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Uranium metallogenesis of the peraluminous leucogranite from the Pontivy-Rostrenen magmatic complex (French Armorican Variscan belt): the result of long-term oxidized hydrothermal alteration during strike-slip deformation

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Christophe Ballouard, Marc Poujol, Julien Mercadier, Etienne Deloule, Philippe Boulvais, et al.. Uranium metallogenesis of the peraluminous leucogranite from the Pontivy-Rostrenen magmatic complex (French Armorican Variscan belt): the result of long-term oxidized hydrothermal alteration during strike-slip deformation. *Mineralium Deposita*, 2018, 53 (5), pp.601-628. 10.1007/s00126-017-0761-5 . insu-01581165

HAL Id: insu-01581165

<https://insu.hal.science/insu-01581165>

Submitted on 27 Jun 2024

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1 **Uranium metallogenesis of the peraluminous leucogranite from the Pontivy-**
2 **Rostrenen magmatic complex (French Armorican Variscan belt): the result**
3 **of long term oxidized hydrothermal alteration during strike-slip**
4 **deformation.**

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15 **Keywords:** Uranium deposits, syntectonic granites, apatite geochemistry and U-
16 Pb dating, fluid-rock interactions, Hercynian, south Armorican shear zone

17 **Abstract**

18 In the French Armorican Variscan belt, most of the economically significant hydrothermal
19 U deposits are spatially associated with peraluminous leucogranites emplaced along the south
20 Armorican shear zone (SASZ), a dextral lithospheric scale wrench fault that recorded ductile
21 deformation from ca. 315 to 300 Ma. In the Pontivy-Rostrenen complex, a composite intrusion,
22 the U mineralization is spatially associated with brittle structures related to deformation along
23 the SASZ. In contrast to monzogranite and quartz monzodiorite ($3 < U < 9$ ppm; $Th/U > 3$), the

24 leucogranite samples are characterized by highly variable U contents (~3 to 27 ppm) and Th/U
25 ratios (~0.1 to 5) suggesting that the crystallization of magmatic uranium oxide in the more
26 evolved facies was followed by uranium oxide leaching during hydrothermal alteration and/or
27 surface weathering. U-Pb dating of uranium oxides from the deposits reveals that they mostly
28 formed between ca. 300 and 270 Ma. In monzogranite and quartz monzodiorite, apatite grains
29 display magmatic textures and provide U-Pb ages of ca. 315 Ma reflecting the time of
30 emplacement of the intrusions. In contrast, apatite grains from the leucogranite display textural,
31 geochemical and geochronological evidences for interaction with U-rich oxidized hydrothermal
32 fluids contemporaneously with U mineralizing events. From 300 to 270 Ma, infiltration of
33 surface-derived oxidized fluids leached magmatic uranium oxide from fertile leucogranite and
34 formed U deposits. This phenomenon was sustained by brittle deformation and by the
35 persistence of thermal anomalies associated with U-rich granitic bodies.

36 **Introduction**

37 Continental scale wrench faults represent common tectonic features in orogenic belts, which
38 act as channels for crustal and mantle derived magmas (Strong and Hanmer 1981; D’Lemos et
39 al. 1992; Hutton and Reavy 1992; De Saint Blanquat et al. 1998; Brown 2010; Pirajno, 2010)
40 as well as hydrothermal fluids (e.g. Sibson 1987, 1990; Faulkner et al. 2010; López-Moro et al.
41 2013; Cao and Neubauer 2016). Major strike-slip faults initiate deep within the crust and the
42 lithospheric mantle due to rheological weakening (Cao and Neubauer 2016). During the
43 exhumation of these tectonic systems, a thermal evolution occurs: brittle deformation
44 (cataclasites, pseudotachylytes) is superimposed on ductile deformation (mylonites) and the
45 ascent of magmas as well as hot lower crustal, and magmatic derived fluids is followed by the
46 downward flow of cold surface-derived water. Therefore, these strike-slip deformation zones
47 can represent an important metallotect for hydrothermal uranium (U) deposits if they affect U
48 fertile lithologies. Among U-rich igneous rocks, felsic volcanic rocks and peraluminous

