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# The post-Variscan tectonic-thermal activity in the southeastern metalliferous province of the French Massif Central revisited with K-Ar ages of illite

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1           **The post-Variscan tectonic-thermal activity in the southeastern**  
2           **metalliferous province of the French Massif Central**  
3           **revisited with K-Ar ages of illite**

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16  
17   **Keywords:** illite-type clays, southeastern French Massif Central, K-Ar dating, Pb-Zn metal  
18   district

19  
20  
21   **Abstract**

22  
23           This study deals with K-Ar data of illite and illite-rich mixed-layers of Cambrian and  
24   Permian metasediments that were subjected to tectono-thermal episodes in the Montdardier  
25   and Mas Lavayre ore districts of the southeastern Massif Central (France). The objective was  
26   to differentiate events that altered the metal-rich deposits from those that affected only barren  
27   host rocks close to the ores. On the basis of combined mineralogical analyses and isotopic  
28   determinations of the clay material, successive tectono-thermal events induced illitization at  
29    $288 \pm 10$ ,  $246 \pm 9$ ,  $197 \pm 6$ ,  $176 \pm 6$  and  $107 \pm 4$  Ma and probably at  $136 \pm 4$  Ma.

30 The lack of major geodynamic activities near the studied ore deposits during these  
31 times hypothesizes periodic long-distance migrations of fluids in the continental crust to  
32 explain their occurrences. Metals seem to have been concentrated at specific places, but  
33 apparently they were not deposited or altered during post-Visean tectono-thermal pulses  
34 recorded by the associated illite. The tectonic-thermal history of the Les Malines district  
35 confirms that repetitive geodynamic re-activations induced local heat transfers. However, the  
36 episode at  $246 \pm 9$  Ma apparently did not affect the earlier deposited ores, while that at 35-40  
37 Ma found in coffinite at Pierre Plantées was probably not induced by a thermal event, but by  
38 an alteration event that did not affect the K-Ar system of the surrounding clay material.

39

40

## 41 **Introduction**

42

43 Isotopic dating represents a valuable contribution to reconstruct the genetic evolution  
44 of sediment-hosted ore deposits. However, the gained information is not straightforward, as  
45 the recent analytical/technical improvements have not solved all uncertainties in ore isotopic  
46 dating. For instance, among the identified difficulties is the lack of the precise knowledge of  
47 the initial Pb isotopic compositions of metals (Romer, 2001), or the tendency of ores to easily  
48 recrystallize when environmental conditions change, which potentially can modify their  
49 chemical and isotopic compositions of the U-Pb system. Also, uranium minerals often contain  
50 large amounts of radiogenic Pb that allow a routine application of the U-Pb dating method;  
51 but their timing of crystallization is sometimes questionable because intermediate isotopes  
52 from uranium decay chain may escape from the host minerals exposed to subsurface  
53 conditions (e.g., Miller and Kulp, 1963; Holliger et al., 1989).

54 Due to these inherent difficulties, indirect isotopic dating of ore deposits has been  
55 explored in recent decades as a complementary approach to direct isotopic dating. This  
56 indirect dating consists of the study of barren minerals associated with the ore concentrations  
57 that record the same evolution (e.g., Clauer and Chaudhuri, 1992). Among barren minerals  
58 often associated with ore deposits in sediments and metasediments are clay minerals that can  
59 be considered potential age markers using basic isotopic methods (e.g., Ineson et al., 1975;  
60 Halliday and Mitchell, 1983). In fact, the information from such studies of barren minerals

61 can be extended beyond strict age comparison by evaluating the physical-chemical parameters  
62 that control the evolution of an entire ore district. Clay minerals have the advantage, which  
63 can sometimes also be a drawback, to be sensitive to discrete changes in the chemical and/or  
64 thermal conditions of a complex regional tectonic-thermal evolution (e.g., Clauer and  
65 Chaudhuri, 1995). Easily identified by appropriate methods including X-ray diffraction and  
66 electron microscopy, their study also has some inconveniences as it is sometimes difficult to  
67 separate authigenic from detrital clay crystals even by sophisticated separation methods. In  
68 fact, many studies of clay minerals from uranium deposits benefitted by such combined  
69 geochronological approaches (e.g., Lee and Brookins, 1978; Bell, 1985; Clauer et al., 1985;  
70 Bray et al., 1987; Brockamp et al., 1987; Turpin et al., 1991). Studies on stable and  
71 radioactive isotope compositions of clay minerals from uranium deposits have also  
72 highlighted how thermal and chemical information registered by such barren minerals can  
73 support and even improve our understanding of ore deposits (e.g., Halter et al., 1987; Wilson  
74 et al., 1987; Kotzer and Kyser, 1995; Kyser et al., 2000; Polito et al., 2006; Laverret et al.,  
75 2010). Since age discordances are often systematically reported in ore deposits and in  
76 associated barren host rocks, a detailed examination of the genetic relationship of both types  
77 of minerals appears justified and timely. In turn, the present study was designed to continue  
78 decrypting the tectonic-thermal evolution of a large ore region to investigate if all events  
79 recorded by the clay material from barren host rocks also occur in the ore deposits. The use of  
80 clay minerals for geochronological information of ore deposits emphasizes the following  
81 concern: since clay minerals potentially record any hydrothermal event (e.g., Velde, 1985),  
82 how can one unequivocally identify those interacting with the metal deposits in an ore district  
83 from those having no impact?

84 To identify the successive affects of such regional evolution on ore concentrations, the  
85 present review provides a compilation of K-Ar data of illite separates of Cambrian and  
86 Permian ore-hosting and barren rocks from southeastern French Massif Central. The attempt  
87 here is to answer the question whether the known ore concentrates had or had not an  
88 independent evolution relative to the surrounding regional host rocks.

89

## 90 **Geological setting and sampling**

91

92 In the southeastern French Massif Central, the Paleozoic basement hosts a major  
93 metalliferous province that produced about 2 million tons of Pb-Zn metals since the initiation  
94 of exploitation. Towards its northern area, the basement consists of late Variscan granites  
95 (Hamet and Mattauer, 1977; Vialette and Sabourdy, 1977), whereas the Causses-Shoal  
96 structural unit to the west and the south results from Triassic-to-Jurassic (Callovia) tectonic  
97 faulting, together with changes in the lithology and sedimentation rates of the cover sediments  
98 (e.g., Clauer et al., 1997). The whole district is located along the major Cévennes fault  
99 system, to the SE of the Massif, active as a normal fault, at least during the Liassic  
100 extensional period (Lemoine, 1984). The plutonic basement is overlain by Cambrian to  
101 Triassic dolomite-rich black shales and Bathonian dolostones followed by Oxfordian dolomite  
102 shales (Le Guen and Combes, 1988).

103 The genesis of the Pb-Zn and U deposits in this southeastern part of the Massif Central  
104 has been explained by various mineralizing processes, from a karstic (Orgeval, 1976; Connan  
105 and Orgeval, 1977; Verraes, 1983) to a syngenetic (Macquar, 1970; Michaud, 1970) and to a  
106 diagenetic type (Bernard, 1958; Fogliérini et al., 1980), as well as by interactions with  
107 migrating hydrothermal fluids (Charef and Sheppard, 1988; Ramboz, 1989; Le Guen et al.,  
108 1991), demonstrating in turn that there is no unanimity about their genesis. Based on Pb  
109 isotopes to trace and constrain the genesis of the Pb-Zn ores, Le Guen et al. (1991) considered  
110 that the Pb-Pb isotopic compositions of the different ore bodies were quite homogeneous,  
111 ruling out a major contribution of external Pb during the successive  
112 concentration/remobilization episodes. In fact, such a model confirms the hypothesis of an  
113 initial stock of Pb that evolved continuously in an almost closed system at a regional scale,  
114 without any significant supply of external Pb or loss of initial Pb.

115 Lancelot et al. (1995) reviewed the U-Pb and Pb-Pb data of three major regional U  
116 deposits: those of the southeastern Lodève and of the central Bertholène and Pierres Plantées.  
117 The results suggest a generalized precipitation of metallic ores during the Liassic (between  
118 195 and 175 Ma), and a common multi-stage evolution. Despite the diversity of the host  
119 rocks, such as Permian sediments above the unconformity with the Variscan basement at  
120 Lodève, the orthogneissic and mylonitic Variscan basement at Bertholène, and the episyenitic  
121 veins cutting post-tectonic Variscan leucogranites at Pierres Plantées, the U-Pb and Pb-Pb  
122 data point to a major Liassic U concentration induced by fluid circulation at temperatures

123 from 140 to 250°C and salinities from 5 to 14% NaCl eq. At Pierres Plantées, a pre-  
124 concentration was also identified at about 270 Ma in leucogranite veins percolated by fluids at  
125 300-350°C. At Bertholène, the U contents were dated at about 170 Ma as tiny spherules  
126 disseminated in altered Oligocene coffinite. Further U remobilization was detected at Lodève  
127 and Pierres Plantées during the Cretaceous, which was confirmed in the southeastern deposits  
128 by K-Ar dating of clays from gouges of the Saint-Julien fault that contain U minerals  
129 (Mendez Santizo et al., 1991).

