm 0.09$ (1 σ , n=12) ‰ and $\delta^{13}\text{C} =$
388 1.86 ± 0.02 (1 σ , n=12) ‰. Results were corrected in accordance with the NBS 19
389 recommended values of 28.65‰ and 1.95‰, for O and C respectively. The analytical
390 uncertainty is estimated at 0.15‰ and 0.1‰ for O and C.

391 VI. Results

392 The isotope compositions for the 48 analyzed samples are presented in table 1 and figure 7.
393 The calcite phase found in clasts, veins and matrices from the Pyrenean ophicalcites and
394 ultramafic-bearing breccias displays a wide range of oxygen isotope compositions with
395 minimum values of 12.6 and 13.8‰ (vs. SMOW) measured in Moncaup and Tos de la
396 Coustette veins (Western ophicalcites), and maximum values of 25.1‰ in the matrices of
397 samples from Etang de Lherz area (fig. 4 & 6; Eastern ophicalcites). The carbon isotope
398 compositions range from -5.8‰ (vs. PDB) in Moncaup samples to 1.5‰ in Lherz samples
399 (matrices). As a whole, the field of isotopic compositions of Pyrenean ophicalcites is
400 displaced from the one of the Iberian margin ophicalcites by lower $\delta^{18}\text{O}$ values and slightly
401 lower $\delta^{13}\text{C}$ values (arrow in figure 7). Whereas no clear distinction can be made when
402 comparing Pyrenean ophicalcites with Alpine and Apenninic ones, it seems that hydrothermal
403 ophicalcite worldwide compare well with ophicalcites from the Western Pyrenees. First order
404 analysis of the distribution of the oxygen and carbon isotope compositions implies to
405 distinguish two separate domains: i) Ophicalcites from the Urdach and Tos de la Coustette
406 ultramafic bodies have rather low and variable values of $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$, ranging from $\delta^{18}\text{O} =$
407 13.8 to 22.1‰ and $\delta^{13}\text{C} = -5.22$ to 1.12‰ and scattered around a mean value of $\delta^{18}\text{O} = 19.1$ ‰
408 and $\delta^{13}\text{C} = -1.0$ ‰. Ophicalcites from Tos de la Coustette body plot into a distinct field having
409 an extremely low value of $\delta^{18}\text{O}$. ii) Ophicalcites from the Eastern Pyrenean bodies display
410 low dispersion (21.3 to 25.1‰) in $\delta^{18}\text{O}$ but variable $\delta^{13}\text{C}$ values (1.53 to -2.47‰). Among the
411 Eastern Pyrenean ophicalcites, very poor discrimination can be made based on the
412 composition of veins and matrices since both display almost similar values with mean $\delta^{18}\text{O}$ of
413 23.4‰ in the veins and 24.1‰ in the matrices. The O and C isotope compositions of the nine
414 samples containing both veins and matrices are reported in figure 8. One can observe good
415 correlations between the compositions of matrices and veins with the exception of samples

416 LHZ8 and LHZ64 whose veins are significantly depleted in ^{18}O and slightly enriched in ^{13}C .
417 We note that the pluri-millimetric size of our sampling drilling spots provide bulk estimates of
418 the isotopic compositions of the veins and matrix but do not allow the complex multistage
419 history recorded in some of the veins to be deciphered (i.e. fig. 6G). The isotope composition
420 of the matrices does not correlate with the variable lithology of the clasts (either ultramafic
421 and/or carbonate), nor with the relative amount of clasts. In contrast the isotopic compositions
422 of the marble clasts are highly variable and plot within a larger field than the matrices and
423 veins (Fig. 7). Also, there is no correlation between the isotopic compositions of clasts and the
424 veins or matrices that host them. Actually, these clasts underwent a strong HT/LP
425 metamorphism and may contain abundant silicates (phyllosilicates, amphiboles, scapolite).
426 Closed-system isotopic equilibration between the carbonate and the silicate phases likely
427 introduced variable isotopic alteration of the carbonate phase, depending on the initial amount
428 of detrital silicates in the sedimentary precursor (see for example Valley, 1986; Boulvais et
429 al., 2000). Also, open-system alteration during syn-metamorphic infiltration possibly caused
430 isotopic shifts, which remain difficult to estimate here because we have no more information
431 on the initial geometry of the clast (for example the distance to a lithological discontinuity).
432 The ophicalcites from the Moncaup body display two distinct generations of carbonates with
433 distinct isotopic compositions (fig. 4C and D). Such differences in isotopic compositions are
434 consistent with the occurrence of two types of textures as described in section IV.

435

436 **VII. Discussion**

437 **VII.1. Origin of the various types of ophicalcites**

438 Based on the petrographic descriptions on the one hand and on the stable isotope
439 compositions on the other, we are able to distinguish three main categories of ophicalcites
440 associated with the subcontinental mantle bodies of the northern Pyrenees (table 2).

441 The first type of ophicalcites or *hydrothermal type*, as defined in the Tos de la Coustette body
442 results from peridotite carbonation by veins and pervasive substitution of the serpentinite
443 minerals by low $\delta^{18}\text{O}$ calcite. The low $\delta^{18}\text{O}$ values of calcite indicate that carbonation
444 occurred from rather hot fluids. Comparable $\delta^{18}\text{O}$ values have been measured in ophicalcites
445 formed in oceanic and ophiolitic hydrothermal systems (Lavoie & Cousineau 1995; Artemyev
446 & Zaykov 2010). Due to its low $\delta^{18}\text{O}$, the first generation of coarse crystalline calcite veins
447 described in the Moncaup peridotite likely corresponds to this type of hydrothermal
448 ophicalcite. The difference in texture types between the Moncaup and the Tos de la Coustette
449 ophicalcites may be explained by the very different rheology and chemical response to fluid
450 circulation of the unserpentinized peridotite at Moncaup compared with the totally
451 serpentinized peridotite of Tos de la Coustette.

452 The second type of ophicalcites or *tectonically-controlled type* is well characterized in the
453 Urdach body. It consists of massive serpentinized peridotites, indifferently lherzolite or
454 harzburgite, crosscut by successive generations of millimetric to centimetric calcite veins with
455 intermediate isotopic compositions. The tectonic control of calcite crystallization is
456 documented by the distribution of veins along preferential planes and their crack-seal
457 geometry. Such calcite crystallizations likely record the arrival of the peridotite close to
458 seafloor environments, directly under the influence of waters with intermediate temperature.
459 Different generations of veins cross-cutting each other with slight variations in isotopic
460 composition may reflect some temperature variations during the successive steps of fracturing
461 / precipitation. In sample URD 1, a first generation of vein with a lower $\delta^{18}\text{O}$ values
462 ($\delta^{18}\text{O}=18.6\text{‰}$) is cut by a later generation with a higher $\delta^{18}\text{O}$ value ($\delta^{18}\text{O}=21.1\text{‰}$). This is

463 consistent with cooling during vein formations, in consequence of progressive exhumation,
464 provided that the isotopic composition of the invading fluid, and then its source, remained
465 constant throughout the history of this sample.

466 The third type of ophicalcites, dominant in the eastern Pyrenees, is *sedimentary ophicalcite*. It
467 consists of a cogenetic association of calcite vein and polymictic breccias. The matrix-
468 supported to clast-supported polymictic breccias are composed of variable proportions of
469 marbles and UM clastic material. Polymictic compositions and typical sedimentary features
470 such as grain-sorting and cross bedding indicate that these ophicalcites have a sedimentary
471 origin (Clerc et al., 2012). Their wide lithological variety, both in UM and metasedimentary
472 material, likely results from sedimentary transport and mixing. By analogy with neptunian
473 veins observed in other extensional settings (Winterer et al. 1991), the micrite-filled brittle
474 fractures have been interpreted as very late, near surface fracturing of the exposed ultramafic
475 basement (Lagabrielle & Auzende 1982; Morgan & Milliken 1996), possibly leading to
476 gravitational instabilities: slumping, slope failure, and landslides as described at ODP site 899
477 by Gibson et al. (1996). The sparite-filled veins and veinlets, either reworked in the breccias,
478 smoothly rooted in the matrix or crosscutting clasts and matrix reveal a multistage deposition
479 history, already implied by the presence of breccias clasts reworked in the breccias (Clerc et
480 al. 2012). Bernoulli & Weissert (1985) describe similar cogenetic and simultaneous sediment
481 infillings and calcitic cement precipitation in Alpine ophicalcites. Such fractures must have
482 allowed the circulation of sedimentary fluids or early diagenetic fluids in domains where
483 restricted dimension hindered the penetration of sedimentary material. The isotopic
484 compositions of matrices and veins (Fig. 8) show a good correlation in both the oxygen and
485 carbon systems, which confirm the idea that both features developed from the same reservoir.
486 Since the relationship is valuable on a rather large range of $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$, one can infer that

487 the system of vein + matrix development was not connected to an external reservoir which
488 would have produced veins with distinct compositions from the ones of the matrices.

