

1 **Origin of zircon megacrysts in alkaline lavas (French Massif Central):**
2 **petrology and in situ U-Pb-Hf isotopes.**

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15 mantle; lower continental crust; French Massif Central.

16
17 **Abstract**

18
19 In the south eastern Devès volcanic domain of the French Massif Central, we sampled
20 red zircon megacrysts in, both, alluvium from a small stream and neighbouring mugearite
21 lava. LA-ICP-MS U-Pb dating results yield similar ages for both suites of zircons at about 2.6
22 Ma. Superchondritic signatures of the Hf isotopes clearly indicate a depleted mantle source
23 for these zircons.

24 The mugearite lava also contains xenoliths of mantle (lherzolites) and lower crustal
25 rocks (granulites). The chemical compositions of the minerals within the mugearite lava are

26 similar to the mineral compositions found in these xenoliths (granulites) and granulite and
27 peridotite xenoliths from other volcanic localities of the Massif Central. The mugearite lava
28 also preserves Variscan zircon crystals dated at 285 Ma, originating from recycled lower
29 continental crust.

30 In spite of their mantle origin, as recorded by in situ Hf isotopes, the geochemistry
31 suggests that these Pliocene zircons do not result from crystallization in the mantle or in
32 mantle-derived mafic magmas. In order to explain this inconsistency, we propose that these
33 zircon grains crystallized from felsic melts produced during the ultimate differentiation of
34 mantle-derived mafic magmas in the mid or upper continental crust. Subsequently, a last
35 basaltic batch forced its way to the surface, scavenging upper mantle and lower crust
36 xenoliths as well as zircon xenocrysts formed in these felsic melts which partly dissolved
37 during their ascent.

38 Large amounts of Cenozoic zircon megacrysts are often associated with sapphires in
39 alluvial deposits in many continental volcanic domains, not only in the French Massif Central
40 but also worldwide. Consequently, a huge volume of felsic differentiates issued from mantle-
41 derived basaltic magmas might be suspected in many alkaline volcanic domains at the global
42 scale.

43

44 **Introduction**

45

46 Experiments on zircon solubility saturation have demonstrated that crystallisation of
47 this mineral in basalts is unlikely (Watson, 1979; Watson and Harrison, 1983) since several
48 percent of ZrO_2 content may be dissolved in the melt at about 1300°C before any
49 crystallisation of zircon (Guo et al., 1996). Consequently, primary crystallisation of zircon in
50 most mafic volcanic melts is rare. Nevertheless, large amounts of zircons and sometimes

51 zircon megacrysts are exposed in alluvial deposits located in the vicinity of large mafic lava
52 flows or occur directly within the volcanic rocks. The presence of zircon, sometimes
53 associated with sapphire, is reported in many alkali basalt fields worldwide (see reviews in
54 Guo et al., 1996; Pin et al., 2006; Simonet, 2000; Sutherland and Fanning, 2001; Sutherland
55 et al., 1998, 2002 and 2008; Upton et al., 1999; Sutthirat et al., 2020 and reference therein). In
56 addition, these rocks frequently include xenolithic enclaves and minerals (Upton et al., 1999;
57 Paquette and Mergoïl-Daniel, 2009; Hurai et al., 2010 and 2013; Rakotosaminazanany et al.,
58 2014; Huraiova et al., 2017). It is generally agreed that zircons found in basaltic rocks must
59 be "exotic minerals" to their mafic hosts, but the origin of these zircon crystals is still
60 debated. They can be derived from differentiated newly-formed magmas emplaced at mantle
61 and/or crustal depth or may be xenocrysts of ancient continental basement carried upwards
62 during the eruption. Constraining their origin is the major goal of this contribution.

63 Zircon megacrysts mostly occur in placers linked to alkali basalt environments in
64 numerous continental volcanic fields around the world. We focus our study in the Velay
65 volcanic province in the south eastern Massif Central in France. This domain is characterized
66 by the occurrence of both zircon gems in sedimentary deposits near the basaltic fields and of
67 zircon crystals directly hosted in alkaline lavas. This area is well known for the size and the
68 abundance of zircon (and sapphire) gems collected since the Middle Ages (Lacroix, 1901).
69 We sampled zircon from both the sedimentary deposits and the lavas in the same volcanic
70 field. The petrography and geochemistry of the lavas and their exotic accessory minerals, and
71 the determination of U-Pb ages and Hf isotopes of the zircons by *in situ* LA-(MC)-ICP-MS,
72 were investigated in order to constrain their age and origin.

73

74 **Geological setting**

75

76 The volcanism in the French Massif Central forms a significant part of the Neogene
77 peri-Alpine intra-continental volcanism. Several large and distinct recent volcanic domains
78 are located in the central and eastern parts of the French Massif Central (Wilson and Downes,
79 1991). Among them, the Velay is a well-individualised entity, where a succession of volcanic
80 eruptions of various types overlies the Variscan continental basement (Fig. 1). The eastern
81 part of the Velay domain is composed of basalts and more differentiated rocks ranging from
82 mugearites and benmoreites to trachytes and phonolites (Willemant et Treuil, 1983; Dautria
83 et al., 2004). Some rare carbonatite lavas are also present (Chazot et al., 2003). The age of
84 this volcanic activity ranges from 14 Ma to 6.5 Ma. In the western part, the Devès province is
85 characterized by abundant mafic lavas forming a large volcanic plateau, mostly emplaced
86 between 6 Ma and 0.6 Ma. The magmas belong to an alkaline series and the differentiated
87 rocks are either silica saturated or undersaturated. The studied samples were collected from
88 the north eastern margin of the Devès plateau (the Puy-en-Velay graben). A detailed
89 geological study of the Velay volcanism can be found in Mergoil and Boivin (1993).