130 Analyzed earlier by Vella (1989) and Mendez Santizo (1990) in their PhDs, the K-Ar  
131 data of varied illite-rich size fractions (<0.2, <0.4, 0.4-1, 0.4-2 and <2 µm), separated from 26  
132 Paleozoic samples from two southeastern locations from French Massif Central are combined  
133 and re-examined here (Fig. 1A to D; Table 1). In the northern part of the selected area, six  
134 Cambrian calcschists were collected in and next to the Montdardier mine about 3 km to the W  
135 of the Les Malines district (Fig. 1C), together with two more argillaceous dolostones taken in  
136 the neighborhood of the mine. In the second sector about 50 km to the SW, four Autunian  
137 samples were collected in the Mas Lavayre mine with five samples from a transect across the  
138 Saint-Julien fault visible in the mine at about 5 km to the SE of the Lodève district, and six  
139 core splits of the exploration drilling MLV located 1 km to the E of the Mas Lavayre mine  
140 (Fig. 1D). In addition to these clay-rich samples, five feldspar separates purified from  
141 interlayered pyroclastic to tuffaceous beds of Autunian sediments from Mas Lavayre mine  
142 were also purified, analyzed for their K-Ar data and the results are included into the  
143 discussion.

144

## 145 Analytical procedure

146

147 As the rock samples were quite indurated, gentle freeze-thaw disaggregation  
148 consisting of repetitive cycles of freezing at -25°C and heating at +25°C applied to 1cm<sup>3</sup> rock  
149 chips sealed in polyethylene bottles filled with de-ionized water (Liewig et al., 1987) proved  
150 to be insufficient. This method was replaced by hand crushing of whole-rock chips in an agate  
151 bowl and separating the <2 µm size fractions from rock powders by dispersion and  
152 sedimentation in de-ionized water and recovery by applying Stoke's law. The complementary  
153 size fractionations (<0.2, <0.4, 0.4-1 and 0.4-2 µm) were completed on the extracted <2 µm

154 fractions by ultracentrifugation. The size fractions were X-rayed (XRD) with determination of  
155 the illite crystallinity index (ICI) of the (001) illite peak following Kübler's (1968; 1997)  
156 concept and called now full width at medium height (FWMH). Theoretically, the FWMH  
157 (ICI) values below 0.25 define epizonal metamorphic conditions, those between 0.25 and 0.37  
158 define anchizonal metamorphic conditions and the higher values correspond to a diagenetic  
159 grade. The limits depend on the analytical conditions and the characteristics of the used XRD  
160 equipment (here operated at 40 KV/20 mA and equipped with a Cu anticathode and a Ni filter  
161 and slits of 1 and 2° for the tube and the counter, respectively). Special attention was given to  
162 identify the potential detrital minerals in the separated size fractions, such as micas and  
163 feldspars, as they potentially bias the isotopic ages of the authigenic minerals. The feldspar  
164 separates were purified by disaggregating the tuffaceous samples with the above freeze-thaw  
165 technique, sizing the slurries by wet-sewing, and purifying the fractions in dense liquids. The  
166 separated size fractions were X-rayed, which showed that they were almost never pure but  
167 always mixed with minute amounts of albite, quartz and muscovite. The two former minerals  
168 have no impact on the K-Ar data, the occurrence of the latter is of some concern, so that only  
169 the size fractions without any muscovite peak in the XRD patterns were analyzed.

170 The K-Ar determinations were obtained following the procedure of Bonhomme et al.  
171 (1975) that has been described in varied publications of the group. The powders of the  
172 different size fractions were preheated at 100°C during 12 hours under vacuum in the  
173 extraction line to remove any atmospheric Ar adsorbed on the clay particles during sample  
174 preparation, handling and size separation. The accuracy of the Ar extraction method was  
175 periodically controlled by analysis of the international GL-O standard mineral with the  
176 radiogenic  $^{40}\text{Ar}$  content averaging  $24.54 \pm 0.15 \times 10^{-6} \text{ cm}^3$  ( $2\sigma$ ) for 4 independent analyses  
177 during the course of the study. The  $^{40}\text{Ar}/^{36}\text{Ar}$  ratio of the atmospheric argon was also  
178 periodically measured, giving an average of  $295.0 \pm 1.9$  ( $2\sigma$ ) for 5 independent analysis, also  
179 during the course of the study. Since the results were analytically consistent and close to the  
180 theoretical values of  $24.85 \pm 0.24 \times 10^{-6} \text{ cm}^3$   $^{40}\text{Ar}$  for the GL-O standard (Odin et al., 1982),  
181 and 295.5 for the atmospheric  $^{40}\text{Ar}/^{36}\text{Ar}$  ratio used at the time the samples were analyzed  
182 (Nier, 1950), no correction was applied to the raw data. As the sample analyses were quite  
183 ancient, the differences with the values of the standards had almost no impact on the final  
184 ages. The blank of the gas extraction line and its coupled mass-spectrometer was also checked

185 weekly before the Ar extraction. Systematically below  $10^{-8} \text{ cm}^3$ , the amounts of radiogenic  
186  $^{40}\text{Ar}$  of the blank were two orders of magnitude lower than the amounts of that extracted from  
187 samples and, therefore, they were considered not to bias the data. The K contents were  
188 determined by flame spectrometry and the K-Ar ages calculated with the usual decay  
189 constants (Steiger and Jäger, 1977). The final precision of the K-Ar ages was estimated to be  
190 better than 2%. To remind the readers, especially those unfamiliar with illite K-Ar isotope  
191 dating but having a preference for the application of the more recent “sister”  $^{40}\text{Ar}/^{39}\text{Ar}$  dating  
192 technique of illite, the specific advantages and drawbacks of the two K-Ar and  $^{40}\text{Ar}/^{39}\text{Ar}$   
193 dating methods were evaluated and compared by a systematic application of both methods on  
194 the same illite-sized separates (Clauer et al., 2012; Clauer, 2013).

195

## 196 **Results**

197

### 198 The XRD results

199 The separated size fractions are quite similar on the basis of the XRD results. They  
200 consist mainly of illite mixed with illite-smectite mixed layers (labeled I-S hereafter) in five  
201 Permian samples of the Mas Lavayre mine and in the deepest sample of the MLV354 drilling,  
202 with chlorite (up to 80-90% in the four deeper Permian samples of the MLV354 drilling), or  
203 kaolinite (traces to 15% in three Montdardier fractions; Table 2). Most FWMH of the finest  
204  $<0.4 \mu\text{m}$  fractions are below 0.36 (Table 2), while only the  $<0.4 \mu\text{m}$  fraction of the  
205 Montdardier M1 sample, of three samples from Mas Lavayre mine and of its associated  
206 MLV354 drilling yield higher values. Consequently, most illite fractions are within the  
207 epizonal (low-grade metamorphism) and the lower anchizonal domains, which suggests  
208 crystallization temperatures between 160 and 280°C (Kübler, 1997).

209 The three F1 feldspar separates contain pyrite, pyrolusite, quartz, and galena for the  
210 80-100  $\mu\text{m}$  fraction, quartz for the 125-200  $\mu\text{m}$  fraction, and apatite, blende and quartz for the  
211 200-400  $\mu\text{m}$  fraction. The 125-200  $\mu\text{m}$  fraction of the F2 feldspar contains pyrite, blende,  
212 quartz and pyrolusite.

