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Christophe Ballouard, Marc Poujol, Armin Zeh. Multiple crust reworking in the French Armorican Variscan belt: implication for the genesis of uranium-fertile leucogranites. International Journal of Earth Sciences, 2018, 107 (7), pp.2317-2336. 10.1007/s00531-018-1600-3. insu-01717290

# HAL Id: insu-01717290 https://insu.hal.science/insu-01717290

Submitted on 26 Feb 2018  $\,$ 

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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

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

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Massif is made up by detritus mainly derived from the West African craton (3500-1600 Ma; 20 21  $T_{DM}$  = 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 22 510-470 Ma (T<sub>DM</sub> = 1.6-0.6 Ga), 410-330 Ma (T<sub>DM</sub> = 1.6-1 Ga) and 320-300 Ma. Furthermore, 23 they reveal that the Late Carboniferous MPGs were mainly formed by partial melting of 24 Brioverian sediments with Cambro-Ordovician and Devonian-Carboniferous granitoids, which 25 are all genetically linked with each other and characterized by Th/U < 4. The new data suggest 26 that the U-fertile MPGs result from multiple reworking of U-rich Brioverian sediments, 27 deposited ca. 550 Ma ago on the northern margin of Gondwana, and partially molten during 28 29 several Paleozoic events, causing a successive increase in U content in the middle-upper crust.

#### 30 Introduction

31 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 32 33 (e.g. Barbarin 1999) including cordierite bearing peraluminous granites (CPGs), muscovite 34 bearing peraluminous granites (MPGs; Barbarin 1996, 1999) and related LCT (lithium-cesiumtantalum) pegmatites (Černý and Ercit 2005). Peraluminous granites and pegmatites are 35 generally considered to form by the partial melting of sedimentary rocks at lower or 36 37 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 38 1999; Shaw et al. 2016) but highly peraluminous melts can also be produced through the partial 39 melting of mica rich igneous rocks (e.g. Turpin et al. 1990; Castro et al. 1999; Villaseca et al. 40 2012; García-Arias et al. 2015; Gao et al. 2016; Laurent et al. 2017; López-Moro et al. 2017). 41 42 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 43 relates significantly to fractional crystallization and hydrothermalism but the pre-concentration 44

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.

48 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 49 element enrichment in sediments prior to partial melting. Epicontinental black shales deposited 50 51 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 52 rocks formed by the partial melting of an enriched lithospheric mantle can also result in the 53 54 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 55 crust. During granulite-facies metamorphism, dehydration of the lower crust can lead to the 56 production of fluids enriched in F, large ion lithophile elements and rare metals. These fluids, 57 preferentially channeled along regional shear zones, may enhance the partial melting of the 58 59 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 60 Lufilian belt where a crustal-scale Ca-Na metasomatism has leached U from the basement, and 61 62 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. 63 Such process is well recorded in the uraniferous Mesoproterozoic western Namaqua province 64 (South Africa) where voluminous granitic magmatism, recorded for a period of about 200 Ma, 65 led to progressive U enrichment of the lower-middle crust (e.g. Andreoli et al. 2006). 66

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 70 French production; UDEPO database: https://infcis.iaea.org) were extracted from the deposits 71 spatially associated with the syntectonic MPGs (leucogranites) emplaced in the southern part 72 73 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; 74 Gapais et al. 2015), appear to be U-fertile, either because they are the source for several 75 76 economically significant U deposits, such as the Mortagne (Cathelineau 1982) and Pontivy 77 (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 78 79 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 80 spatially associated with U deposits (Cathelineau 1981; Ballouard et al. 2017a). The distribution 81 of the U-rich MPGs in the Armorican Massif fits widely the distribution of the so-called high 82 heat production and flow belt (HHPFB; Fig. 2) defined by Jolivet et al. (1989) and Vigneresse 83 et al. (1989). This belt shows a NW-SE orientation, is ca. 50 km wide, and characterized by an 84 elevated heat flux. Granites in this belt have heat production values more than twice the values 85 of the surrounding geological formations (Jolivet et al. 1989; Vigneresse et al. 1989). 86 87 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 88 this crust caused the formation of U-fertile MPGs. 89

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.

#### 100 Geological context

101 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 102 results from the convergence of the Gondwana and Avalonia continents (Fig. 1;) (Ballèvre et 103 al. 2009, 2013, 2014; Kroner and Romer 2013). The oldest rocks from the European Variscan 104 belts are the Icartian orthogneisses that outcrop sporadically to the north of the Armorican 105 106 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 107 108 Cadomian orogeny represents the main Pre-Variscan orogenic event (Linnemann et al. 2008, 109 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 110 Massif (Figs. 1 and 2). The Cadomian-Avalonian orogen is the result of the succession of 111 112 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 113 570 Ma, back-arc basin closure followed by arc-continent collision from ca. 570 to 540 Ma and 114 major granitoid magmatism at ca. 540 Ma (Linnemann et al. 2008, 2014, Koglin et al. 2018). 115 During the Cambro-Ordovician (ca. 510 - 470 Ma), a widespread extension event, leading to 116 117 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 118 volcanic rocks (Fig. 1) (Ballèvre et al. 2012, 2013, 2014; Kroner and Romer 2013; Villaseca et 119