90 The zircon-bearing rocks and the zircon gems from fluvial deposits were both
91 sampled in a narrow zone of few kilometres centred on the Mount Croustet volcano. Mafic
92 rocks containing visible zircon crystals were sampled at or near the "Les Brus" quarry. The
93 zircon gems come from the neighbouring "Les Brus" streams. Very precise locations of the
94 outcrops including detailed maps are provided in Carbonnel and Robin (1972) and Forestier
95 (1993).

96

97 **Petrology and geochemistry of the host rock**

98

99 The studied sample has a porphyritic microlithic texture. The groundmass is mostly
100 made of plagioclase and Fe-Ti oxides with altered glass. Microphenocrysts of clinopyroxene

101 and phenocrysts of amphibole are abundant. These latter are totally weathered but easily
102 recognisable from their characteristic shape.

103 Many xenoliths and xenocrysts are also present in this rock. Large garnets (up to 1.2
104 cm long) contain quartz, clinopyroxene and plagioclase inclusions. They are surrounded by
105 a reaction corona at the contact with the host magma. Granulite xenoliths can be up to 2 cm
106 long and contain quartz, orthopyroxene, clinopyroxene, plagioclase, biotite, oxide and
107 carbonate. Some small and rounded zircons are also present. Some of these xenoliths have
108 been partially disintegrated into the magma, and all their minerals are present as xenocrysts
109 in the rock sample.

110 Despite large amounts of xenoliths and xenocrysts, whole rock analysis of the studied sample
111 indicates a mugearitic composition (Table 1), with 51.2 wt. % of SiO₂ and only 3.6 wt. % of
112 MgO. It is alkali-rich, with Na₂O + K₂O = 8.2 wt. %. The rock is enriched in light rare earth
113 elements, with La content of 88 ppm and La/Yb ratio of 31, which is in the middle range of
114 mafic and intermediate alkaline rocks from the Velay province. However, Zr content is only
115 around 420 ppm, falling in the low range of the Velay volcanic rocks.

116

117 **Analytical techniques**

118

119 *Electron microprobe analyses*

120

121 Major element composition of clinopyroxene was determined by a CAMECA
122 CAMEBAX SX100 electron microprobe at the laboratoire Domaines Océaniques (University
123 of Brest) using a 15 kV and 10 nA beam focused to a spot of ~2 µm in diameter. The peaks
124 were counted for 10 s and backgrounds for 5 s. Light elements were counted first to preclude

125 loss by volatilization. Oxides, natural mineral and synthetic standards have been used. Matrix
126 corrections were performed by the PAP-procedure in the CAMECA software.

127

128 *In situ U-Pb dating*

129

130 Zircons were separated at the Laboratoire Magmas et Volcans (LMV), Clermont-
131 Ferrand (France) using standard techniques of crushing and sieving, followed by Wilfley
132 table, magnetic separation and heavy liquids before handpicking under binocular microscope.
133 They were mounted in epoxy disks and polished to expose crystal interiors.
134 Cathodoluminescence (CL) imaging was conducted at LMV using a JEOL JSM-5910LV
135 SEM. U-Th-Pb isotope data were measured by laser ablation inductively coupled mass
136 spectrometry (LA-ICP-MS) at LMV. Zircons were ablated under pure He using a Resonetics
137 Resolution M-50 system equipped with a 193 nm Excimer laser coupled to a Thermo Element
138 XR sector field ICP-MS. N₂ was supplemented to Ar and He carrier gas for sensitivity
139 enhancement (Paquette et al., 2014). The laser operated with a spot diameter ranging from 47
140 to 60 μm, a repetition rate of 3 Hz, and a fluence of 2.5 J/cm². Complete instrumental
141 operating conditions and data acquisition parameters are described in Table S1.

142 The Element was tuned to maximize the ²³⁸U intensity and minimize ThO⁺/Th⁺ (<1%)
143 using the NIST SRM 612 glass. After 10 pre-ablating cleaning pulses, background levels were
144 measured on-peak with the laser off for ~30 s, followed by ~60 s of measurement with the
145 laser firing and then ~10 s of washout time (see for detail Mullen et al., 2018 and Paquette et
146 al., 2019a). Reduction of raw data was carried out using the GLITTER® software package
147 (van Achterbergh et al., 2001). Isotope ratios were corrected for laser-induced and
148 instrumental mass fractionation via sample-standard bracketing using the GJ-1 zircon
149 reference material (Jackson et al., 2004). The ²³⁵U signal is calculated from ²³⁸U based on the