213

### 214 The K-Ar data

215 At a first glance, the 43 K-Ar data range widely from  $298.3 \pm 13.7$  to  $103.0 \pm 3.5$  Ma  
216 ( $2\sigma$ ; Table 3). The K-Ar ages of the five complementary feldspar separates split into two  
217 groups at  $282 \pm 13$  Ma for three of them and at  $255 \pm 9$  Ma for the two others. Among the  
218 illite ages, 16 were measured on the finer  $<0.2$  and  $<0.4$   $\mu\text{m}$  sub-fractions that were extracted  
219 and analyzed separately to evaluate if the coarser  $<2$   $\mu\text{m}$  fractions are isotopically  
220 homogeneous, or if they consist of several generations of illite-type particles characterized by  
221 an increasing amount of younger generations in the finer fractions and of older generations in  
222 the coarser fractions. In fact, 5 samples yielded K-Ar ages within analytical uncertainty for  
223 their 2 analyzed size fractions. Those of the Cambrian LM1 argillaceous dolomite from Les  
224 Malines mine yield an average age of  $290 \pm 8$  Ma, and those of the MLV4 Permian sample of  
225 the nearby drilling MLV354 provide an average age of  $176 \pm 7$  Ma. In the case of the two SJ3  
226 and SJ4  $<0.2$  and  $<0.4$   $\mu\text{m}$  fractions, the K-Ar values are also within analytical uncertainty at  
227  $136 \pm 4$  and at  $107 \pm 4$  Ma, respectively. This preliminary check of the K-Ar results suggests  
228 episodic illitization at about 290 Ma, 175 Ma, 135 Ma and 105 Ma. On the basis of these  
229 results, it appears that the  $<2$   $\mu\text{m}$  size fractions with identical ages consist of homogeneous,  
230 authigenic illite populations, as is the case for the samples M3, M4, M6, MLV6, and for the  
231 feldspar separates F1a, F1b and F2. The K-Ar data of the 0.4-2  $\mu\text{m}$  size fractions of the M5,  
232 M6, ML1, ML2, SJ2, MLV4 samples also fit these preliminary age groupings, as well as that  
233 of 0.4-1  $\mu\text{m}$  size fraction of SJ3 sample. The 0.4-1, 0.4-2 and  $<2$   $\mu\text{m}$  fractions of the other  
234 samples with older K-Ar data, therefore suggest that they consist of multi-generation material  
235 including detrital components. In summary, it can be considered at this point that half of the  
236 forty-eight available K-Ar data apparently identify four well-defined tectono-thermal  
237 illitization episodes.

238 Harper (1970) plots compare basically the contents of the radiogenic  $^{40}\text{Ar}$  relative to  
239 those of the K of the illite and feldspar fractions. Here, four lines can be drawn through  
240 several alignments of data points that end at the intersection of the coordinates as they should  
241 (Fig. 2). These lines are not strictly isochrons, such as those of  $^{40}\text{Ar}/^{36}\text{Ar}$  vs.  $^{40}\text{K}/^{36}\text{Ar}$   
242 correlations. However, average ages can be calculated by combining the individual ages of all  
243 representative data points of each line. In the detail, the upper line with the steepest slope is  
244 anchored by three feldspar and six illite data points at an average age of  $288 \pm 10$  Ma. The  
245 line below is less constrained with two feldspar and three illite data for an average age of 246

246  $\pm 9$  Ma. The next line below fits through six illite data points, giving an age of  $197 \pm 6$  Ma.  
247 Finally, the line with the lowest slope includes seven data points of illite-rich fractions, giving  
248 an age of  $176 \pm 6$  Ma. The leftover data points that are scattered in between the references  
249 lines, can be assumed to result from a mix of minerals crystallized during at least two of these  
250 episodes (Fig. 2). In summary, by combining the four K-Ar ages obtained from checking  
251 those of the different size fractions of the same samples and the four ages from lines in the  
252 radiogenic  $^{40}\text{Ar}$  vs. K diagram, the ages at about 290 Ma and 175 Ma were found in both  
253 evaluations. The size fractions with the younger ages at about 135 and 105 Ma that were not  
254 obtained in the Harper (1970) plot also contain I-S mixed layers, suggesting some alteration  
255 of illite into K-depleted smectite. This could be due to a hydrothermal influence related to the  
256 motion of the Saint-Julien fault.

257         The coarser size fractions (0.4-1, 0.4-2 and  $<2 \mu\text{m}$ ) of the Cambrian samples range  
258 either around 280-300 Ma or on the older side of the 180-200 Ma group, which confirms the  
259 timing of these episodes and shows also that the 220-260 Ma and the 150-100 Ma events  
260 apparently did not affect the Cambrian metasediments. Conversely, the older K-Ar data of the  
261 coarse Permian size fractions are obviously biased by detrital contaminants with K-Ar data  
262 either similar to the deposition age of the host rocks, or even older. Most of the other K-Ar  
263 data of the Permian material are on the older side of the 170-190 Ma event, which suggests  
264 some resetting (Table 3).

265

## 266 **Discussion**

267

268         The broadly range of the K-Ar ages suggest crystallization of several generations of  
269 illite during distinct thermal episodes in ore-bearing and barren Cambrian and Permian  
270 sediments of the southeastern mining district of the French Massif Central. This evolutionary  
271 sequence occurred along the eastern Cevennes fault system that acted as the regional  
272 circulation drain for advective fluids. Illite of the sampled rocks was altered either once or  
273 several times, completely or incompletely, or even not at all.

274

275 The paleo-thermometric context

276 A paleo-thermometric study showed that bi-phased aqueous inclusions of quartz from  
277 faults in the nearby Upper Permian yield homogenization temperatures from 60 to 300°C  
278 (Staffelbach et al., 1987; Mendez Santizo et al., 1991). In fact, most of the inclusions yield  
279 homogenization temperatures from 150 to 300°C with a maximum frequency at 170-180°C in  
280 the major fractures such as that of Saint-Julien sampled in the Mas Lavayre mine. Those  
281 above 240°C obtained on quartz inclusions correspond to fusion temperatures at salinities  
282 between 16.5 and 12% eq. NaCl. The fractionation coefficient of Co between pyrite and  
283 pyrrhotite being temperature dependent (Bezmen et al., 1975), two sets of temperatures were  
284 obtained for the pyrite-pyrrhotite association in the Saint-Julien fault:  $280 \pm 6^\circ\text{C}$  for the gouge  
285 material, and  $242 \pm 22^\circ\text{C}$  for vein deposits in the host rocks. This latter temperature does not  
286 match a diagenetic impact for the 250-Ma illite resulting from alteration of pyroclastic  
287 material (Brockamp and Clauer, 2013), unless the veins with pyrite-pyrrhotite infillings  
288 formed during moving of the Saint-Julien fault.

289 The obtained large temperature spectrum confirms that various thermal conditions  
290 necessarily occurred in rocks of different stratigraphic ages and locations, including the fact  
291 that the diagenetic/hydrothermal conditions must have happened soon after sediment  
292 deposition during the Permian. The  $185 \pm 11$  Ma episode appears to have had a climax impact  
293 in various stratigraphic strata: in Permian sediments at Lodève, as well as in U-enriched veins  
294 nearby Rabejac (Lancelot and Vella, 1989). Such a paroxysmal activity was reported during  
295 the Rhaetian–Hettangian time in the Cévennes and western Alps, which corresponds in turn to  
296 the development of the Ligurian Tethys (Lemoine et al., 1986; Dromart et al., 1998). The  $105$   
297  $\pm 5$  Ma episode determined in gouge material of the Saint-Julien fault (Mendez Santizo et al.,  
298 1991) has also been considered to correspond to the onset of the first oceanic accretion in the  
299 Biscay Bay (Montadert et al., 1979).

300

### 301 The historical context of the ore genesis in the southeastern Massif Central

302 For Charef and Sheppard (1988), the main ore stage is characterized by the mixture of  
303 fluids with a metal-rich brine at a temperature of about 150°C that the authors identified as a  
304 connate water resulting from dewatering of the basin. Called the “Neo-Variscan stage” by  
305 Merignac and Cuney (1999), this episode of main metal concentration is characterized by a  
306 shift from a compressional to an extensional tectonic regime and a significant heat-flow

307 circulation. Conversely, Early-Jurassic illite ages discussed by Clauer and Chaudhuri (1995)  
308 as representative of tectono-thermal activities in the Les Malines sector were denied by Leach  
309 et al. (2001) who attributed them to a widespread diagenetic event in the host rocks. However,  
310 the alternative model of Leach et al. (2005) based on gravity-driven fluid movements for  
311 some of the known deposits is equally questionable. Indeed, it is because of the high  
312 temperatures determined in fluid inclusions from minerals associated with the ore deposits  
313 evaluated in the previous section, and because of the episodic K-Ar ages obtained here.  
314 Finally, after decades of varied models based on the re-activation of karst infillings, the metal  
315 deposits were also considered by the same authors to result from fluid migrations during a 60-  
316 Ma old compressional tectonic event on the basis of a well-defined remagnetization (Henry et  
317 al., 2001; Rouvier et al., 2001). They attributed this overprint to a chemical process due to a  
318 major uplift in the Pyrenean mountains with fluid migrations from south to north. However,  
319 on the basis of conclusions contradicting the earlier interpretations by Macquar et al. (1990)  
320 of a Triassic origin of the metal concentrations, the authors admitted a tectonic influence in  
321 the evolution of the ores from southeastern Massif Central. The 60-Ma age is, however, far  
322 too young relative to the neo-Visean concentrations and the illite K-Ar ages obtained here.