489 Precipitation of acicular aragonite has commonly been correlated with warm water
490 temperatures, high Mg/Ca ratios, high salinity, and high carbonate concentrations, conditions
491 reached in the uppermost levels of serpentine seamounts (Haggerty 1987; Lagabriele et al.
492 1992). Similar thermal and chemical conditions are associated with fibrous, botryoidal calcite
493 occurrences (Folk 1974; Surour & Arafa 1997), and botryoidal calcite could also represent a
494 replacement texture of acicular radiating aragonite (Ross 1991). More equant and bladed
495 sparry calcite has been correlated with cooler, deeper marine or meteoric settings, typically
496 with low Mg, and carbonate concentrations (Folk 1974; Burton & Walter 1987). Instead,
497 geochemical data (trace element and isotopic signatures) from Iberian margin ophicalcites
498 indicate a seawater imprint at temperatures of 10-20°C, consistent with an early Cretaceous
499 seawater only slightly modified by interaction with serpentinized peridotite basement
500 (Milliken & Morgan 1996). For that reason, Morgan & Milliken (1996) suggested that the
501 temporal evolution in the carbonate phase and morphology, from precipitation of aragonite,
502 followed by fibrous, botryoidal calcite and finally to coarse, bladed sparry calcite may be
503 controlled primarily by fluid flow rates through the vein rather than by variations of the
504 chemical and thermal parameters (fig. 9).

505 The low temperature ophicalcites from the Moncaup body (MP84a, b, c) are peculiar in that
506 they have extremely low $\delta^{13}\text{C}$ (< -5.00 ‰), indicative of interaction with organic material.
507 Such carbon isotope compositions are commonly observed in karstic or calcrete precipitations
508 from continental/meteoric waters transiting through soils and vegetation. These peculiar
509 ophicalcites are located immediately below the cataclastic formations staking out the
510 detachment fault between the peridotites and the overlying marbles. Due to the solubility and
511 permeability contrast between these two formations, this interface is prone to concentrating

512 groundwater. However, we cannot specify if the formations observed are contemporaneous of
513 any very early exhumation of the mantle rocks to subaerial (onland) environments in the
514 Cretaceous or if they result from a later karstification and/or replacement of pre-existing
515 ophicalcites. Regardless the exact explanation, these values strongly differ from the rest of
516 our dataset by a clear shift toward lower $\delta^{13}\text{C}$ values, allowing the distinction to be made
517 between a surface-derived cement precipitated in karstic environments and sedimentary
518 ophicalcites deposited in subaqueous conditions.

519 **VII.2. Environmental conditions for the formation of Central and Eastern Pyrenean** 520 **ophicalcites**

521 The isotopic compositions of the Pyrenean ophicalcites fall into the same field as other
522 ophicalcites from the literature (fig. 7). Only the three samples from Moncaup MP84a, b, and
523 c are out of this range because of their low carbon composition indicative of interactions with
524 carbon from soils, as discussed above. Ophicalcites from the Alps and Apennine were
525 interpreted as having been formed by interaction with seawater moderately heated to 80 to
526 less than 200°C (Barbieri et al. 1979; Barrett & Friedrichsen 1989; Früh-Green et al. 1990;
527 Schwarzenbach 2011) or by sedimentary fluids later re-equilibrated during Alpine
528 metamorphism (Weissert & Bernoulli 1984; Barbieri et al. 1979).

529 Samples from the Eastern Pyrenees are clearly recognized as sedimentary ophicalcites by
530 their textures. Consistently, the Eastern Pyrenean ophicalcites have the highest $\delta^{18}\text{O}$ values
531 measured in our sample set, a feature which is indicative of low temperatures of precipitation
532 at near surface conditions. However, we notice a major difference in the oxygen isotope
533 composition with ophicalcites from the Iberian margin (Agrinier et al. 1988; Evans & Baltuck
534 1988; Agrinier et al. 1996; Milliken & Morgan 1996; Plas 1997; Skelton & Valley 2000).
535 Indeed, in the Iberian ophicalcites, which precipitated from low temperature seawater, the

536 $\delta^{18}\text{O}$ values of calcite are around 31‰ with $\delta^{13}\text{C}$ values varying between -1.7 and 2.2‰.
537 Instead, the Eastern Pyrenean sedimentary ophicalcites analyzed in our study have
538 significantly lower $\delta^{18}\text{O}$ values (around 24‰) with carbon isotope compositions ranging from
539 -2.5 to 1.4‰, so that even if the envelope of Pyrenean sedimentary ophicalcites mimic the
540 Iberian margin ones, it is displaced in the $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ space (grey arrow, fig. 7). At least,
541 three hypotheses can be proposed in order to explain these differences (fig. 10).

542 1. Lowering of the O and C isotope compositions could result from a metamorphic imprint
543 with introduction of externally-derived fluids. Isotopic exchanges between neofomed calcites
544 and the mineral silicates, mainly serpentinite, which form a significant portion of the detrital
545 material associated with the sedimentary ophicalcites, may lower the O isotope composition
546 of the calcite. The breccias and ophicalcites rework clasts of pre-rift material that already bear
547 signs of high-grade recrystallization during the regional HT/LP metamorphism, with the
548 development of scapolite and amphibole. The deposition of the breccias and ophicalcites
549 hence occurs after the peak of metamorphism. But the long-lasting mid-cretaceous thermal
550 anomaly is followed by a lower grade metamorphism that affects the Turonian-Senonian post-
551 rift sediments, with a maximum temperature near 350°C (Ternet et al., 1997; Clerc 2012).
552 This later and lower grade metamorphism may hence have affected the ophicalcites and
553 breccias presented in this study. However, the petrographical effect of metamorphism on
554 these rocks seems rather limited since the matrices show only little recrystallization.
555 Furthermore, we would expect that the oxygen isotope composition would be much more
556 variable depending on the fluid/rock ratio. This is the case for a metamorphic-driven
557 alteration of the isotopic signal as shown from the study of the Alpine ophicalcites (Fig. 7;
558 Weissert & Bernoulli 1984; Früh-Green et al. 1990). Also, one would have expected that the
559 veins show more constant composition instead of displaying delta values that correlate with
560 the values of matrices (Fig. 8).

561 2. The low O and C compositions may be the result of a hot diagenesis from marine porewater
562 during carbonation. This hypothesis, which implies active circulation of relatively hot fluids
563 in the boundary layer between the ultramafic basement and seawater, is consistent with the
564 high geothermal gradients known to characterize the basins of the North Pyrenean Zone
565 during the Albian-Cenomanian period (Dauteuil & Ricou 1989; Golberg & Leyreloup 1990).
566 The thermal gradients for the Albo-Cenomanian metamorphism can be higher than 100°C/km.
567 In such conditions, we may also consider that unconsolidated sediments still soaked with
568 seawater can be rapidly buried and heated to temperatures as high as 50-80°C. At such
569 temperatures, the calcite precipitated from seawater ($\delta^{18}\text{O} = 0\text{‰}$) would have a $\delta^{18}\text{O}$ value of
570 around 23‰ (considering the isotopic fractionation coefficient of Zheng, 2011), a value that
571 compares well with the data of the Eastern ophicalcites. Thermal gradients as high as 160-
572 180°C/km are known in present days, for instance in the Salton Sea geothermal field (Elders
573 et al., 1972; Muffler & White, 1969). Comparable environments can also be found on the top
574 of mantle exhumed in oceanic domains, where hydrothermal fields develop over areas several
575 square kilometerswide (around 2.5km² at the Rainbow hydrothermal site, German et al., 1996;
576 around 2km² at the Lost City hydrothermal site, Kelley et al., 2001, along the Mid-Atlantic
577 Ridge). In similar settings, in the ophiolites of East Liguria, Spooner and Fyfe (1973) describe
578 temperatures as high as 400°C for shallow depth of circa 300 m below the water/rock
579 interface.