150 ratio $^{238}\text{U}/^{235}\text{U} = 137.818$ (Hiess et al., 2012). The 91500 zircon reference material
151 (Wiedenbeck et al., 1995) was analyzed along with the samples to independently monitor the
152 external precision and accuracy of the measurements. Common Pb and initial disequilibria
153 caused by $^{230}\text{Th}/^{238}\text{U}$ and $^{231}\text{Pa}/^{235}\text{U}$ fractionation in the zircon/melt system were corrected
154 according to Sakata et al. (2015) and Sakata (2018) method and related Microsoft Excel
155 spreadsheet. Concentrations of U, Th, and Pb were calculated by normalization to the certified
156 composition of GJ-1 reference material (Jackson et al., 2004). A $\text{Th}/\text{U}_{\text{melt}}$ ratio for the whole
157 rock of 4.1 is considered (see Table 1). The available fractionation factor of Pa/U in a zircon-
158 melt system of rhyolitic composition roughly shows agreement with a value of 2.9 ± 1.0
159 (Sakata, 2018), which was propagated into the calculations. The isotopic ratios and ages
160 together with all of the uncertainties were corrected from elemental and isotopic fractionation,
161 common Pb contribution and Th/U disequilibria correction. Weighted mean $^{206}\text{Pb}/^{238}\text{U}$ ages
162 and $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{238}\text{U}/^{206}\text{Pb}$ diagrams (Tera and Wasserburg, 1972) were generated
163 using Isoplot/Ex v. 2.49 software package by Ludwig (2001). Error ellipses for each point are
164 quoted at the 2σ level and integrate both internal and external uncertainties by quadratic
165 addition.

166

167 *In situ Hf isotopes*

168

169 Hafnium isotope measurements were performed with a Thermo Scientific Neptune
170 Plus multi-collector ICP-MS coupled to the Resonetics M50E 193 nm laser excimer system
171 (LMV). The MC-ICP-MS is equipped with 9 Faraday cups and amplifiers with $10^{11} \Omega$. Data
172 were collected in static mode (^{171}Yb , ^{173}Yb , ^{174}Hf , ^{175}Lu , ^{176}Hf - ^{176}Yb - ^{176}Lu , ^{177}Hf , ^{178}Hf ,
173 ^{179}Hf). The overall operating conditions and instrument settings are reported in Table S1 and
174 described in Moyen et al. (2017) and Paquette et al. (2017). Each LA-MC-ICPMS analysis

175 consisted of 20 s of gas background data followed by 40 s of ablation. With an integration
 176 time of 1 s, each analysis typically produced 36-40 isotopic ratios. Mass bias effects on Hf
 177 were corrected using an exponential law and a true value for $^{179}\text{Hf}/^{177}\text{Hf}$ of 0.7325 (Patchett
 178 and Tatsumoto, 1980; Patchett et al., 1981). Data were reduced in the following order and
 179 based upon user-selected background and sample integration intervals. Firstly, the mean
 180 signal intensity of the background was subtracted from the signal acquired during the laser
 181 firing onto the sample. β_{Yb} was determined using the $^{173}\text{Yb}/^{171}\text{Yb}$ measured during each
 182 analysis and the reference value for $^{173}\text{Yb}/^{171}\text{Yb}$ of 1.132685 (Chu et al., 2002; Fisher et al.,
 183 2011a). The ^{176}Yb and ^{176}Lu isobaric interference on ^{176}Hf were determined using $^{176}\text{Yb}/^{173}\text{Yb}$
 184 of 0.79618 and the measured interference-free ^{175}Lu mass assuming that $\beta_{\text{Lu}} = \beta_{\text{Yb}}$, and using
 185 the reference value for $^{176}\text{Lu}/^{175}\text{Lu}$ of 0.02655 (Vervoort et al., 2004).

186 Both determined ^{176}Lu and ^{176}Yb signals were subtracted from the total 176 signal
 187 intensity. The mass bias is calculated using the true $^{179}\text{Hf}/^{177}\text{Hf}$ ratio of 0.7325 (Patchett and
 188 Tatsumoto, 1980). Finally the interference corrected $^{176}\text{Hf}/^{177}\text{Hf}$ is evaluated according to the
 189 following equation:

$$190 \quad \left(\frac{^{176}\text{Hf}}{^{177}\text{Hf}}\right)_{\text{corrected}} = \left(\frac{^{176}(\text{Hf} + \text{Yb} + \text{Lu})_{\text{meas}} - ^{176}\text{Yb}_{\text{calc}} - ^{176}\text{Lu}_{\text{calc}}}{^{177}\text{Hf}_{\text{meas}}}\right) \times \left(\frac{M176}{M177}\right)^{\beta(\text{Hf})}$$

191 The $^{176}\text{Lu}/^{177}\text{Hf}$ for all samples were corrected for mass bias using β_{Hf} . Outlier rejection of
 192 the Hf isotopic ratio for each analysis were performed using a two-standard deviation
 193 criterion.

194 The $\varepsilon_{\text{Hf}}(t)$ value is defined as the deviation of the $^{176}\text{Hf}/^{177}\text{Hf}$ of a sample from the CHUR in
 195 parts per 10^4 . It is calculated according to the $\lambda^{176}\text{Lu}$ value = $1.865 \times 10^{-11} \text{ yr}^{-1}$ of Scherer et
 196 al. (2001) and the CHUR parameters ($^{176}\text{Hf}/^{177}\text{Hf} = 0.282793$; $^{176}\text{Lu}/^{177}\text{Hf} = 0.0338$) of Iizuka
 197 et al. (2015).

198 In order to certify the Hf isotope results, we checked our analytical reproducibility and
 199 correction procedure with systematic measurement of 20 replicates of four synthetic zircons

200 displaying increasing (Y+Lu) content (Supplementary Table S2). Mean and two standard
201 deviation values obtained on MUNZirc 0-2a ($^{176}\text{Hf}/^{177}\text{Hf} = 0.28213 \pm 3$), MUNZirc 1-2b
202 (0.28214 ± 2); MUNZirc 3-2b (0.28215 ± 2) and MUNZirc 4-2b (0.28216 ± 3) are fully
203 consistent with reference values (Fischer et al., 2011b).