323 The published K-Ar ages of illite from metasediments of the Massif Central are  
324 somewhat of an “uneven” analytical level as many were obtained on  $<2 \mu\text{m}$  illite-rich size  
325 fractions (Bellon et al., 1974; Bonhomme and Millot, 1978; Bonhomme et al., 1983; Bril et  
326 al., 1991; Brockamp and Clauer, 2013). It has long been demonstrated that such coarse clay  
327 fractions often contain detrital minerals that bias the “ages” by increasing the data (Fig. 3).  
328 However, despite this technical aspect, the published ages are never strictly related to the  
329 major Variscan episodes. In other words, they are never older than 300 Ma and, therefore, the  
330 U-Pb and Pb-Pb ages of regional metal deposits of the Variscan period were discarded in this  
331 review. Post-Variscan U concentrations in the region seem to have begun about 270 Ma in  
332 leucogranite veins percolated by fluids at 300-350°C (Lancelot et al., 1995). Later  
333 concentrations were dated at  $188 \pm 12$  Ma in a pitchblende (Respaut et al., 1991), at  $172 \pm 9$   
334 Ma in uraninite (Léveque et al., 1988), at about 100 Ma in a coffinite, and even at 35-40 Ma at  
335 Pierres Plantées, again in a coffinite (Respaut et al., 1991). This last age is the only one that  
336 occurred after the potential Pyrenean activity mentioned just above. Some U concentration  
337 was also detected at about 170 Ma in coffinite at Bertholène. All these recorded ages are far

338 from the Pyrenean activity detected in the southern Massif Central by remagnetization  
339 (Rouvier et al., 2001).

340

#### 341 Meaning of the illite K-Ar ages from southeastern Massif Central

342           Alternatively to the single paleomagnetic record just mentioned, K-Ar illite ages were  
343 reported between 220 and 160 Ma in the Lodève area (Bellon et al., 1974; Bonhomme and  
344 Millot, 1978; Bonhomme et al, 1983). At the Bernardan site, six illite K-Ar ages average  $161$   
345  $\pm 9$  Ma (Clauer, unpublished) with four more values scattered between 240 and 185 Ma.  
346 Lancelot et al. (1994) detected also two U remobilizations by K-Ar dating of illite from  
347 Lodève district at  $173 \pm 6$  and  $108 \pm 5$  Ma. These authors connected the first of these two  
348 events to an extensive thermal event of the continental crust to the south of the Massif  
349 Central. Clauer et al. (1997) reported a K-Ar age of  $190 \pm 20$  Ma for an illite infilling a major  
350 fault in the carbonate-rich, passive paleo-margin along the southeastern Massif Central.

351           It is generally agreed that age identities of two fine-size fractions from the same  
352 samples corroborate with geological meaningful ages, as discussed by Clauer and Chaudhuri  
353 (1998) who illustrated this behavior by a “bench-type” sketch with the same age having a  
354 geological meaning for the fine fractions, and an older meaningless date for the coarser  
355 fraction(s). If the fine ( $<0.2$  or  $<0.4 \mu\text{m}$ ) fractions consist mainly (or only) of authigenic illite  
356 and, depending on the deposition time of their host sediments, a multi-episodic tectonic-  
357 thermal activity can be postulated at  $288 \pm 10$  Ma based on the K-Ar of the pyroclastic  
358 feldspars and of illite separates from sediments of Les Malines deposits, as well as at  $246 \pm 9$   
359 Ma mostly in the Cambrian sediments from the Les Malines district with a further late illite  
360 crystallization at  $105 \pm 5$  Ma in gouge material of the Saint-Julien fault in the Mas Lavayre  
361 mine (Mendez Santizo, 1990; Mendez Santizo et al., 1991).

362           Brockamp and Clauer (2013) studied diagenetic and hydrothermal impacts and their  
363 timing in illite-rich size fractions of Permian shales close to the Lodève U-district located  
364 about 4 km to the NW of the northern sector studied here. There, illite with a mean K-Ar age  
365 of about 260 Ma is the predominant mineral in the shales. It is of early diagenetic origin  
366 relative to deposition and apparently formed directly from deposited pyroclastic materials,  
367 which waives the problem of a detrital contamination as such rock type basically does not  
368 contain detrital minerals. The associated hydrothermally altered shales next to the U-deposit

369 also contain a K-feldspar generation with K-Ar ages clustered around 220 Ma, although  
370 Jurassic and Cretaceous ages were also obtained. The hydrothermal temperatures were  
371 estimated to be below 200° C as the early diagenetic illite is only partially reset. It looks like  
372 Triassic fluids ascended along faults into the northern part of the Lodève basin during an  
373 initial rifting of the nearby western Tethys Ocean. Later fluid pulses were also recorded at  
374 Lodève during the opening of the North Atlantic and separation of Europe from Africa  
375 (Jurassic/Cretaceous). Along with this tectono-thermal activity, successive pulses of hot fluids  
376 of crustal origin apparently also penetrated the Lodève district.

377 In summary, the early thermal event at  $288 \pm 10$  Ma obtained in the pyroclastic  
378 feldspars and in the Montdardier Cambrian sediments was expectedly not recorded in the  
379 Permian sediments that were deposited later. The later  $246 \pm 9$  Ma episode was also recorded  
380 in Permian sediments at Mas Lavayre. By itself, evidence of a volcanic activity does not  
381 necessarily imply regional abnormal thermal conditions in sediments: thermal conditions are  
382 diffusing wider when detected in sediments of varied stratigraphic ages and from different  
383 locations, which strengthens the argument for regional abnormal thermal conditions.

384

#### 385 The regional extent of illite K-Ar ages relative to the evolution of the metallogenic deposits

386 The K-Ar data of the illite-type clay material studied here fit well into the late plutonic  
387 and structural geologic framework of the post-Variscan plutonic history and the Liassic  
388 extensional tectonic episode along the Cévennes fault system. The review by Lancelot et al.  
389 (1995) summarizes the regional ore formation in which the ages obtained here fit well. The  
390 same regional picture applies also to fault gouges filled with clay minerals in plutonic and  
391 sedimentary rocks to the NW of the Massif Central (Cathelineau et al., 2004). There, the  
392 finest  $<0.2 \mu\text{m}$  fractions from deep fractures ( $>570$  m) yield older ages, from 272 to 253 Ma,  
393 than those of 198 to 188 Ma for illite from shallow-depth fractures. This confirms the  
394 occurrence of two distinct episodes, probably of different intensity. Cathelineau et al. (2012)  
395 showed that migrating brines and seawater interacted with both the sedimentary cover and the  
396 crystalline basement in the Poitou High at the northwestern edge of the French Massif  
397 Central, from 156 to 146 Ma based on K-Ar dating of illite and  $^{40}\text{Ar}/^{39}\text{Ar}$  dating of associated  
398 feldspars. This hydrothermal activity induced significant dolomitization and silicification of

399 the sediments, adularization of the granitic basement, and local concentrations of F–Ba (Pb–  
400 Zn) assemblages.

401 All these results confirm the occurrence of geographically dispersed and episodic  
402 hydrothermal activities at different metalliferous sites. Illite crystallization often occurred in  
403 nearby fault systems that acted as drains for migrating fluids that favored or not ore genesis.  
404 As a possible alternative to this scenario, late-Variscan activities in plutonic basement rocks,  
405 especially of the major cratonic basement of Western Europe, were not often reported.

406 In summary, the compilation of the illite K-Ar and ore U-Pb and Pb-Pb ages outline  
407 several age combinations for the regional tectono-thermal activity (Fig. 3). After the Variscan  
408 stage, a regional episode is detected by illite K-Ar ages from NW to SE of the Massif Central  
409 at 305-285 Ma. A following event was also widely detected from NW to SE at 265-245 Ma.  
410 The next episode occurred at 210-190 from NW to SE immediately followed by another in the  
411 NW and the SE areas at 185-160 Ma, assuming these events were not connected. The two last  
412 episodes are less spread out, one to the N at 150-135 Ma and the last to the S at 120-105 Ma.  
413 Only the later of these episodes affected the ore deposits: that at 210-190 Ma at Pierres  
414 Plantées and Rabejac, that at 185-160 Ma again at Pierres Plantées and at Lodève and the final  
415 episode at 120-105 Ma at Lodève.