al. 2016; Koglin et al. 2018). Subduction of oceanic then continental domains in the EVB 120 occurred until ca. 350-340 Ma and transition from a compressive to an extensive regime 121 occurred at ca. 315-310 Ma (Kroner and Romer 2013; Ballèvre et al. 2014). From Middle 122 123 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 124 emplacement of crustal-derived MPGs and CPGs as well as hybrid or mantle-derived K-rich 125 K-feldspar porphyritic calc-alkaline (KCGs) and amphibole-rich calc-alkaline (ACGs) 126 granitoids (Barbarin 1999) (Fig. 1). The EVB represents an important metallogenic province 127 for U where vein, episyenite-type, breccia-hosted or shear zone-hosted U deposits occur 128 129 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 130 Permian from ca. 300 to 260 Ma (e.g. Cathelineau et al. 1990; Cuney et al. 1990; Tischendorf 131 132 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 133 134 zone (NASZ) and the south Armorican shear zone (SASZ), two dextral crustal to lithosphericscale strike-slip faults (Fig. 2) (e.g. Gumiaux et al. 2004b). The northern domain is mostly made 135 of Cadomian basement that belonged to the upper crust during Variscan orogeny (Ballèvre et 136 137 al. 2001; Brun et al. 2001). The central domain is dominantly composed of Brioverian (Neoproterozoic-Cambrian) to Lower Carboniferous sediments that were mostly deformed 138 under greenschist facies conditions during strike-slip deformation (Gumiaux et al. 2004a). 139 Brioverian sediments in this domain constitute a more than 1000 m thick monotonous and 140 141 immature detrital succession of wacke-type sandstones (Dabard et al. 1996). The Ordovician to 142 Lower Carboniferous sediments form a heterogeneous mostly siliciclastic succession with a thickness above 3500 m. Carbonates mostly occur in Devonian sediments whereas black shales 143 144 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 145 intense deformation and by the presence of high grade metamorphic rocks (e.g. Gapais et al. 146 2015). Three tectono-metamorphic units can be distinguished in this domain and comprise from 147 top to bottom: (i) HP-LT rocks, composed of Ordovician acid metavolcanic rocks (Ballèvre et 148 al. 2012) and blueschists subducted and exhumed between ca. 370 and 350 Ma (e.g. Bosse et 149 150 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 151 recorded peak P-T condition of 0.8 GPa, 750-850 °C (Jones and Brown 1990; Augier et al. 152 2015). 153

Magmatism in the Armorican Massif occurred during four main periods. During the 154 Cadomian period from ca. 750 to 580 Ma, the northern domain was affected by acid and mafic 155 arc magmatism followed by voluminous granitoid plutonism at ca. 540 Ma (Ballèvre et al. 2001, 156 2013). In the Early Ordovician times (ca. 490 - 470 Ma), intracontinental rifting was 157 responsible for the emplacement of tholeitic mafic magmas and voluminous acid metavolcanics 158 159 (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 160 Carboniferous period is marked by the emplacement of gabbros and various granitoids 161 162 (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 163 Variscan subduction and initial collision. During the Late Carboniferous (from ca. 320 to 300 164 Ma), dextral wrenching of the central domain and crustal extension in the southern domain, led 165 to the exhumation of migmatitic core complex (Gapais et al. 2015), coeval with the 166 167 emplacement of three main types of granitoids (Fig. 2).

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

Guérande (309.7  $\pm$  1.3 Ma: Ballouard et al. 2015a) MPGs, or along the SASZ such as the 170 Questembert (316.1  $\pm$  2.9 Ma: Tartèse et al. 2011b), Lizio (316.4  $\pm$  5.6 Ma: Tartèse et al. 2011a) 171 and Pontivy (316.7  $\pm$  2.5 Ma: Ballouard et al. 2017b) MPGs. Some of them were also emplaced 172 173 to the north of the SASZ such as the Langonnet (304.7  $\pm$  2.7 Ma: Ballouard et al. 2017b), Huelgoat (314.0  $\pm$  2.8 Ma: Ballouard 2016) and Saint-Renan MPGs (316.0  $\pm$  2.0 Ma: Le Gall 174 et al. 2014). Previous isotopic studies (Bernard-Griffiths et al. 1985; Euzen 1993; Tartèse and 175 Boulvais 2010; Ballouard et al. 2015a, 2017a, 2017b) revealed that these MPGs are 176 characterized by high  $\delta^{18}$ O ratios (>11), sub-chondritic to chondritic  $\epsilon$ Nd(T) and elevated initial 177  ${}^{87}$ Sr/ ${}^{86}$ Sr (> 0.705) values in agreement with a crustal origin. 178

179 (2) CPGs: the Rostrenen (Euzen 1993; Ballouard et al. 2017b) and Huelgoat (Georget 1986) 180 CPGs were emplaced at  $315.5 \pm 2.0$  Ma (Ballouard et al. 2017b) and  $314.8 \pm 2.0$  Ma (Ballouard 181 2016), respectively. These granites were emplaced synchronously with small stocks of mantle-182 derived mafic to intermediate igneous rocks.

(3) KCGs: these metaluminous biotite ± hornblende granitoids consist of a magnesiopotassic (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 slabtearing (Ballouard et al. 2017b).

In the Armorican Massif, U was mostly mined in the district of Mortagne, Pontivy and 193 Guérande. In the Guérande and Pontivy districts, the MPGs were the main source for the U 194 mineralization found in the intra to perigranitic vein and episyenite type hydrothermal deposits. 195 196 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 197 U oxides from the Guérande, Pontivy and Mortagne district deposits reveal several mineralizing 198 199 events that occurred by pulse from ca. 300 to 260 Ma (Cathelineau et al. 1990; Ballouard et al. 200 2017a, in press).