204

205 **Analytical results**

206

207 *Mineral chemistry and origin*

208

209 Minerals have been analysed in three different occurrences: 1) in granulite xenoliths
210 contained in the studied volcanic rock; 2) in the groundmass of the same volcanic rock; 3) in
211 alluvium associated with the studied zircons. The analysed mugearite contains xenoliths of
212 mantle (lherzolites) and lower crustal rocks (granulites). The chemical composition of the
213 minerals analysed in the groundmass or in alluvium has been compared to the composition of
214 minerals from the xenoliths (granulites) contained in the rock, or from granulites and
215 peridotites found as xenoliths in other volcanic localities from the Massif Central.

216 The composition of clinopyroxene (Cpx) is highly variable and plot within three
217 different groups (Fig. 2a and Supplementary Table S3). One group has low TiO_2 and high
218 FeO_t . These compositions are similar to Cpx analysed in granulite xenoliths included in the
219 volcanic rock, and are identical to those encountered in granulite xenoliths from the French
220 Velay volcanic Province (Leyreloup, 1973). Cpx from granulites are found in thin section and
221 in alluvium, while mantle Cpx are found only in alluvium. The second group has also low
222 TiO_2 content but low FeO_t . These compositions are similar to diopside analysed in mantle
223 peridotite xenoliths from the French Massif Central (Chazot, unpublished data) and confirm
224 the presence of disaggregated mantle xenoliths in the mugearite. The third group of Cpx has

225 high TiO_2 and intermediate FeO_t contents. They are found both in the groundmass and in
226 alluvium and represent Cpx crystallized from the melt. They are high-Ca diopside, with
227 chemical compositions typical of Cpx from alkaline magmatic series (Leterrier et al., 1982).

228 Orthopyroxenes (Opx) have been analysed in two granulite xenoliths and in alluvium.
229 They define two different groups (Fig.2b and Supplementary Table S2). One group with
230 crystals from alluvium, has high silica content and low FeO_t , and is similar to Opx from
231 mantle xenoliths (Chazot, unpublished data). The second group contains minerals from the
232 granulite xenoliths in the studied rock as well as crystals from the alluvium. Their
233 composition, with low silica but higher FeO_t content, is similar to Opx analysed in granulite
234 xenoliths from the French Massif Central (Leyreloup, 1973). Accordingly, no Opx
235 composition corresponds to minerals crystallized from the melt.

236 Olivine has been analysed only in alluvium. Some grains have low FeO_t content (<11
237 wt.%) and are typical mantle olivine, similar to those analysed in mantle xenoliths, while
238 other grains have higher FeO_t and lower silica content and have crystallized from the melt.

239 Garnets have been analysed in alluvium, in granulite xenoliths from the mugearite as
240 well as from free grains in the groundmass. They all have chemical compositions in the range
241 of garnets analysed in granulite xenoliths from the French Massif Central (Leyreloup, 1973);
242 consequently, they all come from the lower crustal granulites.

243 Analyses of feldspar have been obtained from alluvium, from the groundmass and
244 from granulite xenoliths in the mugearite. In the groundmass, they are plagioclase ranging
245 from andesine to labradorite. In granulites and alluvium, the compositions are very variable
246 and range from plagioclase to sodic sanidine.

247 All these results show that the zircon-bearing mugearite contains minerals which
248 crystallized from the melt, or which have been sampled all along the way from the mantle to
249 the surface, and derived either from the upper mantle itself or from the continental crust.

250 Granulite xenoliths, as well as numerous minerals with granulitic composition, indicate a
251 strong interaction between the uprising melt and the lower crust.

252

253 *Zircon morphology*

254

255 All the crystals sampled for this study are large (>200 μm) and red-coloured. Isolated
256 crystals from alluvium deposits are the largest, up to several millimetres long. These are
257 always euhedral to subhedral with frequent slightly rounded pyramids and sometimes
258 corroded faces (Fig. 3A). In cathodoluminescence, these large crystals display an igneous-
259 type fine sector oscillatory zoning which is strictly parallel to the crystal faces (Fig. 3B).
260 Zircon rims, although locally rounded by corrosion or abrasion, display sharp outlines
261 without alteration features.

262 Two different morphologies occur among the zircon crystals separated from the
263 mugearite samples. A scarce population is composed of euhedral to subhedral zircon grains,
264 sometimes broken and displaying a broadly similar morphology to the previous ones (Fig.
265 3C). They are also characterized by sector zoning but their sides are often paler than their
266 centres in the cathodoluminescence images. These crystals can be outlined by a fine alteration
267 rim (Fig. 3D). The main population of zircons from the mugearite is composed of spherical to
268 ovoid crystals. In cathodoluminescence, these round grains are characterised by a sector (Fig.
269 3E and F) or an oscillatory zoning (Fig. 3G), which remains clearly visible. This zoning is
270 always incomplete and crosscut the crystal limits (Fig. 3E, F and G). This spherical shape
271 probably results from the chemical disequilibrium between the zircon and the mafic melt,
272 causing dissolution of previously euhedral or subhedral zoned zircon crystals. Similarly to the
273 euhedral population, a thin and light-coloured rim (Fig. 3G), maybe related to a fluid
274 interaction stage, surrounds the whole grains. In some of these rounded grains, the presence

275 of cores, located close to the centre of the crystals, is suspected (Fig. 3H). Visible inclusions
276 were not detected; this is consistent with the moderate U and Th contents of the zircons
277 megacrysts and their use as gemstones in jewellery.