416

#### 417 Combining U-Pb and Pb-Pb dating of ores with K-Ar dating of associated clay minerals

418 On the basis of a large review of major basin-hosted ore deposits in Europe, Muechez  
419 et al. (2005) suspect that most of them formed in extensional settings. Where extension was  
420 pronounced and heat production elevated, mineralizing fluids were expelled along extensional  
421 faults after having migrated through the sedimentary basin and the basement. This conclusion  
422 does not match with the regional illitization episodes in the Les Malines district, which do not  
423 occur next to major extensional tectonic or rifting areas. Indeed, the results from southeastern  
424 Massif Central confirm the occurrence of two major episodes during the Upper Permian-  
425 Lower Triassic and Liassic times far from common tectonic/geodynamic activities, in rather  
426 quiescent areas where only re-activation of fault systems could be identified. This lack of  
427 physical processes in the neighborhood of the metal deposits needs migrations of hot fluids in  
428 the continental crust, as the metals were concentrated at specific places, but not necessarily  
429 during all detected tectonic-thermal episodes. Many of such deposits were dated during

430 Jurassic and Cretaceous episodes at  $185 \pm 15$ ,  $140 \pm 10$  and  $105 \pm 5$  Ma, which supports a  
431 relationship between fluid movements and geodynamic events, however far from major  
432 tectonic-thermal events.

433 In summary, a pre-concentration episode of U was detected at about 270 Ma ago in  
434 leucogranite veins. Alternatively, the 250-Ma event identified by K-Ar dating of illite  
435 separates obviously did not impact the ores, while the  $170 \pm 15$  Ma episode affected varied  
436 types of deposits in the southeastern Massif Central, as well as the 140-Ma and 100-Ma  
437 events. Even a more recent event was found at Pierres Plantées at about 35-40 Ma ago.  
438 Alternatively, no post 100-Ma event was detected in the clay material, which inclines one to  
439 consider that the 35-40 Ma date for coffinite at Pierres Plantées (Respaut et al., 1991) and the  
440 more or less associated remagnetization episode at 50-60 Ma at the les Malines (Rouvier et  
441 al., 2001) were not generated by thermal events.

442

## 443 **Conclusions**

444

445 Illite-rich size-fractions of Cambrian and Permian calcschists, shales and dolostones of  
446 the metallogenic district from southeastern Massif Central were dated by the K-Ar method.

447 The combined mineralogical and K-Ar determinations support the following conclusions:

448 (1) Illite K-Ar ages suggest crystallization episodes at  $288 \pm 10$ ,  $246 \pm 9$  and  $197 \pm 6$  Ma.

449 However, all these tectonic-thermal events were not monitored by mineralizing fluids, which  
450 seem to have contributed to illitization probably at  $136 \pm 4$  Ma and definitely at  $107 \pm 4$  Ma,  
451 evidenced by metal U-Pb and Pb-Pb dating in faults, and during minor Cretaceous pulses.

452 (2) Fluid events occurred within a segment of a continental margin away from rift zones and  
453 from major deformation areas of Western Europe. The lack of important geodynamic  
454 activities next to the metal-rich deposits leads the author to consider periodic migration of  
455 hot-fluids in the underlying continental crust, as metals were concentrated at specific places,  
456 but not necessarily altered by each tectonic-thermal pulse recorded by illite crystallization.  
457 The 250-Ma episode, for instance, seems not to have altered the metalliferous concentrations.

458 (3) The tectonic-thermal history of the Les Malines ore district confirms repetitive  
459 geodynamic re-activation of previously occurring events that affected local mass, as well as  
460 heat transfer in the plutonic basement and in the overlying sediments. On the other hand, the

461 recent episode at 35-40 Ma ago in coffinite at Pierres Plantées Massif has obviously not been  
462 induced by a thermal episode, since it was not recorded in the illite clays.

463

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465

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473

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### 683 **Figure and table captions**

684

685 **Figure 1:** (A) Geographic distribution of the samples from French Massif Central studied  
686 here and earlier (modified from Merignac and Cuney, 1999, and from Santouil, 1980). (B)  
687 Details of the Montdardier - Les Malines district with the locations of the sampling sites;  
688 (C) Details of the Lodève district with the locations of the sampling sites (modified from  
689 Santouil, 1980, where the rock types are identified).

690

691 **Figure 2:** Radiogenic  $^{40}\text{Ar}$  vs.  $\text{K}_2\text{O}$  histogram (Harper, 1970) of the different illite-rich size  
692 fractions and the feldspar separates of this study.

693

694 **Figure 3:** Synthetic sketch of the regional K-Ar illite ages and U-Pb and Pb-Pb ages of metal-  
695 rich deposits from French Massif Central. The locations are in the figure 1A. The origins  
696 of the results are given below the diagram. The thick age-sector arrows were obtained on  
697 variable illite size fractions, while the thin sector arrows were obtained on  $<2\ \mu\text{m}$  illite size  
698 fractions.