580 3. As a last hypothesis, it may be that the Eastern Pyrenean sedimentary ophicalcites formed
581 in a low-temperature but endorheic environment, dominated by continental waters and
582 possibly disconnected from the ocean. Indeed, the oxygen composition measured here is
583 about 7‰ lower than the present-day marine Iberian ophicalcites, a difference consistent with
584 the difference between marine and unspecific waters with a continental affinity. Note first that
585 a continental environment is not precluded by the existence of marine fauna, which would

586 have been observed in sediments associated with opicalcites. The hypothesis of an endorheic
587 environment dominated by continental waters has to be questioned with respect to the
588 paleogeographic reconstructions of the Pyrenean realm during mid-Cretaceous times. These
589 reconstructions point to the existence of a V-shape opening oceanic domain, narrowing from
590 the Bay of Biscay toward the East where it propagates into the continental crust (Jammes et
591 al. 2009 and references therein). The opening of numerous transtensive basins of limited
592 extension in the central and eastern part of the pre-Pyrenean domain may have been such that
593 these basins were endorheic (Le Pichon et al. 1970; Choukroune & Mattauer 1978), partially
594 disconnected from a marine influence at the time of opicalcite development. This hypothesis
595 is consistent with the stratigraphy of the Albian sediments deposited in disconnected basins
596 separated by positive reliefs (Debroas, 1976, 1990; Souquet et al., 1985). Some of these
597 reliefs such as the future North Pyrenean massifs and the future Axial Zone were emerged, as
598 shown by the outline of the Cenomanian transgression and by evidence of cooling and
599 sedimentary reworking of crustal material (Filleaudeau et al., 2011). Such short wave-length
600 and high amplitude morphology likely resulted from the flexural response of the lithosphere
601 to the extreme crustal stretching due to the extensional Albian-Cenomanian tectonics along
602 the Pyrenean realm. In such conditions, we may envision that the area where mantle has been
603 exhumed was surrounded by subaerial catchments and, at that time possibly disconnected
604 from the sea. A possible present-day analog is represented by the Salton Sea basin, which is
605 an endorheic continental basin located ahead of the propagating oceanic spreading axis of the
606 Gulf of California. Circulations of continental waters within sediments are also described in
607 more opened environments, for example at the foot of the Aden Gulf margins (Lucazeau et al.
608 2010).

609 At this time, it is difficult to select between the three hypotheses even if the last one is the
610 simplest in term of the isotopic composition record. Additional informations like fluid

611 inclusion data is needed to strengthen this hypothesis. It remains clear that, regardless of the
612 exact explanation, sedimentary opicalcites in the Eastern Pyrenees are distinguishable from
613 those in the Central and in the Western Pyrenees.

614 **VII.3. Western and Eastern Pyrenean opicalcites: why are they so different?**

615 The three types of opicalcites identified in this study have to be considered within the frame
616 of the exhumation history of the Pyrenean peridotites presented in section III a and c and as
617 summarized in figure 11. We highlight a clear distinction between the Eastern and Western
618 Pyrenean isotope composition of opicalcites also evidenced by the different degrees of
619 serpentinization of the mantle that host them, by the temperatures of the metamorphic peak in
620 the surrounding metasediments (Choukroune & Seguret, 1973; Golberg & Leyreloup, 1990;
621 Ravier, 1959; Clerc 2012) and by the typologies of opicalcites (fig. 11). Following our
622 observations, and in accordance with phase stability of serpentine mineral (Andreani et al.
623 2007), it appears that the variable serpentinization degree of the Pyrenean peridotites can be
624 linked, primarily, to the thermal anomaly accompanying their exhumation. Since carbonation
625 postdated serpentinization, the degree of serpentinization appears as a key factor influencing
626 the development of opicalcites. Volume increase and rheological softening induced by
627 serpentinization tend to favor the development of numerous fractures, allowing an endogenic
628 precipitation of carbonates as observed in the Western opicalcites. In contrast, the less
629 serpentinized peridotites exposed in Moncaup and in the Eastern Pyrenees must have had a
630 different behavior during uprising to crustal levels. Their contrasting rheology with the
631 surrounding rocks implies that they were probably still massive and competent until
632 exhumation. This could explain the predominance of superficial opicalcites found in these
633 localities. The fact that the Eastern Pyrenean peridotites remained preserved from
634 hydrothermal circulation may explain their scarce serpentinization. In addition, such a lack of
635 fluid activity may also be responsible for the preservation of high temperature mineral

636 assemblage since heat was evacuated by convection. The reason of the limited access of fluids
637 to the exhuming peridotites is not yet understood. We could suggest either i) a blanketing
638 effect of the Mesozoic sedimentary cover that would inhibit water infiltration, or ii) fast
639 exhumation in a continental environment with limited amounts of water available for
640 hydrothermal circulations.

641

642 **Conclusion**

643 On the basis of close fieldwork, petrographic and geochemical considerations, we present the
644 first comprehensive review of the Pyrenean ophicalcites. Our results, in accordance with
645 published studies on worldwide occurrences of ophicalcites allowed us to distinguish and
646 characterize three main types of ophicalcite (table 2): (i) hydrothermal ophicalcites resulting
647 in low $\delta^{18}\text{O}$ calcite (13.8‰) pervasively replacing serpentinite; (ii) intermediate or syn-
648 tectonic ophicalcites developed along with brittle discontinuities in the serpentinitized mantle
649 rocks, with intermediate calcite isotope compositions ($\delta^{18}\text{O}$ around 20.0‰; $\delta^{13}\text{C}$ around -
650 1.06‰); (iii) sedimentary ophicalcites occurring as breccias and neptunian dykes, associated
651 with the circulation of syn-sedimentary fluids. The isotopic compositions for this sedimentary
652 type show the highest $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values of the set, consistent with the cold temperatures of
653 precipitation expected in a sedimentary environment. We note a non-linear distribution of the
654 different ophicalcite type along the Pyrenean range, with dominant endogenic ones in the
655 West and dominant exogenic ones in the East. Such a distribution is clearly linked to a
656 difference in serpentinitization degrees likely related to the different exhumation histories and
657 subsequent variable thermal anomalies.

658 We further investigated the possible origins of the fluid and temperatures at which the calcite
659 may have precipitated in both hydrothermal and sedimentary domains. We present three

660 possible explanations for the relatively low values of the sedimentary ophicalcites: i) a post-
661 sedimentary metamorphic imprint; ii) a hot diagenesis in relation to the high regional thermal
662 gradient; iii) sedimentation in an endorheic basin. This last hypothesis is consistent with the
663 paleogeographic reconstructions of isolated Albo-Cenomanian basins at the tip of a
664 propagating rift. Finally, we highlight a major difference between Eastern and Western
665 ophicalcites, linked primarily to the variable degree of serpentinization. Considering the
666 strong control of serpentinization on the rheology of mantle rocks we propose that the
667 formation of different ophicalcites types is controlled by the degree of serpentinization,
668 depending itself on the rate and modalities of exhumation of the subcontinental mantle during
669 extreme crustal stretching.

670

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672

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1102

1103 **Table Caption**

1104

1105 **Table 1:** C (vs. PDB) an O (vs. SMOW) isotopes compositions determined for the carbonate
1106 fraction of the veins, matrix and clasts from the Pyrenean ophicalcite.

1107 **Table 2:** Schematic representation of the different types of ophicalcites analyzed in this study.

1108

1109 **Figure Caption**

1110

1111 Figure 1. Simplified geological map of the Northern Pyrenean belt with location of the
1112 peridotite bodies sampled in this study.