49 leucogranite represent an ideal source for the formation of hydrothermal U deposits because
50 most of their U is hosted in easily leachable glass and uranium oxide, respectively (Cuney
51 2014). The relationships between U-rich felsic volcanic rocks, strike-slip faults, and
52 hydrothermal uranium deposits are for example well illustrated in South China, along the
53 southern termination of the Tan Lu fault (Li et al. 2001, 2002). Similarly, the association
54 between peraluminous leucogranite, wrench faults and U mineralization exists, for example, in
55 Egypt along the El Sela shear zone (Gaafar et al. 2014; Gaafar 2015) and in the European
56 Variscan belt (EVB): the Alentejo-Plasencia strike-slip fault in Iberia (Pérez Del Villar and
57 Moro 1991) and the north-western part of the Massif Central (France; Cathelineau et al. 1990;
58 Cuney et al. 1990; Gèbelin et al. 2009) (Fig. 1).

59 The Armorican Massif (France) in the EVB (Figs. 1 and 2) represents a historical mining
60 province for U where about 20 000 t U (~20 % of the French production; IRSN 2004) have
61 been extracted before the end of the 90's. Few minor deposits are associated with Late
62 Carboniferous metaluminous granitoids emplaced along the north Armorican shear zone
63 (NASZ; Chauris 1984), a crustal-scale, dextral, strike slip fault with a limited displacement of
64 ~20 km (Fig. 2) (Jégouzo 1980). The majority of the U deposits are spatially associated with
65 Late Carboniferous peraluminous syntectonic leucogranite emplaced either along extensional
66 deformation zones (Guérande leucogranite; Cathelineau 1981; Ballouard et al. 2017a) or along
67 the south Armorican shear zone (SASZ: Mortagne and Pontivy leucogranites; Cathelineau
68 1982; Cathelineau et al. 1990; Cuney et al. 1990), a lithospheric-scale, dextral wrench fault with
69 a displacement of ~200 km (Fig. 2) (Berthé et al. 1979; Gapais and Le Corre 1980; Jégouzo
70 1980, Jégouzo and Rosselo 1988; Judenherc et al. 2003; Gumiaux et al. 2004a, 2004b; Tartèse
71 et al. 2012). Recent studies on mylonite, leucogranite and quartz veins along the SASZ
72 demonstrated that, during Late Variscan times, this fault acted as a major channel for lower
73 crustal but also meteoric-derived oxidized hydrothermal fluids (Tartèse and Boulvais 2010;

74 Tartèse et al. 2012; Lemarchand et al. 2012). These fluids were able to transport an important
75 quantity of uranium in solution (Dubessy et al. 1987).

76 The Pontivy-Rostrenen syntectonic composite intrusion hosts U intragranitic deposits
77 associated with peraluminous leucogranite (Figs. 2 and 3). U was interpreted to originate from
78 leaching of uranium oxides from the surrounding leucogranite (Marcoux 1982), although the
79 metallogenic model remains poorly constrained. In this study, we use geochemical analyses and
80 U-Pb dating of apatite from the granitoids as well as U-Pb dating of uranium oxides from the
81 deposits to determine the timing and conditions of hydrothermal U mobilization and then its
82 precipitation in the deposits. We then discuss this model in the geodynamic and metallogenic
83 frameworks at the scale of the region and the EVB.

