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► **To cite this version:**

De Hoÿm de Marien Luc, Pavel Pitra, Marc Poujol, Nathan Cogné, Florence Cagnard, et al.. Complex geochronological record of an emblematic Variscan eclogite (Haut-Allier, French Massif Central). *Journal of Metamorphic Geology*, In press, 10.1111/jmg.12733 . insu-04101059v1

**HAL Id: insu-04101059**

**<https://insu.hal.science/insu-04101059v1>**

Submitted on 19 May 2023 (v1), last revised 5 Jun 2023 (v2)

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**Complex geochronological record of an emblematic Variscan eclogite (Haut-Allier, French Massif Central)**

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Running title: Petrochronology in polymetamorphic Variscan eclogite, French Massif Central.

Key words: Variscan belt, eclogite, petrochronology, zircon, apatite.

**ABSTRACT**

Two eclogite samples from the Haut-Allier record a prograde evolution from ~20 kbar, 650 °C to 750 °C, 22–23 kbar followed by heating up to 850–875 °C and partial melting.

Incipient decompression in high-pressure granulite facies conditions (19.5 kbar, 875 °C) was

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1002/jmg.12733

followed by exhumation to high-temperature amphibolite facies conditions (<9 kbar, 750–850 °C). Following a detailed geochemical, petrological and geochronological investigation using trace-element data and LA-ICP-MS U-Pb dating of zircon, apatite and rutile, the eclogites reveal an Ordovician (c. 490 Ma) rifting event followed by Devonian (c. 370–360 Ma) subduction and Carboniferous (c. 350 Ma) exhumation in this part of the French Massif Central (FMC). The previously proposed Silurian age for the subduction, that strongly influenced many tectonic models, is definitively rejected. In the light of other geological data from the FMC, including the lithological and geochemical zoning of calc-alkaline Devonian volcanism, we propose a southward polarity of the subduction and question the very existence of the so-called Massif Central Ocean. Furthermore, we infer that following subduction, the eclogites were relaminated to the upper plate and exhumed at the rear of the magmatic arc pointing to similarities with the geodynamics of the Bohemian Massif.

The petrochronological record of zircon is particularly complex. Metamorphic zircon with clear eclogitic REE patterns (no Eu anomaly, flat HREE) and inclusions (garnet, rutile, omphacite) shows concordant apparent ages that spread from c. 380 down to c. 310 Ma. This apparent age pattern strongly contrasts with the well-defined age of apatite and rutile of c. 350 Ma. Apparent zircon ages younger than 350 Ma unequivocally testify that zircon can recrystallize outside the conditions of the eclogite facies, which resets the U–Pb while preserving an apparent eclogitic signature. Local fractures filled by analcite, thomsonite, plagioclase and biotite testify to late interaction of the eclogites with alkaline fluids at relatively low temperatures. This interaction, possibly at c. 310 Ma or later, could lead to the recrystallisation of zircon while leaving apatite unaffected.

## 1. INTRODUCTION

Eclogites in ancient orogens are commonly interpreted as the result of past subduction prior to continental collision (Miyashiro, 1961, 1972; Godard, 2001; Ernst and Liou, 2008). Although eclogites may occasionally form during continental collision, dating eclogite-facies metamorphism commonly provides the best approximation of the onset of convergence, which has critical implications for the formulation of tectonic scenarios.

The French Massif Central (FMC) is one the Western European Palaeozoic massifs and belongs to the high-grade core of the Variscan orogen (e.g., Matte, 1986; Lardeaux et al., 2014; Martínez Catalán et al., 2021; Fig. 1a) that resulted from the convergence between Laurussia and Gondwana. During this convergence, terranes previously rifted from the Gondwana margin during the opening of Ordovician oceanic domains (von Raumer and Stampfli, 2008; Nance and Linnemann, 2008) were amalgamated back to Gondwana (Matte, 1986; Kroner and Raumer, 2013; Stampfli et al., 2013). This event is recorded in the Massif Central by high-grade metamorphism including eclogite-facies metamorphism (e.g., Matte, 1991) that developed in terrains from the thinned Gondwanan continental margin (Lardeaux, 2014). The FMC consequently bears strong similarities with the other massifs of the European Variscan belt (e.g., Matte, 1986; Lardeaux et al., 2014; Martínez-Catalán et al., 2021). However, the necessary correlations at the scale of the orogen are hampered by the lack of modern geochronological data in the FMC. There, only a limited number of studies focused on the geochronology of the eclogites. These studies often adopted different methodologies and proposed different ages for the eclogite-facies metamorphism.

Several studies proposed a Silurian age for the subduction event in the FMC (Pin and Lancelot, 1982; Ducrot et al., 1983; Paquette et al., 1995). This age was inferred principally from U-Pb results obtained by dissolution of a zircon population. A more recent similar

Silurian age obtained by U-Pb LA-ICP-MS dating on zircon has been attributed to the eclogite-facies event only by analogy with the results from the three aforementioned studies (Berger et al., 2010). However, two other recent studies, carried out using a multi-mineral and multi-chronometer approach, and based on a well-constrained petrological framework, rather suggests subduction during the Devonian in the southern part of the FMC (Lotout et al., 2018; 2020). Furthermore, although recent studies have argued that the Silurian ages should be abandoned in the whole FMC given their technical limitations and the complexity of the geochronological record of the rocks (Paquette et al., 2017, Lotout et al., 2018, 2020; Benmammam et al., 2020), both ages (i.e. Silurian and Devonian) are still used in the literature (e.g., Vanderhaeghe et al., 2020; Martínez Catalán et al., 2021). Taking the Silurian ages and their regional distribution into account (Fig. 1) suggests a southward younging of the eclogite-facies metamorphism and a very long-lasting subduction event (430 Ma to 360 Ma; Fig. 1b), with important consequences for the tectonic models and correlation of the FMC with other European Variscan massifs. A reassessment of the eclogite outcrops from which this Silurian age is derived is therefore essential.

This contribution presents geochronological results obtained in two eclogite samples and one closely associated felsic diatexite (Supporting Information 1) from a locality where a previous study suggested a Silurian age for the eclogite-facies metamorphism. U-Pb isotopes of zircon, apatite and rutile separated from the eclogites are studied *in-situ* using LA-ICP-MS. The isotopic results are interpreted in the framework of an already published petrological study (de Hoÿm de Marien et al., 2020) complemented by new petrographic data and the determination of micro-inclusions in zircon by Raman spectroscopy, together with trace-element data in zircon and rutile.

## 2. GEOLOGICAL BACKGROUND

The FMC is interpreted as a Palaeozoic nappe stack intruded by syn- to post-orogenic granitoids, dissected by late-orogenic migmatitic domes and unconformably overlain by Upper Carboniferous sediments (e.g., Faure et al., 2009; Lardeaux, 2014). The nappe stack involves several units established from lithological association and tectonometamorphic consideration (Ledru et al., 1989). The studied area belongs to the eastern part of the FMC where the following succession of units is generally recognized from north to south (Fig. 1b):

The northernmost *Morvan and Brévenne units* consist of low-grade Devonian volcano-sedimentary series respectively interpreted as arc and back-arc series (e.g., Delfour, 1989; Leloix et al., 1999). In the northerly Morvan area, the series include fossiliferous carbonates of the upper Devonian, and andesites (Fig. 1b). In the southerly Brévenne area, the volcanic rocks belong to a typical bimodal spilite-keratophyre association including pillow lavas indicating the marine character of the series (Peterlongo, 1958; 1970). The rocks are of Famennian age (c. 360 Ma) and have been interpreted as a suprasubductional complex emplaced in a settings involving incipient rifting of a volcanic arc built upon a thin continental crust (Pin and Paquette, 1997). The regional lithological zoning of the volcanic rocks and the associated southward decreasing enrichment in incompatible elements suggest that the Devonian subduction was dipping to the south (Pin et al., 1982; Pin and Paquette, 2002). The metamorphic basement of the Morvan and Brévenne area includes (undated) eclogites (Godard, 1990) and is attributed to the Upper Gneiss Unit (e.g., Faure et al., 1997; Leloix et al., 1999). This is however open to discussion and this basement is set apart in Fig. 1b.

*The Upper Gneiss Unit* (UGU) consists of para- and orthogneisses, as well as metamorphosed basic and ultrabasic rocks. The metasediments were deposited during the Ediacaran (Chelle-Michou et al., 2017). The base of the UGU is composed of a bimodal

igneous association referred to as the Leptyno-Amphibolitic Complex (LAC; Forestier, 1961, Santallier et al., 1988) typically considered to record a Cambro-Ordovician rifting event (e.g. Lotout et al., 2017; Pin and Lancelot, 1982; Pin and Marini, 1993). The mafic rocks, locally associated with garnet peridotites (e.g. Gardien et al., 1988; 1990; Lasnier, 1968, 1971), are metamorphosed in the eclogite facies at high temperature (e.g., Lardeaux et al., 2001; Mercier et al., 1989; Nicollet et al., 1977, 1978). The eclogite facies is interpreted in terms of subduction of the so-called Massif-Central Ocean (e.g. Matte, 1986) and consequently the LAC is considered as a suture zone (e.g. Matte and Burg, 1981). As mentioned in the introduction, the dating of the subduction event is controversial with inferred ages between 420 Ma and 360 Ma. The subsequent decompression of the LAC is recorded by a granulitic overprint in various lithologies (e.g. Burg and Leyreloup, 1989; Forestier et al., 1973; Lasnier, 1968, 1977; Marchand, 1974; Mercier et al., 1991) and tentatively dated at c.360 Ma (Schulz, 2014). Further decompression is associated with the emplacement of granitoids at c. 315 Ma (Gardien et al., 2011; Laurent et al., 2017) and final cooling is constrained between 300 and 275 Ma (Gardien et al., 2011).

A *Middle Allochthon Unit*, so far only reported in the Najac area (Fig. 1b), is tentatively recognized by comparison with the neighbouring Limousin and Armorican domains (Benmammar et al., 2020) but its extension in the Eastern FMC is unclear. It contains relicts of eclogites dated at c. 380 Ma (Lu-Hf in garnet, U-Pb in zircon; Lotout et al., 2018).

The *Lower Gneiss Unit* is composed of paragneisses and orthogneisses with varied metamorphic overprint. The paragneisses, of probable Ediacaran protolith (Couzinié et al., 2019; Fig. 1b), like those of the UGU, record a Cadomian orogenic event prior to the Variscan orogeny (Couzinié et al., 2017). Paragneisses south of Marvejols (Fig. 1b) display

an inverted medium-pressure metamorphic zoning attributed to the underthrusting below the UGU during the Variscan orogeny (Burg et al., 1984).

The *Parautochton* is mostly composed of metapelites that display a limited metamorphic imprint ranging from greenschist to low-amphibolite grade (e.g., Faure et al., 1999).

The entire eastern FMC is intruded by abundant Carboniferous granitoids (Fig. 1b), including mantle-derived Mg–K-rich (monzo)diorites and lamprophyres (“vaugnerites”), resulting from polyphase melting from c. 340 to c. 300 Ma (e.g., Barbey et al., 2015; Couzinié et al., 2014; Gardien et al., 2011; Laurent et al., 2017; Moyen et al., 2017).

The studied eclogites were sampled in the Haut-Allier (Fig. 2), where the eclogite-bearing UGU is inferred to thrust over the LGU, devoid of HP metamorphism (e.g., Burg et al., 1984). The metabasic rocks of the LAC have geochemical affinities with ocean-floor tholeiites or back-arc basalts (Giraud et al., 1984). The close association with continental crustal rocks points to a thinned continental margin setting (e.g., Lardeaux, 2014). The protoliths are not dated. The dominant metamorphic grade ranges from the amphibolite to the granulite facies, but eclogite lenses are also present (Lasnier, 1977). Ultramafic rocks include garnet-bearing peridotites (Lasnier, 1968) but available *P–T* conditions from a garnet-pyroxene granulite indicate 20 kbar, 800 °C (Pin and Vielzeuf, 1988). Similar conditions (~800–900 °C at 18 kbar) are recorded in a coronitic gabbro, followed by a LP–HT overprint at ~4 kbar, 650 °C (Nicollet et al., 1993).

The age of the HP metamorphism was estimated in the pioneering study of Ducrot et al. (1983) at  $430 \pm 20$  Ma by U-Pb dating of a dissolved population of zircon from an eclogite found at the locality of La Borie (Fig. 2). Because of the importance of the age of the HP metamorphic stage in the eclogites, and the problematic reliability, discussed above, of the

method employed by Ducrot and collaborators, the age of the eclogite-facies metamorphism of the La Borie eclogite is re-evaluated in this paper. The eclogites occur within a variably retrogressed hectometre lens embedded in a sillimanite-bearing diatexite in close association with a felsic orthogneiss. The eclogites underwent prograde metamorphism in the eclogite facies from 650 °C at 20 kbar to 750 °C at 22–23 kbar and crossed the solidus during an isobaric temperature increase up to 850–875 °C (de Hoÿm de Marien et al., 2020). Following incipient decompression to ~19 kbar in the HP granulite facies, further decompression to 5–10 kbar at 750–850 °C led to partial recrystallisation in the HT amphibolite facies.