1113

1114 Figure 2. A: Simplified geological map of the Urdach and Tos de la Coustette in the Mail
1115 Arrouy and Sarrance *Chaînon Béarnais* with sample sites. B: Simplified geologic map of the
1116 Aulus basin presenting the extent of exposure of the peridotite bearing deposits surrounding
1117 the *Etang de Lherz* area with the location of the Freychinède, Fontête Rouge, and Berqué
1118 samples sites.

1119

1120 Figure 3: Photograph of an ultramafic olistolith in the Aulus basin illustrating the progressive
1121 transition from polymictic breccias to massive peridotite penetrated by calcitic veins

1122

1123 Figure 4: Macroscopic aspects of some of the western Pyrenean ophicalcites. A and B:
1124 development of calcites veins along tectonic discontinuities in the Urdach peridotite body. C:
1125 Mesh texture in highly serpentinized peridotite of Tos de la Coustette. D: Pervasive
1126 carbonation and veins in Tos de la Coustette peridotite. Calcite veins (E) and cavities
1127 infillings (F) in the Moncaup ultramafic body.

1128

1129 Figure 5: Macroscopic aspects of some of the eastern Pyrenean ophicalcites. A: Bimodal
1130 litharenite presenting slumps and syn-sedimentary normal faults from Lherz. B: Grain sorting

1131 in polymictic litharenites from Lherz. C and D: Breccia reworking fresh (orange to green) and
1132 serpentinized (dark green to black) peridotites in a calcitic matrix from the Lherz ophicalcites.
1133 E: Close association of matrix and veins in a typical ophicalcite from Vicdessos. F: Exposure
1134 of an ultramafic body presenting a centimetric orange-brown oxidation ring on the contact
1135 between peridotites and carbonates (Ercé-Angladure) G: Metric-sized mesh texture in the
1136 Bestiac peridotites. H: Detail of F showing calcite veins cross-cutting the latest serpentinite
1137 veins.

1138

1139 Figure 6: Microscopic aspects of the Pyrenean ophicalcites. A: Seal crack calcite vein from
1140 Urdach in cathodoluminescence (CL) and redrawn. B: Micrometric veinlets from Urdach. C:
1141 Botryoidal calcite in a vein from Urdach, in transmitted light and CL. D: Replacement texture
1142 of the ophicalcites from Tos de la Coustette, polarized light. F: Clear sparry calcite in veins
1143 from Lherz. G: Close vein/matrix association in transmitted light and CL. H: Dogtooth calcite
1144 ghosts in recrystallized veins, in polarized light and Redrawn.

1145

1146 Figure 7: $\delta^{13}\text{C}$ vs. $\text{d}18\text{O}$ diagram showing the isotopic compositions of the Pyrenean
1147 ophicalcites (veins, matrices and clasts). Shaded areas represent values from the literature for
1148 ophicalcites from the Iberian margin and Galicia bank (Evans & Baltuck 1988; Milliken &
1149 Morgan 1996; Plas 1997; Skelton & Valley 2000); the Alps and Apennines (Brotzu et al.
1150 1973; Barbieri et al. 1979; Weissert & Bernoulli 1984; Barrett & Friedrichsen 1989; Demeny
1151 et al. 2007) and from other hydrothermal ophicalcites (Lavoie & Cousineau 1995; Artemyev
1152 & Zaykov 2010).

1153

1154 Figure 8: Comparison of the C and O isotope compositions of calcitic veins and matrices in
1155 the ophicalcites from Eastern Pyrenees.

1156

1157 Figure 9: Comparison of calcite microtextures in veins and matrices from this study and from
1158 the Iberian margin (Morgan & Milliken, 1996).

1159

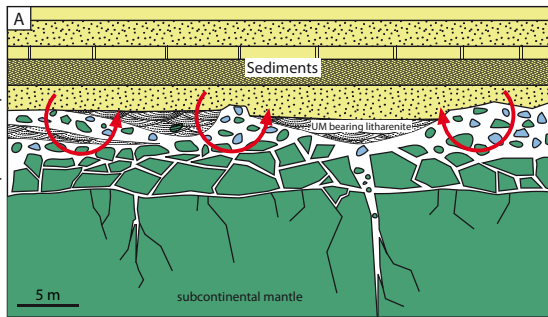
1160 Figure 10: Cartoons illustrating the three possible mechanisms responsible the low O
1161 composition of the sedimentary ophicalcites from the Eastern Pyrenees.

1162

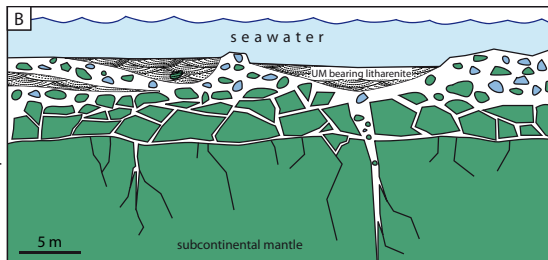
1163 Figure 11: Sketches presenting the exhumation history of the Eastern and Western Pyrenean
1164 peridotites in the light of our isotope study. Serpentinization processes are represented by
1165 green colors and the formation of ophicalcites by blue colors.

1166

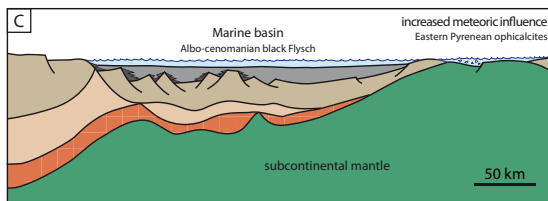
Post-sedimentary
metamorphic imprint



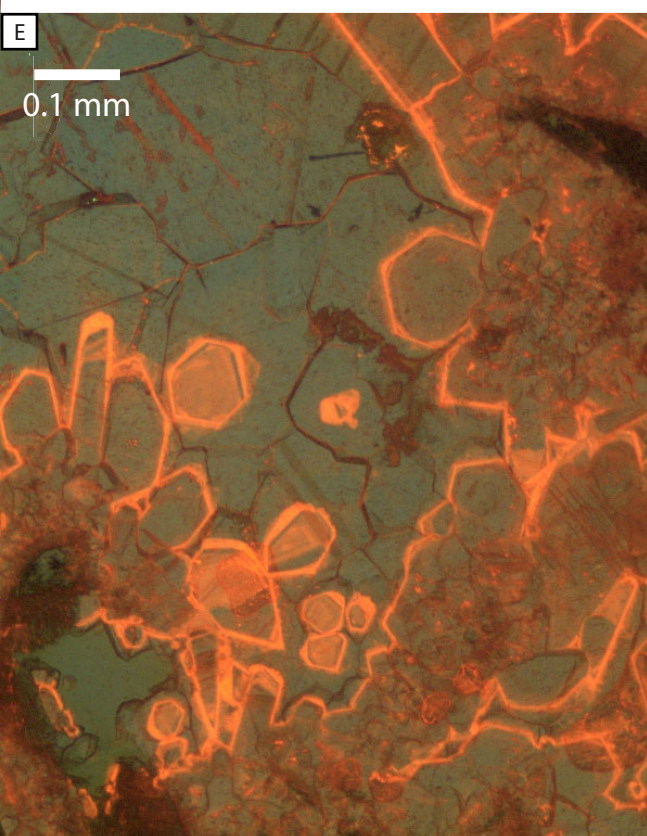
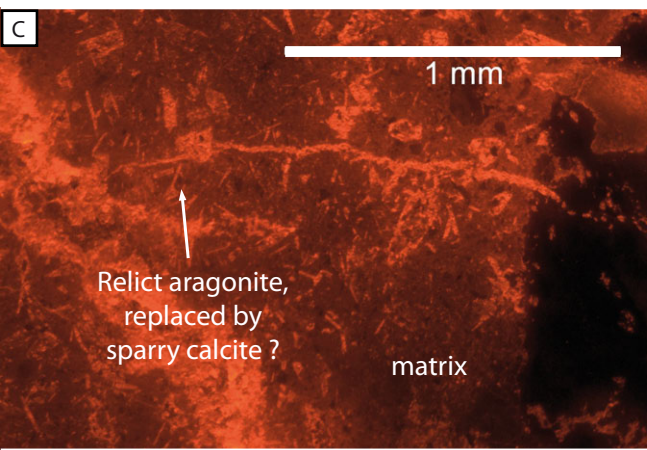
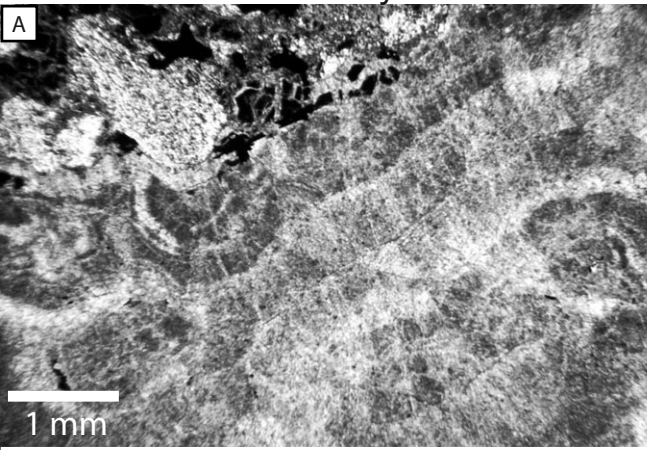
Precipitation from
hot pore-water