#### 201 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 206 mineralized Guérande and Pontivy MPGs as well as the Questembert, Lizio, Langonnet and 207 Hulgoats MPGs that are not spatially associated with U deposits (Fig. 2). Most sandstone or 208 siltstone samples (Brioverian to Devonian) were collected in the region of Crozon in the western 209 edge of the central Armorican domain (Fig. 2). In addition, a Lower Carboniferous (this study) 210 and Devonian sandstone (Ducassou et al. 2014) were sampled in the Châteaulin basin (western 211 212 part of the central domain, Fig. 2) and the Chalonnes region (eastern part of the central domain, 213 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 214 orthogneissic complex (central domain, Fig. 2), as well as one metatonalite (sample QIMP-1) 215 from Moëlan (southern domain, Fig. 2) were also selected for zircon U-Pb dating. 216

#### 217 Whole rock isotopic, major and trace element analyses

Large samples (5 to 10 kg) were crushed in the Geosciences Rennes Laboratory using agate 218 mortars. Chemical analyses were performed by the Service d'Analyse des Roches et des 219 220 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). 221 222 The results of the analyses are provided in Supplementary file 2. Whole rock Sr and Sm-Nd 223 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 224 Carboniferous sandstone sample from the Châteaulin Basin. The methodology and analytical 225 226 conditions are described in Ballouard et al. (2017b) and the results of the isotope analyses are provided in Supplementary file 3. 227

#### 228 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.

233 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 234 235 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 236 in Ballouard et al. (2015a) and Supplementary file 4. The analyses with a degree of concordance 237 between 90 and 110% are provided in Supplementary file 5 with errors listed at  $1\sigma$ . Histograms 238 (50 Ma bins) and Kernel density estimates (KDE, 25 Ma bands) of the zircon U-Pb dates were 239 realized using the DensityPlotter software (Vermeesch 2012). <sup>206</sup>Pb/<sup>238</sup>U dates were used for 240

241 zircon with a  ${}^{207}$ Pb/ ${}^{206}$ Pb date < 1.0 Ga whereas  ${}^{207}$ Pb/ ${}^{206}$ Pb dates were used for zircon with a 242  ${}^{207}$ Pb/ ${}^{206}$ Pb date > 1.0 Ga.

Hafnium (Hf) isotope analyses were performed at Goethe-University Frankfurt using a 243 244 Thermo-Finnigan NEPTUNE multicollector ICP-MS coupled to a Resolution M-50 (Resonetics) 193 nm ArF Excimer laser (ComPexPro 102F, Coherent), using the procedure 245 outlined in detail in Gerdes and Zeh (2006, 2009) and summarized in Supplementary file 4. The 246 247 epsilon Hf values [EHf(t)] were calculated using the chondritic uniform reservoir (CHUR) as recommended by Bouvier et al. (2008;  $^{176}Lu/^{177}Hf = 0.0336$  and  $^{176}Hf/^{177}Hf = 0.282785$ ) and a 248 decay constant of  $1.867 \times 10^{-11}$  yr<sup>-1</sup> (Scherer et al. 2001; Söderlund et al. 2004). The initial 249  $^{176}$ Hf/ $^{177}$ Hft,  $\epsilon$ Hf(t) values and hafnium model ages (T<sub>DM</sub>) (for zircon grains with a degree of 250 concordance between 90 and 110%) were calculated using <sup>206</sup>Pb/<sup>238</sup>U ages for zircon with 251  $^{206}$ Pb/ $^{207}$ Pb ages < 1.0 Ga and  $^{206}$ Pb/ $^{207}$ Pb ages for zircon with  $^{206}$ Pb/ $^{207}$ Pb ages > 1.0 Ga. The 252 result of analyses and additional information are provided in Supplementary file 6. 253

#### 254 **Results**

#### 255 Geochemical features and U-Th distribution

All investigated Late Carboniferous MPGs of the Armorican Massif are highly peraluminous and characterized by A/CNK  $[Al_2O_3/(CaO + Na_2O + K_2O), molar proportions]$ 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 wholerock 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 265 generally show much higher Th/U ratios (between ~4 and 7), i.e., above the average value of 266 the upper continental crust (UCC ~ 4), compared to most Brioverian sediments (Th/U ratio 267 mostly between ~2 and 4). Most Ordovician (meta)volcanics (Lower Paleozoic) are 268 characterized by U contents between ~1 and 7 ppm and variable Th/U ratios between ~2 and 269 20 whereas Cambro-Ordovician (Lower Paleozoic) and Early Carboniferous (Pertre MPG) 270 granitoids show more variable U contents between ~2 and 40 ppm and are generally 271 272 characterized by relatively low but variable Th/U ratios ranging from ~0.5 to 4.

#### 273 Whole-rock Sr-Nd isotope data

Most MPGs are characterized by elevated initial <sup>87</sup>Sr/<sup>86</sup>Sr [I<sub>Sr</sub> (315 Ma] ratios ranging from 274 0.706 (Pontivy MPG) and 0.719 (Guérande MPG) and negative  $\epsilon$ Nd (315 Ma) values (-2.3 > 275 276  $\epsilon$ Nd (315 Ma) > -8.0) with two stage Nd model ages (T<sub>DM</sub>) between 1.2 (Pontivy and Langonnet MPG) and 1.8 Ga (Guérande MPG, Fig. 5). However, two samples from the Pontivy MPG 277 278 display super-chondritic ɛNd (315 Ma) values (1.2 and 2.2) with relatively low Isr (0.704 and 279 0.706) ratios and young T<sub>DM</sub> (1.0 and 0.9 Ga). As noticed by Bernard-Griffiths et al. (1985), 280 the Isr (315 Ma) and ENd (315 Ma) values of MPG increase and decrease, respectively, from north to south with an evolution from the Pontivy-Langonnet-Lizio MPG, Questembert MPG 281 282 to Guérande MPG (Figs. 2 and 5).

283 U-Pb dating

284 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). UPb 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).