## **METHODS**

### **2.1 Geochemistry**

Samples for whole-rock chemistry were cleaned from weathering alteration, successively crushed in a jaw and a roller crusher and reduced in powder with a planetary crusher in an agate bowl. Powders were analysed at the Geochemical and Petrographical Research Center in Nancy, France (SARM laboratory, CNRS-CRPG) for major and trace elements using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) and Mass Spectrometry (ICP-MS) respectively. FeO (vs. Fe<sub>2</sub>O<sub>3</sub>) was analysed by wet titration. Whole-rock geochemical data are accessible in the Supporting information 2.

### **2.2 U-Pb trace element petrochronology**

A mineral separation procedure has been applied to concentrate zircon and apatite grains for U-Pb dating using the facilities available at Géosciences Rennes (University of Rennes). The samples were crushed and only the powder fraction with a diameter < 250 µm was kept. Heavy minerals were first concentrated by Wilfley table and magnetic minerals were removed with an isodynamic Frantz separator. Non-magnetic heavy minerals were afterwards

separated with heavy liquids. The concentrated zircon and apatite grains were handpicked under a binocular microscope. The selected minerals were embedded in epoxy mounts, grounded to the middle of the grain and then polished on a lap wheel.

Separated zircon and apatite grains were imaged by cathodoluminescence (CL) using a Reliotron CL system equipped with a digital colour camera available at the GeOHeLiS analytical platform (OSUR, University of Rennes). Thin section imaging of zircon was performed at Clermont-Ferrand using a scanning electron microscope equipped with a CL system (JEOL JSM-6400, Laboratoire Magmas et Volcans, Clermont-Ferrand). Rutile crystals in thin section were imaged using back-scattered electrons (BSE; JEOL IT 300 LA EDS scanning electron microscope, CMEBA platform, University of Rennes ).

U-Pb geochronology of zircon, rutile and apatite grains was conducted by in-situ laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at the GeOHeLiS analytical platform (OSUR, University of Rennes) using an ESI NWR193UC Excimer laser coupled to an Agilent quadrupole 7700x ICP-MS equipped with a dual pumping system to enhance sensitivity (Paquette et al., 2014). The instrumental conditions are reported in the Table S1-3 in the file Supporting information 1 and details for each analytical sessions may be found in the first sheet of the files Supporting information 3, 4 (BR18 & BR19 – eclogite samples; BR60 – diatexites sample). Further information on the dating protocol can be found in Nosenzo et al. (2022) for zircon, Lotout et al. (2020) for rutile and in Pochon et al. (2016) for apatite.

Zircon, apatite and rutile were analysed during several analytical sessions with distinct analytical conditions (beam size, repetition rate, fluence and analysed masses) summarized in Supplementary tables (Supporting information 3, 4). Data were corrected for U-Pb and Th-Pb fractionation and for the mass bias by standard bracketing with repeated measurements of the

GJ-1 zircon standard (Jackson et al., 2004), Madagascar apatite standard (Thomson et al., 2018) or R10 rutile standard (Luvizotto et al., 2009). In addition, zircon, apatite and rutile reference materials were measured along with the unknowns to monitor the accuracy of the analyses. For the sake of clarity, the ages indicated for the standard in the following text (and Table S1-3) were calculated using all the data (obtained during different analytical session) and the ages produced during individual analytical session are indicated in Supporting information 3 & 4. McClure (Schoene and Bowring, 2006;  $524.5 \pm 4.5$ ;  $n = 13$ ; MSWD = 0.66) and Durango (McDowell et al., 2005;  $32 \pm 0.5$ ;  $n = 14$ ; MSWD = 1.2) apatite reference materials were measured along with the apatite analyses. Accuracy during the rutile analyses was monitored using rutile R19 (Zack et al., 2011;  $484.5 \pm 9$ ;  $n = 6$ ; MSWD = 0.1) and R632 (Axelsson et al., 2018;  $500 \pm 6$  Ma;  $n = 10$ ; MSWD = 1.3) reference materials. Data for Zr in rutile were corrected using the rutile R10 (Luvizotto et al., 2009) and rutile R19 (Zack et al., 2011) was measured along with the unknown as a secondary reference material.

Zircon crystal domains have various shapes and sizes (see below). In order to ensure that all the different domains were analysed without compromising the precision and accuracy of the geochronological results, different analytical procedures were implemented. First, the zircon grains were characterized for trace element and isotopic data at the same time, using a small rectangular beam size ( $37 \times 14 \mu\text{m}$ ). Data for trace elements in zircon were corrected using NIST612 glass (Kent et al., 2004) and zircon standard 91500 (Wiedenbeck et al., 1995; Wiedenbeck et al., 2004;  $1080 \pm 7.5$  Ma;  $n = 23$ ; MSWD = 2.5) was measured along with the unknowns as a secondary standard following the procedure described in Nosenzo et al. (2022). In a second time, large zircon crystals of known trace-element chemistry (i.e. already analyzed with small-sized beam) were targeted for U-Pb isotope analysis using a larger rounded beam size ( $50 \mu\text{m}$ ) in order to enhance analytical precision. The accuracy of the

analyses was monitored using the zircon standard Plešovice (Sláma et al., 2008;  $337 \pm 1.3$  Ma;  $n = 13$ ;  $MSWD = 0.5$ ) as a secondary reference material.

Data reduction was carried out with Iolite v4 (Paton et al., 2011). Reproducibility and age uncertainties of reference material Plešovice and 91500 were propagated by quadratic sum to the analysed zircon grains according to Horstwood et al. (2016). Concordia and lower intercept ages and diagrams were generated using IsoplotR (Vermeesch, 2018). All errors given in Supporting information 3 & 4 are listed at two sigmas.

Raman spectroscopy was performed at the Laboratoire Magmas et Volcans (Clermont-Ferrand, France). Raman spectra were obtained using a Renishaw InVia confocal Raman microspectrometer, equipped with a 532 nm diode laser (output power of 200 mW), a Peltier-cooled CCD detector, a motorized XY stage and a Leica DM 2500M optical microscope. Back-scattered geometry allowed to collect the scattered light and a dielectric edge filter to reject reflected laser radiation as well as Rayleigh scatter. Analyses were performed with a varying laser power of  $\sim 1$  to 15 mW depending on the analysed mineral phase, and the slit aperture set to 20  $\mu\text{m}$  in a high confocality setting. A 100x objective and a 2400 l/mm grating were used for the analyses resulting in spatial and spectral resolution of  $\sim 1$   $\mu\text{m}$  and  $1$   $\text{cm}^{-1}$  respectively. Calibration of the spectrometer was based on the Si  $520.5 \pm 0.5$   $\text{cm}^{-1}$  peak. The spectra were recorded using the Wire 4.2 software in the aluminosilicate network domain ( $\sim 100$  to  $1300$   $\text{cm}^{-1}$  Raman shifts), and the water domain ( $\sim 3000$  to  $3800$   $\text{cm}^{-1}$ ) was checked when needed. Acquisition times varied from 10 to 300 s depending on the laser power condition and the nature of the analysed mineral phase.

### **3. PETROGRAPHY**

Both eclogite samples display comparable petrography. A detailed description of the sample BR18 is presented in de Hoÿm de Marien et al. (2020). In summary, the textural relationships in both samples point to a peak mineral assemblage of garnet (g1), omphacite (cpx1), kyanite (ky), phengitic muscovite (mu) and accessory quartz (q), rutile (ru), sulphide, apatite and zircon. Garnet 1 porphyroblasts are anhedral, up to 3 mm in diameter (Fig. 3a). Numerous inclusions of epidote (ep1), amphibole (amp1) and rutile as well as scarcer inclusions of omphacite, kyanite, phengite, quartz, apatite and zircon are present in garnet 1 (Fig. 3b-d). Epidote (ep1) and amphibole (amp1) are common in garnet 1 core and mantle but lack in its rim. A thin (up to 250 µm thick) rim of garnet 2 (g2) overgrows garnet 1 and contains inclusions of amphibole (amp2; Fig. 3 in de Hoÿm de Marien et al., 2020). Rare anhedral crystals of albite observed in the matrix are significantly smaller (~100 µm) than garnet 1 but larger than the crystals in the matrix symplectites (~10 µm). They are inferred to be associated with garnet 2 because they are separated from the symplectites by a corona of diopside.

The matrix is mostly composed of various fine-grained symplectites (1 to 10 µm) that replace the peak metamorphic assemblage. Anhedral garnets are surrounded by a corona of amphibole (amp3) and plagioclase (pl). Matrix omphacite is replaced by fine-grained symplectites of diopside (cpx2), amphibole (amp3), plagioclase and quartz. Kyanite is replaced by corundum-plagioclase and spinel-plagioclase symplectites, surrounded by an aureole of plagioclase locally containing small flakes of muscovite. Former phengitic muscovite is replaced by fine-grained aggregates involving biotite and low-Si muscovite. Rutile is surrounded by or juxtaposed to ilmenite, both being locally surrounded by a corona of titanite.

Many inclusions in garnet are large polymineralic inclusions with varied mineralogy, but consistently containing plagioclase (Fig. 3c, f-g). The shape of these inclusions is usually

anhedral with locally complicated intergrowths between the host garnet and the inclusion (Fig. 3g). Despite the overall anhedral shape of the inclusion, garnet locally forms isolated faceted crystals in contact with the inclusion (blue arrow in Fig. 3h). At the contact with the polyinclusions, garnet has generally a modified composition (lower Mg and Ca, and higher Fe and Mn; see X-Ray maps in Fig. S1).

The individual crystals forming the inclusions are usually anhedral (Fig. 3f). Their grain boundaries may be straight with the local presence of granoblastic texture (in Fig. 3f – middle and top inclusion). In other cases, the grain boundaries are winding (Fig. 3f – lower inclusion) and amphibole and clinopyroxene are rimmed by plagioclase at the contact with garnet. The garnet-plagioclase contact is curved. Plagioclase forms embayments and locally sharp offshoots along fractures within the host garnet (Fig. 3f) as well as films with low dihedral angle terminations at the amphibole-garnet grain boundary (red arrows in Fig. 3g; Fig. S2).

Garnet displays several sets of fractures. They are not systematically associated with inclusions, but garnet around the polyinclusions is always cracked. These fractures radiate around isolated inclusions (Fig. 3e) or connect inclusion clusters (Fig. 3f). The radial disposition of the fractures suggests a volume increase of the inclusions. Some fractures are filled with minerals similar to those of the neighbouring inclusion. Other fractures are healed with new garnet (white arrow in Fig. 3g) and show fluid inclusion trails suggesting the presence of fluids possibly related to devolatilization.

Within the polyinclusions, the following mineral associations were observed: amphibole-plagioclase; amphibole-plagioclase-clinopyroxene; amphibole-plagioclase-corundum-spinel-K feldspar; amphibole-kyanite-plagioclase-spinel-sulfide; ilmenite-clinopyroxene-plagioclase-K feldspar-sulfide; K feldspar-plagioclase-orthopyroxene-titanite-calcite; phengite-K feldspar.

Another set of fractures, cutting through both the garnet and the symplectitic matrix and filled by analcite is locally observed (Fig. 3j). Pools filled with analcite, thomsonite (a zeolite group mineral), subordinate plagioclase and biotite developed along the fractures. Plagioclase is rounded and biotite forms partially chloritized flakes. The pools are observed in the vicinity of the sp-cor-pl symplectites after kyanite where the Ca-rich plagioclase of the symplectites is embayed by analcite and thomsonite (Fig. 3k). When the fractures occur next to garnet, the latter is penetrated by offshoots also filled by analcite, thomsonite, biotite and plagioclase (Fig. 3j).

#### 4. GEOCHEMISTRY

The samples have a basaltic composition ( $\text{SiO}_2 = 46.9\text{--}46.4\%$ ) with high  $\text{Al}_2\text{O}_3$  (19.9–17.7 %) and CaO (10.8–10.2 %) contents. Both samples show enrichment in the most incompatible elements marked by a positive  $(\text{Nb}/\text{Yb})_N$  ratio (4–6; Fig. 4a). However, BR19 is strongly depleted in Th and LREE with respect to BR18. This difference approaches an order of magnitude, resulting in an apparent Nb anomaly that is either slightly negative in BR18, or positive in BR19 (Fig. 4a). This could suggest contrasting petrogenesis of the two samples, which is surprising given the identical sampling locality, comparable mineralogy and major-element geochemistry. Alternatively, however, it could also point to the modification of one sample with respect to the other. Indeed, whereas Th and LREE are mostly immobile with respect to fluid alteration, they are mobile in melts (Klimm et al., 2008; Luginbühl, 2015), which could suggest that BR19 was depleted in melt with respect to BR18. This is consistent with the inferred presence of melt in the samples, the  $P$ – $T$  evolution, as well as the typical igneous textures of some metamorphic zircons (see below).

