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Multiple crust reworking in the French Armorican Variscan belt: implication for the genesis of uranium-fertile leucogranites

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9 **Keywords: peraluminous granites, zircon U-Pb dating, Hf-Nd isotope analyses, Variscan**
10 **orogeny, uranium metallogenesis**

11 **Abstract**

12 Muscovite peraluminous granites (MPGs) form by partial melting of the continental crust and
13 can be related to metalliferous deposits such as tin, tungsten and uranium (U). Metal enrichment
14 in MPGs commonly results from fractional crystallization, but the metal contents of the source
15 play a major role for their fertility. Between ca. 320 and 300 Ma (Late Carboniferous), the
16 French Armorican Variscan belt was intruded by numerous U-fertile MPGs that contain
17 inherited zircon grains with a wide range of ages from Archean-to-Carboniferous. U-Pb and Hf
18 isotopic data of zircon grains from Brioverian-to-Carboniferous sediments, Cambrian-to-Early
19 Carboniferous granitoids and Late Carboniferous MPGs indicate that the crust of the Armorican

Massif is made up by detritus mainly derived from the West African craton (3500-1600 Ma; $T_{DM} = 3.8-2.3$ Ga), Grenvillian belt (1200-900 Ma; $T_{DM} = 2.7-1.2$ Ga) and Avalonian-Cadomian belt (800-550 Ma; $T_{DM} = 2.5-0.8$ Ga), and that the crust was affected by magmatic events at 510-470 Ma ($T_{DM} = 1.6-0.6$ Ga), 410-330 Ma ($T_{DM} = 1.6-1$ Ga) and 320-300 Ma. Furthermore, they reveal that the Late Carboniferous MPGs were mainly formed by partial melting of Brioverian sediments with Cambro-Ordovician and Devonian-Carboniferous granitoids, which are all genetically linked with each other and characterized by $Th/U < 4$. The new data suggest that the U-fertile MPGs result from multiple reworking of U-rich Brioverian sediments, deposited ca. 550 Ma ago on the northern margin of Gondwana, and partially molten during several Paleozoic events, causing a successive increase in U content in the middle-upper crust.

Introduction

In late orogenic setting, the partial melting of the crust during extensional and/or transcurrent tectonic regimes commonly lead to the emplacement of peraluminous granitoids (e.g. Barbarin 1999) including cordierite bearing peraluminous granites (CPGs), muscovite bearing peraluminous granites (MPGs; Barbarin 1996, 1999) and related LCT (lithium-cesium-tantalum) pegmatites (Černý and Ercit 2005). Peraluminous granites and pegmatites are generally considered to form by the partial melting of sedimentary rocks at lower or intermediate crustal levels (e.g. Bernard-Griffiths et al. 1985; Le Fort et al. 1987; Vielzeuf and Holloway 1988; Patiño-Douce and Johnston 1991; Montel and Vielzeuf 1997; Patiño-Douce 1999; Shaw et al. 2016) but highly peraluminous melts can also be produced through the partial melting of mica rich igneous rocks (e.g. Turpin et al. 1990; Castro et al. 1999; Villaseca et al. 2012; García-Arias et al. 2015; Gao et al. 2016; Laurent et al. 2017; López-Moro et al. 2017). MPGs and related pegmatites can be associated with various metalliferous deposits such as tin (Sn), tungsten (W), uranium (U), lithium (Li) and tantalum (Ta). The genesis of these deposits relates significantly to fractional crystallization and hydrothermalism but the pre-concentration

of these lithophile elements in the source region submitted to partial melting is a necessary requirement for the genesis of peraluminous granites and pegmatites associated with economically significant metal deposits.

Recent studies (Romer and Kroner 2015, 2016) suggested that the formation of granites associated with Sn-W deposits and LCT pegmatites may result from weathering-related rare element enrichment in sediments prior to partial melting. Epicontinental black shales deposited after major glaciation events also have the capacity to trap a large amount of U (Cuney 2010) and their partial melting can lead to the formation of U rich melts. Emplacement of igneous rocks formed by the partial melting of an enriched lithospheric mantle can also result in the enrichment of incompatible element in the continental crust (Cuney 2010). Furthermore, crustal reworking can achieve significant metal enrichment at the scale of the lower, middle and upper crust. During granulite-facies metamorphism, dehydration of the lower crust can lead to the production of fluids enriched in F, large ion lithophile elements and rare metals. These fluids, preferentially channeled along regional shear zones, may enhance the partial melting of the middle crust resulting in the formation of magmas enriched in rare metals and U (Cuney and Barbey 2014). A comparable model was proposed by Eglinger et al. (2016) in the Pan African Lufilian belt where a crustal-scale Ca-Na metasomatism has leached U from the basement, and transported and redistribute it into supracrustal rocks along the brittle-ductile transition. Finally, crustal-derived magmatism can also induce significant metal mobility at the scale of the crust. Such process is well recorded in the uraniferous Mesoproterozoic western Namaqua province (South Africa) where voluminous granitic magmatism, recorded for a period of about 200 Ma, led to progressive U enrichment of the lower-middle crust (e.g. Andreoli et al. 2006).

In the European Variscan belt (EVB), a large proportion of the hydrothermal U deposits are spatially associated with uraninite bearing low-Ca Late Carboniferous MPGs (e.g. Cathelineau et al. 1990; Cuney et al. 1990; Tischendorf and Förster 1994; Ballouard et al. 2017a, in press)

(Fig. 1) and in the Armorican Massif (French part of the EVB) 20.000 t of U (20 % of the French production; UDEPO database: <https://infcis.iaea.org>) were extracted from the deposits spatially associated with the syntectonic MPGs (leucogranites) emplaced in the southern part of the massif (Fig. 2). Overall, most of the MPGs emplaced along the south Armorican shear zone (SASZ), a lithospheric scale strike-slip fault (e.g. Jégouzo 1980; Gumiaux et al. 2004a, b; Gapais et al. 2015), appear to be U-fertile, either because they are the source for several economically significant U deposits, such as the Mortagne (Cathelineau 1982) and Pontivy (Marcoux 1982; Ballouard et al., in press) MPGs or because they liberated an important amount of U during post-magmatic hydrothermal alteration, such as the Questembert MPG (Tartèse et al. 2013). In contrast, MPGs that were emplaced in extensional setting to the south of the SASZ (Gapais et al. 2015), are barren. The only exception is the U-fertile Guérande MPG, which is spatially associated with U deposits (Cathelineau 1981; Ballouard et al. 2017a). The distribution of the U-rich MPGs in the Armorican Massif fits widely the distribution of the so-called high heat production and flow belt (HHPFB; Fig. 2) defined by Jolivet et al. (1989) and Vigneresse et al. (1989). This belt shows a NW-SE orientation, is ca. 50 km wide, and characterized by an elevated heat flux. Granites in this belt have heat production values more than twice the values of the surrounding geological formations (Jolivet et al. 1989; Vigneresse et al. 1989). Vigneresse et al. (1989) therefore suggested that the HHPFB reflects the presence of an upper to intermediate continental crust enriched in radioactive elements and that partial melting of this crust caused the formation of U-fertile MPGs.

Recent studies focused mainly on the detailed deciphering of intramagmatic and hydrothermal processes leading to the genesis of the U deposits associated with the Guérande and Pontivy MPGs (Ballouard et al. 2015a, 2017a, 2017b, in press). However, the more global reasons behind the U-fertility of the MPGs in the Armorican Massif, and particularly the role of their source(s), remain unclear. To place new constraints on this open issue new sets of data

are presented in this study, comprising whole rock trace elements and Nd-Sr isotopic analyses, U-Pb ages and Hf isotopic data of inherited zircon grains from Late Carboniferous MPGs, detrital zircon from Brioverian to Carboniferous sedimentary rocks and zircon crystals from pre-Late Carboniferous igneous rocks surrounding the MPGs. Finally, the implication of the new data sets for the genesis of U-fertile MPGs is discussed.