296 Detrital zircon from sediments

U-Pb dates obtained on detrital zircon grains from the main sedimentary formations of theArmorican 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 299 300 appear zoned or homogeneous in CL images (Fig. 6). The detrital grains reflect similar age 301 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 302 sediments compared to the MPGs, especially in Silurian and Devonian sediments (Fig. 8 and 303 Table 1). The most significant difference is that some samples contain abundant zircon grains 304 of Grenvillian age (population 2), e.g., the Silurian and Devonian sediments from Crozon (24-305 32%), whereas they are completely absent or very scares in the Brioverian sandstone from 306 Crozon (1%) and the Lower Carboniferous sediments from Châteaulin (0%). It is important to 307 308 note that zircon age spectra similar to those of Silurian and Devonian sediments were obtained 309 for an Ordovician sandstone from Crozon (Matteini et al. 2014). A minor Grenvillian population is also observed in the Devonian sandstone from Chalonnes (10%; Fig. 8 and Table 310 1). 311

312 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. 313 2) intruded between 504.5  $\pm$  1.8 (PLG-2) and 466.2  $\pm$  3.6 Ma (QIMP-1) – (for details see text 314 and diagrams in Supplementary file 7), and that most of them contain inherited components 315 (Fig. 9a and Table 1). Zircon shapes are euhedral to sub-euhedral, and CL images reveal well-316 developed grow zonation for most grains (Fig. 6). Xenocrystic zircon crystals in the 317 (meta)granitoids overlap in age with the previously defined populations 1 (4%) and 3 (18%, 318 319 Table 1). Zircon analyses with apparent Silurian to Devonian ages (7%) are interpreted to result 320 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 321 322 (meta)volcanics from the southern domain (Vendée Porphyroids, Fig. 2) are reported in Figure 323 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 324 (population 4: 93%) brackets the age of intrusion from  $494 \pm 4$  to  $472 \pm 4$  Ma (Ballèvre et al. 325 2012). These ages are much older than those obtained on zircon grains from the Pertre MPG 326 (Vernhet et al. 2009) in the central domain (Fig. 2) that yield  ${}^{206}$ Pb/ ${}^{238}$ U ages between  $362 \pm 2$ 327 328 and  $330 \pm 4$  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 329 belongs to the population 3 defined in the Late Carboniferous MPGs whereas two zircon grains 330 with apparent ages of  $323 \pm 3$  and  $321 \pm 4$  Ma, respectively, reflect Pb loss (Vernhet et al. 2009). 331

## 332 Hf analyses

The results of Hf isotope analyses on inherited zircon grains from MPGs are reported in a  $\epsilon$ 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 ( $\epsilon$ Hf(t) = 3.9 to -11.5), whereas

Mesoproterozoic-to-Carboniferous xenocrysts (1035-329 Ma) yield mostly superchondritic 337  $\epsilon$ Hf(t) up to +9.1 (> 60%). Superchondritic xenocrysts are predominant in the Pontivy (-14.3 to 338 +8.3), Lizio (-4 to +8.2) and Langonnet (-10.3 to +8.4) MPGs, and mostly characterized by 339 hafnium model ages (T<sub>DM</sub>) between 1.6 and 0.9 Ga. In contrast, the Questembert MPG (-12.1 340 to 7) is dominated by subchondritic grains, even though most of them also show  $T_{DM}$  between 341 1.6 and 0.9 Ga. Then, zircon grains from the Guérande MPG (-14.2 to 9.1) show a wide and 342 343 relatively continuous range of EHf(t) from sub- to superchondritic (Fig. 10a). About 70% of zircon are characterized by T<sub>DM</sub> between 1.6 and 0.9 Ga whereas others show older T<sub>DM</sub> between 344 1.60 and 2.3 Ga. 345

346 The *ɛ*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-347 Mesoproterozoic zircon grains (3080-1430 Ma) show T<sub>DM</sub> from 3.7 to 2.2 Ga and about 75 % 348 of the Neoproterozoic-Carboniferous grains (851-329 Ma) are characterized by T<sub>DM</sub> between 349 and 1.6 and 0.9 Ga. In contrast, detrital zircon grains in the Silurian and Devonian sedimentary 350 351 rocks show significantly different Mesoproterozoic-to-Cambrian EHf(t) spectra characterized by a wide range in  $\varepsilon$ Hf(t) at 1200-900 Ma (-38 to +10.8; T<sub>DM</sub> = 3.8-1.2 Ga), and by about 70 % 352 of subchondritic  $\epsilon$ Hf(t) for ages < 900 Ma (-37 to +8; T<sub>DM</sub> = 3.6–1.1 Ga; Fig. 10b). This 353 354 difference is also reflected by the generally much older Hf model ages for the Neoproterozoicto-Cambrian population, with more than 50 % between 1.6 and 2.5 Ga (Fig. 10b). The Hf 355 isotopic data of the Archean-to-Paleoproterozoic grains (2922-1841 Ma;  $T_{DM} = 3.5-2.5$  Ga) in 356 the Silurian-Devonian sediments overlap with those in the Brioverian-Carboniferous sediments 357 and in the MPGs. 358

The  $\epsilon$ Hf(t) values of magmatic zircon grains from the Cambro-Ordovician metaigneous 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<sub>DM</sub> overlap with those obtained 362 from most Neoproterozoic-to Carboniferous xenocrysts in the MPGs (Figs. 10a and 10d). 363 However, there is also an important number of grains showing significantly higher  $\epsilon$ Hf(t), very 364 close to the depleted mantle. Such grains were mostly found in sample PLG-1 from the 365 Plouguenast orthogneissic complex (Fig. 10c).