The differences between the two samples may be further illustrated with chondrite-normalized rare-earth-element (REE) patterns (Fig. 4b). BR18 shows an overall higher REE

concentration ( $\Sigma\text{REE}$  – BR18: 56.61 ppm; BR19: 18.54 ppm) and a negative slope of the chondrite-normalized pattern stronger than BR19. The difference between the two samples is best shown when BR19 is normalized with respect to BR18 (Fig. 4c). According to the resulting pattern, BR19 displays significantly lower LREE from La to Nd, the magnitude of the difference rapidly decreases between Nd to Tb and is minimum in HREE between Tb to Lu. This pattern compares perfectly with that presented by Wang et al. (2014, their Fig. 7d) for a restite after eclogite-facies melting, suggesting again that partial melting in the eclogite facies and subsequent melt loss could account for the chemical composition of BR19.

## 5. GEOCHRONOLOGY

Zircon, apatite and rutile were selected for geochronology because they are characterized by significantly different closure temperatures and may consequently provide geochronological constraints on distinct thermal events or parts of the  $P$ – $T$  history. In this study, the rather straightforward results obtained on apatite and rutile are used to interpret the more complex results obtained on zircon and are consequently presented first. For comparison, zircon and apatite from the felsic sillimanite-bearing diatexite, that encloses the eclogite lenses, were also analysed and the results are presented in the Supporting information 1 (section "Geochronology of the embedding migmatite" including Figs. S5-8).

### 5.1 Apatite

Apatite is common in the matrix of both samples and can be occasionally found as inclusions either in garnet (Fig. 5a) or zircon. In the matrix, apatite occurs as mm-sized aggregates of subhedral crystals 200–500  $\mu\text{m}$  long (Fig. 5b). These aggregates are elongated and parallel with the fabric marked by former kyanite crystals. Under transmitted light, separated crystals are limpid and locally contain tiny inclusions. Under cathodoluminescence

(CL), apatite crystals are homogeneous at the scale of a single grain but the colour of the whole population is variable (Fig. 5c), reflecting variation in trace element contents.

Seventy-two analyses were performed on 33 crystals hand-picked from BR18 and 40 analyses on 21 crystals hand-picked from BR19. Apatite in BR18 has a variable U content (0.1–84 ppm; av = 20 ppm; Supporting information 3, sheet #10), higher than BR19 (0.1–4 ppm; av = 1.37 ppm; Supporting information 3, sheet #11).

Apatite separates analysed from the two eclogites yield homogeneous results. In a Tera-Wasserburg diagram, the data plot in a discordant position indicative of a significant proportion of common Pb (Fig. 6a,b), higher in BR19 than in BR18. Characterization of the whole population together with repeated measurement of the grains with higher U contents in BR18 allow to define a discordia yielding a lower intercept date of  $353 \pm 1.5$  Ma (MSWD = 1.4, n = 72) with a  $^{207}\text{Pb}/^{206}\text{Pb}$  initial value of  $0.857 \pm 0.002$  (Fig. 6a). The spots in apatite grains from the sample BR19 define a less precise lower intercept date of  $355 \pm 12.5$  Ma (MSWD = 1.2, n = 40) with a  $^{207}\text{Pb}/^{206}\text{Pb}$  initial value of  $0.856 \pm 0.003$  (Fig. 6b). Both  $^{207}\text{Pb}/^{206}\text{Pb}$  initial values are in a good agreement with the  $^{207}\text{Pb}/^{206}\text{Pb}$  value of 0.859 calculated following the Pb evolution model of Stacey and Kramers (1975) for an age of 355 Ma. These dates are comparable within error with the lower intercept date of  $346 \pm 5$  Ma (MSWD = 0.86, n = 24) with a  $^{207}\text{Pb}/^{206}\text{Pb}$  initial value of  $0.826 \pm 0.017$  obtained on apatite from the surrounding migmatites (Fig. S8).

## 5.2 Rutile

Rutile is common both in the matrix and as inclusions in garnet (Fig. 7a-d). The crystals are anhedral with size ranging from 50 to 250  $\mu\text{m}$ . Forty-two in-situ analyses were performed on 18 grains directly in-context in thin section. U and Pb contents are low (U = 0.03–

34.3 ppm,  $av = 2.0$ ;  $Pb = 0.01\text{--}3.3$  ppm;  $av = 0.2$ ) whatever their textural positions.

Zirconium contents vary from 137 to 362 ppm with an average of 250 ppm.

Rutile was analysed only in BR18. Forty-two analyses were performed directly in-context in thin section on 17 different rutile crystals with distinct textural positions. Because of low U and Pb contents in most grains, only six analyses from four different rutile grains found in the matrix could be used for geochronological purpose. Plotted in a Tera-Wasserburg diagram, the data are discordant indicating the presence of various amounts of common Pb (Fig. 6c). A lower intercept date of  $348 \pm 9$  Ma can be calculated ( $MSWD = 2.2$ ,  $n = 6$ ) yielding an initial  $^{207}Pb/^{206}Pb$  ratio of  $0.80 \pm 0.11$ , compatible with the  $^{207}Pb/^{206}Pb$  value of 0.859 calculated following the Pb evolution model of Stacey and Kramers (1975) for an age of 350 Ma.

### 5.3 Zircon

Textural relationships between zircon and other minerals were investigated optically directly in thin section and through successful identification of 23 micro-inclusions out of over ~ 100 analyses carried out by Raman spectroscopy on ~ 250 zircon crystals separated and mounted in epoxy. Internal textures were investigated by CL imaging. Each zircon internal domain was analysed for trace elements and U-Pb isotopes. Potential control of zircon position (e.g., in garnet or in the matrix) on U-Pb results was also checked by performing analyses directly in-context in thin section.

A first population of zircon crystals is characterised by CL-dark cores that show in general patchy zoning but may be unzoned or display oscillatory growth zoning. These cores are surrounded by CL-bright rims; the interface between the dark cores and the bright rims is irregular, commonly convex toward the cores and cutting across the oscillatory zoning when present. CL-bright zones also form along cracks and around inclusions within the CL-dark

zones. The zircon crystals are in general yellowish, cracked, inclusion-rich and display stocky rounded habitus (Fig. 7e), although translucent elongated crystals have also been observed. They represent ~ 75 % of the zircon population in BR18, but only ~ 10–15 % in BR19.

Zircon crystals from the second population are in the vast majority rounded, colourless, translucent, and contain few minute (up to 50  $\mu\text{m}$ ) faceted inclusions (Fig. 7f). Compared with the zircons from the first population, they are less fractured but may contain few discrete and thin fractures. Trails of minute ( $\sim 1 \mu\text{m}$ ) inclusions can be observed in the extension of the fractures suggesting that the zircon crystals may have partially healed after fracturing (Fig. 8). However, the fractures are not distinguishable under CL, possibly because the contrast is not strong enough. Under CL, these zircon grains are bright, comparable to the rims of the crystals of the first population (Fig. 7e, f). They display various internal textures that encompass flow textures, patchy, oscillatory or sector zoning (Fig. 7f). Zircon crystals with flow textures are common and in general contain inclusions, whereas those with oscillatory or sector zoning are rarer and generally inclusion-free. In thin section, CL-bright zircon is commonly associated with rutile, included in garnet, rutile or in the cpx-amp-pl-q symplectites after omphacite. In garnet, zircon generally forms small clusters (Fig. 7a) or trails of tiny and rounded crystals around inclusions of rutile (Fig. 7b). By contrast, zircon associated with rutile in the matrix forms single larger crystals (Fig. 7c,d). Raman spectroscopy investigation of inclusions in these CL-bright zircon crystals demonstrates mutual inclusion relationships with garnet ( $n = 13$ ), rutile ( $n = 5$ ) and clinopyroxene (possibly omphacite;  $n = 2$ ) suggesting that these minerals crystallized contemporaneously. In addition, inclusions of apatite ( $n = 1$ ), titanite corona around rutile in a cracked zircon ( $n = 1$ ), hematite ( $n = 1$ ), and a zig-zag tubular shaped - feldspar ( $n = 1$ ) were also determined.

The two zircon populations display contrasting trace-element characteristics well illustrated by chondrite-normalized (Boynnton, 1984) trace-element patterns, and in the Th/U vs  $\text{Lu}_N/\text{Gd}_N$  plot (Fig. 9). The CL-dark sectors from BR18 display variable uranium content (114–556 ppm; av = 284 ppm), Th/U (0.44–2.16; av = 0.78) and REE contents ( $\Sigma\text{REE} = 251\text{--}1079$  ppm). One spot displays similarities with the CL-bright zircons described below and is considered as an outlier. Chondrite-normalized REE patterns (Fig. 9a) are characterized by negative Eu anomalies ( $\text{Eu}_N/\text{Eu}^* = 0.11\text{--}0.16$ ; av = 0.26) and moderate HREE enrichment ( $\text{Lu}_N/\text{Gd}_N = 13\text{--}31$ ; av = 21). Ti content is variable (2.5–15 ppm; av = 8 ppm).

CL-bright crystal domains display strikingly homogeneous chemical compositions in both samples. Compared to the CL-dark domains, they are characterized by lower U (BR18: 14–58 ppm, av = 28; BR19: 19–99 ppm, av = 47.5), Th/U ratios (BR18: 0.01–0.1, av = 0.03; BR19: 0.01–0.06, av = 0.02) and REE contents (BR18: 5.2–20.2 ppm, av = 11.3 ppm; BR19: 4.6–25.6 ppm, av = 11.3 ppm). Their chondrite-normalised REE patterns show average Eu anomalies close to unity (BR18:  $\text{Eu}_N/\text{Eu}^* = 0.55\text{--}1.19$ , av = 0.94; BR19:  $\text{Eu}_N/\text{Eu}^* = 0.4\text{--}1.7$ , av = 1.1), and flat HREE patterns (BR18:  $\text{Lu}_N/\text{Gd}_N = 0.35\text{--}8$ , av = 4; BR19:  $\text{Lu}_N/\text{Gd}_N = 0.4\text{--}2.1$ , av = 1.0). The Ti content is less variable than in the CL-dark sectors (BR18: Ti = 1.8–5.1 ppm, av = 3.2 ppm with one analysis at 20 ppm; BR19: Ti = 0.89–7.5 ppm, av = 3.73 ppm; with one analysis at 10 ppm).

The thin CL-bright rims or patches around or within the CL-dark domains display heterogeneous chemical characteristics intermediate between those resembling the CL-dark domains and the CL-bright crystals (Fig. 9b) and may be considered in most cases as mixed analyses.

The isotopic results obtained using the small spot size (see Methods) from each sample were plotted in two Tera-Wasserburg diagrams (Figs. 10a, b) except for spots with high

analytical errors. Data obtained in BR18 are dispersed but form two main clusters of concordant to slightly discordant analyses around c. 500 Ma and c. 350 Ma. Data from the CL-dark domains are shown in deep blue and compose the c. 500 Ma cluster, although a few spots plot in younger positions, down to c. 400 Ma. An apparent age of  $490 \pm 3$  Ma ( $MSWD_{C+E} = 1.6$ ;  $n = 19$ ; Fig. 11) can be calculated using the data from the main cluster. In BR19, the only spot obtained in a CL-dark zircon domain yields a younger apparent age of 420 Ma suggesting mixing with an underlying CL-bright domain. Data obtained in the CL-bright zircons from both samples (red and orange ellipses in Fig. 10a, b) cluster at c. 350 Ma close to the apparent age obtained on apatite (see above). Results for the spots from the CL-bright rims analysed in BR18 are shown in light blue (Fig. 10a). They plot in a concordant to very discordant position and strongly scatter, mostly between the two main clusters, but they may also plot in older positions.

CL-bright zircon crystals commonly yield apparent ages (350–320 Ma) equivalent to or even younger than apatite (c. 355–350 Ma), which is commonly not expected (see discussion below). Despite the important cluster around 350 Ma (Fig. 10b), zircon data scatter between c. 380 and 320 Ma suggesting a more complex distribution of the age data, that could be disclosed through greater precision on individual spots. Consequently, both the CL-bright crystals from sample BR18 already characterized for trace elements, and the zircon grains in thin sections, were (re)analyzed using a larger beam-size in order to ensure the ablation of a larger volume of material (almost 4 times). The resulting data are equivalent (Fig. 10c) to the ones obtained with small spot size but display a better individual analytic precision. Data obtained directly in thin section do not reveal any clear textural control and are shown in the same figure. Fig. 10d shows that the main cluster of concordant to slightly discordant spots plots again between c. 350–320 Ma. A secondary population of concordant and discordant spots (red coloured ellipses) plots along a discordia yielding a lower intercept date of

360.5 ± 2.5 Ma (MSWD = 1.6; n = 25) and pointing to an initial Pb isotopic composition of 0.96 ± 0.35. Despite the large error, this initial Pb isotopic composition is broadly consistent with the  $^{207}\text{Pb}/^{206}\text{Pb}$  value of 0.8597 calculated following the terrestrial Pb evolution model of Stacey and Kramers (1975) for an age of 360 Ma. Apart from two older spots around 390–380 Ma (dashed-rim ellipses), the rest of the data plots in a younger position (down to c. 310 Ma).