Geological context

The Armorican Massif belongs to the European Variscan Belt (EVB), a Paleozoic orogenic belt that extends from Western (Iberian Massif) to Central Europe (Bohemian Massif) and results from the convergence of the Gondwana and Avalonia continents (Fig. 1;) (Ballèvre et al. 2009, 2013, 2014; Kroner and Romer 2013). The oldest rocks from the European Variscan belts are the Icartian orthogneisses that outcrop sporadically to the north of the Armorican Massif (Vidal et al. 1981). These, ca. 2 Ga old metagranitoids, are the witnesses of a Paleoproterozoic continental crust (Samson and D'lemos 1998). The Neoproterozoic-Cambrian Cadomian orogeny represents the main Pre-Variscan orogenic event (Linnemann et al. 2008, 2014). Strong Cadomian imprints are preserved in the Saxo-Thuringian domains of the Bohemian and Iberian Massifs, and the main type locality is the northern part of the Armorican Massif (Figs. 1 and 2). The Cadomian-Avalonian orogen is the result of the succession of variable geodynamic events occurring along the northern margin of the West African craton (Gondwana). These events include the formation of a continental magmatic arc from ca. 750 to 570 Ma, back-arc basin closure followed by arc-continent collision from ca. 570 to 540 Ma and major granitoid magmatism at ca. 540 Ma (Linnemann et al. 2008, 2014, Koglin et al. 2018). During the Cambro-Ordovician (ca. 510 – 470 Ma), a widespread extension event, leading to the separation of continental micro-blocks (Ibero-Armorica) from the northern Gondwana margin, induced an important magmatism related to the emplacement of various granitoids and volcanic rocks (Fig. 1) (Ballèvre et al. 2012, 2013, 2014; Kroner and Romer 2013; Villaseca et

al. 2016; Koglin et al. 2018). Subduction of oceanic then continental domains in the EVB occurred until ca. 350-340 Ma and transition from a compressive to an extensive regime occurred at ca. 315-310 Ma (Kroner and Romer 2013; Ballèvre et al. 2014). From Middle Carboniferous to Early Permian (e.g. Romer et al. 2007; Martínez Catalán et al. 2014; Laurent et al. 2017; Ballouard et al. 2017b), the EVB experienced an important magmatism leading to emplacement of crustal-derived MPGs and CPGs as well as hybrid or mantle-derived K-rich K-feldspar porphyritic calc-alkaline (KCGs) and amphibole-rich calc-alkaline (ACGs) granitoids (Barbarin 1999) (Fig. 1). The EVB represents an important metallogenic province for U where vein, episyenite-type, breccia-hosted or shear zone-hosted U deposits occur throughout the belt (Fig. 1). A large proportion of these hydrothermal deposits are spatially associated with Late Carboniferous MPGs and mineralizing events mostly occurred during the Permian from ca. 300 to 260 Ma (e.g. Cathelineau et al. 1990; Cuney et al. 1990; Tischendorf and Förster 1994; Cuney and Kyser 2008; Romer et al. 2010; Ballouard et al. 2017a, in press).

The Armorican Massif is separated into three main domains by the north Armorican shear zone (NASZ) and the south Armorican shear zone (SASZ), two dextral crustal to lithospheric-scale strike-slip faults (Fig. 2) (e.g. Gumiaux et al. 2004b). The northern domain is mostly made of Cadomian basement that belonged to the upper crust during Variscan orogeny (Ballèvre et al. 2001; Brun et al. 2001). The central domain is dominantly composed of Brioverian (Neoproterozoic-Cambrian) to Lower Carboniferous sediments that were mostly deformed under greenschist facies conditions during strike-slip deformation (Gumiaux et al. 2004a). Brioverian sediments in this domain constitute a more than 1000 m thick monotonous and immature detrital succession of wacke-type sandstones (Dabard et al. 1996). The Ordovician to Lower Carboniferous sediments form a heterogeneous mostly siliciclastic succession with a thickness above 3500 m. Carbonates mostly occur in Devonian sediments whereas black shales are characteristic of the Silurian and the Upper Devonian (Pelhate 1994; Vidal et al. 2001). The

southern domain, that belongs to the internal part of the Variscan belt, is characterized by an intense deformation and by the presence of high grade metamorphic rocks (e.g. Gapais et al. 2015). Three tectono-metamorphic units can be distinguished in this domain and comprise from top to bottom: (i) HP-LT rocks, composed of Ordovician acid metavolcanic rocks (Ballèvre et al. 2012) and blueschists subducted and exhumed between ca. 370 and 350 Ma (e.g. Bosse et al. 2005), (ii) micaschists, and (iii) migmatite-bearing units (Fig. 2). Migmatites, mostly formed by partial melting of pre-Carboniferous sediments and Early Paleozoic orthogneisses and recorded peak P-T condition of 0.8 GPa, 750-850 °C (Jones and Brown 1990; Augier et al. 2015).

Magmatism in the Armorican Massif occurred during four main periods. During the Cadomian period from ca. 750 to 580 Ma, the northern domain was affected by acid and mafic arc magmatism followed by voluminous granitoid plutonism at ca. 540 Ma (Ballèvre et al. 2001, 2013). In the Early Ordovician times (ca. 490 – 470 Ma), intracontinental rifting was responsible for the emplacement of tholeiitic mafic magmas and voluminous acid metavolcanics (Vendée porphyroids) in the southern domain (Ballèvre et al. 2012), whereas Late Cambrian- Early Ordovician granitoids intruded the whole massif (Fig. 2). The Devonian-Early Carboniferous period is marked by the emplacement of gabbros and various granitoids (Marcoux et al. 2009; Capdevila 2010) (Fig. 2) as well as by the development of an extensive network of mafic dikes at ca. 360 Ma (Pochon et al. 2016). This period is related to Early Variscan subduction and initial collision. During the Late Carboniferous (from ca. 320 to 300 Ma), dextral wrenching of the central domain and crustal extension in the southern domain, led to the exhumation of migmatitic core complex (Gapais et al. 2015), coeval with the emplacement of three main types of granitoids (Fig. 2).

(1) MPGs: they were mostly emplaced either along extensional deformation zone in the internal domain of the Armorican Variscan belt, such as the Quiberon (Gapais et al. 2015) and

Guérande (309.7 ± 1.3 Ma: Ballouard et al. 2015a) MPGs, or along the SASZ such as the Questembert (316.1 ± 2.9 Ma: Tartèse et al. 2011b), Lizio (316.4 ± 5.6 Ma: Tartèse et al. 2011a) and Pontivy (316.7 ± 2.5 Ma: Ballouard et al. 2017b) MPGs. Some of them were also emplaced to the north of the SASZ such as the Langonnet (304.7 ± 2.7 Ma: Ballouard et al. 2017b), Huelgoat (314.0 ± 2.8 Ma: Ballouard 2016) and Saint-Renan MPGs (316.0 ± 2.0 Ma: Le Gall et al. 2014). Previous isotopic studies (Bernard-Griffiths et al. 1985; Euzen 1993; Tartèse and Boulvais 2010; Ballouard et al. 2015a, 2017a, 2017b) revealed that these MPGs are characterized by high $\delta^{18}\text{O}$ ratios (> 11), sub-chondritic to chondritic $\epsilon\text{Nd}(\text{T})$ and elevated initial $^{87}\text{Sr}/^{86}\text{Sr}$ (> 0.705) values in agreement with a crustal origin.

(2) CPGs: the Rostrenen (Euzen 1993; Ballouard et al. 2017b) and Huelgoat (Georget 1986) CPGs were emplaced at 315.5 ± 2.0 Ma (Ballouard et al. 2017b) and 314.8 ± 2.0 Ma (Ballouard 2016), respectively. These granites were emplaced synchronously with small stocks of mantle-derived mafic to intermediate igneous rocks.

(3) KCGs: these metaluminous biotite $\pm$ hornblende granitoids consist of a magnesian-potassic (Mg-K) and ferro-potassic (Fe-K) suite emplaced between 325 and 300 Ma and associated with mantle-derived mafic to intermediate rocks (Peucat et al. 1984; Capdevila, 2010; Caroff et al. 2015; Ballouard et al. 2015b).