84 **Geological framework**

85 *The Armorican Massif as part of the European Variscan belt*

86 The Armorican Massif belongs to the EVB, a Paleozoic orogenic belt which extends
87 throughout the western (Iberian Massif) and central Europe (Bohemian Massif) and results from
88 the collision of supercontinents Laurussia and Gondwana (Ballèvre et al. 2009, 2013, 2014;
89 Kroner and Romer 2013) (Fig.1). The structure of the EVB is a consequence of a complex
90 interaction between low-strain and high-strain domains (Kroner and Romer 2013). Low strain
91 domains, such as Ibero-Armorica, represent blocks of thick continental crust drifted from the
92 northern Gondwana margin during Cambrian-Ordovician extension whereas high strain
93 domains, such as the Massif Central and southern part of the Armorican Massif, that were
94 highly deformed during Variscan orogeny, represent domains of Gondwana crust thinned
95 during Lower Paleozoic extension (Kroner and Romer, 2013). Continental subduction events
96 in the Variscides occurred until ca. 350-340 Ma and transition from a compressive to an
97 extensive regime occurred ca. 315-300 Ma ago (Kroner and Romer, 2013; Ballèvre et al. 2014).

98 The Armorican Massif is separated into three main domains by the NASZ and the SASZ
99 (Fig. 2). The northern domain is mostly made of a Proterozoic basement (e.g. Brun et al. 2001),
100 locally intruded by Variscan granitoids. The central domain is composed of Late Proterozoic
101 (Brioverian) to Lower Carboniferous sedimentary rocks mostly deformed under greenschist
102 facies conditions during dextral wrenching along the NASZ and SASZ in Carboniferous times
103 (Gumiaux et al. 2004a). The deformation in this area is marked by a vertical foliation associated
104 with a sub-horizontal stretching lineation (Jégouzo 1980). The southern domain, which belongs
105 to the internal part of the Variscan belt, is characterized by a higher degree of deformation and
106 by the presence of high grade metamorphic rocks (Gapais et al. 2015). Three tectono-
107 metamorphic units can be distinguished in this domain and include, from top to bottom, HP-LT
108 rocks, composed of blueschists and metavolcanic rocks subducted and exhumed during early
109 tectonic events from 370 to 350 Ma (Bosse et al. 2005), micaschists, and migmatite bearing
110 units (Fig. 2). Between 315 and 300 Ma (Tartèse et al. 2012), the SASZ acted as a transfer zone
111 between the southern domain, where crustal extension lead to the exhumation of core
112 complexes cored by migmatites and synkinematic leucogranites, and the central domain
113 deformed by pervasive dextral wrenching (Gapais et al. 2015).

114 During the Late Carboniferous, the Armorican Massif was intruded by various granitoids
115 ranging from peraluminous to metaluminous in composition (Capdevilla 2010; Fig. 2). To the
116 south, two mica peraluminous leucogranites are characteristic. They were emplaced either
117 along extensional deformation zone in the southern domain, such as the Quiberon (Gapais et al.
118 1993, 2015), Sarzeau (Turrillot et al. 2009) and Guérande (309.7 ± 1.3 Ma: leucogranites, or
119 along, the SASZ such as the Lizio (316.4 ± 5.6 Ma: Tartèse et al. 2011a), Questembert (316.1
120 ± 2.9 Ma : Tartèse et al. 2011b) and Pontivy (316.7 ± 2.5 Ma: Ballouard et al. 2017b)
121 leucogranites. To the north, a bimodal magmatism occurred as expressed by the emplacement
122 of Bt \pm cordierite (Cd) peraluminous granites, associated with intermediate rocks of mantle

123 origin, such as the Rostrenen (315.5 ± 2.0 Ma: Ballouard et al. 2017b) and Huelgoat intrusions
124 (314.8 ± 2.0 Ma: Ballouard 2016) and two suites of biotite \pm hornblende metaluminous
125 granitoids. These granitoids consist of a magneso-potassic and a ferro-potassic suite emplaced
126 between 320 and 300 Ma and associated with mantle-derived mafic to intermediate rocks
127 (Ballouard et al. 2017b). On a regional scale, the crustal magmatism to the south of the SASZ
128 was triggered by late-orogenic crustal extension. In contrast, to the north, partial melting of the
129 crust and mantle, enriched during earlier subduction events, were triggered by an asthenosphere
130 upwelling during strike-slip deformation and subsequent slab-tearing (Ballouard et al. 2017b).