366 Discussion

#### 367 Pre- to Early Variscan zircon record

368 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 369 Cadomian-Avalonian belt, an Andean-type cordillera located at the northern margin of 370 Gondwana, e.g. to the north of the Armorican Massif (Fig. 2a), between approximately 800 and 371 540 Ma (e.g. Nance and Murphy 1994; Dabard et al. 1996; Ballèvre et al. 2001; Brun et al. 372 373 2001; Zeh et al. 2001; Linnemann et al. 2008, 2014; Koglin et al. 2018). This hypothesis is 374 supported by the vertical array in the  $\varepsilon$ 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 375 376 the West Africa craton (EHft down to -22), and mantle-derived melts (EHft up to +13). Then, most Archean-to-Mesoproterozoic zircon grains (3080 - 1430 Ma; Fig. 8a) likely originated 377 from the erosion of the West African craton (Fig. 10d), that represented the hinter land of the 378 Cadomian orogeny (e.g. Zeh et al. 2001; Linnemann et al. 2008, 2014), whereas the ca. 2000 379 Ma old Icartian orthogneisses (e.g. Samson and D'Lemos 1998) only represented a minor 380 381 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 386 Paleozoic sediments throughout the EVB, commonly associated with a predominant 387 Neoproterozoic-Cambrian (800-500 Ma), and a subordinate Archean-to-Palaeoproterozoic 388 zircon population (1700-3500 Ma), and with a pronounced age gap between 1.2 and 1.7 Ga 389 (e.g. Zeh et al. 2001; Linnemann et al. 2008, 2014). In fact, these are the same features observed 390 in the Armorican sediments during this study (Fig. 10b). In contrast, zircon grains derived from 391 392 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 393 Mid-German Crystalline Zone (northwestern edge of the Saxo-Thuringian in Fig. 1) of the 394 eastern EVB (Geisler et al. 2005; Zeh and Gerdes 2010). 395

The overlap in zircon Age-Hf isotopic data suggests that the Cambro-Ordovician 396 (meta)granitoids of the Armorican Massif were predominantly formed by partial melting of 397 Brioverian sediments (Fig. 10d). However, the highly superchrondritic  $\varepsilon$ Hf(t) (near to depleted 398 mantle) signature of some zircon grains from the granitoids of the Plouguenast orthogneissic 399 400 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 401 crustal melting) is not restricted to the Armorican Massif (e.g. Bernard-Griffiths et al. 1986; 402 403 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-404 Michou et al. 2017) and Saxothuringian Domain (e.g. Bankwitz et al. 1994; Höhn et al. 2017), 405 where it is related to the breakup of continental microterranes from the northern Gondwanan 406 margin (e.g. Kroner and Romer 2013). The relatively wide variation in EHf(t) of the Cambro-407 408 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 409

410 melts (depleted mantle-related) and crustal-derived melts, formed by the partial melting of411 Brioverian sediments.

412 The Age-Hf-isotopic data furthermore suggest that the Brioverian sediments and the 413 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). 414 Moreover, a significant addition from zircon grains derived from Early Variscan granitoids 415 416 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 417 418 Carboniferous sediments are characterized by T<sub>DM</sub> from 1.2 to 1.6 Ga that overlap with those 419 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 420 Brioverian sediments. This hypothesis is in agreement with the presence of a ca. 625 Ma old 421 zircon xenocryst in the Pertre granite (Fig. 9c, Table 1). 422

#### 423 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  $\varepsilon$ Nd(315 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). 434 The ɛNd(315 Ma) values of the Questembert MPG (-3.9 to -5.7) overlap with those obtained 435 from the Brioverian sediments from the central domain as well as with Cambro-Ordovician acid 436 437 (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-438 Ordovician (meta)granitoids and (meta)volcanic rocks. The super-chondritic ɛNd(315 Ma) 439 composition of two Pontivy MPG samples could be explained by the partial melting of Cambro-440 Ordovician (meta)granitoids. However, the Nd isotopic values on their own are ambiguous, and 441 do not allow a detailed source characterization. Two reasons may be proposed at this stage. 442 443 First, the MPG melts can be derived from several sources and, second, some sources show a wide range in ENd values (Fig. 5), e.g., the Cambro-Ordovician acid (meta)volcanics rocks, 444 which themselves were formed by melting of even older crustal components such as Brioverian 445 446 sediments (Ballèvre et al. 2012).

Combined U-Pb and Hf isotopic data of zircon xenocrysts (2890-329 Ma; Table 1; Figs. 7 447 and 10d) suggest that the Late Carboniferous MPGs were formed by partial melting of a 448 heterogeneous crust mainly consisting of Brioverian sediments, Cambro-Ordovician acid 449 igneous rocks and Devonian to Early Carboniferous granitoids. In contrast, Ordovician to 450 451 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 (1.2-0.9 452 Ma: this study, Matteini et al. 2014) with a wide range in  $\varepsilon$ Hf(t) (-38 to 10.8), which is barely 453 seen in the xenocrysts from the MPGs (Fig. 10d). The only exceptions are four grains (8%) in 454 the Guérande MPG and one grain in the Lizio (3%) and Pontivy (1%) MPGs, respectively 455 456 (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 (< 457 10%, Table 1). Third, detrital zircon grains from Silurian to Devonian sediments with ages < 458

0.9 Ga mostly show subchondritic EHf(t) values (ca. 70%), corresponding to Hf model ages of 459 2.5 to 1.6 Ga, which are much older than those of most MPGs (0.9 to 1.6 Ga). On the other 460 hand, a significant number of Neoproterozoic to Carboniferous (750-329 Ma) zircon with old 461 T<sub>DM</sub> from 1.6 to 2.5 Ga (Fig. 10a) together with some Grenvillian grains (1080-974 Ma; Fig. 462 7f) in the Guerande MPG suggests a minor contribution of Ordovician to Devonian sediments 463 in its source, in agreement with its high sub-chondritic ENd composition (Fig. 5). Moreover, the 464 contribution of Lower Carboniferous sediments in the source of all MPGs is unlikely and 465 difficult to explain structurally. Indeed, the central domain that was mainly deformed under 466 strike-slip deformation regime during the Carboniferous was poorly thickened during the 467 Variscan orogeny (Gumiaux et al. 2004a; Gapais et al. 2015) and the burying, and subsequent 468 partial melting, of Lower Carboniferous sediments probably never occurred in the central 469 domain. Then, it is unlikely that important partial melting of the Lower Carboniferous 470 471 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 472 473 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 474 observed in all the MPGs (population 5, Table 1). Finally, metatexites and diatexites derived 475 from pre-Carboniferous sediments and Ordovician orthogneisses are abundant in the southern 476 Armorican domain (e.g. Augier et al. 2015) (Fig. 2). These rocks reached peak P-T conditions 477 of 0.8 GPa, 750-850 °C followed by near isothermal decompression down to 0.4 GPa, 750 °C 478 and melting occurred, during the Late Carboniferous, from water saturated solidus to biotite 479 breakdown conditions (Augier et al. 2015; Jones and Brown, 1990), in agreement with the role 480 of Brioverian sediments and Paleozoic orthogneisses as a major source for the MPGs. 481