At a global scale, crustal partial melting to the south of the SASZ, during the Late Carboniferous, was triggered by lithospheric thinning during late orogenic extension. In contrast, to the north of the SASZ, partial melting of the crust and of a lithospheric mantle, metasomatized during previous subduction events, were likely induced by asthenosphere upwelling during diffuse strike-slip deformation of the central domain and subsequent slab-tearing (Ballouard et al. 2017b).

In the Armorican Massif, U was mostly mined in the district of Mortagne, Pontivy and Guérande. In the Guérande and Pontivy districts, the MPGs were the main source for the U mineralization found in the intra to perigranitic vein and episyenite type hydrothermal deposits. Leaching of magmatic U oxides from the MPGs was promoted by the infiltration at depth of oxidized surface-derived hydrothermal fluids (Ballouard et al. 2017a, in press). U-Pb dating of U oxides from the Guérande, Pontivy and Mortagne district deposits reveal several mineralizing events that occurred by pulse from ca. 300 to 260 Ma (Cathelineau et al. 1990; Ballouard et al. 2017a, in press).

Samples and methods

A description and the GPS coordinates of Late Carboniferous MPGs, sedimentary rocks, and Cambro-Ordovician granitoid samples selected for whole-rock major and trace element analyses as well as zircon U-Pb and Hf isotope analyses are provided in Table 1 and Supplementary file 1.

MPGs selected for U-Pb (and Hf) isotope analyses on inherited zircon grains include the U mineralized Guérande and Pontivy MPGs as well as the Questembert, Lizio, Langonnet and Hulgoats MPGs that are not spatially associated with U deposits (Fig. 2). Most sandstone or siltstone samples (Brioverian to Devonian) were collected in the region of Crozon in the western edge of the central Armorican domain (Fig. 2). In addition, a Lower Carboniferous (this study) and Devonian sandstone (Ducassou et al. 2014) were sampled in the Châteaulin basin (western part of the central domain, Fig. 2) and the Chalonnès region (eastern part of the central domain, Fig. 2), respectively. One metagranite (sample PLG-1), two metatonalites (samples PLG-2 and PLG-4) and one undeformed granite (sample PLG-3) from the Lower Paleozoic Plouguenast orthogneissic complex (central domain, Fig. 2), as well as one metatonalite (sample QIMP-1) from Moëlan (southern domain, Fig. 2) were also selected for zircon U-Pb dating.

Whole rock isotopic, major and trace element analyses

Large samples (5 to 10 kg) were crushed in the Geosciences Rennes Laboratory using agate mortars. Chemical analyses were performed by the Service d'Analyse des Roches et des Minéraux (SARM; CRPG-CNRS, Nancy, France) using a ICP-AES for the major elements and a ICP-MS for the trace elements following the techniques described in Carignan et al. (2001). The results of the analyses are provided in Supplementary file 2. Whole rock Sr and Sm-Nd isotope analyses were performed at Geosciences Rennes, on two samples from the Guérande MPG (muscovite-tourmaline coarse grained facies, see Ballouard et al. 2015a) and one Lower Carboniferous sandstone sample from the Châteaulin Basin. The methodology and analytical conditions are described in Ballouard et al. (2017b) and the results of the isotope analyses are provided in Supplementary file 3.

Zircon U-Pb and Hf isotope analyses

A classical mineral separation procedure was applied to concentrate zircon grains using the facilities available at Geosciences Rennes (Ballouard et al. 2015a). Zircon grains were imaged by cathodoluminescence (CL) using a Reliotron CL system equipped with a digital color camera available at the Geosciences Rennes laboratory.

U-Pb dating of zircon was performed by in-situ laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) using a ESI NWR193UC Excimer laser coupled to a quadrupole Agilent 7700 x ICP-MS equipped with a dual pumping system to enhance sensibility. The methodology and analytical protocol used to perform the analyses can be found in Ballouard et al. (2015a) and Supplementary file 4. The analyses with a degree of concordance between 90 and 110% are provided in Supplementary file 5 with errors listed at 1σ . Histograms (50 Ma bins) and Kernel density estimates (KDE, 25 Ma bands) of the zircon U-Pb dates were realized using the DensityPlotter software (Vermeesch 2012). $^{206}\text{Pb}/^{238}\text{U}$ dates were used for

zircon with a $^{207}\text{Pb}/^{206}\text{Pb}$ date < 1.0 Ga whereas $^{207}\text{Pb}/^{206}\text{Pb}$ dates were used for zircon with a $^{207}\text{Pb}/^{206}\text{Pb}$ date > 1.0 Ga.

Hafnium (Hf) isotope analyses were performed at Goethe-University Frankfurt using a Thermo-Finnigan NEPTUNE multicollector ICP-MS coupled to a Resolution M-50 (Resonetics) 193 nm ArF Excimer laser (ComPexPro 102F, Coherent), using the procedure outlined in detail in Gerdes and Zeh (2006, 2009) and summarized in Supplementary file 4. The epsilon Hf values [$\epsilon\text{Hf}(t)$] were calculated using the chondritic uniform reservoir (CHUR) as recommended by Bouvier et al. (2008; $^{176}\text{Lu}/^{177}\text{Hf} = 0.0336$ and $^{176}\text{Hf}/^{177}\text{Hf} = 0.282785$) and a decay constant of $1.867 \times 10^{-11} \text{ yr}^{-1}$ (Scherer et al. 2001; Söderlund et al. 2004). The initial $^{176}\text{Hf}/^{177}\text{Hf}$, $\epsilon\text{Hf}(t)$ values and hafnium model ages (T_{DM}) (for zircon grains with a degree of concordance between 90 and 110%) were calculated using $^{206}\text{Pb}/^{238}\text{U}$ ages for zircon with $^{206}\text{Pb}/^{207}\text{Pb}$ ages < 1.0 Ga and $^{206}\text{Pb}/^{207}\text{Pb}$ ages for zircon with $^{206}\text{Pb}/^{207}\text{Pb}$ ages > 1.0 Ga. The result of analyses and additional information are provided in Supplementary file 6.

Results

Geochemical features and U-Th distribution

All investigated Late Carboniferous MPGs of the Armorican Massif are highly peraluminous and characterized by A/CNK [$\text{Al}_2\text{O}_3 / (\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O})$, molar proportions] values > 1.1 (Fig. 3). However, the Huelgoat MPG plot on a different trend than the others MPGs, with relatively higher A/NK values at low A/CNK ratio, reflecting a higher CaO content. In Figure 4, these MPGs are mostly characterized by low and highly variable whole-rock Th/U ratio mostly from ~0.1 to 2, as well as variable U content from 1 to 27 ppm.

The whole-rock composition of silicoclastic sediments and peraluminous igneous rocks representing potential source for the Late Carboniferous MPGs were also reported in the U versus Th diagram (Fig. 4). The U contents of the Brioverian and Paleozoic sediments

(Ordovician to Devonian) range between ~1 and 7 ppm. However, the Paleozoic sediments generally show much higher Th/U ratios (between ~4 and 7), i.e., above the average value of the upper continental crust (UCC ~ 4), compared to most Brioverian sediments (Th/U ratio mostly between ~2 and 4). Most Ordovician (meta)volcanics (Lower Paleozoic) are characterized by U contents between ~1 and 7 ppm and variable Th/U ratios between ~2 and 20 whereas Cambro–Ordovician (Lower Paleozoic) and Early Carboniferous (Pertre MPG) granitoids show more variable U contents between ~2 and 40 ppm and are generally characterized by relatively low but variable Th/U ratios ranging from ~0.5 to 4.

Whole-rock Sr-Nd isotope data

Most MPGs are characterized by elevated initial $^{87}\text{Sr}/^{86}\text{Sr}$ [I_{Sr} (315 Ma)] ratios ranging from 0.706 (Pontivy MPG) and 0.719 (Guérande MPG) and negative ϵNd (315 Ma) values ($-2.3 > \epsilon\text{Nd}$ (315 Ma) > -8.0) with two stage Nd model ages (T_{DM}) between 1.2 (Pontivy and Langonnet MPG) and 1.8 Ga (Guérande MPG, Fig. 5). However, two samples from the Pontivy MPG display super-chondritic ϵNd (315 Ma) values (1.2 and 2.2) with relatively low I_{Sr} (0.704 and 0.706) ratios and young T_{DM} (1.0 and 0.9 Ga). As noticed by Bernard-Griffiths et al. (1985), the I_{Sr} (315 Ma) and ϵNd (315 Ma) values of MPG increase and decrease, respectively, from north to south with an evolution from the Pontivy–Langonnet-Lizio MPG, Questembert MPG to Guérande MPG (Figs. 2 and 5).