131 The EVB represents an important metallogenic province for U where vein, episyenite-type,
132 breccia-hosted or shear zone-hosted U deposits occur all over the belt. Major occurrences are
133 located in the Iberian Massif, the Armorican Massif, the Massif Central and the Bohemian
134 Massif (Fig. 1). A large proportion of these hydrothermal U deposits are spatially associated
135 with Late Carboniferous low Ca peraluminous two mica (or biotite) granite (Fig. 1) and main
136 U mineralizing events in the EVB occurred during the Permian from 300 to 260 Ma (Cuney et
137 al. 1990; Cathelineau et al 1990; Tischendorf and Förster 1994; Cuney and Kyser, 2008; Romer
138 et al. 2010; Ballouard et al. 2017a).

139 In the Armorican Massif, U has been mostly mined in the Guérande, Pontivy and Mortagne
140 districts (Fig. 2). In the Guérande district, the most important vein deposit (Pen Ar Ran) is
141 perigranitic and localized above the apical zone of the Guérande leucogranite (Cathelineau
142 1981). The Guérande leucogranite itself was the main source for U (Ballouard et al. 2017a).
143 Trace elements and oxygen isotope compositions suggest that hydrothermal leaching of the
144 magmatic uranium oxides in deformed facies of the apical zone was promoted by the infiltration
145 of surface-derived oxidized fluids. The leached U was then precipitated in the reducing
146 environment represented by black shales and graphitic quartzites (Ballouard et al. 2017a). The
147 age of the U mineralizing events in the Guérande area (ca. 300 to 275 Ma) is comparable with

148 that in the Mortagne district, and with other U deposits from the EVB (Cathelineau et al. 1990;
149 Ballouard et al. 2017a). The Questembert leucogranite (Fig. 2) is not associated with U
150 mineralization but the petro-geochemical and geochronological study of Tartèse et al. (2013)
151 suggests that this intrusion liberated an important amount of uranium during a sub-solidus
152 alteration event at depth, by surface-derived oxidized fluids.

153 *The Pontivy-Rostrenen magmatic complex.*

154 *General framework*

155 Gravimetric data reveals that the Pontivy-Rostrenen complex represents a continuous
156 intrusion with the main root (~6 km depth) localized to the north (Vignerresse and Brun 1983;
157 Vignerresse 1999). The southern part of the complex is composed almost exclusively of
158 peraluminous leucogranite, whereas peraluminous leucogranite and monzogranite outcrop to
159 the north with small stocks of mantle-derived metaluminous quartz monzodiorite (Fig. 3)
160 (Euzen 1993; Ballouard et al. 2017b). To the south, leucogranite intrudes Late-Proterozoic
161 (Brioverian) sedimentary rocks, whereas to the north leucogranit, monzogranite and quartz-
162 monzodiorite intrude Late-Proterozoic and Paleozoic (Ordovician to Lower Carboniferous)
163 sedimentary formations causing contact metamorphism (Fig. 3). Vignerresse (1999) estimated
164 that these intrusions were emplaced at a depth around 6 – 8 km. The shape of the Pontivy
165 leucogranite intrusion to the south marks the dextral shearing of the SASZ (Figs. 2 and 3) and,
166 in the southern edge, syn-cooling shearing is revealed by the development of C/S structures
167 (Gapais 1989) and mylonites in 100 m wide dextral shear zones, oriented N 100-110° (Jégouzo
168 1980). The oxygen isotope study carried out on mylonites from the Guilligomarch quarry (Fig.
169 3) in the southern edge of the complex showed that some of these rocks experienced
170 hydrothermal alteration by low $\delta^{18}\text{O}$ meteoric-derived fluids (Tartèse et al. 2012).