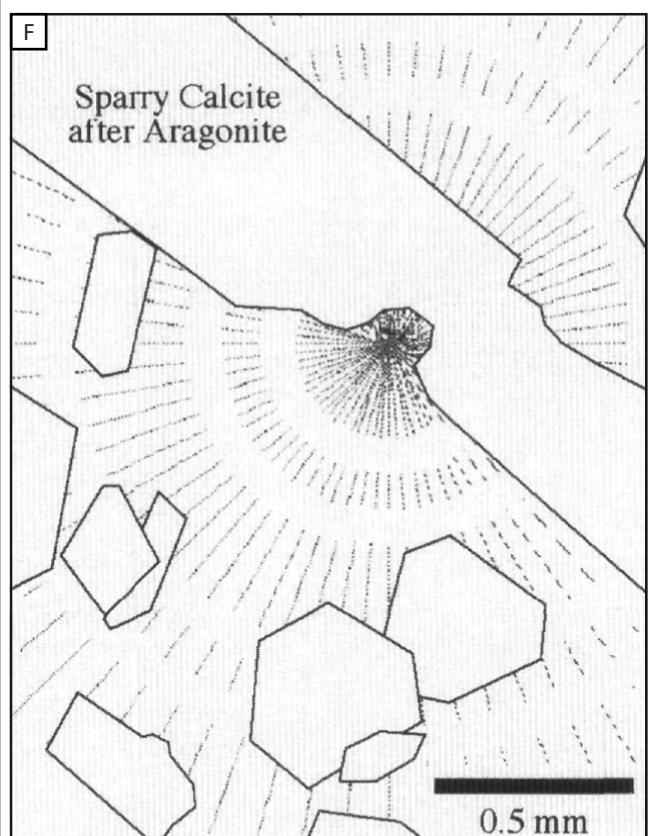
Precipitation from
continental waters

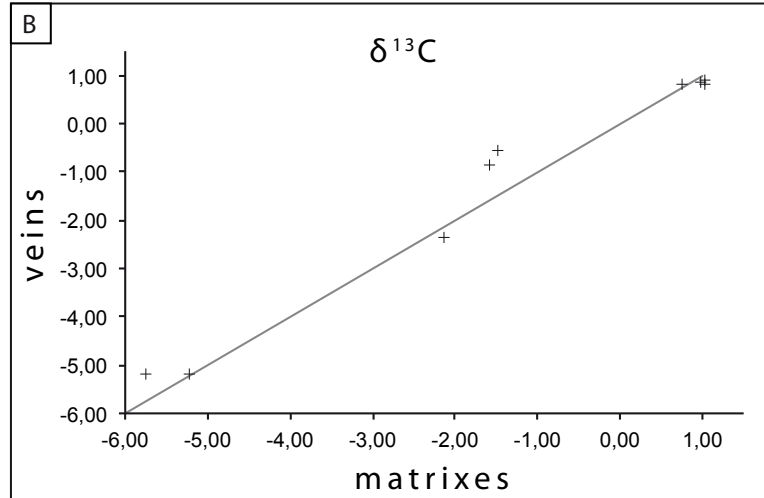
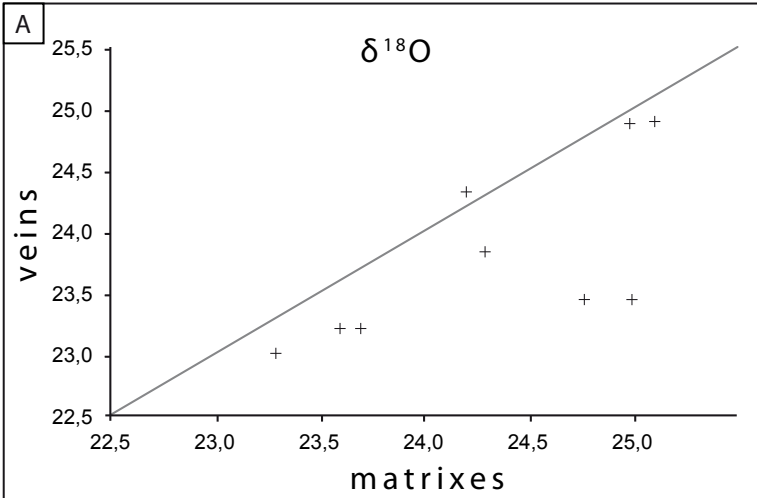