U-Pb dating

Inherited zircon from MPG

Zircon grains from all investigated MPGs of the Armorican Massif are generally euhedral. They commonly show inherited cores surrounded by magmatic rims in CL images (Fig. 6). U-Pb ages obtained on inherited zircon cores or grains from the MPGs are reported in form of histograms and KDE in Figure 7. In general, five ages populations can be distinguished which,

however do not occur in all MPGs: (1) an Archean-Mesoproterozoic population at 3500-1400 Ma, (2) a Grenvillian population at 1200-900 Ma, (3) a Neoproterozoic population at 900-540 Ma, (4) a Cambro-Ordovician population at 540-440 Ma and (5) a Silurian–Lower Carboniferous population at 440-330 Ma (Table 1). All MPG samples are dominated by zircon grains of the populations (3), (4) and (5). A significant amount of population (1) zircon occurs only in the Pontivy MPG (8%), and of population (2) zircon in the Guérande MPG (8%; see Fig. 7 and Table 1).

Detrital zircon from sediments

U-Pb dates obtained on detrital zircon grains from the main sedimentary formations of the Armorican Massif are reported in Figure 8 and summarized in Table 1.

Zircon grains from the different sedimentary rocks show an euhedral to rounded shape and appear zoned or homogeneous in CL images (Fig. 6). The detrital grains reflect similar age populations (1 to 5) than the ones obtained from the xenocrysts in the MPGs (Table 1). However, the amount of Archean-to-Mesoproterozoic grains (population 1) is higher in the sediments compared to the MPGs, especially in Silurian and Devonian sediments (Fig. 8 and Table 1). The most significant difference is that some samples contain abundant zircon grains of Grenvillian age (population 2), e.g., the Silurian and Devonian sediments from Crozon (24-32%), whereas they are completely absent or very scarce in the Brioverian sandstone from Crozon (1%) and the Lower Carboniferous sediments from Châteaulin (0%). It is important to note that zircon age spectra similar to those of Silurian and Devonian sediments were obtained for an Ordovician sandstone from Crozon (Matteini et al. 2014). A minor Grenvillian population is also observed in the Devonian sandstone from Chalonnès (10%; Fig. 8 and Table 1).

Zircon from Cambrian to Lower Carboniferous peraluminous (meta)igneous rocks

Results of U-Pb dating reveal that the (meta)granitoids from Plouguenast and Moëlan (Fig. 2) intruded between 504.5 ± 1.8 (PLG-2) and 466.2 ± 3.6 Ma (QIMP-1) – (for details see text and diagrams in Supplementary file 7), and that most of them contain inherited components (Fig. 9a and Table 1). Zircon shapes are euhedral to sub-euhedral, and CL images reveal well-developed grow zonation for most grains (Fig. 6). Xenocrystic zircon crystals in the (meta)granitoids overlap in age with the previously defined populations 1 (4%) and 3 (18%, Table 1). Zircon analyses with apparent Silurian to Devonian ages (7%) are interpreted to result from Pb loss (see Supplementary file 7 for details).

The result of zircon U-Pb dating performed by Ballèvre et al. (2012) on acid peraluminous (meta)volcanics from the southern domain (Vendée Porphyroids, Fig. 2) are reported in Figure 9b and Table 1. These (meta)volcanics contains Neoproterozoic xenocrysts (6%), belonging to the population 3 defined in the Late Carboniferous MPGs, whereas the Early Paleozoic grains (population 4: 93%) brackets the age of intrusion from 494 ± 4 to 472 ± 4 Ma (Ballèvre et al. 2012). These ages are much older than those obtained on zircon grains from the Pertre MPG (Vernhet et al. 2009) in the central domain (Fig. 2) that yield $^{206}\text{Pb}/^{238}\text{U}$ ages between 362 ± 2 and 330 ± 4 Ma (population 5, 93 %) with a marked peak at ca. 340 Ma, interpreted to reflect the time of intrusion (Fig. 9c and Table 1) (Vernhet et al. 2009). One Neoproterozoic xenocryst belongs to the population 3 defined in the Late Carboniferous MPGs whereas two zircon grains with apparent ages of 323 ± 3 and 321 ± 4 Ma, respectively, reflect Pb loss (Vernhet et al. 2009).

Hf analyses

The results of Hf isotope analyses on inherited zircon grains from MPGs are reported in a $\epsilon\text{Hf}(t)$ versus age diagram (Fig. 10a). The Hf isotope compositions of inherited zircon grains in all investigated MPGs are similar. The rare Archean-to- Paleoproterozoic grains (2890-1590 Ma) show superchondritic to subchondritic compositions ($\epsilon\text{Hf}(t) = 3.9$ to -11.5), whereas

Mesoproterozoic-to-Carboniferous xenocrysts (1035-329 Ma) yield mostly superchondritic $\epsilon\text{Hf}(t)$ up to +9.1 (> 60%). Superchondritic xenocrysts are predominant in the Pontivy (-14.3 to +8.3), Lizio (-4 to +8.2) and Langonnet (-10.3 to +8.4) MPGs, and mostly characterized by hafnium model ages (T_{DM}) between 1.6 and 0.9 Ga. In contrast, the Questembert MPG (-12.1 to 7) is dominated by subchondritic grains, even though most of them also show T_{DM} between 1.6 and 0.9 Ga. Then, zircon grains from the Guérande MPG (-14.2 to 9.1) show a wide and relatively continuous range of $\epsilon\text{Hf}(t)$ from sub- to superchondritic (Fig. 10a). About 70% of zircon are characterized by T_{DM} between 1.6 and 0.9 Ga whereas others show older T_{DM} between 1.60 and 2.3 Ga.

The $\epsilon\text{Hf}(t)$ values of detrital zircon grains from the Brioverian and Lower Carboniferous sediments shows a remarkable overlap with those of the MPGs (Figs. 10b and 10d): Archean-Mesoproterozoic zircon grains (3080-1430 Ma) show T_{DM} from 3.7 to 2.2 Ga and about 75 % of the Neoproterozoic-Carboniferous grains (851-329 Ma) are characterized by T_{DM} between 1.6 and 0.9 Ga. In contrast, detrital zircon grains in the Silurian and Devonian sedimentary rocks show significantly different Mesoproterozoic-to-Cambrian $\epsilon\text{Hf}(t)$ spectra characterized by a wide range in $\epsilon\text{Hf}(t)$ at 1200-900 Ma (-38 to +10.8; T_{DM} = 3.8-1.2 Ga), and by about 70 % of subchondritic $\epsilon\text{Hf}(t)$ for ages < 900 Ma (-37 to +8; T_{DM} = 3.6-1.1 Ga; Fig. 10b). This difference is also reflected by the generally much older Hf model ages for the Neoproterozoic-to-Cambrian population, with more than 50 % between 1.6 and 2.5 Ga (Fig. 10b). The Hf isotopic data of the Archean-to-Paleoproterozoic grains (2922-1841 Ma; T_{DM} = 3.5-2.5 Ga) in the Silurian-Devonian sediments overlap with those in the Brioverian-Carboniferous sediments and in the MPGs.

The $\epsilon\text{Hf}(t)$ values of magmatic zircon grains from the Cambro-Ordovician metagneous rocks (510-460 Ma) vary from -0.7 to +14.2, corresponding to hafnium model ages between 1.4 and 0.6 Ga (Fig. 10c). It is pertinent to note, that these T_{DM} overlap with those obtained

from most Neoproterozoic-to Carboniferous xenocrysts in the MPGs (Figs. 10a and 10d). However, there is also an important number of grains showing significantly higher $\epsilon_{\text{Hf}}(t)$, very close to the depleted mantle. Such grains were mostly found in sample PLG-1 from the Plouguenast orthogneissic complex (Fig. 10c).