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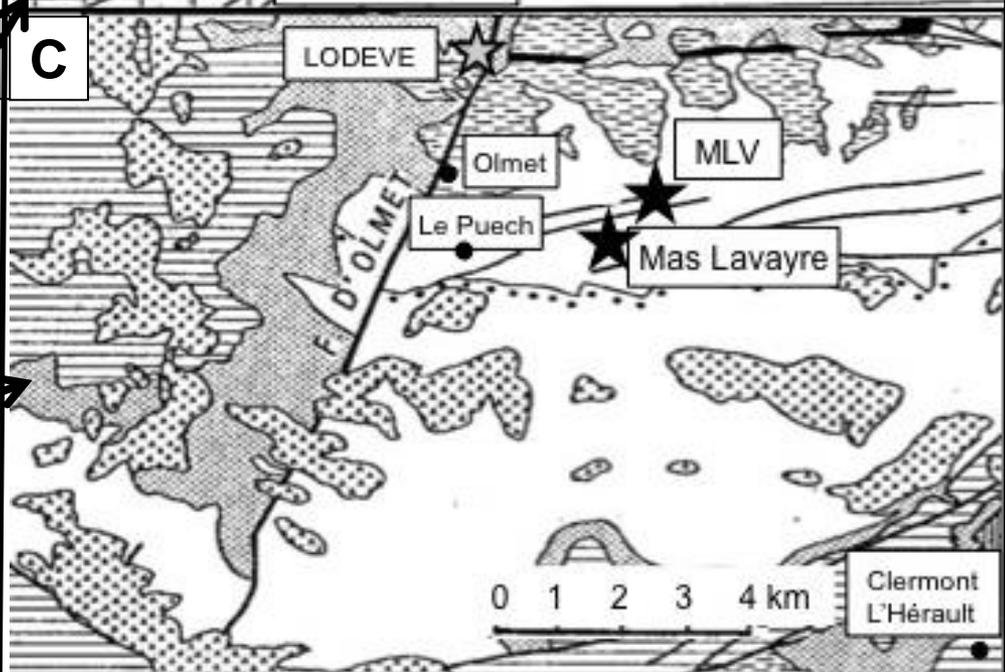
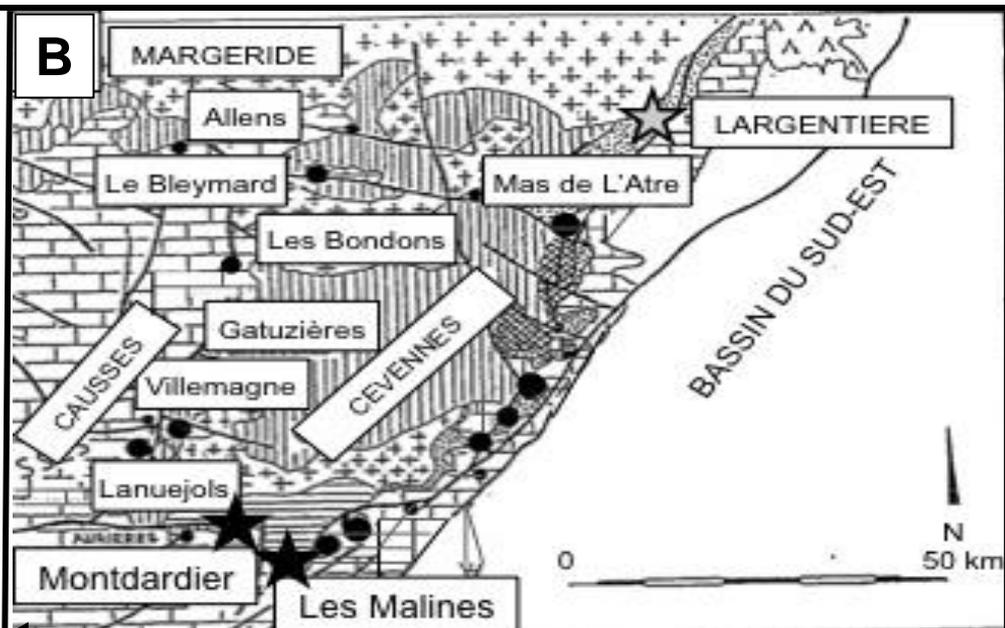
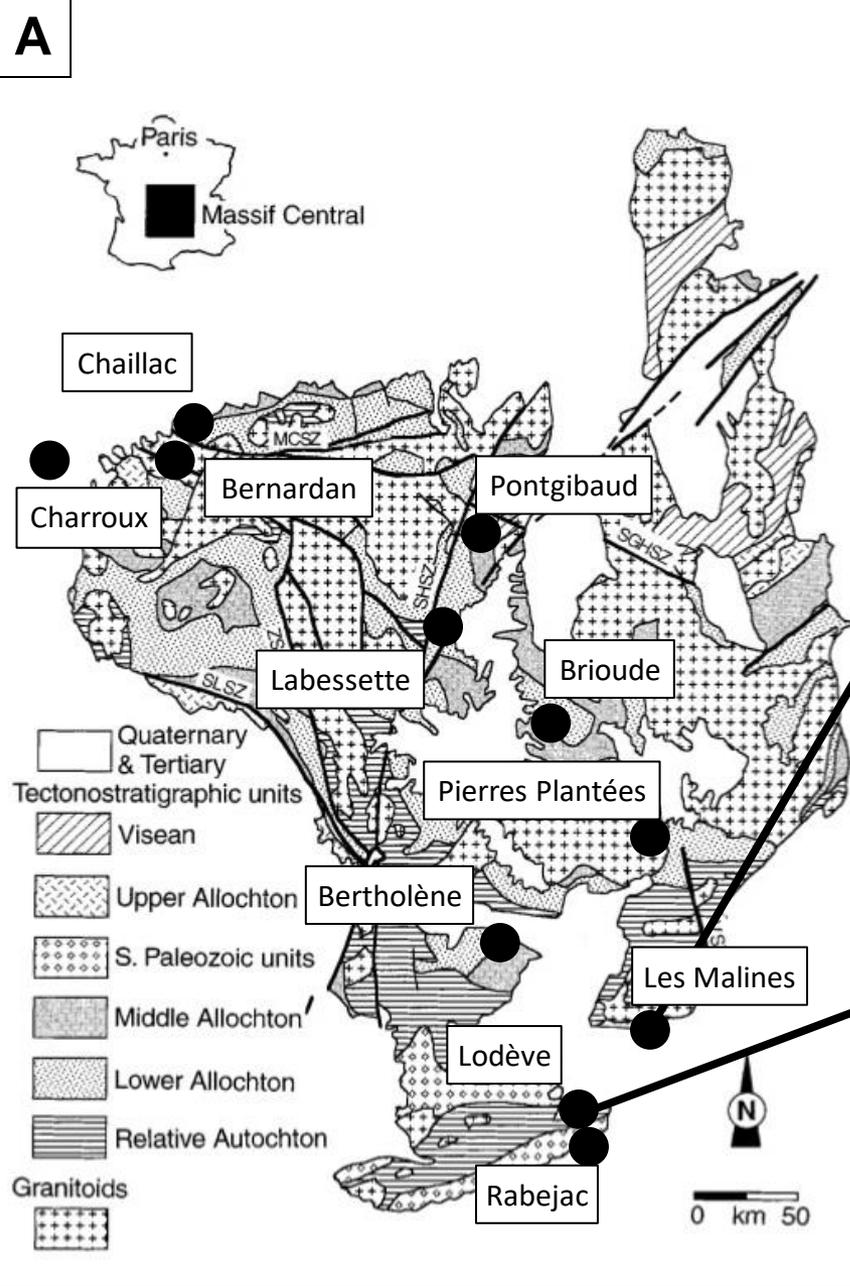
701 **Table 1:** Location, stratigraphic position and description of the studied samples.

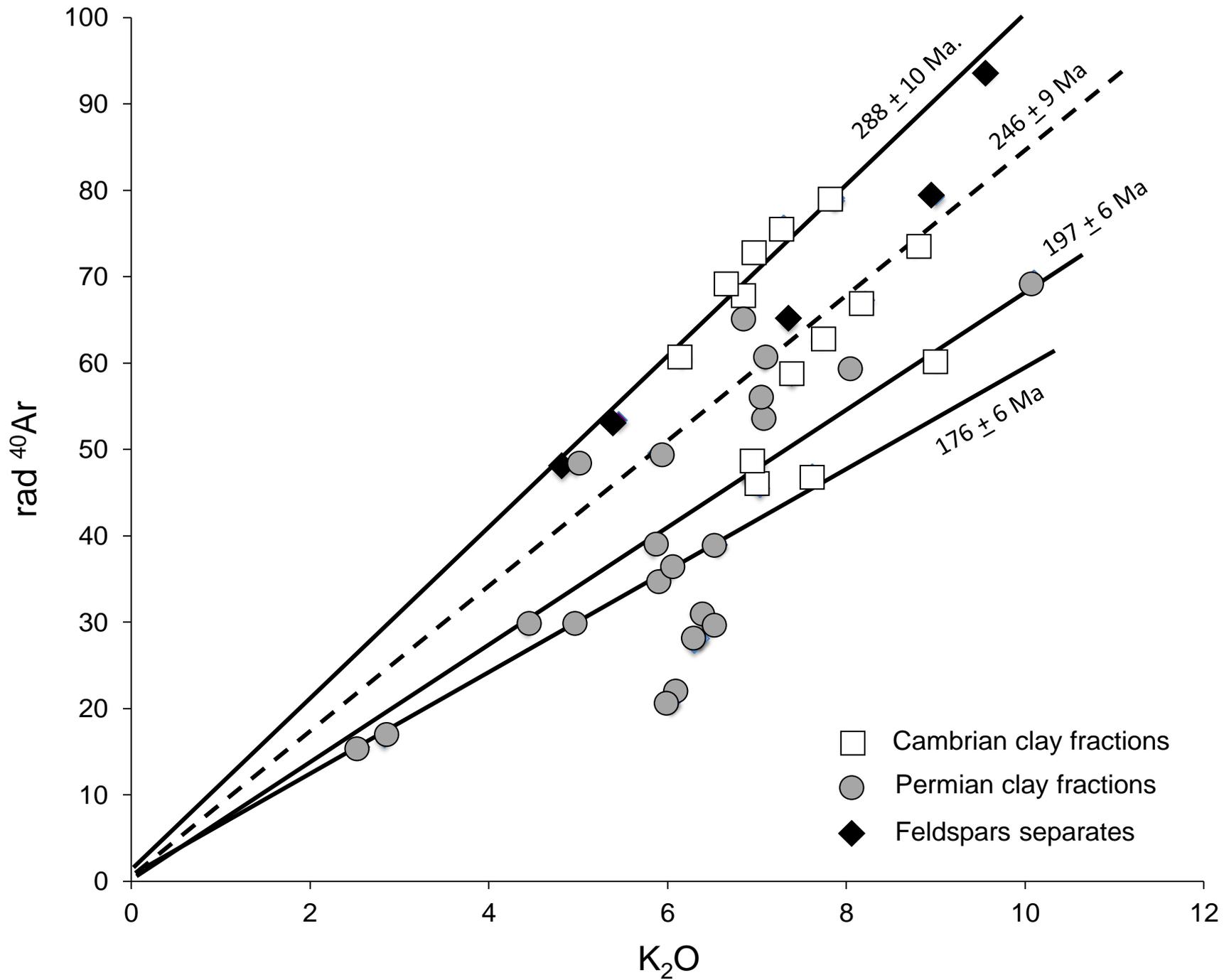
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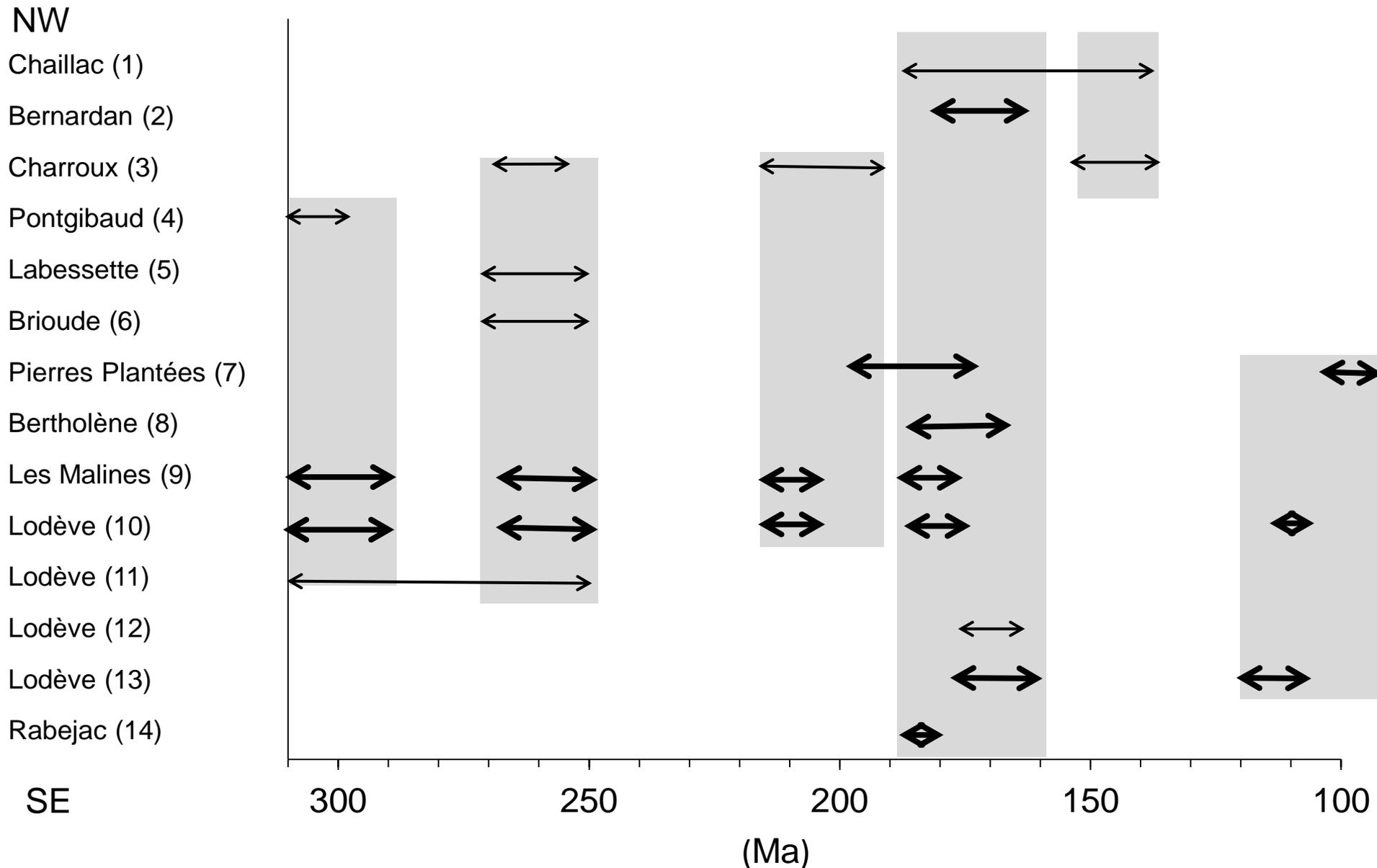
703 **Table 2:** XRD mineralogical data of the studied size fractions.