This study

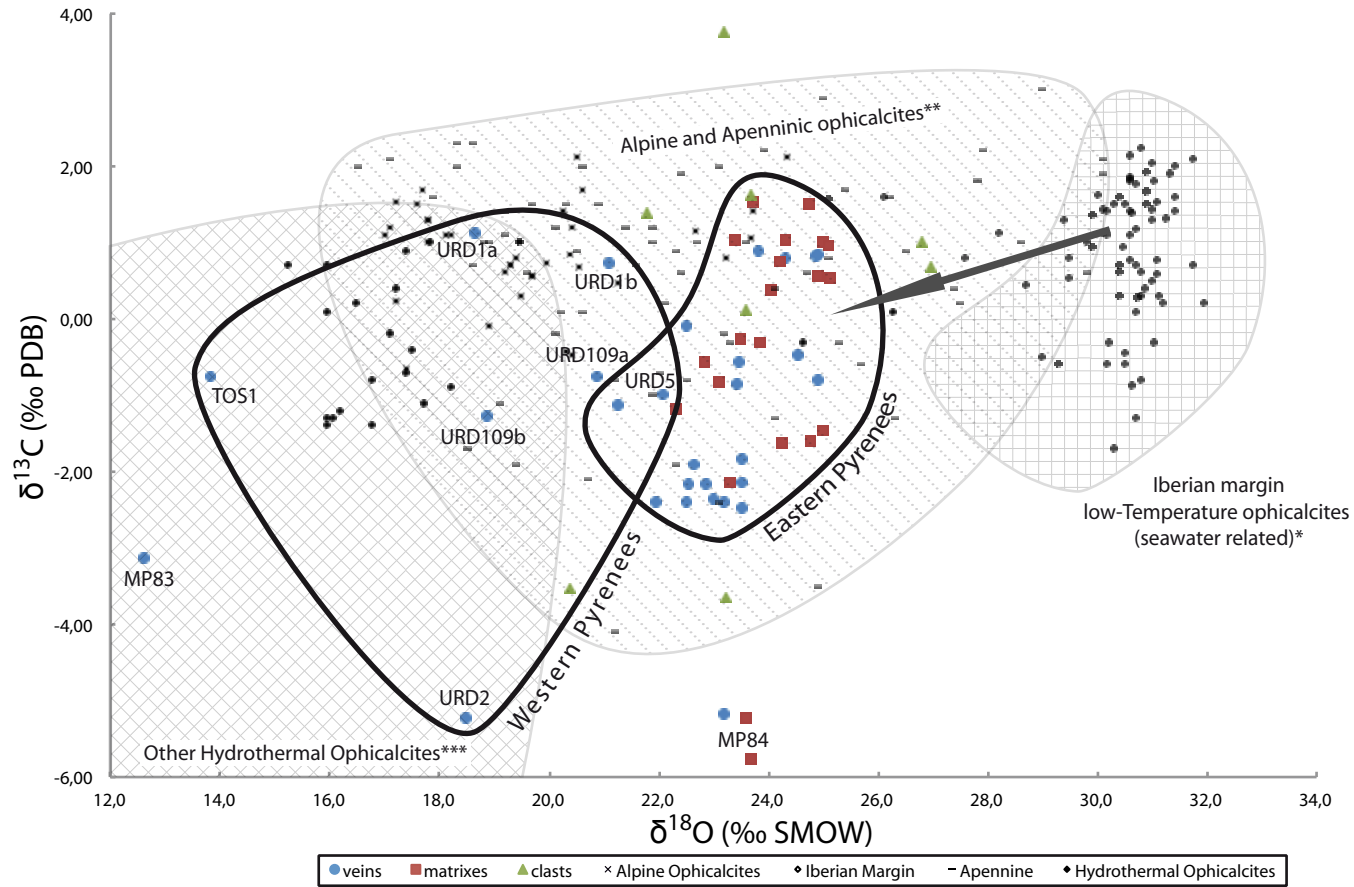


Morgan & Milliken (1996)

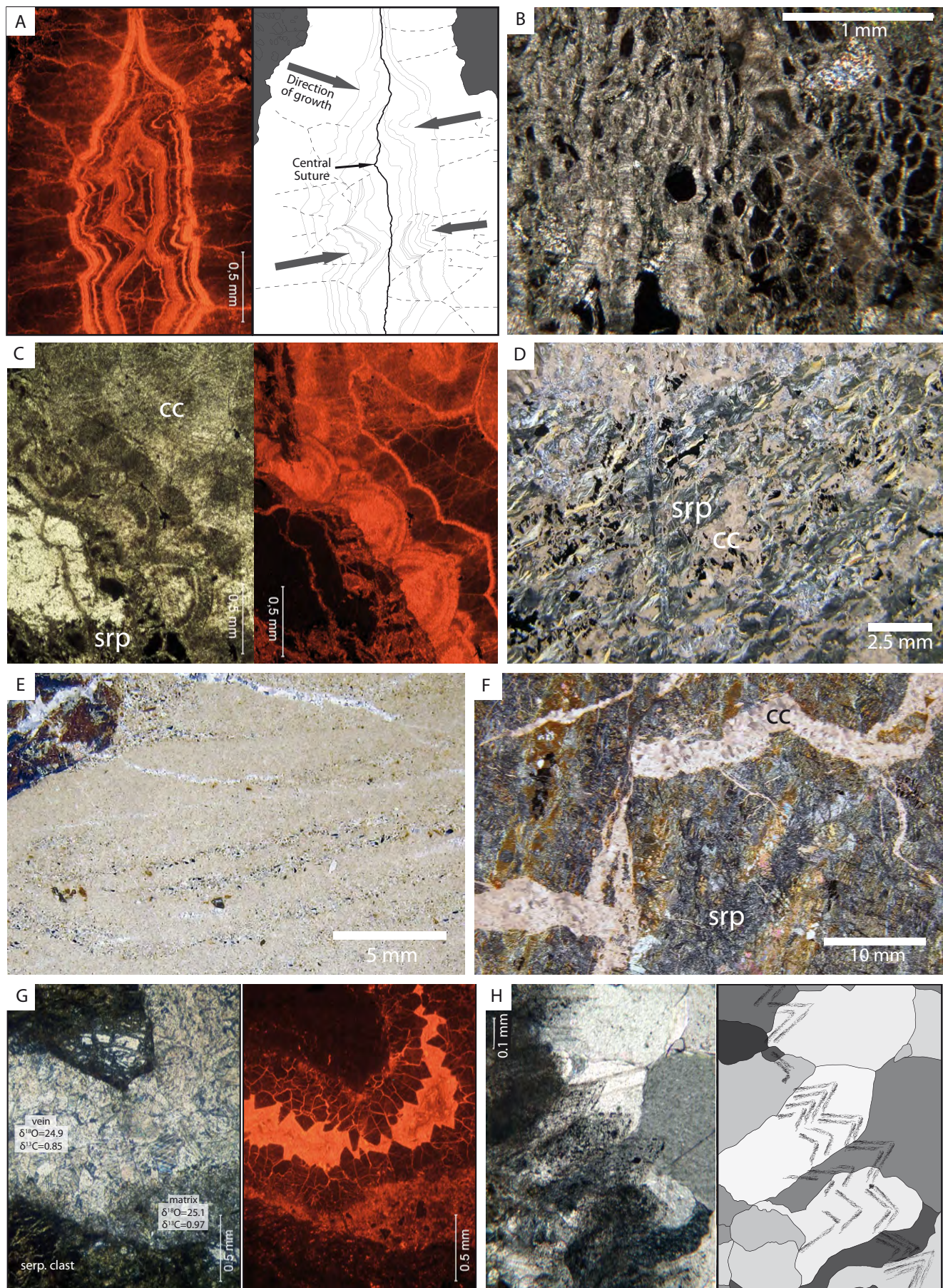




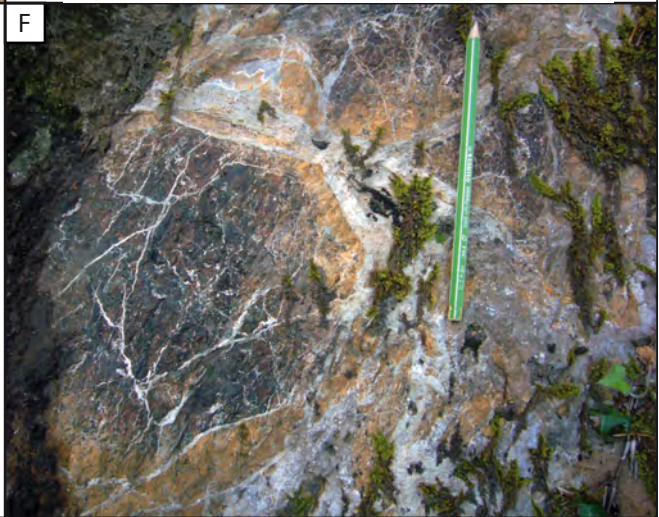
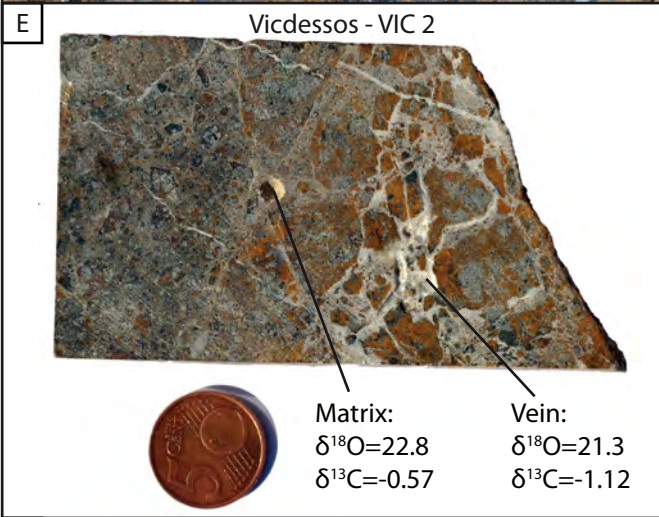
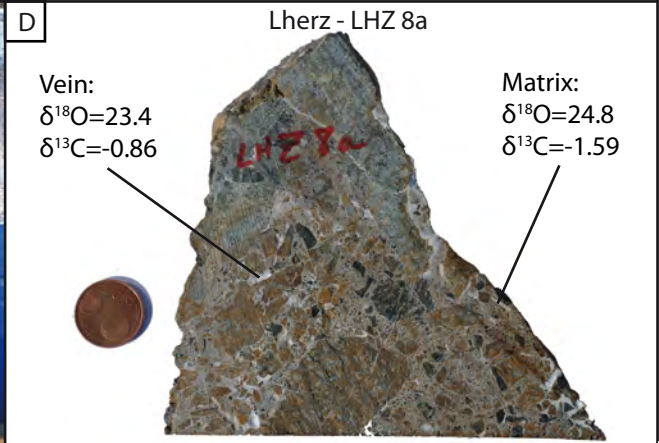
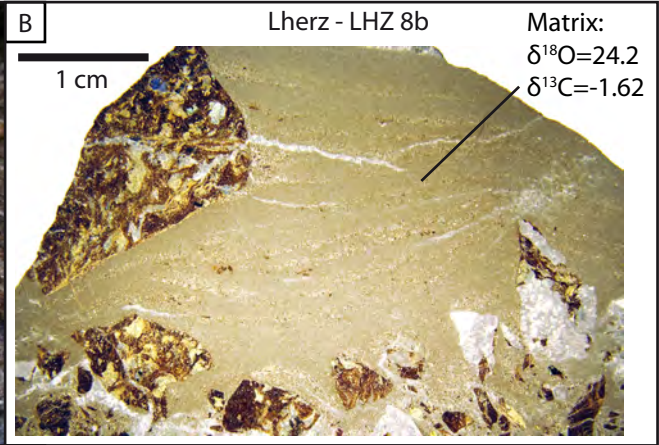
Clerc et al - IJES - Submitted - Figure 8