278

279 *U-Pb geochronology*

280

281 The dataset for ESP-2 zircon megacrysts from Les Brus stream comprises 55 laser
282 spots performed on thirteen crystals. 52 pooled measurements on twelve of the thirteen
283 analysed zircons yield a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ concordant age of 2.629 ± 0.024 Ma (Fig.
284 4A and Table 2) after correcting for the common Pb and Th/U disequilibria. The three
285 remaining analyses on the last zircon grain yield a significantly older age of 10.06 ± 0.39 Ma.
286 Both ages, 10.06 Ma and 2.63 Ma, are interpreted as reflecting the crystallization ages of two
287 distinct episodes of zircon formation, the younger one being better sampled. The U content of
288 the main population ranges from 39 to 664 ppm but is variable from grain to grain and
289 particularly from zone to zone inside each grain up to a factor of five. Th/U ratios of 1.2 ± 0.5
290 (1 standard deviation) are systematically high and typical of magmatic zircons from alkaline
291 volcanic environments (Paquette and Mergoïl-Daniel, 2009; Hurai et al., 2012 and 2013,
292 Huraiova et al., 2017; Paquette et al., 2019b).

293 A second set of 78 spots was performed in 49 zircon grains separated from the ESP-1
294 mugearite. These zircon crystals are mostly fully rounded, about ten are broken with sharp
295 angles between faces, consequently their original shape cannot be distinguished, and two are
296 subhedral. Among these different morphologies, the U-Pb dating results can be easily divided
297 into two groups. The first one comprised rounded, broken and euhedral zircon grains yielding
298 together a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ concordant age of 2.579 ± 0.033 Ma (Fig. 4B and Table
299 2) corrected for the common Pb and Th/U disequilibria. Similarly to ESP-2 zircon

300 megacrysts, U and Th content are highly variable, ranging from 75 to 1340 ppm and from 50
301 to 2706 ppm, respectively. U content may increase by a factor of four between dark and light
302 cathodoluminescence zones inside each grain. These Pliocene zircons provide systematically
303 high Th/U ratios of 1.4 ± 0.6 (1 standard deviation), which agree with a similar magmatic
304 environment as the ESP-2 zircon megacrysts sampled in the neighbouring stream. The
305 subhedral morphology of some synchronous grains possibly represents crystals that have been
306 better preserved from dissolution. The second dated group is also composed of rounded
307 zircons but yield an older lower intercept age of 284.7 ± 3.1 Ma (Fig. 4C and Table 2). Lower
308 U content and Th/U ratios than those of the Pliocene zircons, range from 53 to 538 ppm and
309 from 0.04 to 1.0, respectively, with a mean value for the latter of 0.5 ± 0.3 . We never
310 observed any Permian core-Pliocene rim combination, each zircon type being consistent in
311 age.

312

313 *Lu-Hf isotopes*

314 All the Lu-Hf analyses of ESP-2 zircon megacrysts yield super-chondritic initial $\epsilon_{(\text{Hf})}$
315 values (Supplementary Table S4 and Fig. 5) ranging from +7.0 to +12.9 with a mode of +9.2
316 ± 1.4 (1 standard deviation). $\epsilon_{(\text{Hf})}$ values may evolve significantly above uncertainties between
317 different spots in several individual grains. Similar variability within single grains and
318 between different zircon megacrysts occurring in comparable geological context has already
319 been reported (e.g. Huraiová et al., 2017; Paquette et al., 2019b). This has been correlated
320 with heterogeneous and metasomatized mantle, affected only marginally by recycled
321 continental crust. Depleted Mantle Hf model ages $T_{\text{DM}}(\text{Hf})$ are very consistent around a mean
322 value of 348 ± 56 Ma (1 standard deviation). The Miocene #A01 zircon grain provides an
323 initial Hf isotope composition which is at the lower limit of those of the Pliocene crystals.

324 In the ESP-1 mugearite sample, both zircon types identified by U-Pb dating are still
325 valid. The Pliocene zircon population from the lava displays super-chondritic initial $\epsilon_{(\text{Hf})}$
326 values (Supplementary Table S4 and Fig. 5) ranging from +7.2 to +14.8 with a mode of +10.1
327 ± 1.8 (1 standard deviation). This variability is interpreted similarly than for ESP-2 zircon
328 megacrysts and demonstrates a significant heterogeneity of the mantle source, possibly
329 weakly affected by contribution of continental crust. $T_{\text{DM}}(\text{Hf})$ model ages are also consistent
330 around a mean age of 307 ± 68 Ma. Consequently, all the Neogene zircon grains, both those
331 from the stream sediment and those extracted from the mugearite, provide similar super-
332 chondritic initial $\epsilon_{(\text{Hf})}$ values. This implies that the parental magmas of the source rocks for
333 these zircon grains were juvenile and formed from a depleted mantle source characterized in
334 both cases by Carboniferous $T_{\text{DM}}(\text{Hf})$ model ages. The second age group comprising the
335 Permian zircons yield significantly different Hf initial isotope compositions with slightly
336 super-chondritic to mostly sub-chondritic initial $\epsilon_{(\text{Hf})}$ values ranging from +0.7 to -14.1. These
337 negative initial $\epsilon_{(\text{Hf})}$ values document that the zircon parental rocks were derived from sources
338 dominated by recycled crustal material. In other words, the Neogene zircon group is mantle-
339 derived whereas the Permian one is continental crust-derived.