704

705 **Table 3:** K-Ar results of the studied illite-rich size fractions and feldspar separates. Ar\*  
706 stands for radiogenic Ar.







Data (1) and (3) of Cathelineau et al. (2004; 2012); data (2) of Clauer (unpublished); data (4, 5 and 6) of Bril et al. (1991); data (7) of Respaut et al. (1991); data (8) of Schmitt et al. (1984) and Lévêque et al. (1988); data (9 and 10) of this review; data (11) of Brockamp and Clauer (2013); data (12) of Bellon et al. (1974); data (13 and 14) of Lancelot and Vella (1989).

Sample ID	Stratigraphy	Sample description
Montdardier Mt1	Cambrian	Mine, eastern wall, argillaceous dolomite
Montdardier Mt2		Mine, northern wall, calcschist sampled 1 m below a pyrite layer
Montdardier Mt3		Mine, the same unit at the contact with the pyrite layer
Montdardier Mt4		Mine, the same location, pyrite layer
Montdardier Mt5		Mine, the same location, breccia at the top of the pyrite layer
Montdardier Mt6		Mine, sampled 1 m above in the same facies
Montdardier MS1		Mine, excavation surface close to an important ore concentration
Montdardier F1		Core, mineralized sample from F18 drilling into an excavation
Montdardier F2		Core, sample from F19 drilling to the N of Montardier mine
Les Malines LM1		Permian
Les Malines LM2	Mine, argillaceous dolomite in a cavity of dolostones	
Mas Lavayre ML1	Mine, greyish silt with some bituminous inclusions	
Mas Lavayre ML2	Mine, grey-to-red sandy shale	
Mas Lavayre ML3	Mine, redish pelite with bituminous stratiform accumulations	
Mas Lavayre ML4	Mine, redish pelite	
Mas Lavayre SJ2	Mine, eastern wall Saint Julien fault, 4 m away from main fault	
Mas Lavayre SJ3	Mine, eastern wall, Saint Julien fault, 1 m from a lateral fault	
Mas Lavayre SJ4	Mine, eastern Saint Julien fault, within a lateral fault	
Mas Lavayre SJ5	Mine, eastern wall, Saint Julien fault, 0.5 m off the lateral fault	
Mas Lavayre SJ6	Mine, eastern Saint Julien fault, 5 m from lateral fault	
MVL354 (93m) MVL1	Drilling core, barren shale	
MLV354 (94m) MLV2	Drilling core, barren shale on top of a bituminous dolomitic shale	
MLV354 (236m) MLV3	Drilling core, sandy pelite	
MLV354 (286m) MLV4	Drilling core, dolomitic shale above an uraniferous bitumen concentration	
MLV354 (286.5m) MLV5	Drilling core, bituminous shale	
MLV354 (287m) MLV6	Drilling core; shale with uranium concentrates	

Sample ID	Stratigraphy	Size ( $\mu\text{m}$ )	Illite (%)	I-S (%)	Kaolinite (%)	Chlorite (%)	Acc.	FWHM	
Montdardier M1	Cambrian	<0.4	100		tr		-	0.50	
		0.4-2	85		15		Q, F	0.29	
Montdardier M2		<2	100				Q, F	0.23	
Montdardier M3		<2	90			10	Q, F	0.27	
Montdardier M4		<2	100				Q	0.36	
Montdardier M5		<0.4	100				-	0.35	
		0.4-2	100				Q, F	0.21	
Montdardier M6		<2	100				Q, F	0.29	
Montdardier DS1		<2	100				-	n.d.	
Montdardier F1		<2	100				Q	n.d.	
Montdardier F2		<2	100				-	n.d.	
Les Malines LM1		<2	100				q	0.28	
Les Malines LM2		<2	100				q, F	0.30	
Mas Lavayre ML1		Permian	<2	25	15		60	f	n.d.
Mas Lavayre ML2	<2		55			45	Q, f	n.d.	
Mas Lavayre ML3	<2		35			65	Q, F	n.d.	
Mas Lavayre ML4	<2		40		15	45	F	n.d.	
Mas Lavayre SJ2*	<0.4		0		70	30	-	n.d.	
Mas Lavayre SJ3*	<0.4		60		10	30	-	n.d.	
Mas Lavayre SJ4*	<0.4		35		50	15	-	n.d.	
Mas Lavayre SJ5*	<0.4		70			30	-	0.52	
Mas Lavayre SJ6*	<0.4		60			40	-	n.d.	
MLV354 (93m) MLV1	<2		80				20	f	0.45
MLV354 (94m) MLV2	<2		80				20	F	0.45
MLV354 (236m) MLV3	<2		20				80	Q, F	n.d.
MLV354 (286m) MLV4	<2		10				90	F	n.d.
MLV354 (286.5m) MLV5	<2		10				90	f, q	n.d.
MLV354(287m) MLV6	<2	0		15		85	f	n.d.	