A weighted mean  $^{206}\text{Pb}/^{238}\text{U}$  apparent age of 360.5 ± 1.5 Ma (MSWD = 1.1; n = 16; Fig. 11) can be calculated using only the concordant analyses (concordance > 90 %) out of the data aligned along the discordia. Most of the other data spread along the concordia curve from c. 350 to c. 310 Ma and three older spots yield an apparent weighted mean age of 370 ± 3.5 Ma (MSWD = 1.0). The data set obtained in the other sample (BR19) displays an overlap between individual analyses due to the small size of the analytical spots and does not bring a clear geochronological information. However, apparent ages comparable to the results of BR18 can be calculated. The three oldest spots yield an apparent age of c. 381 Ma. Within the main population, the oldest spots allow the calculation of an apparent age of c. 360 Ma (Fig. 11).

## 6. THERMOMETRY

The coeval crystallisation of zircon, rutile and quartz, inferred above and supported by the mutual inclusion relations, and the inclusion of all three minerals in garnet, is suitable for the application of the Zr-in-rutile and Ti-in-zircon geothermometry. The pressure-dependent calibrations of Hofmann et al. (2014) and Tomkins et al. (2007) were applied to the CL-bright zircon domains, and rutile, respectively.

Ti-in-zircon and Zr-in-rutile data were checked for systematic trends with respect to the position of the spot in a single crystal and for textural control in thin section, but no correlation could be observed. The thermometry results were plotted against P and T using the minimum and maximum values of Ti in zircon and Zr in rutile (Fig. 12). Ti-in-zircon from BR19 yields temperatures between 600 and 850 °C (at  $P > 19$  kbar), a wide interval that encompasses the whole temperature range modelled for the eclogite- and granulite-facies metamorphism (de Hoÿm de Marien et al., 2020). The results obtained on zircon from BR18, indicate a narrower range of temperatures ( $T = 770\text{--}860$  °C at  $P = 20\text{--}22.5$  kbar, inferred from the phase-diagram modelling) above the modelled solidus of the sample. Application of the thermometer using the averaged Ti-in-zircon content results in temperatures of  $833 \pm 15$  °C and  $845 \pm 10$  °C for BR18 and BR19, respectively. Minimum and maximum Ti-in-rutile thermometry results intersect with the  $P\text{--}T$  path between  $\sim 620$  and  $\sim 700$  °C ( $P > 19$  kbar) corresponding to the conditions of crystallization of the garnet cores (de Hoÿm de Marien et al., 2020).

## 7. INTERPRETATION OF THE POLYMINERALIC INCLUSIONS

The studied eclogites have been inferred to have undergone partial melting during the peak temperature part of their evolution (de Hoÿm de Marien et al., 2020; Fig. 12). Additional arguments are presented here, since the presence of melt is important for the interpretation of the geochronological and geochemical data, as zircon or apatite might have interacted with or crystallized directly from the melt (e.g., Yakymchuk et al., 2019; Harley et al., 2007) and the geochemical signature of the samples may have been modified by partial melting (e.g., Palin et al., 2016).

One of the interesting features of the studied eclogites is the presence of large polymineralic inclusions in garnet crystals. Such inclusions have been described from natural

samples (e.g., Lang and Gilotti, 2007; Cao et al., 2020) and HT experiments (Perchuk et al., 2005, 2008), and were interpreted as the result of partial melting of primary monomineralic inclusions. In the present case, they may represent former inclusions of amphibole, entrapped during the prograde subsolidus growth of garnet. *P–T* pseudosection modelling of such amphibole melting (Fig. S9) suggests the formation of peritectic garnet and clinopyroxene, in agreement with the observations, alongside with the recrystallisation of amphibole. The presence of melt along grain boundaries is supported by the plagioclase films with low dihedral terminations (Fig. 3g; black arrows), interpreted as melt pseudomorphs (e.g., Holness and Sawyer, 2008; Holness et al., 2011; Ferrero et al., 2012). The associated volume increase of the inclusions would account for the radial fractures in garnet.

## 8. INTERPRETATION OF THE GEOCHRONOLOGICAL RESULTS

The textural relations described above suggest that all dated minerals, zircon, rutile and apatite belong to the eclogite-facies metamorphic stage. Whereas the geochronological record of zircon is relatively complex, it can be decrypted using the comparatively simple data from rutile and apatite.

Rutile and apatite from both samples yield a homogeneous, coherent apparent age of c. 350 Ma ( $350 \pm 7.5$  Ma,  $353 \pm 1.5$  Ma,  $355 \pm 12.5$  Ma; Fig. 6). The same date is also recorded by apatite from the surrounding felsic migmatite ( $346 \pm 5$  Ma; Fig. S8). Although apatite belongs to the HP assemblage, it is reputed for a relatively low closure temperature for Pb diffusion (350–600 °C; Cherniak et al., 1991; Chamberlain and Bowring, 2001; Harrison et al., 2002; Cochrane et al., 2014) and significant reactivity in the presence of fluids (e.g., Harlov, 2015). Since, a significant part of the post-eclogite evolution occurred at high temperatures (750–850 °C; Fig. 12), and involved a fluid as indicated by the replacement of

anhydrous omphacite and garnet by amphibole-bearing assemblages, the well-defined c. 350 Ma apparent ages of apatite are interpreted as the age of cooling.

Rutile also belongs to the HP assemblage in agreement with the results of thermodynamic modelling and Zr-in-rutile thermometry. However, the close apparent age and closure temperature of apatite and rutile ( $\sim 600$  °C; Cherniak, 2000) suggest that the  $350 \pm 7.5$  Ma apparent age obtained in rutile records the same cooling process (cf. Lotout et al., 2020).

Zircon, on the other hand, yields apparent ages both significantly older and younger than the rutile and apatite cooling age of c. 350 Ma (Figs. 10-11). However, the closure temperature with respect to Pb diffusion in zircon ( $\sim 900$  °C; e.g., Cherniak & Watson, 2001; Kramers et al., 2009; Lee et al., 1997) is higher than that of rutile, apatite and the maximum temperature reached by the rocks ( $\sim 875$  °C, Fig. 12). Consequently, the "young" apparent ages cannot be interpreted in terms of simple diffusive Pb loss.

The texturally oldest CL-dark zircon cores display internal textures including oscillatory growth zoning pointing to a magmatic origin (Vavra, 1990; Corfu et al., 2003) in agreement with the trace-element data showing  $\text{Th/U} > 0.1$ , negative Eu anomaly and HREE-enriched REE patterns (e.g., Rubatto and Gebauer, 2000; Rubatto, 2002; 2017). The  $490 \pm 3$  Ma concordia date obtained with these zircon crystals is accordingly interpreted as the crystallization age of the gabbroic protolith of the eclogite.

CL-bright zircon crystals consistently display low  $\text{Th/U}$  ( $< 0.1$ ), an absence of negative Eu anomaly and a flat HREE pattern (Fig. 9a) suggesting they are metamorphic and crystallized in the presence of garnet but absence of plagioclase (e.g., Rubatto, 2002; 2017), characteristics typical of the eclogite facies. They show two major groups of internal textures – flow textures vs. oscillatory or sector zoning. Zircon crystals with flow textures commonly

contain minute inclusions of rutile, garnet and possibly omphacite, consistently with the trace-element data (Supporting information 3; sheet #6). The inclusions are faceted and much smaller than the crystal size of the minerals of the eclogitic assemblage. This suggests that they were captured during an important nucleation stage likely related to the onset of the eclogite facies. This is consistent with Ti-in-zircon temperatures that span a large interval but may be as low as 630 °C at 19 kbar. Furthermore, the crystallisation of zircon along the prograde evolution is compatible with the observation of clusters or trails of zircon beads around rutile included in garnet, interpreted as a prograde product of destabilisation of a magmatic ilmenite (cf. Bingen et al., 2001; Beckman and Möller, 2018). However, flow-textured zircons have been interpreted as modified primary zoning (Vavra et al., 1996; Harley et al., 2007). The large spread of apparent ages ranging from c. 380 to 310 Ma (with one older spot at c. 390 Ma) obtained from zircons with flow textures could be consequently interpreted in term of partial to total recrystallisation of early eclogite-facies crystals.

Zircon crystals with sector or oscillatory zoning are less common, mostly lack inclusions and show a slightly less important spread of apparent ages (c. 365–310 Ma, i.e. they do not yield the older ages recorded in the flow-textured crystals; Fig. 11). This type of zoning is usually interpreted as pristine (Vavra et al., 1996; Corfu et al., 2003; Harley et al., 2007; Rubatto, 2017) igneous textures (Schaeltegger et al., 1999; Vavra, 1990). It suggests that these grains crystallised from melt, which is compatible with the Ti-in-zircon thermometry results and the inferred  $P$ – $T$  evolution of the studied samples in the presence of melt (Fig. 12). As a general rule, in partially molten rocks, zircon grows during melt crystallisation (e.g., Kelsey et al., 2008; Kohn et al., 2015). Along the  $P$ – $T$  path of the eclogite (Fig. 12), melt crystallises during decompression at ~875 °C and ~19 kbar, at the transition between the eclogite and the HP granulite facies, and it is inferred that the CL-bright oscillatory-zoned zircons crystallised at this stage. The oldest recorded apparent age (c. 360 Ma) could be

associated with this crystallisation event. However, many of these zircon domains yield apparent ages younger than the age of apatite (inferred to represent cooling below ~600 °C) and must have undergone subsequent recrystallisation.

Consequently, zircon ages older than apatite and rutile (i.e. older than 350 Ma) can be interpreted in terms of zircon crystallisation along the prograde  $P$ – $T$  path or at the eclogite-facies peak, and further crystallisation or recrystallisation during the incipient decompression at high temperatures, in the presence of melt. The age of c. 360 Ma that can be calculated using different methods (cf. Fig. 10 and 11) on both samples is the best estimate for the age of the eclogite-facies  $P$ – $T$  peak. On the other hand, apparent ages younger than c. 350 Ma, must be attributed to the late cooling and associated retrogression of the eclogites. Since diffusion of radiogenic Pb is essentially impossible in zircon at these temperatures, the results are interpreted to reflect zircon recrystallisation, which appears to preserve the REE signature while resetting the U-Pb system (cf. Hoskin & Black, 2000; Martin et al., 2008; Schmädicke et al., 2018; Pitra et al., 2022). Zircon is known to readily recrystallize in the presence of hydrothermal fluids (e.g., Ayers et al., 2003; Spandler et al., 2004; Geisler et al., 2007), in particular if they have an alkaline character (e.g., Rubatto et al., 2008; Soman et al., 2010), from greenschist-facies conditions (250–450 °C; Chen and Zhou, 2017) up to the conditions of the granulite facies (750–1000 °C; Taylor et al., 2014 and references therein). Such a process is consistent with the common presence of planar fluid-inclusion trails in zircon (Fig. 8), which points to zircon fracturing and subsequent healing. The alkaline character of the fluids that interacted with the eclogite at low temperatures is supported by the occurrence of analcite and thompsonite in some of the fractures that cut through the rock. On the other hand, apatite is sensitive to alkaline fluids at high temperatures (700–900 °C; Antignano and Manning, 2008), but the presence of Na in the fluid inhibits apatite recrystallisation at low temperatures (Harlov et al., 2002; 2015). The circulation of a relatively low-temperature

sodic fluid during cooling then probably results in selective recrystallisation of zircon, whereas apatite remained unaffected.

The apparent ages of these recrystallised zircon domains spread down to c. 310 Ma (Figs. 10-11). Given the shape of the concordia curve in this time interval, a 360 Ma old zircon crystal affected by partial resetting at say 310 Ma, would produce data that are concordant within error although the individual apparent ages are meaningless (Pitra et al., 2022). This could suggest that the youngest recorded date (c. 310 Ma) would reflect the maximum age of the fluid circulation. At the regional scale these fluid circulations may be related to the widespread granitoid plutonism between c. 340 and c. 310 Ma (Laurent et al., 2017).

## 9. GEOLOGICAL HISTORY OF THE LA BORIE ECLOGITE

The eclogites occur closely associated with biotite-sillimanite metatexites, felsic diatexites and migmatitic felsic orthogneisses suggesting emplacement of the protolith in a continental setting rather than a mature oceanic domain. U-Pb dating of the magmatic core of zircons indicates that the protolith was emplaced during the late Cambrian ( $490 \pm 3$  Ma). This event can be related to the thinning and fragmentation, accompanied by widespread bimodal magmatism, that affected the northern margin of Gondwana during the Cambrian and Ordovician (e.g., Ballèvre et al. 2012; Paquette et al., 2017; Chelle-Michou et al., 2017; Lotout et al., 2017; Sánchez-García et al., 2003; Linneman et al., 2008; Košler et al., 2004).