340

341 **Discussion: origin and formation of the zircons**

342

343 On the basis of petrological observations, in-situ geochemistry on minerals, U-Pb
344 geochronology, and Hf isotopic data, the origin and formation of the zircon crystals can be
345 outlined as follows.

346 First, Hf isotopes demonstrate that all Neogene zircons crystallized from a melt
347 derived from a depleted mantle source. However, this depleted mantle source is different from

348 the depleted mantle sources for mid-ocean ridge basalts which are characterized by initial $\epsilon_{(\text{Hf})}$
349 values around +18. Rather, the intermediate initial $\epsilon_{(\text{Hf})}$ values ranging from +7.0 to +14.8
350 suggest a metasomatized sub-continental lithospheric mantle. Subsequent enrichment of the
351 mantle in incompatible trace elements such as HFSE, Th and U (Aulbach et al., 2008;
352 Kalfoun et al., 2002; Pfänder et al., 2012) by carbonatite metasomatism can be considered
353 beneath the French Massif Central (Chazot et al., 2003; Paquette and Mergoïl-Daniel, 2009;
354 Chazot and Mergoïl-Daniel, 2012). This is probably responsible for some variability of the
355 $\epsilon_{(\text{Hf})}$ values, as well as the possibility of a weak continental crust assimilation. Such a
356 metasomatized lithospheric mantle source is favoured beneath the Velay area. According to
357 Carboniferous $T_{\text{DM}}(\text{Hf})$ model ages, this lithospheric mantle may have been fertilized during
358 the Cenozoic from an asthenospheric mantle fragment formed during Variscan times.

359 Important modifications in the mantle beneath the French Massif Central during
360 Variscan times have already been demonstrated from mantle studies. Hf isotopes in
361 clinopyroxene from mantle xenoliths lie along a 360 Ma “isochron” (Wittig et al., 2006;
362 2007), indicating extensive partial melting of a mantle wedge becoming the new lithospheric
363 mantle in this area. Nd isotopes are consistent with these observations. Available data on
364 mantle clinopyroxene from various volcanic centres in the Massif Central (Downes and
365 Dupuy, 1987; Downes et al., 2003; Touron et al., 2008; Xu et al., 1998; Yoshikawa et al.,
366 2008; Zangana et al., 1997) plot on a similar “isochron” from which the slope defines an age
367 of 345 Ma (Fig. 6). The initial $^{143}\text{Nd}/^{144}\text{Nd}$ value of the isochron is 0.51267, very close to the
368 Nd isotopic value of the depleted mantle recalculated at the same age ($^{143}\text{Nd}/^{144}\text{Nd}_i = 0.51263$,
369 Gale et al., 2013). This confirms the conclusions obtained by Wittig et al. (2006 and 2007)
370 that the depleted mantle beneath the French Massif Central underwent partial melting during
371 or at the end of the Hercynian orogeny. This part of the mantle was then incorporated into the
372 lithosphere and subsequently metasomatized and melted during Cenozoic and Quaternary

373 volcanism. The chemical evolution of asthenospheric mantle towards lithospheric mantle may
374 occur during or at the end of Hercynian continental collision. Detachment and sinking of the
375 lithospheric mantle with or without a portion of the lower crust results in asthenosphere
376 upwelling and cooling to replace the missing mantle root (Kay and Kay, 1993; Gutiérrez-
377 Alonso et al., 2011). Partial melting during upwelling of the asthenospheric mantle is often
378 advocated to explain heat and magma transfer to the crust during and at the end of an orogeny
379 (e.g. Schott and Schmelting, 1998; Lustrino, 2005; Li et al., 2016). This model is in good
380 agreement with the Hercynian model ages calculated with Hf isotopes in our zircons, and
381 provides more evidence that these zircons crystallized from melts formed in the lithospheric
382 mantle.

383

384 The origin of zircon megacrysts in mafic or intermediate volcanic rocks has been
385 debated for a long time. Old zircons can be sampled throughout the crust during magma
386 ascent. In that case, they are older than the volcanic host-rock and genetically unrelated to the
387 volcanic event. The Permian zircon population sampled in the ESP-1 mugearite and dated at
388 285 Ma, can be related to the late metamorphic evolution of the Variscan belt, when felsic
389 granulites crystallized in the lower continental crust (e.g. Pin and Vielzeuf, 1983; Paquette et
390 al., 2003). Enclaves from Variscan granulitic metasediments scavenged by basaltic volcanism
391 in the northern Massif Central were already sampled and studied (Rossi et al., 2006).

392 Several studies have demonstrated that volatile fluxes interacting with the lithospheric
393 mantle in an environment enriched in incompatible elements promote the generation of felsic
394 melts by low degree partial melting (Aspen et al., 1990; Hinton and Upton, 1991). Large
395 zircon grains can crystallize from these melts within small dykes in the mantle (Upton et al.,
396 1999; Pin et al., 2001 and 2006; Sutherland et al., 2002). Despite such lithologies have rarely
397 been identified in mantle xenoliths worldwide (Shimizu et al., 2004; Avanzinelli et al, 2020),

398 minerals from these rocks could be scavenged by ascending magmas and carried up to the
399 surface. In those situations, one can expect associated minerals to be transported along with
400 the zircons. Furthermore, the age of the zircons must be older than the age of the eruption and
401 in some cases, the zircons should be genetically unrelated to the host magmas. Our studied
402 samples contain many xenocrysts (garnet, orthopyroxene, clinopyroxene), but none of these
403 minerals can be associated with felsic rocks formed in the mantle and we have no indication
404 that the studied zircons were formed at mantle depth. Xenocrysts contained in our samples
405 have a crustal origin, as is probably the case for the associated zircons.