Discussion

Pre- to Early Variscan zircon record

Neoproterozoic to Early Cambrian zircon grains (850 – 530 Ma, Fig. 7a) in the Brioverian sediments likely result from magma mixing, which occurred during the formation of the Cadomian-Avalonian belt, an Andean-type cordillera located at the northern margin of Gondwana, e.g. to the north of the Armorican Massif (Fig. 2a), between approximately 800 and 540 Ma (e.g. Nance and Murphy 1994; Dabard et al. 1996; Ballèvre et al. 2001; Brun et al. 2001; Zeh et al. 2001; Linnemann et al. 2008, 2014; Koglin et al. 2018). This hypothesis is supported by the vertical array in the $\epsilon_{\text{Hf}}(t)$ vs. age diagram (array CA in Figure 10d), reflecting the interaction of crustal melts formed by reworking of Archean-to-Paleoproterozoic crust of the West Africa craton ($\epsilon_{\text{Hf}}(t)$ down to -22), and mantle-derived melts ($\epsilon_{\text{Hf}}(t)$ up to +13). Then, most Archean-to-Mesoproterozoic zircon grains (3080 – 1430 Ma; Fig. 8a) likely originated from the erosion of the West African craton (Fig. 10d), that represented the hinter land of the Cadomian orogeny (e.g. Zeh et al. 2001; Linnemann et al. 2008, 2014), whereas the ca. 2000 Ma old Icartian orthogneisses (e.g. Samson and D’Lemos 1998) only represented a minor source (e.g. Dabard et al. 1996).

The Silurian and Devonian sediments might result from internal reworking of a very heterogeneous Grenvillian crust (indicated by the vertical array G in Fig. 10d), with minor contribution from the Cadomian-Avalonian crust. The Grenville zircon grains are very likely to be derived from a southern Gondwana-connected source, whereas a northern Baltica

provenance can be excluded. Indeed, Grenvillian zircon populations were found in many Paleozoic sediments throughout the EVB, commonly associated with a predominant Neoproterozoic-Cambrian (800-500 Ma), and a subordinate Archean-to-Palaeoproterozoic zircon population (1700-3500 Ma), and with a pronounced age gap between 1.2 and 1.7 Ga (e.g. Zeh et al. 2001; Linnemann et al. 2008, 2014). In fact, these are the same features observed in the Armorican sediments during this study (Fig. 10b). In contrast, zircon grains derived from Baltica, have ages mostly between 1.0 and 1.8 Ga, as it has been demonstrated for Paleozoic (meta)sediments from the Rhenohercynian Domain (Avalonia-Laurussia in Fig. 1), and the Mid-German Crystalline Zone (northwestern edge of the Saxo-Thuringian in Fig. 1) of the eastern EVB (Geisler et al. 2005; Zeh and Gerdes 2010).

The overlap in zircon Age-Hf isotopic data suggests that the Cambro-Ordovician (meta)granitoids of the Armorican Massif were predominantly formed by partial melting of Brioverian sediments (Fig. 10d). However, the highly superchondritic $\epsilon\text{Hf}(t)$ (near to depleted mantle) signature of some zircon grains from the granitoids of the Plouguenast orthogneissic complex additionally reveals that new (juvenile) crust was formed at the same time. In this context it is pertinent to note that Cambro-Ordovician juvenile crust formation (in addition to crustal melting) is not restricted to the Armorican Massif (e.g. Bernard-Griffiths et al. 1986; Ballèvre et al. 2012) but has been reported from many other areas throughout the EVB, e.g., from the NW Iberian Massif (e.g. Arenas et al. 2016), French Massif Central (e.g. Chelle-Michou et al. 2017) and Saxothuringian Domain (e.g. Bankwitz et al. 1994; Höhn et al. 2017), where it is related to the breakup of continental microterranes from the northern Gondwanan margin (e.g. Kroner and Romer 2013). The relatively wide variation in $\epsilon\text{Hf}(t)$ of the Cambro-Ordovician metaigneous rocks (array R in Fig. 10d) is ambiguous. It could be explained by heterogeneities of the source (Brioverian sediments), but, more probably, by mixing of juvenile

melts (depleted mantle-related) and crustal-derived melts, formed by the partial melting of Brioverian sediments.

The Age-Hf-isotopic data furthermore suggest that the Brioverian sediments and the Cadomian-Avalonian basement together with Cambro-Ordovician igneous rocks were the main sources for the detrital zircon grains in the Lower Carboniferous sediments (Fig. 10b). Moreover, a significant addition from zircon grains derived from Early Variscan granitoids emplaced between ~400 and 330 Ma (array EV in Fig. 10d) such as the ca. 340 Ma old Pertre granite (Fig 9c), is revealed by isotopic data. Most 390 to 330 Ma old zircon grains in the Lower Carboniferous sediments are characterized by T_{DM} from 1.2 to 1.6 Ga that overlap with those of Neoproterozoic zircon in the Brioverian sediments (Figs 10b and d), suggesting that these zircon grains crystallized in a crustal-derived granitic magma formed by the partial melting of Brioverian sediments. This hypothesis is in agreement with the presence of a ca. 625 Ma old zircon xenocryst in the Pertre granite (Fig. 9c, Table 1).

Crustal sources of Late Carboniferous MPGs

The highly peraluminous character ($A/CNK > 1.1$, Fig. 3) of the MPGs is in agreement with a crustal origin and suggest that they were produced by the partial melting of mica-rich rocks with a peraluminous composition (e.g. Gao et al. 2016).

Whole-rock Nd and Sr isotope analyses indicate a significant difference in composition between the MPGs emplaced to the north of the SASZ (i.e. Pontivy, Langonnet, Lizio) and the Guérande MPG intruded to south of the SASZ (Figs. 2 and 5). The Questembert MPG, which emplaced between the two branches of the SASZ, has an intermediate composition. The negative $\epsilon Nd(315 \text{ Ma})$ values of the Guérande MPG (-7.6 to -9.5) could be explained by the partial melting of Cambro-Ordovician acid (meta)volcanic rocks as well as Ordovician to Devonian sediments from the south Armorican domain, although some samples plot at the

lower boundary of the range defined by Brioverian sediments from the central domain (Fig. 5). The $\epsilon\text{Nd}(315 \text{ Ma})$ values of the Questembert MPG (-3.9 to -5.7) overlap with those obtained from the Brioverian sediments from the central domain as well as with Cambro-Ordovician acid (meta)volcanic rocks. Then, the composition of the Pontivy, Langonnet and Lizio MPGs (-2.3 to -4.7) is similar to that of Brioverian sediments from the central domain as well as the Cambro-Ordovician (meta)granitoids and (meta)volcanic rocks. The super-chondritic $\epsilon\text{Nd}(315 \text{ Ma})$ composition of two Pontivy MPG samples could be explained by the partial melting of Cambro-Ordovician (meta)granitoids. However, the Nd isotopic values on their own are ambiguous, and do not allow a detailed source characterization. Two reasons may be proposed at this stage. First, the MPG melts can be derived from several sources and, second, some sources show a wide range in ϵNd values (Fig. 5), e.g., the Cambro-Ordovician acid (meta)volcanics rocks, which themselves were formed by melting of even older crustal components such as Brioverian sediments (Ballèvre et al. 2012).