I-S stands for illite-smectite mixed layer, F for feldspar, Q for quartz, both in small lettering when in trace amounts, n.d. f not determined ; Acc. for accessory minerals and FWHM for Full Width at Half Maximum

Sample ID	Stratigraphy	Sizes ( $\mu\text{m}$ )	K <sub>2</sub> O (%)	Ar* (%)	40Ar* (10 <sup>-6</sup> cm <sup>3</sup> /g)	K-Ar ages (Ma $\pm$ 2 $\sigma$ )
<b>Clay fractions</b>						
Montdardier M1	Cambrian	<0.4	9.01	95.81	60.27	196.5 (4.2)
		0.4-2	8.21	96.65	67.32	238.0 (5.1)
Montdardier M2		<2	7.72	88.17	62.89	236.6 (6.6)
Montdardier M3		<2	7.87	66.25	78.79	286.7 (9.8)
Montdardier M4		<2	6.81	45.57	67.68	284.7 (13.3)
Montdardier M5		<0.4	8.77	94.90	73.59	243.2 (5.3)
		0.4-2	7.30	96.80	76.09	297.5 (6.4)
Montdardier M6		<2	6.97	46.40	72.85	298.3 (13.7)
Montdardier DS1		<2	7.62	92.40	47.31	183.0 (4.1)
Montdardier F1		<2	7.01	87.80	48.44	202.6 (4.8)
Montdardier F2		<2	7.03	89.30	45.55	190.6 (4.4)
Les Malines LM1		<0.4	6.16	54.74	60.72	282.6 (10.5)
		0.4-2	6.66	84.74	69.05	296.1 (7.3)
Les Mlines LM2		<0.4	7.39	54.92	58.40	229.9 (8.5)
Mas Lavayre ML1	Permian	<0.4	6.30	79.20	27.46	130.4 (3.9)
		0.4-2	6.05	90.50	36.35	177.4 (4.1)
		<2	5.89	71.60	30.67	154.8 (4.5)
Mas Lavayre ML2		<2	8.06	66.50	59.56	215.8 (6.6)
Mas Lavayre ML3		<2	4.43	46.70	29.62	196.4 (8.6)
Mas Lavayre ML4		<2	10.10	91.20	69.71	202.4 (4.5)
Mas Lavayre SJ2		<0.4	6.50	75.92	29.71	136.5 (3.7)
		0.4-1	6.56	86.45	38.96	175.5 (4.2)
Mas Lavayre SJ3		<0.2	6.36	71.65	28.07	132.0 (4.3)
		<0.4	6.43	78.07	30.24	140.3 (3.7)
		0.4-1	5.93	78.06	35.17	175.3 (4.7)
Mas Lavayre SJ4		<0.2	6.04	58.84	20.67	103.0 (3.5)
		<0.4	6.10	76.97	22.31	110.1 (3.0)
		0.4-1	6.36	76.93	31.23	146.3 (3.9)
Mas Lavayre SJ5		<0.4	6.93	85.73	46.80	198.3 (4.8)
		0.4-1	7.05	86.65	53.85	222.7 (5.3)
Mas Lavayre SJ6		<0.2	7.05	85.99	55.95	230.8 (6.5)
		<0.4	7.09	90.15	60.53	247.2 (5.7)
		0.4-1	6.85	89.70	65.92	276.3 (6.9)
MLV354 (93m) MLV1		<2	5.81	91.10	48.22	240.8 (5.5)
MLV354 (94m) MLV2		<0.4	5.88	66.50	49.53	244.1 (8.3)
		0.4-2	4.98	75.50	48.20	277.8 (7.8)
MVL354 (236m) MLV3		<0.4	4.95	42.70	30.00	178.9 (8.7)
		0.4-2	5.84	54.30	38.81	195.2 (7.3)
MLV354 (286m) MLV4	<0.4	2.83	69.90	16.57	173.1 (7.5)	
	0.4-2	2.56	85.40	15.53	179.1 (4.8)	
MLV354 (286m) MLV5	<2	2.56	80.60	15.98	184.1 (5.0)	
MLV354 (287m) MLV6	<2	4.11	84.70	23.56	169.7 (4.3)	
<b>Feldspar separates</b>						
Mas Lavayre F1a		80-100	4.82	45.22	47.71	283.6 (13.0)
Mas Lavayre F1b		100-125	5.45	52.43	53.38	280.9 (11.6)
Mas Lavayre F1c		125-200	9.00	76.90	79.29	254.5 (7.5)
Mas Lavayre F1d		200-400	7.36	62.18	64.82	254.5 (8.9)
Mas Lavayre F2		125-200	9.56	85.32	93.67	281.0 (7.6)

Ar\* stands for radiogenic Ar, and SJ for Saint-Julien fault

NW

Chaillac (1)

Bernardan (2)

Charroux (3)

Pontgibaud (4)

Labessette (5)

Brioude (6)

Pierres Plantées (7)

Bertholène (8)

Les Malines (9)

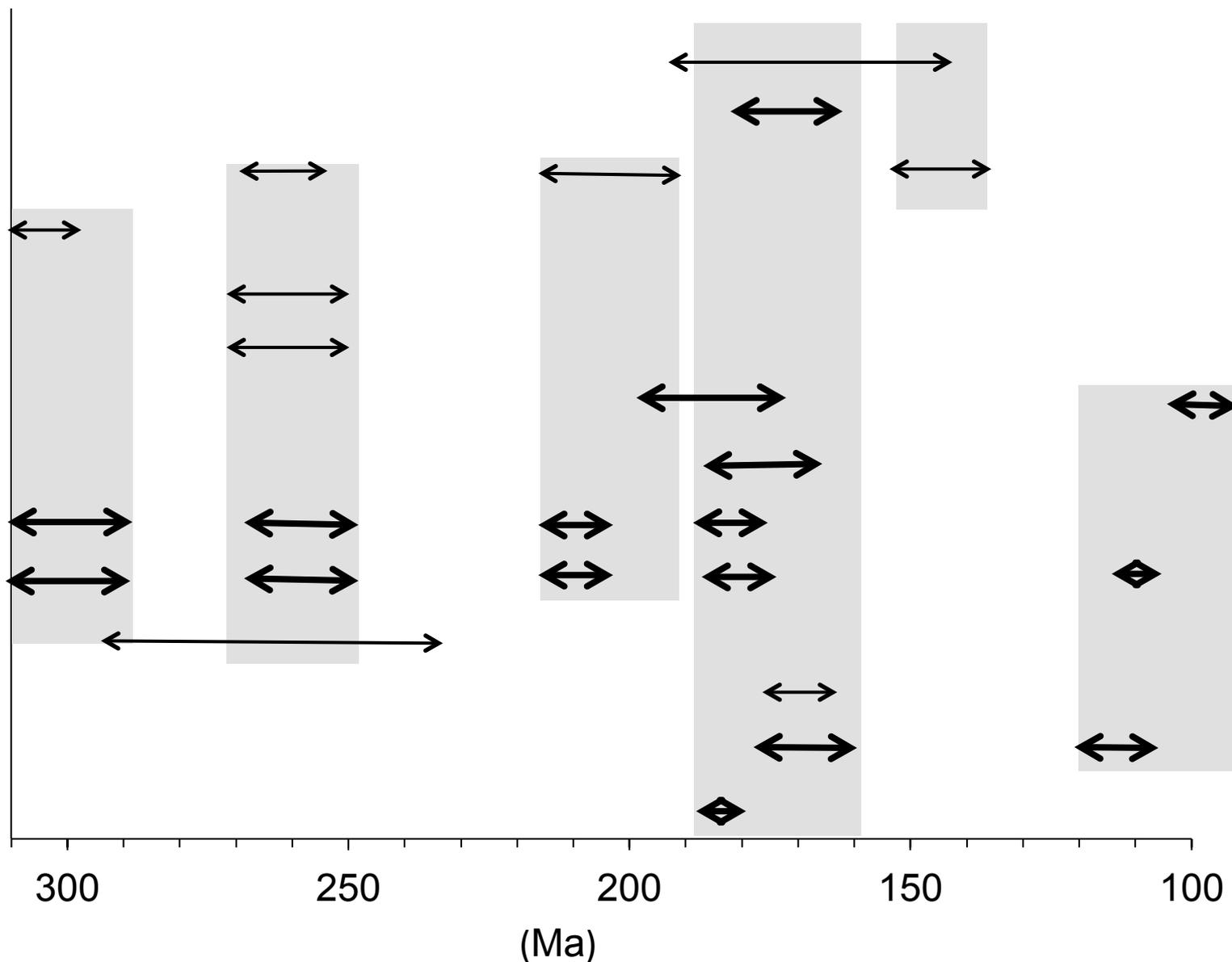
Lodève (10)

Lodève (11)

Lodève (12)

Lodève (13)

Rabejac (14)



Data (1) and (3) of Cathelineau et al. (2004; 2012); data (2) of Clauer (unpublished); data (4, 5 and 6) of Bril et al. (1991); data (7) of Respaut et al. (1991); data (8) of Schmitt et al. (1984) and Lévêque et al. (1988); data (9 and 10) of this review; data (11) of Brockamp and Clauer (2013); data (12) of Bellon et al. (1974); data (13 and 14) of Lancelot and Vella (1989).