406 Paquette and Mergoïl-Daniel (2009) reported the occurrence of zircon-bearing
407 nepheline syenite xenoliths formed from differentiated mantle-derived magmas in
408 neighbouring areas from the French Massif Central. These felsic magmas can be derived
409 through fractional crystallization in crustal magma chamber from ascending alkaline mafic
410 magmas formed in the lithospheric mantle. Such alkaline felsic melts may generate the large
411 volumes of zircon megacrysts discovered worldwide in secondary deposits within continental
412 basaltic provinces (e.g. Garnier et al., 2004 and 2005). Scavenging of the crystallizing melt
413 during recharge of the magma chamber by a new batch of more mafic magma can explain the
414 presence of these zircon megacrysts in our studied samples. The discovery of sodic sanidine
415 crystals associated with the zircons in the alluvium samples is in good agreement with the
416 presence of crystallizing syenitic bodies in the crust, through which the mafic magma passed,
417 en route to the surface. This kind of shallow magma chamber with a zircon-rich syenitic
418 magma has already been described in Quaternary Eifel volcanoes of Germany where zircon
419 are mostly contained in lapilli-sized plutonic ejecta (Schmitt et al., 2017).

420

421 With more than 51% SiO₂ and less than 60 ppm Ni, the host magma is far from a
422 primitive melt in equilibrium with mantle lithology. This indicates that this magma has

423 already undergone crystal fractionation at greater depth in the crust. It is probably during this
424 earlier differentiation episode that mantle xenoliths were partly disaggregated and mantle
425 minerals such as olivine, Opx and Cpx incorporated into the magma. The presence of
426 granulite xenoliths, and xenocrysts from granulite lithologies, may indicate that differentiation
427 occurred at great depth in the lower crust. Magma transfer towards the surface would then
428 follow a similar pathway previously used and bring the magma into a shallow syenitic magma
429 chamber where zircon and sanidine were incorporated into the ascending magma before
430 eruption.

431

432 Zircons from both stream sediments and lavas originated from a common source by
433 similar petrogenetic processes and crystallized synchronously within uncertainties during a
434 single event in the mid-upper Pliocene (Piacenzian-Gelasian boundary). A comparable U-Pb
435 age of about 2.51 Ma (uncorrected from Th-Pa disequilibria; Wiedenbeck et al., 1995;
436 Cocherie et al., 2009) was obtained on a quite similar zircon megacryst from the National
437 Muséum d'Histoire Naturelle in Paris and recorded as belonging to a deposit at Croustet
438 (Haute Loire, France), probably close to our sampling area. Furthermore, all the zircon
439 crystals in the sedimentary deposit are large and euhedral without any partial dissolution
440 illustrating a lack of - or very limited - dissolution time and disequilibrium between the
441 mineral and the magma. In contrast, most zircons from the lava are rounded which edges that
442 crosscut the zoning of the crystals. A very thin luminescent rim at the border of the grains
443 coupled with irregular small dissolved gulfs can be linked to a late fluid activity during the
444 cooling of the lava flow. All these features are related to the incomplete equilibration between
445 zircon xenocrysts, or zircon-bearing xenoliths, and the mafic melt. Consequently, the major
446 difference between the zircons of both samples is essentially related to their extraction from
447 the felsic magmas and uplift to the surface, which preserves the euhedral megacrysts and not

448 the rounded zircons. These latter are obviously connected to mafic lava flows scavenging, and
449 partly dissolving, both mantle and lower-crustal enclaves. The rare subhedral zircon crystals
450 discovered in the mugearite sample were probably shielded in larger felsic xenoliths from the
451 corrosion of the mafic magma.

452 The preserved morphology of zircon megacrysts from sedimentary deposits implies a
453 different sampling and uplift mechanism, without any dissolution by interaction with a mafic
454 magma. Basically, the scavenging of enclaves and their uplift may also be performed by more
455 felsic lavas. Trachyte domes preserving deep xenoliths and xenocrysts are known in the
456 French Massif Central (e.g. Brousse and Varet, 1966). A major drawback of this hypothesis is
457 that zircon megacrysts are sampled in secondary deposits, which would imply that the
458 trachytes would be entirely dismembered by erosion. Indeed, the Devès volcanic domain is
459 essentially basaltic in composition, without significant occurrence of felsic lavas.
460 Furthermore, there is no evidence and geologic reason for global differential erosion of the
461 felsic lavas and preservation of synchronous mafic ones. Consequently, this hypothesis seems
462 rather unlikely. Another opportunity to restrict the time of interaction between lavas, xenoliths
463 and xenocrysts is the occurrence of phreatomagmatic eruptions. Such events do not preserve
464 significant volcanic structures 2.6 Myrs after eruption (e.g. Paquette et al., 2019b), whereas
465 sediments related to ancient maar craters often contain large amounts of euhedral zircon
466 megacrysts (e.g. Hurai et al., 2010; 2012 and 2013; Huraiova et al., 2017). In our present
467 knowledge, we consider that hypothesis as the most likely. We propose a two-steps process in
468 which a magma containing mantle and lower crust xenoliths was scavenging a zircon-bearing
469 felsic assemblage in a crystallizing shallow magma chamber during its ascent. The first part of
470 the eruption was characterized by phreatomagmatic activity and a very brief interaction
471 between the magma and the zircon megacrysts. These zircons can thus preserve their euhedral
472 shape. The second part of the eruption was less violent, with emplacement of a lava flow and

473 longer interaction between the magma and the zircon megacrysts. Those zircons acquire their
474 rounded shape during this effusive part of the eruption.