171 *Petrogeochemical characteristics*

172 The petro-geochemistry and geochronology is summarized from Ballouard et al. (2017b).
173 Leucogranite contains quartz-feldspar-muscovite with a variable amount of biotite that can be
174 either absent or dominant over muscovite. Tourmaline is rarely observed. Common accessory
175 minerals are zircon, apatite, monazite and Fe-Ti oxides. Uranium oxides were not observed in
176 our samples. This absence is likely the consequence of the instability of this mineral during
177 post-crystallization alteration and/or weathering, because uranium oxides were commonly
178 observed in the drill cores from leucogranite associated with U deposits, such as Guérande
179 (Oudou 1984) (Fig. 1), or in the northwestern part of the Massif Central (Friedrich et al. 1987).
180 The leucogranite was divided into three main sub-facies (Fig. 3):

181 (1) The porphyritic leucogranite is characterized by the abundance of K-feldspar
182 megacrysts and a higher amount of biotite over muscovite.

183 (2) The equigranular leucogranite represents the most common type of leucogranite in the
184 complex, and is characterized by a low abundance (or the absence) of K-feldspar
185 megacrysts. The proportion of biotite over muscovite is variable, biotite being even
186 absent in some cases.

187 (3) The late equigranular leucogranite (Langonnet leucogranite) forms an elliptic stock
188 which crosscuts the other facies. This leucogranite is rarely porphyritic and generally
189 contains a low proportion of biotite.

190 In terms of alteration, chloritization of biotite is common and secondary muscovitization
191 affects more particularly the muscovite > biotite equigranular facies. Several veins of pegmatite
192 and aplite crosscut the leucogranite. Moreover, pegmatite stocksheider were described along
193 the western edge of the late equigranular leucogranite and greisenization locally affects the most
194 evolved parts of the equigranular and late equigranular leucogranites (Euzen 1993; Bos et al.
195 1997).

196 The monzogranite (Rostrenen granite) crops out in the northern part of the complex (Fig.
197 3). This facies contains a quartz-feldspar-biotite assemblage with a small amount of muscovite
198 and, locally, cordierite. The most common accessory minerals include zircon, monazite and Fe-
199 Ti oxides. Mafic enclaves with a composition similar to quartz-monzodiorite are commonly
200 observed in this facies (Euzen 1993). The quartz-monzodiorite facies mostly appears as small
201 stocks of a few km² in the eastern part of the monzogranitic intrusion (Fig. 3). This facies
202 generally contains quartz-feldspar-biotite-amphibole ± clinopyroxene as well as apatite,
203 titanite, zircon and Fe-Ti oxide as accessory minerals. Ocellar quartz is frequently observed in
204 this facies and interpreted as the result of a mixing with a felsic magma. Mingling features are
205 visible at the contact between the quartz-monzodiorite and the monzogranite.

206 U-Pb dating of magmatic zircon grains revealed that the three magmatic facies of the
207 complex were emplaced synchronously between 316.7 ± 2.5 Ma and 310.3 ± 4.7 Ma, whereas
208 the late equigranular leucogranite was emplaced later at 304.7 ± 2.7 Ma. The peraluminous
209 leucogranite ($A/CNK > 1.1$) has mostly sub-chondritic $\epsilon Nd(t)$ values (-4.79 to 2.08) and contain
210 numerous inherited zircon grains with Archean to Paleozoic ages. It formed by partial melting
211 of Neoproterozoic metasedimentary rocks and Paleozoic peraluminous orthogneisses. The
212 moderately peraluminous monzogranite ($1.0 < A/CNK < 1.3$) has a sub-chondritic $\epsilon Nd(t)$
213 composition (-3.95 to -3.22), and contains magmatic zircon with sub-chondritic $\epsilon Hf(t)$ values
214 and scarce Paleozoic inherited zircon. It formed by partial melting of an orthogneiss with a
215 probable metaluminous composition. The metaluminous quartz monzodiorite ($0.7 < A/CNK <$
216 1.1) has sub-chondritic $\epsilon Nd(t)$ values (-3.19 to -2.17) and contains magmatic zircon grains with
217 sub- to slightly superchondritic $\epsilon Hf(t)$ values. It formed by partial melting of a metasomatized
218 lithospheric mantle.