After the protolith emplacement, the samples lack chronological or petrological record until the onset of the high-pressure metamorphism. The earliest metamorphic record is found as garnet cores that crystallized around 650 °C, 20 kbar (Fig. 12; de Hoym de Marien et al., 2020). The breakdown of magmatic ilmenite to rutile occurred around the same time as

indicated by the results of Zr-in-rutile thermometry (c. 620–700 °C) and was accompanied by the formation of first metamorphic zircon (Fig. 7a,b). This possibly corresponds to the oldest ages recorded in the metamorphic zircon (c. 380–370 Ma). Metamorphic zircons subsequently recrystallized during the prograde metamorphism documented otherwise by the growth and chemical zoning of garnet porphyroblasts. Garnet zoning suggests an initial pressure and temperature increase to 700 °C, 22–23 kbar prior to a roughly isobaric temperature increase likely up to 875 °C, associated with partial melting (de Hoym de Marien et al., 2020). At this moment, amphibole inclusions in garnet partially melted resulting in the formation of polyinclusions with features such as plagioclase pseudomorphs after melt films along grain boundaries. The high temperature recorded by the Ti-in-zircon thermometry (up to ~ 875 °C) points to a major recrystallization of the pre-existing zircons during this stage, close to the peak  $P$ – $T$  conditions. This recrystallisation event was accompanied or closely followed by progressive crystallisation of the melt and the concomitant formation of new zircon with igneous textures at c. 360 Ma. Overgrowth of new garnet around the eclogitic garnet is associated with incipient crystallization of plagioclase in the matrix and records decompression in the HP granulite facies. Further decompression is marked by the pervasive replacement of matrix omphacite by HT amphibole-plagioclase symplectites around 750–850 °C and less than 9 kbar. During subsequent cooling, the U-Pb system of apatite and rutile closed around c. 350 Ma. Later lower-temperature fluid infiltration promoted a major recrystallization of the zircon crystals at c. 310 Ma or younger. Hence, important decompression and subsequent cooling were achieved around 10 Ma after the peak  $P$ – $T$  conditions.

## 10. TECTONIC IMPLICATIONS

The results obtained in the studied eclogites document the emplacement of the protoliths during a late Cambrian (c. 490 Ma) rifting of a continental margin, subsequently subducted in the late Devonian (c. 370–360 Ma), partly exhumed during the early Carboniferous (c. 350 Ma) and finally affected by late hydrothermal alteration (possibly at c. 310 Ma or later). Our results do not reproduce the Silurian age for the eclogite-facies metamorphism and this age must consequently be abandoned in the tectonic reconstructions as already proposed by other authors (Paquette et al., 2017, Lotout et al., 2018, 2020; Benmammar et al., 2020).

The Devonian age obtained from the studied eclogite is equivalent to the age obtained in other eclogites associated with migmatites in southern parts of the FMC (Lotout et al., 2020; Pitra et al., 2022). Overall, the results show, that the Variscan subduction in the Massif Central was much shorter than previously thought (max. 20 Ma instead of ~70 Ma), and do not support any significant north-south diachrony of the HP metamorphism (cf. Fig. 1b).

### **10.1 Implications for the tectonic evolution of the FMC**

Our data show that the age of the eclogite-facies metamorphism in the Upper Gneiss Unit (c. 360 Ma) is equivalent to the age of the suprasubduction calc-alkaline magmatism in the Morvan and Brévenne areas pointing to a possible genetic relationship (Fig. S10). This suggests that the basement of the the Morvan and Brévenne area represents a part of the Devonian upper plate rather than a piece of the UGU (lower plate). It can be proposed that the thinning of the arc domain in the Brévenne area, and the associated magmatism, have been produced by the retreat of a subducting slab around c. 360 Ma. This was contemporaneous with the incipient decompression following melting at the eclogite-facies peak. Partial melting of the subducted rocks could have triggered their detachment from the subducting slab (Labrousse et al., 2011; Závada et al., 2018) and account for the onset of the exhumation of the eclogites that reached crustal conditions shortly after (c. 350 Ma).

As mentioned above, the magmatic rocks of the Morvan and Brévenne area display a regional lithological zoning from andesite in the north (Morvan) to the bimodal spillite-keratophyre association in the south (Brévenne). This lithological zoning is associated with southward decreasing enrichment in incompatible elements of these magmatic rocks, suggesting their emplacement during a southward subduction (Pin et al., 1982; Pin and Paquette, 2002; Fig. 13). The Morvan would represent the arc and the Brévenne the transition to the back-arc region. The similarity between the age of the volcanic rocks and the eclogite-facies metamorphism suggests that the eclogites belonged to this south-dipping slab during the Devonian. However, eclogites exposed in the FMC are commonly inferred to originate from a northward subducting slab (e.g., Matte and Burg, 1981; Faure et al., 2009; Lardeaux, 2014; Vanderhaeghe et al., 2020). This questions the polarity of the subduction.

The continental collision model for the FMC was built in reference to the Himalayas (Matte, 1986). It assumes the development of the crustal wedge to be related with a crust-mantle décollement developed in response to a strong slab-pull (e.g. Mattauer, 1986). The inversion of the isograds considered as the hallmark of crustal-scale thrusts would – as in the Himalayas – require the thrust to be synthetic with the previous subduction (e.g. Le Fort, 1975). Considering the LAC as a major suture zone (Matte and Burg, 1981) and the south-vergent superposition of the UGU over the LGU, where an inverted medium-pressure gradient is developed (Briand et al., 1978; Burg et al., 1984), the subduction was inferred to be dipping to the north. In this case, the eclogites were exhumed in the forearc position (e.g. Vanderhaeghe et al., 2020). This model, however, is in contradiction with the results of detrital-zircon studies, which show that all the units in the FMC have Gondwanan affinity (e.g. Chelle-Michou et al., 2017), and the distribution of trilobite fauna across Western Europe (Ballèvre, 2016), which indicates the closing of a major oceanic domain to the north of, rather than within, the FMC. This precludes the interpretation of the eclogite-bearing

leptyno-amphibolitic complex (LAC) as a major suture zone. From this perspective, the lithological and geochemical zoning of the Morvan-Brévenne area suggesting a southward subduction may be regarded as carrying a first-order message. The suture zone situated to the north of the FMC (e.g. Ballèvre et al., 2009; Baptiste et al., 2016; Schulmann et al., 2022) would result from a late Devonian subduction dipping to the south (e.g. Pin et al., 1982; Faure et al., 1997), which promoted the development of the arc and back-arc domains in the Brévenne and Morvan area (Pin and Paquette, 2002). The Devonian-Carboniferous age of the eclogites determined here suits within the proposed timing of a north-lying, south-dipping subduction. In this hypothesis, the eclogites, that appear today to the south of the arc, are situated in a back-arc position. A similar situation is described in the Bohemian Massif (see after). By comparison, the LAC and the UGU would represent a piece of the northern Gondwana margin teared from the continent and eventually dragged into the subduction zone. Following the detachment of the crust from the subducting lithospheric mantle, this buoyant partially melted material would accumulate below the upper plate (i.e. relamination; cf. Hacker et al., 2011) and flow at the rear of the magmatic arc to the present-day location. Partial melting of the subducted material enhanced lateral migration and promoted the exhumation in the back-arc domain undergoing active rifting in the Devonian (Pin and Paquette, 1997). In this model, it is considered that thrusting of the UGU over the LGU is the result of the inversion of the back-arc basin and that the UGU and the LAC represent a portion of the relaminated material pinched between the margins of the closed back-arc domain – namely, the Morvan-Brévenne and LGU.

From the perspective of this model invoking a southward subduction and subsequent southward migration of the partially exhumed material, it is interesting to note that a similar southward flow of migmatite-hosted eclogites has been proposed in the southernmost part of the FMC, between the Lévézou and the Montagne Noire massifs (Pitra et al., 2022). This

crustal flow is remarkably correlated with the progressive migration of the magmatism during the comparable 350–310 Ma time interval (Laurent et al., 2017).

## 10.2 Implications for orogen-scale correlations

Although several correlations between the Variscan massifs exposed in western Europe have been proposed recently (e.g., Ballèvre et al., 2014; Lardeaux et al., 2014; Martínez Catalán et al., 2020), modern orogen-scale correlations stumbled on the Silurian age previously inferred for the Variscan subduction in the FMC. Using these data, Martínez Catalán et al. (2021) proposed that the FMC was a part of the so-called Mid-Variscan Allochthon, a nappe stack, where the units with the oldest high-pressure metamorphic age (c. 400 Ma) reached the highest metamorphic grade (Fig. S11). However, our new data combined with a summary of modern  $P$ – $T$  and radiometric data show that in the FMC, the eclogites that underwent the highest-grade conditions are the youngest (Fig. S12). Hence, the FMC does not fit into the framework of the Mid-Variscan Allochthon, but rather compares with some of the high-pressure rocks exposed in the Bohemian Massif (BM; Fig. 14). There, (U)HP granulitic complexes (e.g., O'Brien et al., 1993; Medaris et al., 2006) occur several hundreds of kilometres away from the plate interface (e.g., Schulmann et al., 2009). Counterintuitively, the lower crust underlying the region between the suture zone and the granulitic complexes is characterised by low-densities suggesting a felsic composition (Guy et al., 2011). Consequently, the crustal material in the (U)HP complexes is regarded as molten pieces of the subducted (Saxothuringian) continental crust detached from the downgoing slab, having laterally flowed behind the arc and having been relaminated to and exhumed in the core of the upper (Moldanubian) plate (Schulmann et al., 2014; Nahodilová et al., 2020; Maierová et al., 2020, 2021). The position of the studied eclogites at the rear of the

arc in the FMC reminds the location of the (U)HP granulitic complexes of the BM, providing a more consistent picture of the Variscan orogen.

## 11. CONCLUSIONS

The main conclusions of this study are as follow:

1) Eclogites from the Haut-Allier (northern Massif Central) record an Ordovician (c. 490 Ma) rifting event, a Devonian (c. 370–360 Ma) subduction and a Carboniferous (c. 350 Ma) exhumation, followed by a late interaction (c. 310 Ma or later) with alkaline fluids. This study brings compelling evidence against the Silurian age previously proposed for the subduction.

2) Metamorphic zircon with clear eclogitic REE patterns (no Eu anomaly, flat HREE) and inclusions (garnet, rutile and probably omphacite) shows concordant apparent ages that spread from c. 380 down to c. 310 Ma. Apparent ages younger than the cooling age of apatite (c. 350 Ma) unequivocally testify that zircon can recrystallize outside the conditions of the eclogite facies, reset its U–Pb system but preserve an apparent eclogitic signature. It is suggested that interaction with alkaline fluids at relatively low temperatures would lead to the recrystallisation of zircon while leaving apatite unaffected.

3) The age of the eclogite-facies metamorphism (c. 370–360 Ma) compares with that of the suprasubductional calc-alkaline Devonian volcanism in the northern Massif Central. Active thinning of the arc domain during the exhumation of the eclogites was likely driven by the roll-back of the subducting lithospheric slab. The lithological and geochemical zoning of the volcanic rocks is consistent with a southward polarity of the subduction, at variance with the commonly accepted northward subduction of the so-called Massif Central Ocean. The present-day location of the eclogites suggests that they were exhumed at the rear of that arc after this southward subduction. The age and the architecture of the system question the

existence of a major suture zone in the Massif Central and point to similarities with the geodynamics of the Bohemian Massif.

## **ACKNOWLEDGMENTS**

This work benefited the support of the French geological survey (BRGM) as well as an internal Czech Geological Survey grant (#311190), accorded to L. de Hoÿm de Marien. X. Le Coz is acknowledged for the production of the thin sections and Y. Lepagnot for the mineral separation. We are grateful for the assistance in SEM observations and EDS analyses of the staff of the CMEBA facility (ScanMAT, University of Rennes), which received financial support from the Région Bretagne and the European Union (CPER-FEDER 2007-2014). We are also grateful to J. Langlade and F. Schiavi, for their assistance during the EPMA analyses (Microsonde Ouest, IFRMER, Brest-Plouzané, France) and Raman spectroscopy analyses (Laboratoire Magmas et Volcans, Clermont-Ferrand, France) respectively. Additional SEM observations were performed at the Faculty of Science, Charles University in Prague, with the help of M. Racek. L. de Hoÿm de Marien also thanks K. Schulmann, S. Collett and A. Guy for exciting discussions about the correlation across the Variscan belt and guidance in the discovery of the geology of the Bohemian Massif. The paper benefited from the comments of two anonymous reviewers.

## **CONFLICT OF INTEREST**

None of the authors have a conflict of interest to disclose.

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## SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

**SuppInfo1.pdf** – See details below

Table S1. Operating conditions for the LA-ICP-MS equipment during zircon analyses.

Table S2. Operating conditions for the LA-ICP-MS equipment during apatite analyses.

Table S3. Operating conditions for the LA-ICP-MS equipment during rutile analyses.

Figure S1. X-Ray maps of a large polyinclusion in garnet.

Figure S2. Examples of plagioclase with low dihedral angle in amphibole-plagioclase and kyanite-rutile-plagioclase polyinclusions in garnet.

Figure S3. Th/Yb vs Nb/Yb plot after Pearce (2008) and Pearce (2014).

Figure S4. Representative Raman spectra of micro inclusions in zircon.