#### 482 Implication for the U-fertility of MPGs

The results of this study show that the U-fertile Pontivy and Questembert MPGs, and the 483 484 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 485 486 of Brioverian sediments, Cambro-Ordovician acid igneous rocks and Devonian-to-Carboniferous granitoids. Similar hafnium model ages furthermore suggest that all these rocks 487 are genetically linked, and result from the successive internal reworking of Brioverian 488 sediments (Fig. 10d). Age-Hf isotopic data also suggest that the U-fertile Guérande MPG, 489 emplaced to the south of the SASZ, result from the partial melting of a similar crust with only 490 a minor contribution from Ordovician to Devonian sediments. 491

Thus, fertility or non-fertility of the Late Carboniferous MPGs from the Armorican Massif 492 is obviously controlled primarily by differences during the reworking process, comprising local 493 variations of source rocks (sediment vs. orthogneiss), degree of melting and fractionation, and 494 the melting history itself (unique or multiple), as well as hydrothermal processes during and 495 after each magmatic cycle. The complex interplay of all these processes has been demonstrated 496 497 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 498 the former traveled a greater vertical distance in the crust, enhancing crystal segregation from 499 500 the melts and U enrichment in the residual liquids (Tartese and Boulvais 2010; Tartèse et al. 2013). 501

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 Ufertile 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 508 509 crust (UCC Th/U value = 3.8, Rudnick and Gao 2003), suggest that most U is hosted in Thbearing, highly refractory, minerals such as monazite (xenotime and zircon), whereas Th/U 510 ratios < 3.8 suggest that a significant amount of U is localized along microcracks or in 511 adsorption on the surfaces of rock forming minerals. In most favorable cases, peraluminous 512 513 igneous rocks with Th/U < 1 and U contents > ~10 ppm can host U oxides (Peiffert et al. 1996, 514 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 515 lithologies characterized by Th/U <  $\sim$ 4 and U content >  $\sim$ 2.7 ppm (average UCC values). Such 516 517 rocks comprise the Brioverian sediments, but also Cambro-Ordovician and Devonian-Early Carboniferous peraluminous igneous rocks (Fig. 4). In contrast, the Ordovician to Devonian 518 sediments and Cambro-Ordovician acid volcanic rocks with Th/U > 4 and U < 2.7 ppm, can be 519 520 considered as less fertile. Our new data also suggest that U-fertility of a crust increases if oneand-the same (peraluminous) crust is affected by multiple melting events, which obviously 521 522 occurred in the Armorican Massif during the internal reworking of already U-rich (low Th/U) 523 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 524 scale shear zones. More specifically, multiple crustal reworking can induce progressive U 525 enrichment of some crustal domains (i.e. middle crust) and in late orogenic context, the melting 526 of these domains can lead to the formation of U-fertile granites. These interpretations are in 527 good agreement with the fact that peraluminous orthogneisses of Cambro-Ordovician, and 528 529 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 530 et al. 2017), the Iberian Massif (e.g. Villaseca et al. 2012; López-Moro et al. 2017), and the 531 Bohemian Massif in Germany and the Czech Republic (Tichomirowa et al. 2012), three 532

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.

#### 538 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  $(\sim 2 < Th/U < \sim 4)$ , Cambrian-Early Carboniferous granitoids ( $\sim 0.5 < Th/U < \sim 4$ ) to Late Carboniferous MPGs ( $\sim 0.1 < Th/U < \sim 2$ ). 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, 546 well known for hosting U deposits with natural fission reactors such as Oklo. 547 Ultimately, the U from the deposits likely originates from the leaching of U-rich 548 minerals present in the basin, such as monazite, thorite and possibly uraninite, 549 themselves coming from the erosion of the surrounding Archean basement (e.g. 550 Gauthier-Lafaye and Weber 2003). In the basement, granitoid plutonism occurred from 551 2.94 to 2.71 Ga with an evolution from TTG gneiss, grey granitoids, pink granitoids to 552 Neoarchean granites and pegmatites (Thiéblemont et al. 2009). 553

(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 (Robbet al. 1999).

Despite the different degree of enrichment, by one order of magnitude, the U (and Th) 559 560 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 561 granitoids) to ~1-4 ppm (pink granites), and in the Namaqualand province, from ~1-10 ppm 562 (Little Namaqualand suite), 3-60 ppm (Early Spektakel suite) to 3-800 ppm (Late Spektakel 563 suite). In the two provinces, fractionation of U over Th, a process that can lead, ultimately, to 564 the crystallization of magmatic uraninite (Cuney, 2014), is only recorded by the youngest 565 566 granites and pegmatites (Neoarchean 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 567 melting events seems to be an important mechanism for the concentration of U, the fractionation 568 of U over Th, and finally for the genesis of U-fertile granites. 569

#### 570 Conclusion

(1) Age-Hf isotopic data of detrital zircon grains in Brioverian to Carboniferous sediments, 571 magmatic and inherited zircon crystals in Cambrian to Early Carboniferous (meta)igneous 572 rocks, and xenocrysts in the Late Carboniferous MPGs indicate that the crust of the 573 Armorican Massif is made up by detritus derived from three major sources: West African 574 craton (3500-1600 Ma;  $T_{DM}$  = 3.8 to 2.2 Ga), Grenville belt/domains (1200-900 Ma;  $T_{DM}$ 575 = 2.7 to 1.2 Ga), and predominately Avalonian-Cadomian belt (800-550 Ma;  $T_{DM}$  = 2.5 to 576 577 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 1.0 578 579 Ga), and 320-300 Ma (Late Carboniferous).