Clerc et al - IJES - Submitted - Figure 7



Clerc et al - IJES - Submitted - Figure 6



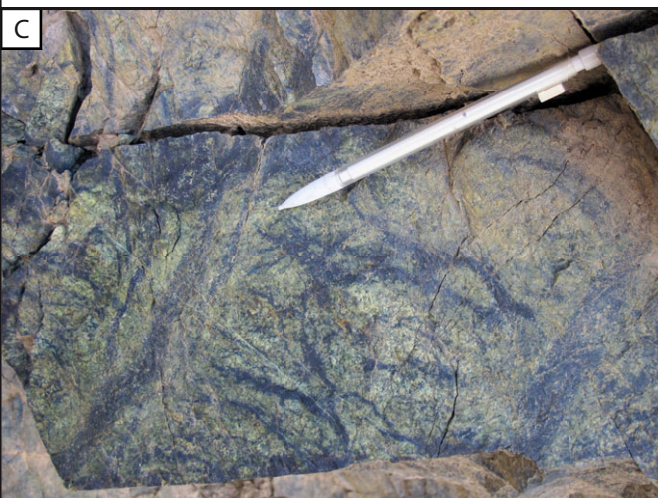
A**B**

Urdach - URD 1

Vein:
 $\delta^{18}\text{O}=21.1$
 $\delta^{13}\text{C}=0.73$

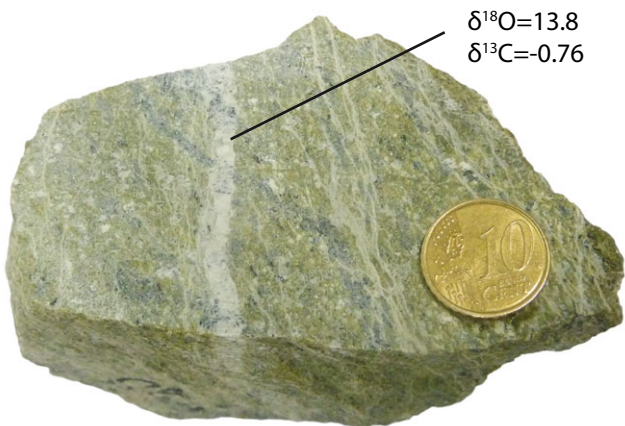


Vein:
 $\delta^{18}\text{O}=18.6$
 $\delta^{13}\text{C}=1.12$

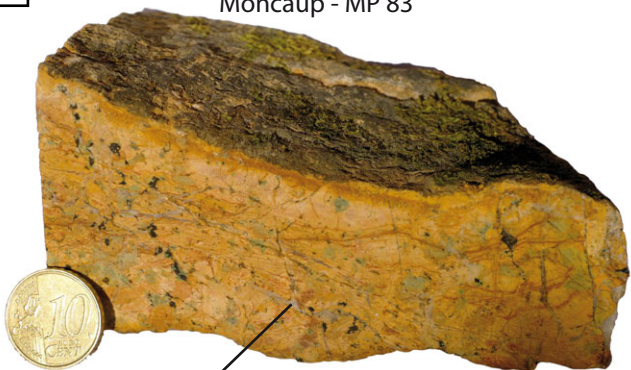
C**D**

Tos de la Coustette - TOS 1

Vein:
 $\delta^{18}\text{O}=13.8$
 $\delta^{13}\text{C}=-0.76$

**E**

Moncaup - MP 83



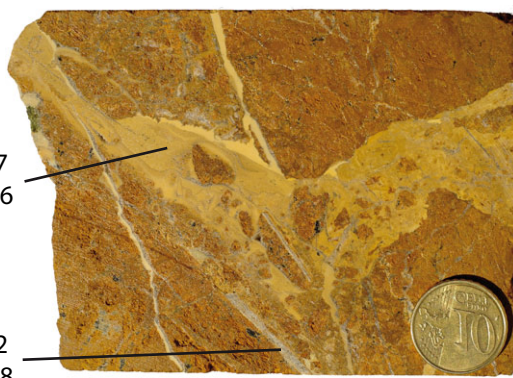
Vein:
 $\delta^{18}\text{O}=12.6$
 $\delta^{13}\text{C}=-3.12$