Figure S5. Representative CL images of zircon in the diatexite embedding the studied eclogites.

Figure S6. Terra-Wasserburg and KDE plots of the U-Pb data in the zircon from the diatexite.

Figure S7. Representative CL images of apatites in the diatexite.

Figure S8. Terra-Wasserburg plot of the U-Pb data in the apatites from the diatexite.

Figure S9.  $P$ - $T$  pseudosection constructed to study partial melting of amphiboles.

Figure S10. Summary of the radiometric data in the eclogites and calc-alkaline magmatism in the Morvan area.

Figure S11. Summary of the  $P$ - $T$  paths and radiometric data in the Allochthonous units of the Variscan belt.

Figure S12: Summary of  $P$ - $T$  paths and ages of the main metamorphic events in the French Massif Central and the Bohemian Massif.  $P$ - $T$  paths for the FMC after the compilation of Benmamar et al. (2020) actualized with the data from Pitra et al. (2022). References for the ages in the FMC are this study, Lotout et al. (2018, 2020), Faure et al. (2014). See Figure S10 for details.  $P$ - $T$  paths and ages for the Bohemian Massif after the compilation in Maierová et al. (2021). Note that the peak-temperature conditions in the FMC usually coincide with the peak-pressure conditions, but post-date a significant decompression from UHP conditions in the BM.

**SuppInfo2\_BulkRocks.xlsx** – Bulk rock compositions of the studied samples.

**SuppInfo3\_U-Pb\_Eclogites.xlsx** – Summary of the operating conditions; U-Pb and trace element data for the standard and eclogites samples.

**SuppInfo4\_U-Pb\_Diatexite.xlsx** – Summary of the operating conditions; U-Pb data for the standard and diatexite sample.

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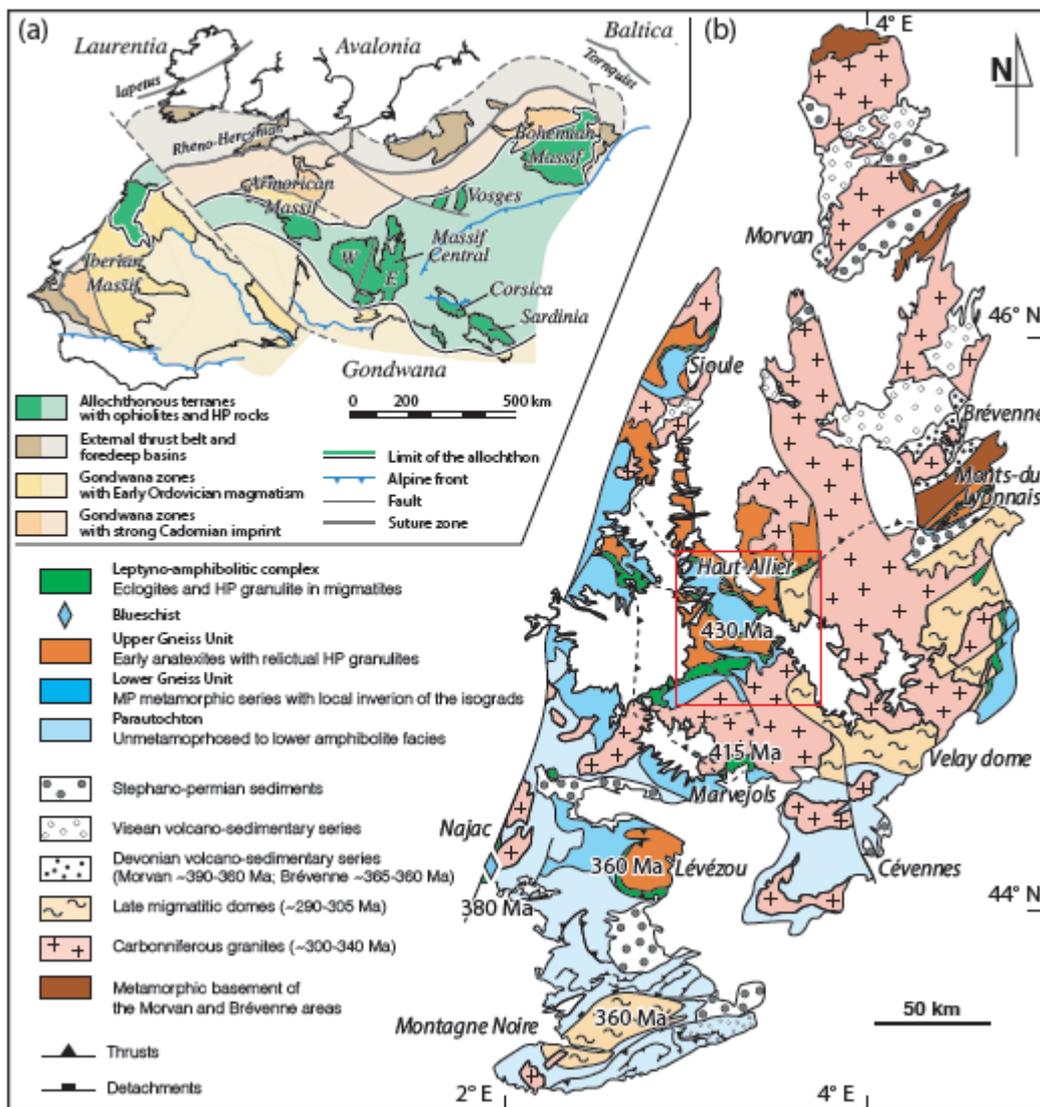


Figure 1: (a) Sketch of the main Variscan massifs and terranes of the European Variscan orogen (modified after Martínez Catalán et al., 2007 and 2021) with the position of the French Massif Central (E and W denote its eastern and western part, respectively). (b) Simplified tectonic map of the eastern part of the French Massif Central (modified after Ledru et al., 1989). The ages interpreted as the age of the eclogite-facies metamorphism are reported (see main text for references). Ages for the Devonian and Carboniferous magmatism are also reported in the text. Age of the emplacement of the late migmatite domes after

Barbey et al., 2015, Roger et al., 2015 & Poujol et al., 2016. The red rectangle shows the area covered by Fig. 2.

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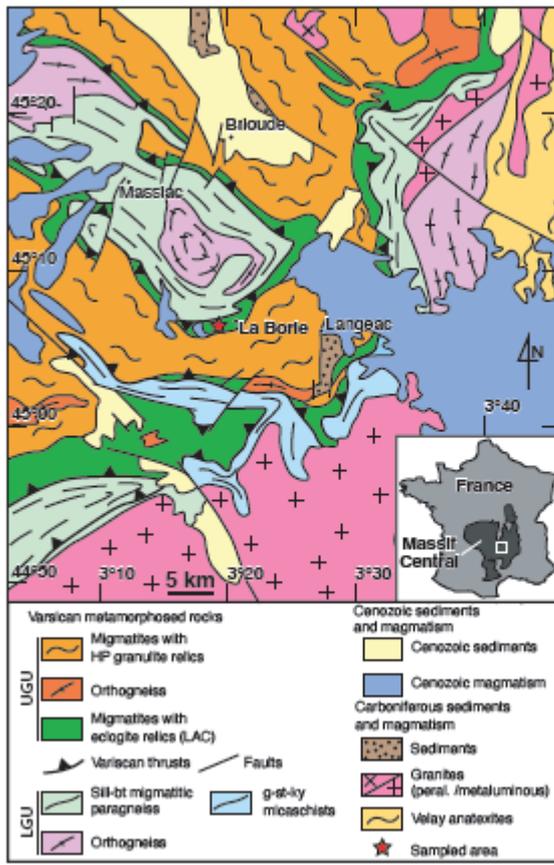


Figure 2: Geological map of the study area and its location in the French Massif Central (white rectangle in the inset). The samples have been gathered west of la Borie ( $45^{\circ}6'45.29''\text{N}$ ;  $3^{\circ}19'55.83''\text{E}$ ; WGS84) in the LAC.

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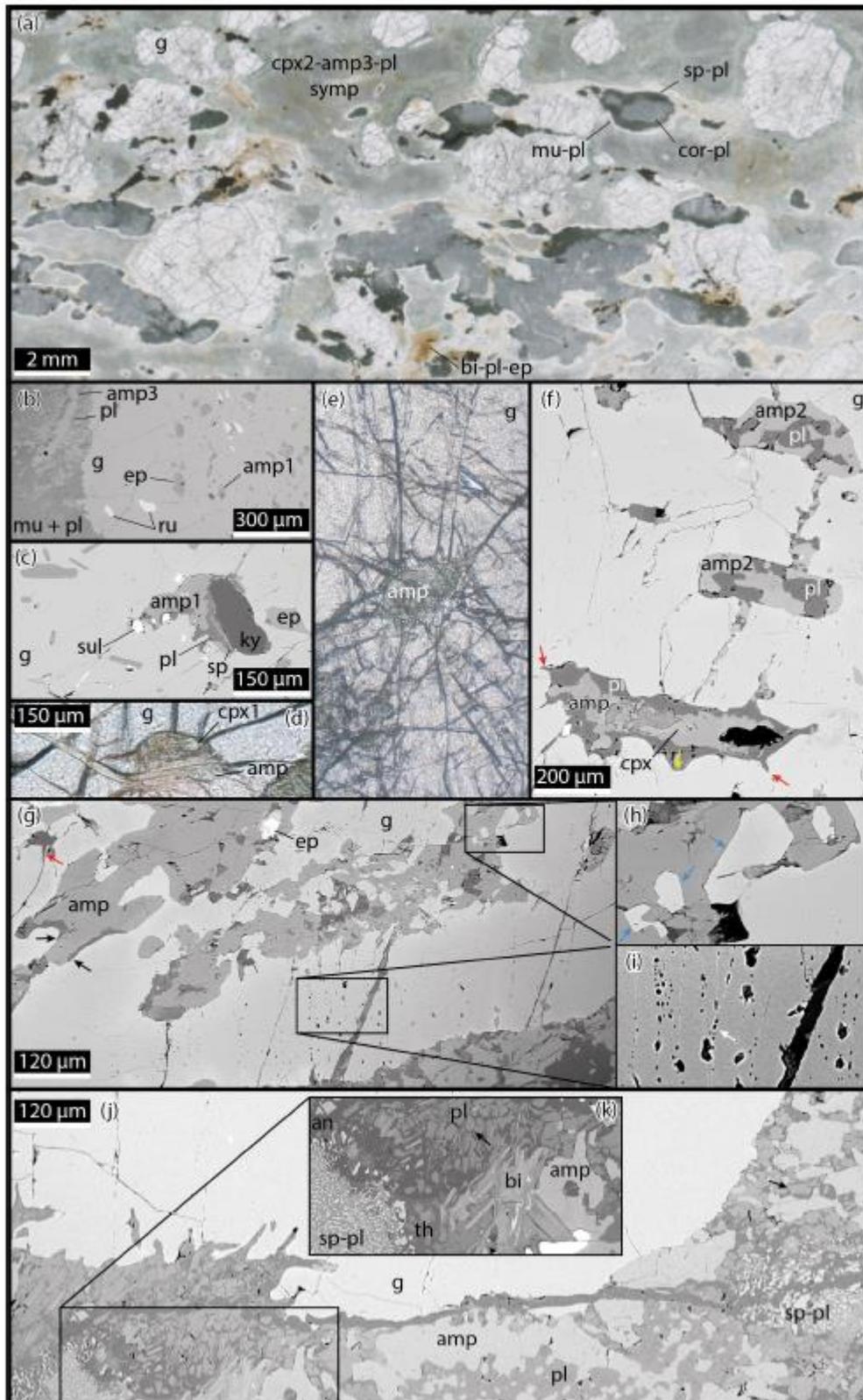


Figure 3: Photomicrographs and BSE images of the main textural features of the eclogites. (a) Photomicrograph of anhedral garnet crystals within a matrix of various symplectites after the eclogite-facies minerals. Note that the shape of the corundum-spinel-plagioclase symplectites after kyanite shows a preferred orientation, but the crystals within the symplectites are undeformed. Garnet contains inclusions of (b) epidote, amphibole, rutile; (c) kyanite and (d) omphacite (cpx1). Garnet around the amphibole inclusions may be radially fractured (e). Amphibole inclusions are commonly associated with plagioclase (c, f, g). Note the presence of garnet crystals with straight grain boundaries with amphibole (*blue arrows*; h) and the alignment of small fluid inclusions, along sealed fractures in garnet (i). (j) Fracture filled by analcite (an) and thomsonite (th; center) associated with biotite and rounded crystals of plagioclase close to spinel-plagioclase symplectite (left and right). At the vicinity of the fracture, garnet is dissolved and penetrated by channels also filled by analcite, thomsonite and biotite. Note the interstitial character of analcite in (k). *Red arrows* indicate plagioclase offshoots within garnet, *yellow arrow*, embayment in garnet, *black arrow* plagioclase with low dihedral angle.