(2) These data also reveal that the Late Carboniferous MPGs (300-320Ma) were mainly formed 580 by partial melting of Brioverian sediments, Cambro-Ordovician igneous rocks and 581 Devonian-Early Carboniferous granitoids, which are all genetically linked with each other, 582 as is indicated by a remarkable overlap in their Hf model ages between 1.6 to 0.8 Ga. In 583 contrast, Ordovician, Silurian and Devonian sediments of the Armorian Massif contain a 584 significant Grenvillian zircon population (~ 30%) with mostly older Hf model ages 585 between 1.6 and 2.5 Ga, which are barely seen in the Late Carboniferous MPGs (Huelgoat, 586 Langonnet, Pontivy, Lizio and Questembert), except for a minor amount in the Guérande 587 MPG (~ 10%). 588

(3) In contrast to Ordovician to Devonian sediments (Th/U > 4), Brioverian sediments and Cambrian to Early Carboniferous granitoids characterized by U > 2.7 ppm and 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 592 593 multiple internal reworking of relatively U-rich Brioverian sediments, which were 594 originally deposited along the northern Gondwanan margin at ca. 550 Ma, and became 595 partially molten during several Paleozoic events, causing a successive increase in Ucontent by crystal fractionation, and intermediate hydrothermal processes in the middle-596 597 upper crust. Similar multiple, internal reworking processes of Neoproterozoic-Cambrian sediments is recorded by rocks from many areas across the EVB hosting economically 598 599 important U deposits, such as Iberia, French Massif Central and Bohemian Massif.

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

#### 603 Acknowledgment

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This work was supported by the 2012-2013 NEED-CNRS (AREVA-CEA) and 2015-CESSUR-INSU (CNRS) grants attributed to M. Poujol. We are very grateful to M.P. Dabard as well M. Jolivet, R. Tartèse and P. Bessin who helped for the sampling. We also want to thanks Y. Lepagnot and D. Vilbert (Geosciences Rennes) for crushing the samples and realizing whole-rock radiogenic isotope analyses (Sm-Nd and Sr), respectively. Comments from C. Villaseca and A.S. André-Mayer significantly improved the quality of the manuscript. Finally, this study is dedicated to the memory of M.P. Dabard who left us too early.

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#### 973 Captions

974 Fig.1: Schematic representation of the west European Variscan belt representing the main terranes 975 (Ballèvre et al. 2009, 2014; Ballèvre 2016) and showing the distribution of U deposits (Cuney and Kyser 976 2008), Variscan granitoids and Early Paleozoic felsic to intermediate igneous rocks (after the 1:1 000 977 000 geological maps of France [Chantraine et al. 2003], Portugal and Spain [Civis Llovera (2015)] and 978 the 1:500 000 geological maps of Czechoslovakia [Fusán et al. 1967] and Czech Republic [Cháb et al. 979 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 980 981 calc-alkaline granitoids. ACG: amphibole-rich calc-alkaline granitoids. NASZ: North Armorican shear 982 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 983 984 (1983).

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986 Fig. 2: (a) principal structural domains from the Armorican Massif. (b) General geological map of the 987 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 988 989 Capdevila (2010) and localizing the U deposits (http://infoterre.brgm.fr). The high heat production and 990 flow belt (HHPFB) from Vigneresse et al. (1989) and Jolivet et al. (1989) is indicated. Locality numbers 991 are referenced in Table 1. NASZ: north Armorican shear zone; NBSASZ: northern branch of the South 992 Armorican shear zone; SBSASZ: southern branch of the south Armorican shear zone. Fe-K granites: 993 ferro-potassic granites. Mg-K granites: magneso-potassic granites. Mineral abbreviations according to 994 Kretz (1983).

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Fig. 3: A/NK  $[Al_2O_3/(Na_2O + K_2O)]$  versus A/CNK  $[Al_2O_3/(CaO + Na_2O + K_2O)]$  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.

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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), Béchennec et al. (1996, 1999, 2001), Trautmann and Carn (1997), Dabard
and Peucat (2001), Le Hébel (2002), Tartèse and Boulvais (2010), Béchennec 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.

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1008 Fig. 5: Initial Sr and Nd isotopic composition calculated at 315 Ma of Late Carboniferous MPGs from 1009 the Armorican Massif (Data from this study; Ballouard et al. 2015a, 2017b; Tartèse and Boulvais 2010). 1010 The yellow double arrow represents the ENd(315 Ma) composition of the Langonnet MPG (Ballouard 1011 et al. 2017b) whereas other vertical bars represent the cNd(315 Ma) composition of Cambro-Ordovician 1012 peraluminous igneous rocks [acid (meta)volcanics: Ballèvre et al. 2012; (meta)granitoids: Ballouard et 1013 al. 2017b] as well as Brioverian (Dabard et al. 1996; Dabard 1997) and Paleozoic (Michard et al. 1985; 1014 Dabard and Peucat 2001; this study) sediments. The composition of ca. 2000 Ma Icartian orthogneisses 1015 from the Channel Islands (black arrow) is out of the diagram with  $\varepsilon Nd(315 \text{ Ma})$  values between -18 and 1016 -14 (Samson and D'Lemos 1998). The grey dashed arrow represents the north-south evolution of the 1017 MPG isotopic composition. Sm-Nd isotopic composition of CHUR from Bouvier et al. (2008).