Combined U-Pb and Hf isotopic data of zircon xenocrysts (2890-329 Ma; Table 1; Figs. 7 and 10d) suggest that the Late Carboniferous MPGs were formed by partial melting of a heterogeneous crust mainly consisting of Brioverian sediments, Cambro-Ordovician acid igneous rocks and Devonian to Early Carboniferous granitoids. In contrast, Ordovician to Devonian sedimentary rocks seems to play only a subordinate role during U-fertile MPGs formation for several reasons. First, they contain an important Grenvillian population (1.2-0.9 Ma: this study, Matteini et al. 2014) with a wide range in $\epsilon\text{Hf}(t)$ (-38 to 10.8), which is barely seen in the xenocrysts from the MPGs (Fig. 10d). The only exceptions are four grains (8%) in the Guérande MPG and one grain in the Lizio (3%) and Pontivy (1%) MPGs, respectively (Table 1, Figs 7). Second, the Silurian and Devonian sediments contain a pronounced Archean to Mesoproterozoic population (3.5-1.4 Ga; 23-36%) which is not observed in the MPGs (< 10%, Table 1). Third, detrital zircon grains from Silurian to Devonian sediments with ages <

0.9 Ga mostly show subchondritic $\epsilon\text{Hf}(t)$ values (ca. 70%), corresponding to Hf model ages of 2.5 to 1.6 Ga, which are much older than those of most MPGs (0.9 to 1.6 Ga). On the other hand, a significant number of Neoproterozoic to Carboniferous (750-329 Ma) zircon with old T_{DM} from 1.6 to 2.5 Ga (Fig. 10a) together with some Grenvillian grains (1080-974 Ma; Fig. 7f) in the Guerande MPG suggests a minor contribution of Ordovician to Devonian sediments in its source, in agreement with its high sub-chondritic ϵNd composition (Fig. 5). Moreover, the contribution of Lower Carboniferous sediments in the source of all MPGs is unlikely and difficult to explain structurally. Indeed, the central domain that was mainly deformed under strike-slip deformation regime during the Carboniferous was poorly thickened during the Variscan orogeny (Gumiaux et al. 2004a; Gapais et al. 2015) and the burying, and subsequent partial melting, of Lower Carboniferous sediments probably never occurred in the central domain. Then, it is unlikely that important partial melting of the Lower Carboniferous sediments occurred in the region without partial melting of Ordovician to Devonian sediments that are structurally below. Alternatively, the partial melting of the Devonian to Early Carboniferous granitoids such as the 391-385 Ma old peraluminous Plounévez-Lochrist orthogneiss (Marcoux et al. 2009) (Fig. 2) can explain the younger range of inheritance observed in all the MPGs (population 5, Table 1). Finally, metatexites and diatexites derived from pre-Carboniferous sediments and Ordovician orthogneisses are abundant in the southern Armorican domain (e.g. Augier et al. 2015) (Fig. 2). These rocks reached peak P-T conditions of 0.8 GPa, 750-850 °C followed by near isothermal decompression down to 0.4 GPa, 750 °C and melting occurred, during the Late Carboniferous, from water saturated solidus to biotite breakdown conditions (Augier et al. 2015; Jones and Brown, 1990), in agreement with the role of Brioverian sediments and Paleozoic orthogneisses as a major source for the MPGs.

Implication for the U-fertility of MPGs

The results of this study show that the U-fertile Pontivy and Questembert MPGs, and the U-unfertile Lizio, Langonnet and Hulgoat MPGs, all emplaced to the north of the southern branch of the SASZ, result from the melting of the same heterogeneous crust, which consisted of Brioverian sediments, Cambro-Ordovician acid igneous rocks and Devonian-to-Carboniferous granitoids. Similar hafnium model ages furthermore suggest that all these rocks are genetically linked, and result from the successive internal reworking of Brioverian sediments (Fig. 10d). Age-Hf isotopic data also suggest that the U-fertile Guérande MPG, emplaced to the south of the SASZ, result from the partial melting of a similar crust with only a minor contribution from Ordovician to Devonian sediments.

Thus, fertility or non-fertility of the Late Carboniferous MPGs from the Armorican Massif is obviously controlled primarily by differences during the reworking process, comprising local variations of source rocks (sediment vs. orthogneiss), degree of melting and fractionation, and the melting history itself (unique or multiple), as well as hydrothermal processes during and after each magmatic cycle. The complex interplay of all these processes has been demonstrated by recent studies suggesting that the Lizio and Questembert MPGs represent “twin” leucogranites but that the Questembert MPG is more differentiated than the Lizio MPG because the former traveled a greater vertical distance in the crust, enhancing crystal segregation from the melts and U enrichment in the residual liquids (Tartese and Boulvais 2010; Tartèse et al. 2013).

The formation of U rich peraluminous liquid is favored by an elevated U content of its sedimentary and/or igneous sources. However, the proportion of U located outside the lattice of accessory minerals poorly soluble in peraluminous melts, such as zircon and monazite (Watson and Harrison 1983; Montel 1993), has a primordial importance for the genesis of U-fertile MPGs (e.g. Cuney 2014). In the absence of a detailed petrographic study, the whole-rock Th/U ratio can be used as a preliminary indicator of the fertility of a rock for the genesis of U-

rich melts during anatexis. High Th/U ratios, above the average value of the upper continental crust (UCC Th/U value = 3.8, Rudnick and Gao 2003), suggest that most U is hosted in Th-bearing, highly refractory, minerals such as monazite (xenotime and zircon), whereas Th/U ratios < 3.8 suggest that a significant amount of U is localized along microcracks or in adsorption on the surfaces of rock forming minerals. In most favorable cases, peraluminous igneous rocks with Th/U < 1 and U contents > ~10 ppm can host U oxides (Peiffert et al. 1996, 1994). By taking these points into account, we suggest that the U-fertile MPGs (~0.1 < Th/U < ~2, Fig. 4) of the Armorican Massif were formed by partial melting of peraluminous mica-rich lithologies characterized by Th/U < ~4 and U content > ~2.7 ppm (average UCC values). Such rocks comprise the Brioverian sediments, but also Cambro-Ordovician and Devonian-Early Carboniferous peraluminous igneous rocks (Fig. 4). In contrast, the Ordovician to Devonian sediments and Cambro-Ordovician acid volcanic rocks with Th/U > 4 and U < 2.7 ppm, can be considered as less fertile. Our new data also suggest that U-fertility of a crust increases if one-and-the same (peraluminous) crust is affected by multiple melting events, which obviously occurred in the Armorican Massif during the internal reworking of already U-rich (low Th/U) Brioverian sediments during the Cambro-Ordovician, Devonian-Lower Carboniferous and finally during the Upper Carboniferous, in particular along or close to the crustal to lithospheric scale shear zones. More specifically, multiple crustal reworking can induce progressive U enrichment of some crustal domains (i.e. middle crust) and in late orogenic context, the melting of these domains can lead to the formation of U-fertile granites. These interpretations are in good agreement with the fact that peraluminous orthogneisses of Cambro-Ordovician, and Devonian-Early Carboniferous ages occur in the source regions of nearly all Late Carboniferous MPGs throughout the EVB, comprising the French Massif Central (Turpin et al. 1990; Laurent et al. 2017), the Iberian Massif (e.g. Villaseca et al. 2012 ; López-Moro et al. 2017), and the Bohemian Massif in Germany and the Czech Republic (Tichomirowa et al. 2012), three

historical mining provinces for granite-related hydrothermal U deposits (Fig. 1). In this context, it is interesting to note that, the Cambro-Ordovician Saint-Goueno MPG from the Plouguenast orthogneissic complex in the Armorican Massif (Fig. 2) hosts intragranitic U occurrences (Carric et al., 1980), suggesting that it contained magmatic U oxides, and reached U-fertility long before the Variscan orogeny.

Comparison with other uraniferous provinces worldwide

In the Armorican Variscan belt, multiple partial melting of the crust is associated with progressive increase of the U-fertility of crustal-derived granitoids with time (Fig. 4). This is revealed by, in general, a progressive decrease of the Th/U ratio from the Brioverian sediments ($\sim 2 < \text{Th/U} < \sim 4$), Cambrian-Early Carboniferous granitoids ($\sim 0.5 < \text{Th/U} < \sim 4$) to Late Carboniferous MPGs ($\sim 0.1 < \text{Th/U} < \sim 2$). To see if this behavior can be generalized to other crustal domains, we report in Figure 11 the evolution of the Th-U contents of granitoids from two major U provinces for comparison, from:

(1) the Archean Gabonese craton that surrounds the Paleoproterozoic Franceville basin, well known for hosting U deposits with natural fission reactors such as Oklo. Ultimately, the U from the deposits likely originates from the leaching of U-rich minerals present in the basin, such as monazite, thorite and possibly uraninite, themselves coming from the erosion of the surrounding Archean basement (e.g. Gauthier-Lafaye and Weber 2003). In the basement, granitoid plutonism occurred from 2.94 to 2.71 Ga with an evolution from TTG gneiss, grey granitoids, pink granitoids to Neoarchean granites and pegmatites (Thiéblemont et al. 2009).

