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

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Abstract

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

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

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

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

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

Introduction

Experiments on zircon solubility saturation have demonstrated that crystallisation of this mineral in basalts is unlikely (Watson, 1979; Watson and Harrison, 1983) since several percent of $Zr0_2$ content may be dissolved in the melt at about 1300°C before any crystallisation of zircon (Guo et al., 1996). Consequently, primary crystallisation of zircon in most mafic volcanic melts is rare. Nevertheless, large amounts of zircons and sometimes

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

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

Geological setting

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

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

Petrology and geochemistry of the host rock

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

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

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

Despite large amounts of xenoliths and xenocrysts, whole rock analysis of the studied sample indicates a mugearitic composition (Table 1), with 51.2 wt. % of SiO_2 and only 3.6 wt. % of MgO. It is alkali-rich, with $Na_2O + K_2O = 8.2$ wt. %. The rock is enriched in light rare earth elements, with La content of 88 ppm and La/Yb ratio of 31, which is in the middle range of mafic and intermediate alkaline rocks from the Velay province. However, Zr content is only around 420 ppm, falling in the low range of the Velay volcanic rocks.

Analytical techniques

Electron microprobe analyses

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

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

In situ U-Pb dating

Zircons were separated at the Laboratoire Magmas et Volcans (LMV), Clermont-Ferrand (France) using standard techniques of crushing and sieving, followed by Wilfley table, magnetic separation and heavy liquids before handpicking under binocular microscope. They were mounted in epoxy disks and polished to expose crystal interiors. Cathodoluminescence (CL) imaging was conducted at LMV using a JEOL JSM-5910LV SEM. U-Th-Pb isotope data were measured by laser ablation inductively coupled mass spectrometry (LA-ICP-MS) at LMV. Zircons were ablated under pure He using a Resonetics Resolution M-50 system equipped with a 193 nm Excimer laser coupled to a Thermo Element XR sector field ICP-MS. N_2 was supplemented to Ar and He carrier gas for sensitivity enhancement (Paquette et al., 2014). The laser operated with a spot diameter ranging from 47 to 60 µm, a repetition rate of 3 Hz, and a fluence of 2.5 J/cm^2. Complete instrumental operating conditions and data acquisition parameters are described in Table S1.

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

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

In situ Hf isotopes

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

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

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

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

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

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

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

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

Analytical results

Mineral chemistry and origin

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

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

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

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

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

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

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

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

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

Zircon morphology

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

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

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

U-Pb geochronology

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

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

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

Lu-Hf isotopes

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

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

Discussion: origin and formation of the zircons

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

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

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

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

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

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

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

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

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

With more than 51% SiO_2 and less than 60 ppm Ni, the host magma is far from a primitive melt in equilibrium with mantle lithology. This indicates that this magma has

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

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

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

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

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

Conclusion

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

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

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

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

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

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

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Figure Captions:

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

Figure 2. a: FeO_t vs. TiO_2 diagram (weight %) displaying the chemical composition of the clinopyroxenes from the samples. b: FeO_t vs. SiO_2 diagram (weight %) displaying the chemical composition of the orthopyroxenes from the samples. In both diagrams, the light and dark gray arrays represent typical compositions of these minerals in granulite (Leyreloup, 1973) and mantle xenoliths (Chazot, unpublished data) from the Massif Central, respectively.

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

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

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

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

Tables content:

Table 1. Major and trace element content of the ESP-1 mugearite sample.

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

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

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

Supplementary Table S3. Chemical composition of minerals of the alluvium and mugearite sample.

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