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1019 Fig. 6: Representative cathodoluminescence pictures of zircon grains from MPGs, sediments and 1020 Cambro-Ordovician metagranitoids of the Armorican Massif analyzed during this study. Dashed circles 1021 represent the location of LA-ICP-MS U-Pb analyses. The corresponding U-Pb dates ( $1\sigma$  uncertainty) 1022 and  $\epsilon$ Hf(t) values are indicated. All zircon grains are shown at the same scale and the scale bar is 1023 indicated in the upper right corner. Carbon. = Carboniferous. 1024

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

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

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

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1039 Fig. 10: EHf(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 from Late Carboniferous MPGs and their potential sources. In (d), the composition of magmatic zircon 1041 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 = Grenvillian belt, CA = Cadomian-Avalonian belt, R = Cambro-Ordovician Rift, EV = Early Variscan. 1044 The crustal evolution trends are calculated using a <sup>176</sup>Lu/<sup>177</sup>Hf ratio of 0.0113 (Taylor and McLennan 1045 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

- Fig.11: U versus Th diagram reporting the whole-rock composition of granitoids from (a) the Archean Gabonese craton and (b) Mesoproterozoic western Namaqualand province, South Africa. Geochronological and Geochemical data for Gabon are from Bouton et al. (2009) and Thiéblemont et al. (2009) whereas data for Namaqualand are from Robb (1986, 1999), Raith (1995), Thomas et al. (1996), Clifford et al. (2004), Andreoli et al. (2006), Bailie et al. (2007) and Duchesne et al. (2007). The average U content (2.7 ppm) and Th/U value (~4) of the upper continental crust (UCC; Rudnick and Gao 2003) are indicated.
- 1056
- 1057 Table 1: summary of zircon U-Pb data. Pop. = population.

























Reference	Sample	Locality / intrusion name	Reference N° in Figure 2	Rock type	Intrusion / deposition age	Number U-Pb / Hf	Pop. 1 (%) 3500 – 1400 Ma	Pop. 2 (%) 1200 – 900 Ma	Pop. 3 (%) 900 - 540 Ma	Pop. 4 (%) 540 - 440 Ma	Pop. 5 (%) 440 - 330 Ma
This study	LOC-1	Châteaulin basin	12	Sandstone	Lower Carboniferous	107 / 45	22	0	23	22	32
This study	CRO-2 CRO-1 CRO-12 CRO-11	Crozon	10	Sandstone Sandstone-siltstone Sandstone	Upper Devonian Middle Devonian Lower Devonian	300 / 27	23	24	46	6	0
Ducassou et al. (2014)	DevSA2	Chalonnes	11	Sandstone	Devonian	45 / 0	24	11	42	7	13
This study	CRO-6 CRO-3	Crozon	10	Sandstone Sandstone-siltstone	Silurian	213 / 54	36	32	29	3	0
This study	CRO-9	Crozon	10	Sandstone	Brioverian	105 / 48	17	1	78	4	0
This study	PLG-1 PLG-2 PLG-3 PLG-4 QIMP-1	Plouguenast Moëlan	9	Metagranitoid (MPG) Metagranitoid (tonalite) MPG Metagranitoid (tonalite) Metagranitoid (tonalite)	Ordovician (477.9 ± 2.9 Ma) Cambrian (504.5 ± 1.8 Ma) Ordovician Ordovician (486.7 ± 8.8 Ma) Ordovician (466.2 ± 3.6 Ma)	216/37	4	0	18	71	7
Ballèvre et al. (2012)	FL-31 CG13a CG5 CG09 Fl2a	Bréthomé Chie-Loup Mareuil / Lay La Sauzaie Sauzon		Felsic (meta)volcanics	Ordovician $(472 \pm 4 \text{ Ma})$ Ordovician $(486 \pm 4 \text{ Ma})$ Ordovician $(478 \pm 2 \text{ Ma})$ Ordovician $(471 \pm 12 \text{ Ma})$ Ordovician $(494 \pm 4 \text{ Ma})$	71/0	1	0	6	93	0
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) This study	GUE-3 GUE-4 GUE-5	Guérande	6	MPG	Upper Carboniferous (309.4 ± 1.9 Ma) Upper Carboniferous (309.7 ± 1.3 Ma) Upper Carboniferous (302.5 ± 1.6 Ma)	52 / 32	4	8	50	13	25
Ballouard et al. (2017b) This study Ballouard et al. (2017b)	PONT-1 PONT-10 PONT-14 PONT-15 PONT-26	Pontivy	5	MPG	Upper Carboniferous (316.7 ± 2.5 Ma) Upper Carboniferous Upper Carboniferous (310.3 ± 4.7 Ma)	83 / 24	8	1	39	20	31
Ballouard et al. (2017b)	PONT-20	Langonnet	4	MPG	Upper Carboniferous $(304.7 \pm 2.7 \text{ Ma})$	28/18	4	0	39	32	25
This study	QRT-08	Questembert	3	MPG	Upper Carboniferous $(316.1 \pm 2.9 \text{ Ma})$	19 / 15	5	0	21	47	26
This study, Tartèse et al. (2011a) Tartèse et al. (2011a)	LRT-10 LRT-15	Lizio	2	MPG	Upper Carboniferous (316.4 ± 5.6 Ma) Upper Carboniferous (319 ± 15 Ma)	29 / 18	0	3	59	28	10
This study	HUEL-3	Huelgoat	1	MPG	Upper Carboniferous $(314.0 \pm 2.8 \text{ Ma})$	18 / 0	6	0	50	17	28

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