(2) the Mesoproterozoic western Namaqualand province, South Africa, representing a granulite- to amphibolite-facies metamorphic terrane characterized by the presence of granites and charnokites highly enriched in U and Th (Andreoli et al. 2006). In the

region, crustal-derived granitic magmatism occurred over a period of ca. 200 Ma (Robb et al. 1999).

Despite the different degree of enrichment, by one order of magnitude, the U (and Th) contents of the granitoids from both provinces increase progressively and significantly with time. In the Gabonese craton, U increase from ~0.1-0.2 ppm (TTG), ~0.2-1 ppm (grey granitoids) to ~1-4 ppm (pink granites), and in the Namaqualand province, from ~1-10 ppm (Little Namaqualand suite), 3-60 ppm (Early Spektakel suite) to 3-800 ppm (Late Spektakel suite). In the two provinces, fractionation of U over Th, a process that can lead, ultimately, to the crystallization of magmatic uraninite (Cuney, 2014), is only recorded by the youngest granites and pegmatites (Neoproterozoic granites and pegmatites in Fig. 11a and granites of the Late Spektakel suite in Fig. 11b). To summarize, the recycling of the crust by multiple partial melting events seems to be an important mechanism for the concentration of U, the fractionation of U over Th, and finally for the genesis of U-fertile granites.

Conclusion

(1) Age-Hf isotopic data of detrital zircon grains in Brioverian to Carboniferous sediments, magmatic and inherited zircon crystals in Cambrian to Early Carboniferous (meta)igneous rocks, and xenocrysts in the Late Carboniferous MPGs indicate that the crust of the Armorican Massif is made up by detritus derived from three major sources: West African craton (3500-1600 Ma; $T_{DM} = 3.8$ to 2.2 Ga), Grenville belt/domains (1200-900 Ma; $T_{DM} = 2.7$ to 1.2 Ga), and predominately Avalonian-Cadomian belt (800-550 Ma; $T_{DM} = 2.5$ to 0.8 Ga). Moreover, the crust was affected by magmatic events at 510-460 Ma (Cambro-Ordovician; $T_{DM} = 1.6$ to 0.6 Ga), 410-330 Ma (Devonian-Carboniferous; $T_{DM} = 2.1$ to 1.0 Ga), and 320-300 Ma (Late Carboniferous).

(2) These data also reveal that the Late Carboniferous MPGs (300-320Ma) were mainly formed by partial melting of Brioverian sediments, Cambro-Ordovician igneous rocks and Devonian-Early Carboniferous granitoids, which are all genetically linked with each other, as is indicated by a remarkable overlap in their Hf model ages between 1.6 to 0.8 Ga. In contrast, Ordovician, Silurian and Devonian sediments of the Armorican Massif contain a significant Grenvillian zircon population (~ 30%) with mostly older Hf model ages between 1.6 and 2.5 Ga, which are barely seen in the Late Carboniferous MPGs (Huelgoat, Langonnet, Pontivy, Lizio and Questembert), except for a minor amount in the Guérande MPG (~ 10%).

(3) In contrast to Ordovician to Devonian sediments ($\text{Th/U} > 4$), Brioverian sediments and Cambrian to Early Carboniferous granitoids characterized by $\text{U} > 2.7$ ppm and $\text{Th/U} < 4$ represent an ideal source for the generation of U-fertile MPGs.

(4) All data suggests that the U-fertile MPGs of the Armorican Massif result mainly from multiple internal reworking of relatively U-rich Brioverian sediments, which were originally deposited along the northern Gondwanan margin at ca. 550 Ma, and became partially molten during several Paleozoic events, causing a successive increase in U-content by crystal fractionation, and intermediate hydrothermal processes in the middle-upper crust. Similar multiple, internal reworking processes of Neoproterozoic-Cambrian sediments is recorded by rocks from many areas across the EVB hosting economically important U deposits, such as Iberia, French Massif Central and Bohemian Massif.

(6) Comparison with other U provinces of different ages worldwide suggest that crustal reworking by multiple partial melting events is a major mechanism for U concentration, fractionation of U over Th, and the genesis of U-fertile granites.

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Captions

Fig.1: Schematic representation of the west European Variscan belt representing the main terranes (Ballèvre et al. 2009, 2014; Ballèvre 2016) and showing the distribution of U deposits (Cuney and Kyser 2008), Variscan granitoids and Early Paleozoic felsic to intermediate igneous rocks (after the 1:1 000 000 geological maps of France [Chantraine et al. 2003], Portugal and Spain [Civis Llovera (2015)] and the 1:500 000 geological maps of Czechoslovakia [Fusán et al. 1967] and Czech Republic [Cháb et al. 2007]). The granitoids typology is from Barbarin (1999). MPG: muscovite-bearing peraluminous granites; CPG: cordierite-bearing peraluminous granitoids; KCG: K-rich and K-feldspar porphyritic calc-alkaline granitoids. ACG: amphibole-rich calc-alkaline granitoids. NASZ: North Armorican shear zone; NBSASZ: northern branch of the south Armorican shear zone; SBSASZ: southern branch of the south Armorican shear zone. NEF: Nort-sur-Erdre fault. Mineral abbreviations according to Kretz (1983).

Fig. 2: (a) principal structural domains from the Armorican Massif. (b) General geological map of the Armorican Massif [modified from Chantraine et al. (2003), Gapais et al. (2015) and Ballouard et al. (2017b, in press)] identifying the different types of Carboniferous granitoids after Barbarin (1999) and Capdevila (2010) and localizing the U deposits (<http://infoterre.brgm.fr>). The high heat production and flow belt (HHPFB) from Vignerresse et al. (1989) and Jolivet et al. (1989) is indicated. Locality numbers are referenced in Table 1. NASZ: north Armorican shear zone; NBSASZ: northern branch of the South Armorican shear zone; SBSASZ: southern branch of the south Armorican shear zone. Fe-K granites: ferro-potassic granites. Mg-K granites: magnesio-potassic granites. Mineral abbreviations according to Kretz (1983).

Fig. 3: A/NK [$\text{Al}_2\text{O}_3 / (\text{Na}_2\text{O} + \text{K}_2\text{O})$] versus A/CNK [$\text{Al}_2\text{O}_3 / (\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O})$] diagram (molar proportion) reporting the composition of Late Carboniferous MPGs from the Armorican Massif. Data

are from Ballouard et al. (2015a, 2017a), Euzen (1993), Georget (1986), Tartèse and Boulvais (2010), Tartèse et al. (2012) and this study.

Fig. 4: U versus Th diagram reporting the whole-rock composition of Late Carboniferous MPGs from the Armorican Massif and their potential sources. Data are from this study, Georget (1986), Vigneresse et al. (1989), Euzen (1993), Béchenec et al. (1996, 1999, 2001), Trautmann and Carn (1997), Dabard and Peucat (2001), Le Hébel (2002), Tartèse and Boulvais (2010), Béchenec and Thiéblement (2013), Tartèse et al. (2012) and Ballouard et al. (2015a, 2017b). The average U content (2.7 ppm) and Th/U value (~4) of the upper continental crust (UCC; Rudnick and Gao 2003) are indicated.

Fig. 5: Initial Sr and Nd isotopic composition calculated at 315 Ma of Late Carboniferous MPGs from the Armorican Massif (Data from this study; Ballouard et al. 2015a, 2017b; Tartèse and Boulvais 2010). The yellow double arrow represents the $\epsilon\text{Nd}(315 \text{ Ma})$ composition of the Langonnet MPG (Ballouard et al. 2017b) whereas other vertical bars represent the $\epsilon\text{Nd}(315 \text{ Ma})$ composition of Cambro-Ordovician peraluminous igneous rocks [acid (meta)volcanics: Ballèvre et al. 2012; (meta)granitoids: Ballouard et al. 2017b] as well as Brioverian (Dabard et al. 1996; Dabard 1997) and Paleozoic (Michard et al. 1985; Dabard and Peucat 2001; this study) sediments. The composition of ca. 2000 Ma Icartian orthogneisses from the Channel Islands (black arrow) is out of the diagram with $\epsilon\text{Nd}(315 \text{ Ma})$ values between -18 and -14 (Samson and D'Lemos 1998). The grey dashed arrow represents the north-south evolution of the MPG isotopic composition. Sm-Nd isotopic composition of CHUR from Bouvier et al. (2008).