475

476 **Conclusion**

477

478 In the south eastern Devès volcanic domain of the French Massif Central, we report
479 the rare occurrence of zircon megacrysts in, both, alluvium of a small stream and within a
480 neighbouring alkaline lava. The crystals from the alluvial deposit display a subhedral
481 morphology while the zircons in the lavas are completely rounded.

482 Both zircon types share common U-Pb ages of about 2.6 Ma, synchronous within
483 uncertainties, and mantle-derived super chondritic Hf isotopic signatures.

484 The analysed mugearite also contains xenoliths of mantle and lower crustal rocks
485 (granulites). The chemical composition of the minerals analysed in the groundmass is similar
486 to the composition of those from the xenoliths (granulites), or from granulites and peridotites
487 found as xenoliths in other volcanic localities from the Massif Central. The mugearite lava
488 also preserves rounded Variscan zircon crystals dated at 285 Ma and originating from lower
489 continental crust.

490 In spite of their mantle origin recorded by in situ Hf isotopes, the Pliocene zircons
491 cannot have crystallized directly in the mantle or in mantle-derived mafic melts. We propose
492 that these zircons crystallized from felsic melts produced during extensive differentiation of
493 mantle-derived magmas within the continental crust. Finally, a last more mafic magma batch
494 en route to the surface scavenged upper mantle and lower crust xenoliths as well as zircon
495 xenocrysts from the felsic melts during its ascent.

496 Many continental volcanic basaltic fields are associated to alluvial deposits containing
497 Cenozoic zircon megacrysts as well as sapphire gemstones. Such occurrences are reported in

498 the western and central European volcanic province which includes the French Massif
499 Central. This would imply that significant volumes of felsic differentiate issued from mantle-
500 derived basaltic magmas, and consequently huge amounts of zircon megacrysts, may have
501 been produced in the alkaline volcanic domains at the global scale.

502

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513

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757

758 **Figure Captions:**

759

760 **Figure 1.** Geological sketch map of the sampled area. 1: Variscan basement; 2: Cenozoic
761 grabens and sedimentary basins; 3: Cenozoic and Quaternary volcanism; blue square: sampled
762 area.

763

764 **Figure 2.** a: FeO_t vs. TiO₂ diagram (weight %) displaying the chemical composition of the
765 clinopyroxenes from the samples. b: FeO_t vs. SiO₂ diagram (weight %) displaying the
766 chemical composition of the orthopyroxenes from the samples. In both diagrams, the light and
767 dark gray arrays represent typical compositions of these minerals in granulite (Leyreloup,
768 1973) and mantle xenoliths (Chazot, unpublished data) from the Massif Central, respectively.

769

770 **Figure 3.** Scanning Electron Microscope and binocular microscope images of the zircon
771 crystals; A: subhedral zircons megacrysts from ESP-2 sample stream; B: fine oscillatory
772 zoning parallel to the zircon faces (ESP-2); C: euhedral zircon crystal from the lava flow
773 (ESP-1); D: sector-zoned and euhedral zircon crystal from the lava flow (ESP-1) partly
774 outlined by a fine alteration rim; E and F: sector zoned and rounded Pliocene zircons from the
775 lava flow (ESP-1); G: oscillatory zoned and rounded Permian zircon from the lava flow (ESP-
776 1), partly outlined by a fine alteration rim; H: inherited core in a Permian rounded zircon from
777 the lava flow (ESP-1).

778

779 **Figure 4.** Tera and Wasserburg (1972) $^{238}\text{U}/^{206}\text{Pb}$ vs. $^{207}\text{Pb}/^{206}\text{Pb}$ diagrams of the dated
780 samples A: alluvium deposit from Les Brus stream (ESP-2), mugearite lava flow (ESP-1) B:
781 Pliocene zircons and C: Permian zircons.

782

783 **Figure 5.** Hf-isotope evolution versus time diagram for zircons of the alluvium (blue circles)
784 and of the mugearite lava flow (red circles).

785

786 **Figure 6.** Sm-Nd isochron diagram for mantle clinopyroxenes from various volcanic centres
787 of the Massif Central (see text for references).

788

789 **Tables content:**

790

791 **Table 1.** Major and trace element content of the ESP-1 mugarite sample.

792

793 **Table 2.** Zircon U-Th-Pb data from the dated samples obtained by in *situ* Laser Ablation ICP-
794 MS.

795 **Supplementary Table S1.** Detail of analytical conditions for In situ U-Th-Pb and Lu-Hf
796 isotope analysis.

797

798 **Supplementary Table S2.** Lu-Hf isotope data obtained by in *situ* Laser Ablation MC-ICP-
799 MS on synthetic zircons doped with hafnium and rare earth elements (Fischer et al., 2011b).

800

801 **Supplementary Table S3.** Chemical composition of minerals of the alluvium and mugarite
802 sample.

803

804 **Supplementary Table S4.** Zircon Lu-Hf isotope data obtained by in *situ* Laser Ablation MC-
805 ICP-MS.

806