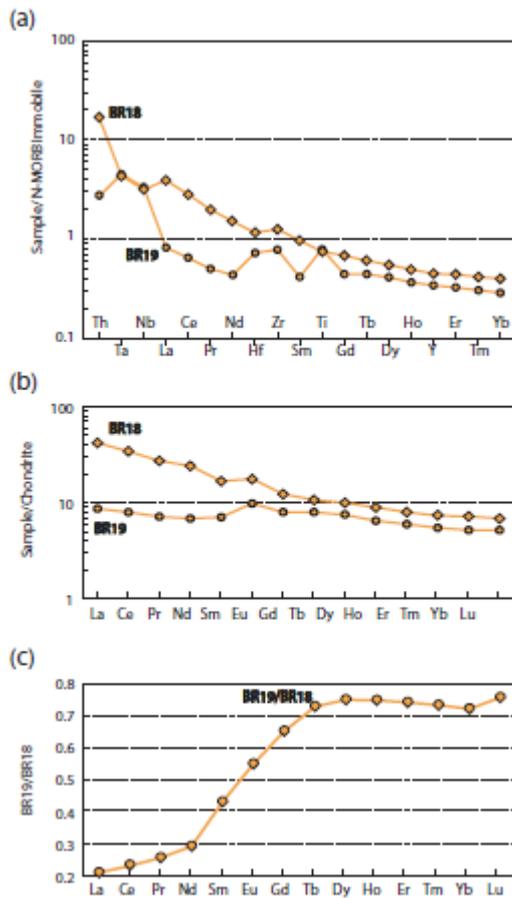


Figure 4: Geochemical plots. (a) Immobile-element N-MORB-normalized spidergram. (b) Chondrite-normalized REE pattern. Note the depleted pattern of BR19 relative to BR18. (c) Sample/sample normalized REE pattern. This pattern is very similar to eclogite residue after partial melting documented by Wang et al. (2014). The plots were generated with GCDKit (Janoušek et al., 2006). Normalizing chondritic and N-MORB values after Sun and McDonnough (1989).

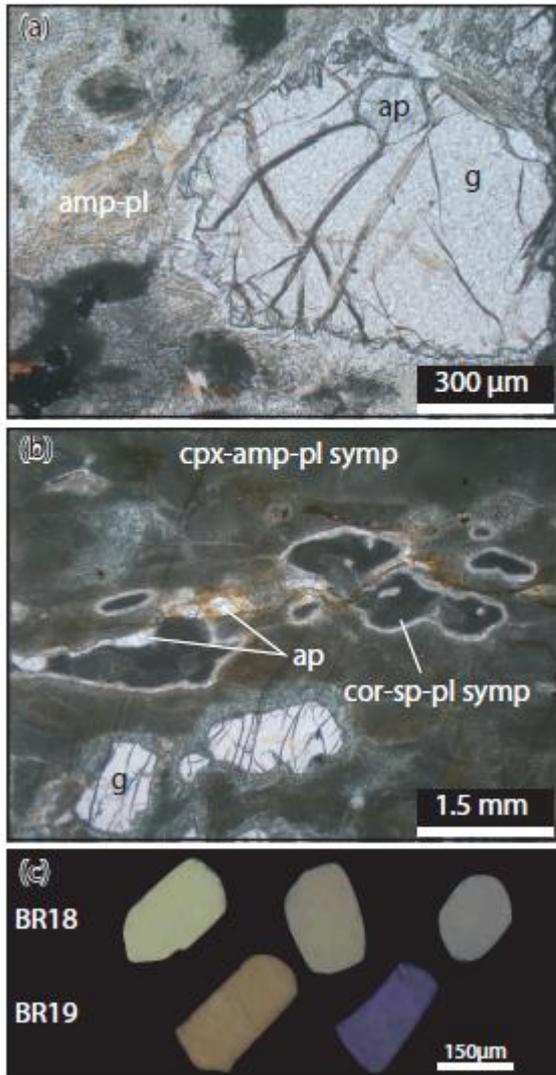


Figure 5: Photomicrographs (PPL) of an apatite inclusion in garnet (sample BR18) (a) and an apatite cluster oriented parallel with former kyanite crystals (sample BR18) (b). (c) CL images of separated apatite crystals.

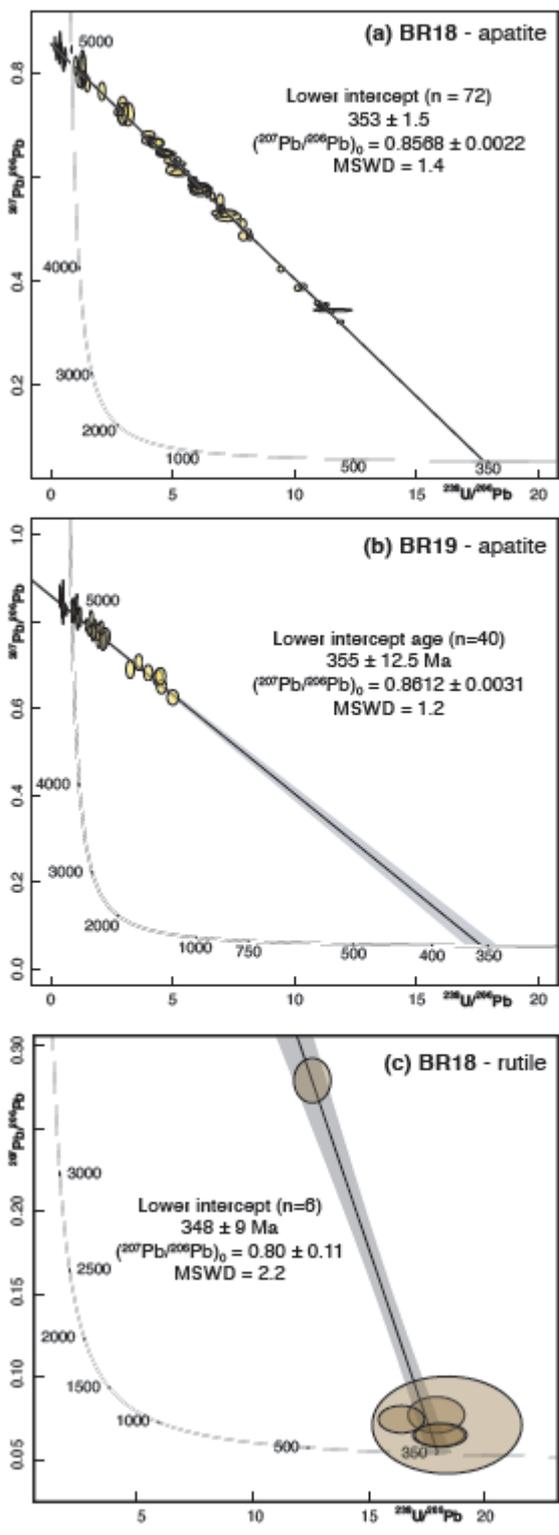


Figure 6: Tera-Wasserburg plots of U-Pb data from apatite from samples BR18 (a) and BR19 (b) and rutile from BR18 (c).

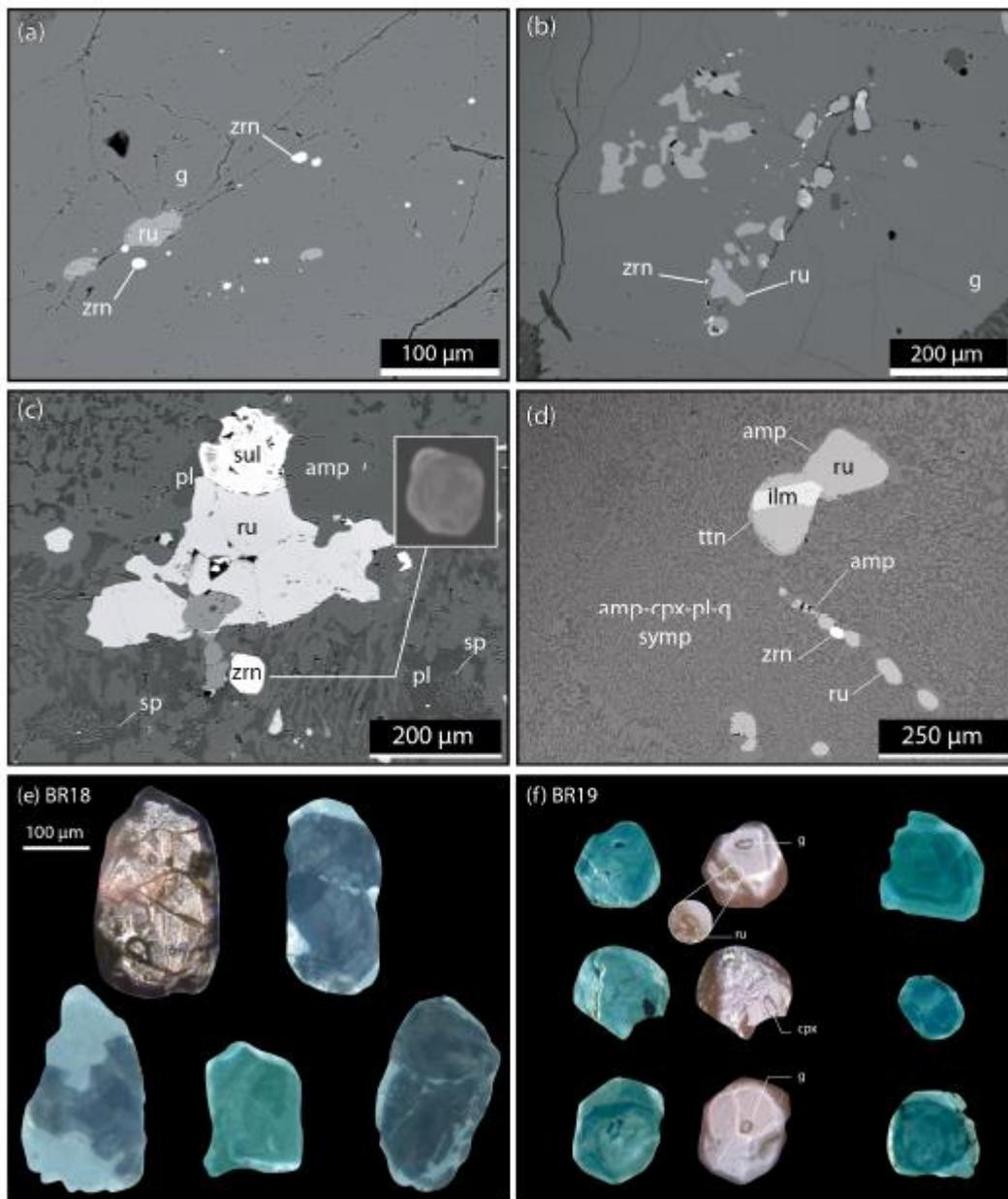


Figure 7: Zircon internal textures and textural relations. (a-b) BSE images of zircon included in garnet. Note the spaced coronitic textures of zircon around rutile in (b). (c-d) BSE images of zircon associated with rutile in the matrix. Inset in (c) is a CL image of the zircon. Compare (a-b) and (c-d) and note that numerous tiny zircons are included in garnet but single and larger zircons are observed in the matrix. (e-f) CL images and photomicrographs of selected zircons. (e) Note that the CL-bright zircon forms rims and embayments, or seals

fractures in the CL-dark zircon domains. CL-bright crystals are present in both samples. (f)  
Note the coexistence of patchy or flow textures, sector and oscillatory growth zoning in the  
CL-bright zircons. Microinclusions were determined by Raman spectroscopy.

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Figure 8: Photomicrograph (PPL) of planar inclusion trails in zircon. The white arrows mark the tip of the inclusion trails.

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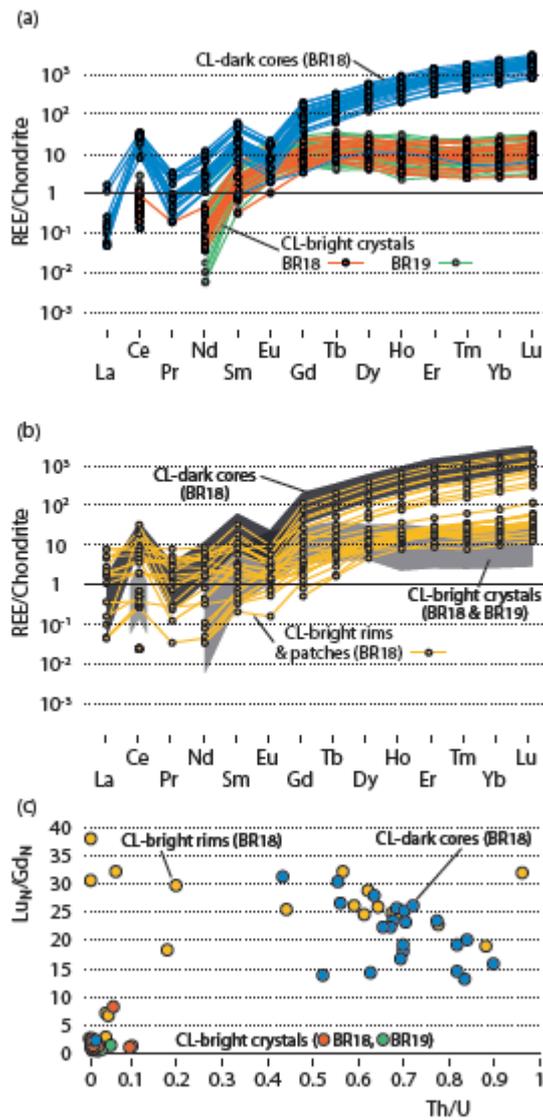


Figure 9: Trace-element characteristics of zircon. Chondrite-normalized trace-element patterns in the CL-dark cores and CL-bright crystals (a), and the CL-bright rims (b). (c) Lu<sub>N</sub>/Gd<sub>N</sub> vs Th/U plot of spots in BR18 showing two distinct endmembers and scattering of spots in the CL-bright crystals. Same colours as (a, b).