Fig. 6: Representative cathodoluminescence pictures of zircon grains from MPGs, sediments and Cambro-Ordovician metagranitoids of the Armorican Massif analyzed during this study. Dashed circles represent the location of LA-ICP-MS U-Pb analyses. The corresponding U-Pb dates (1 σ uncertainty) and $\epsilon\text{Hf}(t)$ values are indicated. All zircon grains are shown at the same scale and the scale bar is indicated in the upper right corner. Carbon. = Carboniferous.

1024

1025 Figure 7: Histogram and Kernel density estimates (KDE) of U-Pb dates obtained on inherited zircon
1026 grains from Late Carboniferous MPGs of the Armorican Massif. Number in grey inside the diagrams
1027 corresponds to the age of peaks (Ma) obtained by KDE. Zones in color represent the main period of
1028 inheritance.

1029

1030 Fig. 8: Histogram and Kernel density estimates (KDE) of U-Pb dates obtained on detrital zircon grains
1031 from sedimentary rocks of the Armorican Massif. Zones in color represent the main period of inheritance
1032 recorded by zircon from the Late Carboniferous MPGs.

1033

1034 Fig. 9: Histogram and Kernel density estimates (KDE) of U-Pb dates obtained on zircon grains from
1035 Cambro-Ordovician and Early Carboniferous peraluminous igneous rocks of the Armorican Massif.
1036 Zones in color represent the main period of inheritance recorded by zircon from the Late Carboniferous
1037 MPGs.

1038

1039 Fig. 10: $\varepsilon\text{Hf}(t)$ versus U-Pb ages for (a) inherited zircon grains from Late Carboniferous MPGs, (b)
1040 detrital zircon from sediments (c) magmatic zircon from Early Paleozoic metagranitoids and (d) zircon
1041 from Late Carboniferous MPGs and their potential sources. In (d), the composition of magmatic zircon
1042 from Early Variscan granitoids is extrapolated using Devonian-Carboniferous zircon from Lower
1043 Carboniferous sediments. The main origin of zircon grains is indicated: WA = West African craton, G
1044 = Grenvillian belt, CA = Cadomian-Avalonian belt, R = Cambro-Ordovician Rift, EV = Early Variscan.
1045 The crustal evolution trends are calculated using a $^{176}\text{Lu}/^{177}\text{Hf}$ ratio of 0.0113 (Taylor and McLennan
1046 1985; Wedepohl 2016). Data for the Pontivy and Langonnet MPG are from Ballouard et al. (2017b)
1047 whereas other data are from this study.

1048

46

1049 Fig.11: U versus Th diagram reporting the whole-rock composition of granitoids from (a) the Archean
1050 Gabonese craton and (b) Mesoproterozoic western Namaqualand province, South Africa.
1051 Geochronological and Geochemical data for Gabon are from Bouton et al. (2009) and Thiéblemont et
1052 al. (2009) whereas data for Namaqualand are from Robb (1986, 1999), Raith (1995), Thomas et al.
1053 (1996), Clifford et al. (2004), Andreoli et al. (2006), Bailie et al. (2007) and Duchesne et al. (2007). The
1054 average U content (2.7 ppm) and Th/U value (~ 4) of the upper continental crust (UCC; Rudnick and
1055 Gao 2003) are indicated.

1056

1057 Table 1: summary of zircon U-Pb data. Pop. = population.

Figure 1

Figure 2

[Click here to download Figure 2.eps](#)

Figure 3

Figure 4

Figure 5

Figure 7

Figure 8

Figure 9

- Silurian - Lower Carboniferous (440 - 330 Ma)
- Lower Paleozoic (540 - 440 Ma)
- Neoproterozoic (900 - 540 Ma)
- Archean - Mesoproterozoic (3500 - 1400 Ma)

Figure 10

Table 1

Reference	Sample	Locality / intrusion name	Reference N° in Figure 2	Rock type	Intrusion / deposition age	Number U-Pb / Hf	Pop. 1 (%)	Pop. 2 (%)	Pop. 3 (%)	Pop. 4 (%)	Pop. 5 (%)
							3500 – 1400 Ma	1200 – 900 Ma	900 – 540 Ma	540 – 440 Ma	440 – 330 Ma
This study	LOC-1	Châteaulin basin	12	Sandstone	Lower Carboniferous	107 / 45	22	0	23	22	32
This study	CRO-2	Crozon	10	Sandstone	Upper Devonian	300 / 27	23	24	46	6	0
	CRO-1			Sandstone-siltstone	Middle Devonian						
	CRO-12			Sandstone	Lower Devonian						
Ducassou et al. (2014)	DevSA2	Chalonnès	11	Sandstone	Devonian	45 / 0	24	11	42	7	13
This study	CRO-6	Crozon	10	Sandstone	Silurian	213 / 54	36	32	29	3	0
	CRO-3			Sandstone-siltstone							
This study	CRO-9	Crozon	10	Sandstone	Brioverian	105 / 48	17	1	78	4	0
This study	PLG-1	Plouguenast	9	Metagranitoid (MPG)	Ordovician (477.9 ± 2.9 Ma)	216 / 37	4	0	18	71	7
	PLG-2			Metagranitoid (tonalite)	Cambrian (504.5 ± 1.8 Ma)						
	PLG-3			MPG	Ordovician						
	PLG-4	Metagranitoid (tonalite)	Ordovician (486.7 ± 8.8 Ma)								
	QIMP-1	Moëlan	8	Metagranitoid (tonalite)	Ordovician (466.2 ± 3.6 Ma)						
Ballèvre et al. (2012)	FL-31	Bréthomé		Felsic (meta)volcanics	Ordovician (472 ± 4 Ma)	71 / 0	1	0	6	93	0
	CG13a	Chie-Loup			Ordovician (486 ± 4 Ma)						
	CG5	Mareuil / Lay			Ordovician (478 ± 2 Ma)						
	CG09	La Sauzaie			Ordovician (491 ± 12 Ma)						
	FI2a	Sauzon			Ordovician (494 ± 4 Ma)						
Vernhet et al. (2012)	Pertre 1 Pertre 4 Pertre 6	Pertre	7	MPG	Lower Carboniferous (345 ± 3 Ma) Lower Carboniferous (348 ± 19 Ma) Lower Carboniferous (340 ± 2 Ma)	41 / 0	0	0	2	0	93
Ballouard et al. (2015)	GUE-3	Guérande	6	MPG	Upper Carboniferous (309.4 ± 1.9 Ma)	52 / 32	4	8	50	13	25
This study	GUE-4				Upper Carboniferous (309.7 ± 1.3 Ma)						
	GUE-5				Upper Carboniferous (302.5 ± 1.6 Ma)						
Ballouard et al. (2017b)	PONT-1	Pontivy	5	MPG	Upper Carboniferous (316.7 ± 2.5 Ma)	83 / 24	8	1	39	20	31
This study	PONT-10										
	PONT-14										
	PONT-15										
Ballouard et al. (2017b)	PONT-26				Upper Carboniferous (310.3 ± 4.7 Ma)						
Ballouard et al. (2017b)	PONT-20	Langonnet	4	MPG	Upper Carboniferous (304.7 ± 2.7 Ma)	28/ 18	4	0	39	32	25
This study	QRT-08	Questembert	3	MPG	Upper Carboniferous (316.1 ± 2.9 Ma)	19 / 15	5	0	21	47	26
This study, Tartèse et al. (2011a)	LRT-10	Lizio	2	MPG	Upper Carboniferous (316.4 ± 5.6 Ma)	29 / 18	0	3	59	28	10
Tartèse et al. (2011a)	LRT-15				Upper Carboniferous (319 ± 15 Ma)						
This study	HUEL-3	Huelgoat	1	MPG	Upper Carboniferous (314.0 ± 2.8 Ma)	18 / 0	6	0	50	17	28

Table 1: summary of zircon U-Pb data. Pop. = population.