F

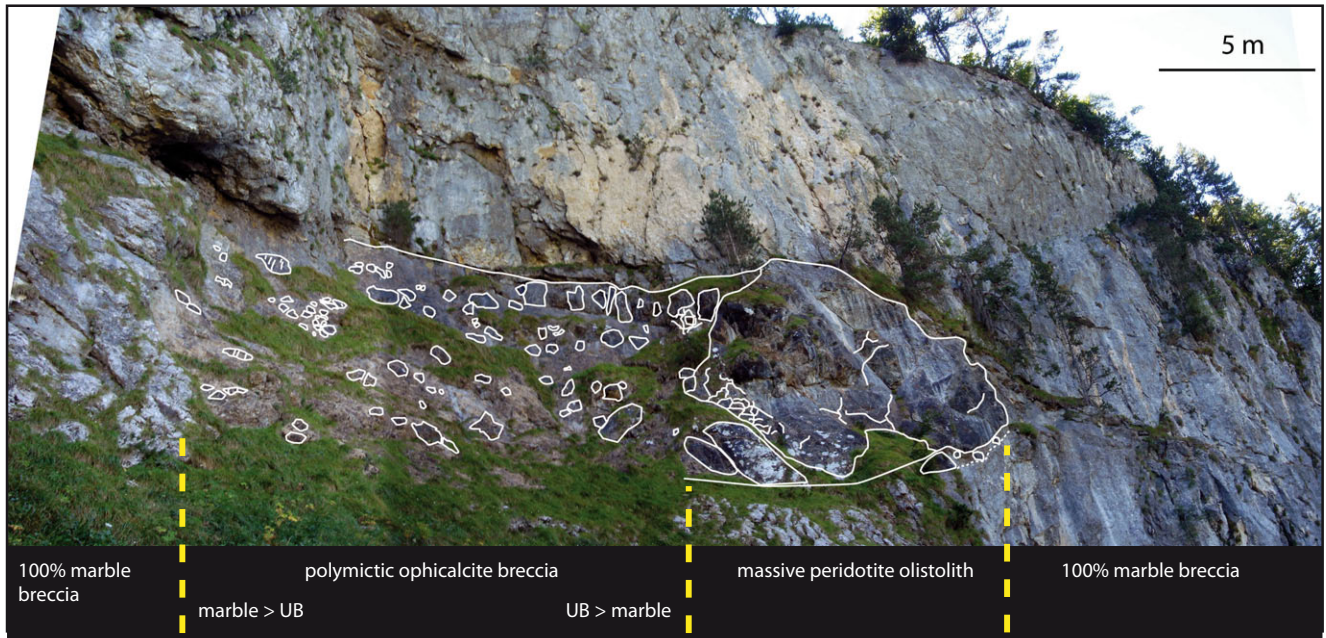
Moncaup - MP 84

Matrix:
 $\delta^{18}\text{O}=23.7$
 $\delta^{13}\text{C}=-5.76$

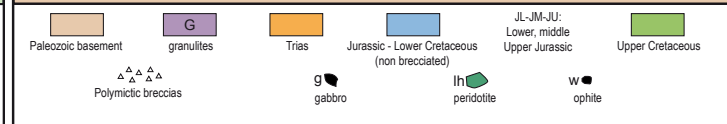
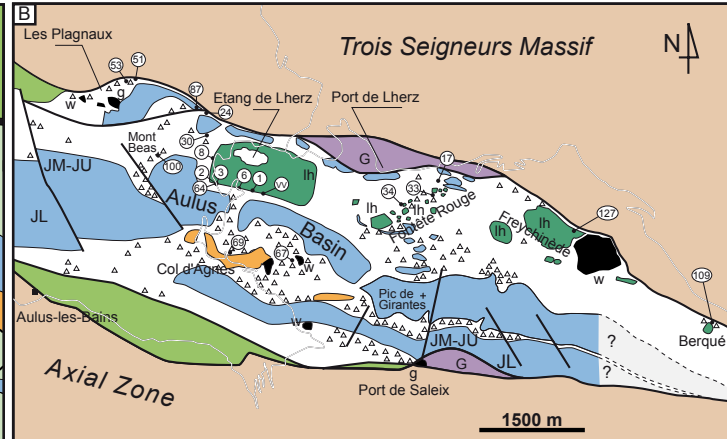
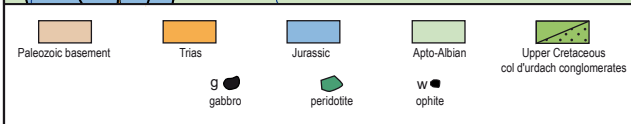
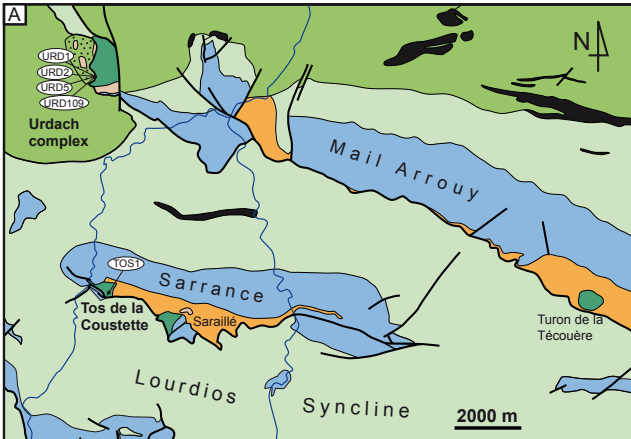
Vein:
 $\delta^{18}\text{O}=23.2$
 $\delta^{13}\text{C}=-5.18$



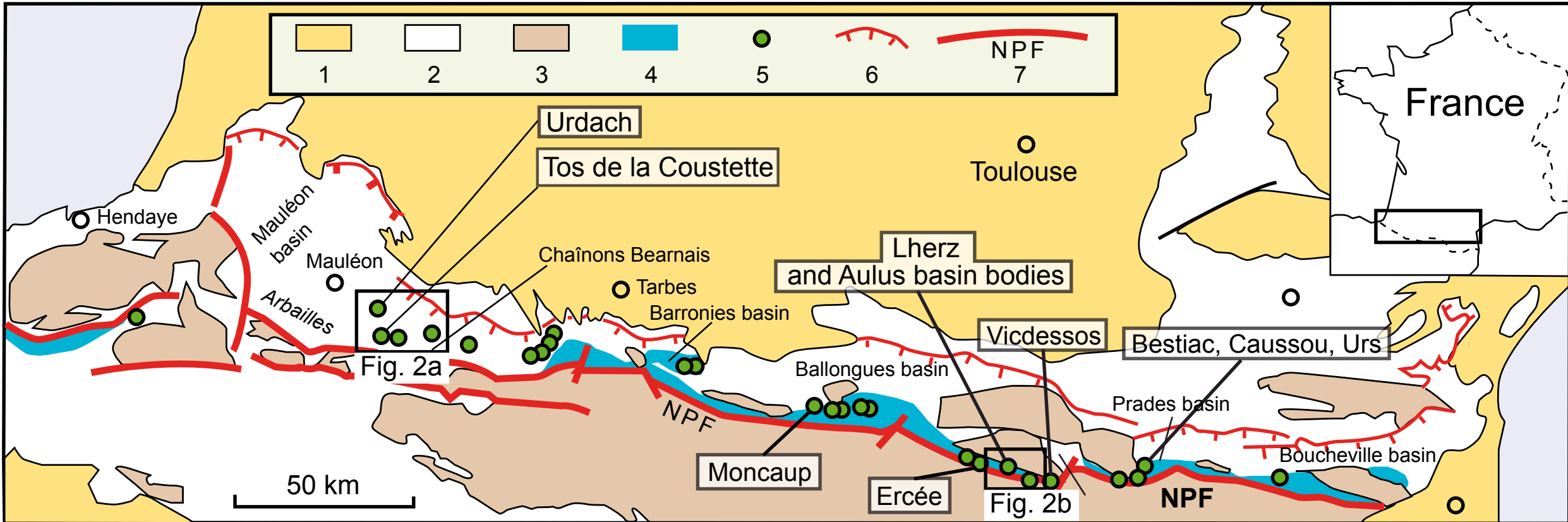
5 m



Clerc et al - IJES - Submitted - Figure 3

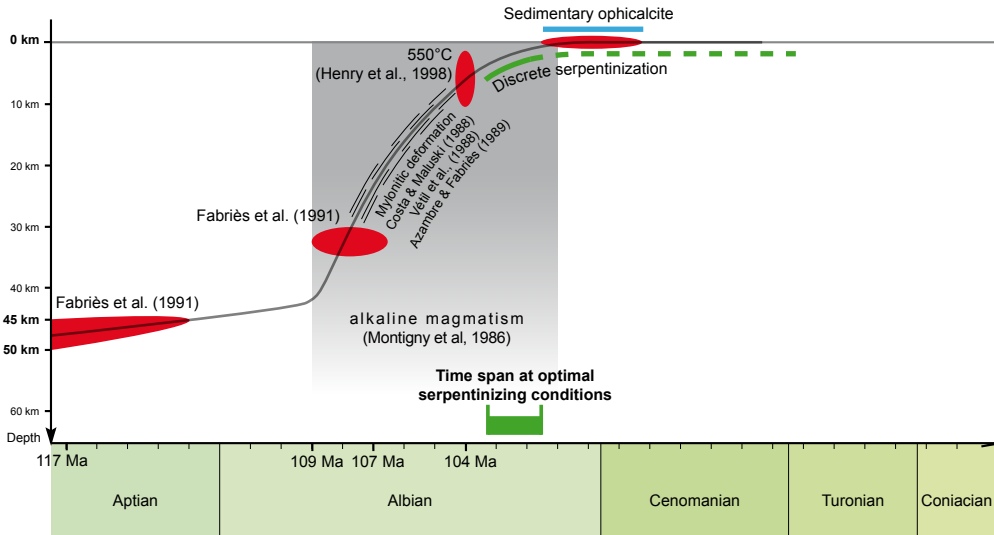


Clerc et al - IJES - Submitted - Figure 2



1, Oligocène and post-Oligocène; 2, Mesozoic and Eocene; 3, Paleozoic Basement; 4, area of HT-LP Pyrenean metamorphism; 5, peridotite; 6, main external thrusts; 7, North Pyrenean Fault (NPF)

EASTERN Peridotite



WESTERN Peridotite