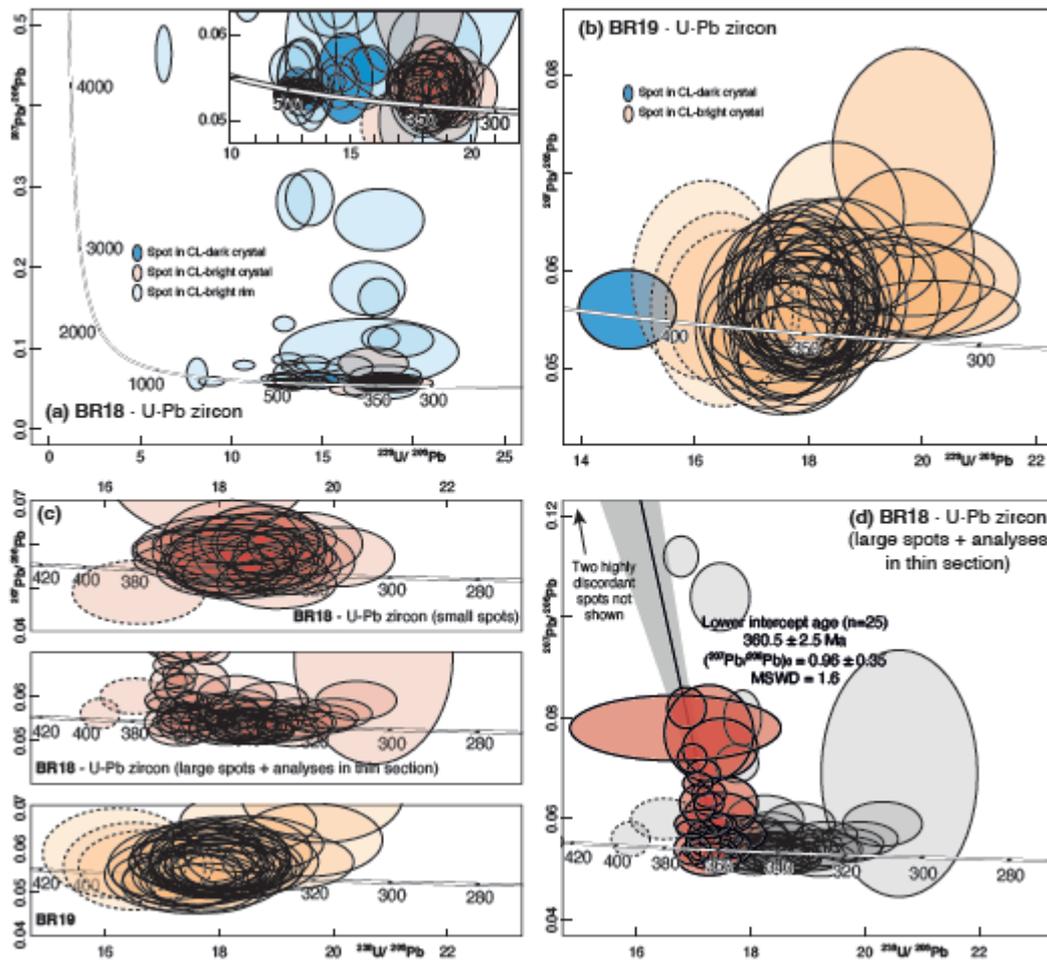


Figure 10: Tera-Wasserburg plots of zircon U-Pb data. (a) All zircon data from BR18 and zoom on the two main cluster. (b) All zircon data from BR19. (c) Comparison between the data obtained in BR18 with small spots (top plot), large spots and analyses in thin section (middle plot) and in BR19 (bottom plot). (d) Zircon data obtained using large spot size and directly in thin section. The ellipses coloured in red are used to calculate the lower intercept age. Note also the important spread along the concordia (grey ellipse with thin contour), the scarce apparently older spots (grey ellipses with dashed contour) and that two highly discordant spots (that are not used in the calculation of the discordia) are not shown.

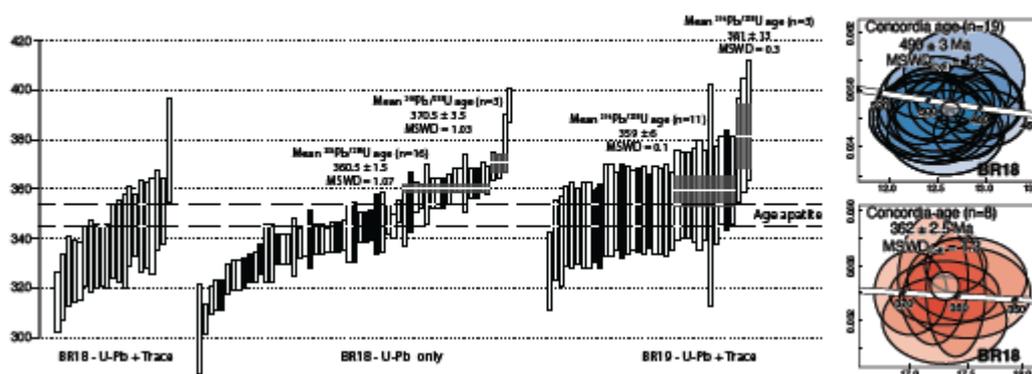


Figure 11: Weighted Mean Average (WMA) and concordia age plots of the U-Pb data in zircon. The long-dashed lines on the WMA plot correspond to the interval of the discordia ages obtained from apatite. Analyses in zircons with flow textures are shown as white bars and those with oscillatory and sector zoning as black bars. The concordia ages have been calculated using the data obtained in the CL-dark zircon cores (blue ellipses) and the CL-bright crystals (red ellipses).  $MSWD_{C+E}$  for concordance + equivalence.

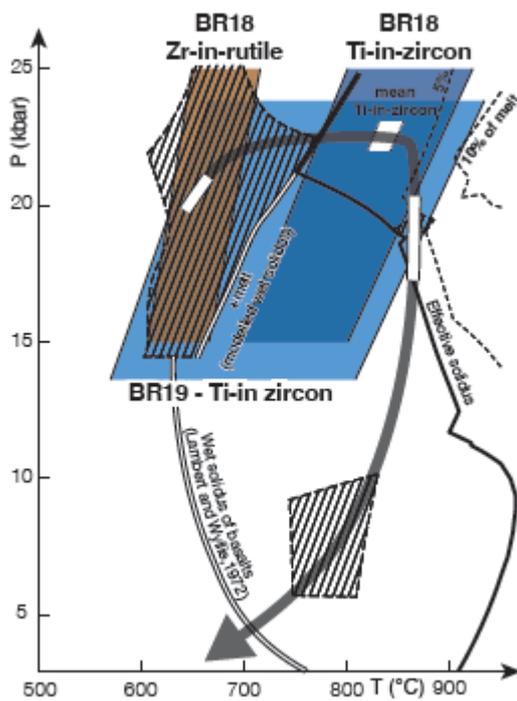


Figure 12: Ti-in-zircon and Zr-in-rutile thermometry results. The  $P$ - $T$  path is drawn after pseudosection modelling in de Hoÿm de Marien (2020). The hatched areas correspond to the stability fields of the critical mineral assemblages. White portions on the  $P$ - $T$  path indicate recognized periods of zircon growth.

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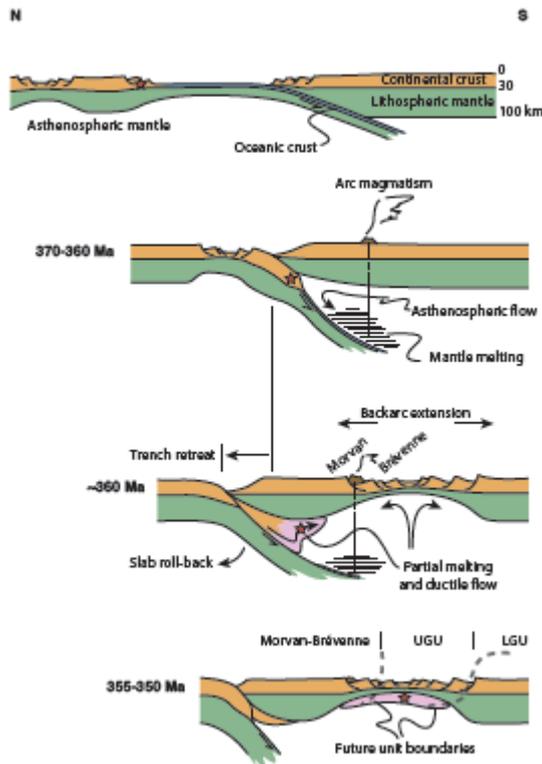


Figure 13: Sketch of the Devonian to early Carboniferous evolution of the French Massif Central. The red star represents the studied eclogite. Following subduction (>370 Ma) and the arrival of buoyant continental crust in the subduction zone, the slab steepens providing space for asthenospheric flow (~370) resulting in heating of the subducted crust up to partial melting and incipient upward detachment from the subducting slab (360 Ma). Note the contemporaneous development of the arc magmatism (Morvan). Detachment of the buoyant crust from the subducting lithosphere results in shallowing the dip of the slab by moving the trench toward the subducting plate (slab roll-back). The trench retreat in turn causes extension, thinning and incipient rifting of the upper lithosphere (including the magmatic arc) and associated magmatism (Brévenne; 360 Ma). The partially molten portion of the subducted lithosphere then flows toward the spreading center and emplace at crustal depths around 355-350 Ma.

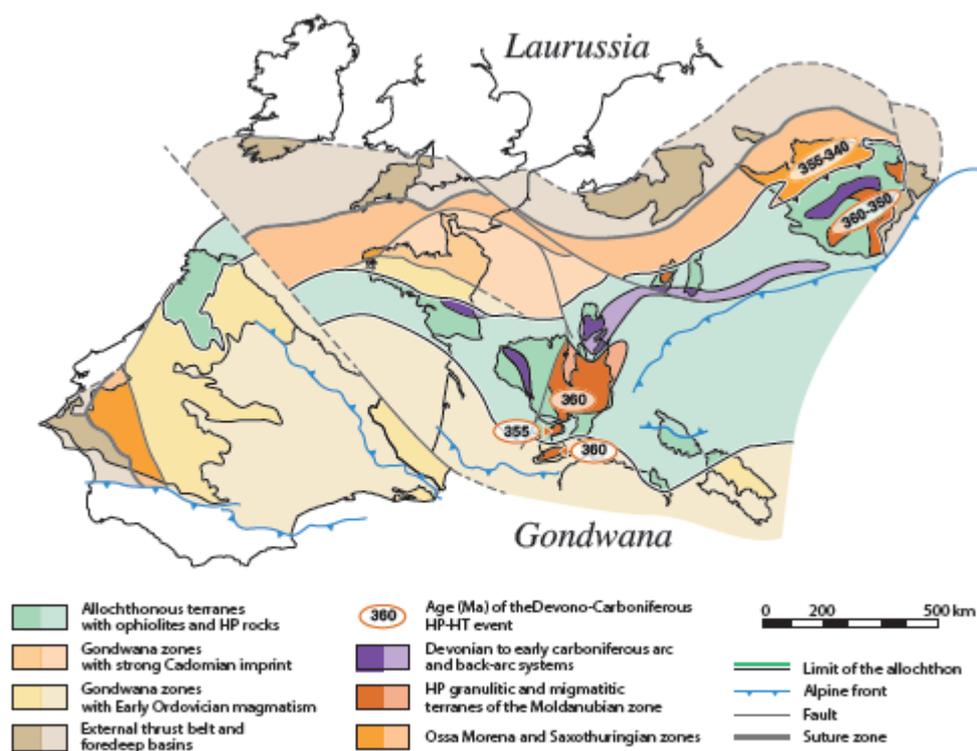


Figure 14: Sketch of the main Variscan massifs and terranes of the European Variscan orogen (modified after Martínez Catalán et al., 2007; 2021 and Schulmann et al., 2022) highlighting the possible correlations between the eastern FMC and the Bohemian Massif based on the occurrence and age of the Devonian-Carboniferous HP-HT rocks and the location of the associated arc and back-arc systems. Differences between the correlations before and after the present contribution can be seen by comparison with Fig. 1a.

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