219 The evolution from high (~70 wt.%) to very high (~75 wt.%) SiO₂ leucogranite is likely
220 explained by the fractional crystallization of a cumulate composed of Bt + Kfs + Pl as well as

221 accessory minerals such as Ap + Zrn + Mnz. The chemical evolution of monzogranite from
222 high (~71 wt.%) to low (~65 wt.%) SiO₂ may reflect entrainment of peritectic minerals from
223 the source (i.e. Cpx + Grt + Pl + Ilm) and/or a mixing with a mantle derived melt. The evolution
224 of the quartz monzodiorite from ~ 54 wt.% to ~60 wt.% SiO₂ is likely the consequence of
225 fractionation of a cumulate made of Pl + Bt + Cpx and mixing with a felsic magma with a
226 probable monzogranitic composition.

227 *U mineralization*

228 Most of the U deposits in the Pontivy-Rostrenen complex (~2000 t of U extracted; IRSN
229 2004) are spatially associated with the equigranular leucogranite facies (Fig. 3). They are
230 generally localized close to contact with the sedimentary country rock, or to micaschist enclaves
231 (Marcoux 1982; Alabosi 1984; Cuney 2006). The most important U deposits occur as
232 polyphase, commonly hematized, quartz veins mostly oriented N170° and interpreted as tension
233 gashes accommodating dextral wrenching along the SASZ (Marcoux 1982; Alabosi 1984) such
234 as the Bonote (~400 t U extracted) or the Kerlech-Lignol deposits (~1000 t U extracted; Fig.
235 3). A second type of U deposit, with a main orientation of N120-130°, occurs generally as
236 brecciated quartz veins, such as in the Guern area where ~40 t U were extracted (e.g. Quistiave
237 and Kerroch deposits), in relation with second order faults which also likely developed due to
238 deformation along the SASZ (Marcoux 1982; Alabosi 1984). The third type corresponds to
239 episyenite-hosted deposits such as in the Prat Mérrien and Poulprio area (~ 100 t U extracted;
240 Fig. 3), where the mineralized bodies follow N130-160° oriented faults (Alabosi 1984). The
241 episyenitization of leucogranite during hydrothermal alteration resulted in the dissolution of
242 magmatic quartz, the destabilization of plagioclase, the development of secondary muscovite
243 and the geodic crystallization of adularia, quartz, montmorillonite and carbonate, U being
244 disseminated in clay or in major minerals (Alabosi 1984). A fourth type of deposits occurs as
245 fracture fillings within Brioverian micaschist xenoliths (e.g., Kerségalec; Cuney 2006) (Fig. 3).

246 **Analytical techniques**

247 *Whole-rock major and trace elements analyses*

248 Three samples of episyenites collected by Alabosi (1984) in the Prat Mérien and Poulprio
249 quarries (Fig. 3) were crushed in the Géosciences Rennes Laboratory using agate mortars.
250 Chemical analyses were performed by the Service d'Analyse des Roches et des Minéraux
251 (SARM; CRPG-CNRS, Nancy, France) using an ICP-AES for major elements and an ICP-MS
252 for trace elements following the techniques described in Carignan et al. (2001). The results of
253 the whole-rock analyses are provided in Table 1.

254 *Apatite chemistry and U-Pb dating*

255 Apatite crystals from the different magmatic facies forming the complex as well as an
256 episyenite sample were separated using classical magnetic and heavy liquid methods in the
257 Géosciences Rennes Laboratory. Apatite grains were handpicked under a binocular microscope
258 before being embedded in epoxy resin and polished. Apatite grains were imaged by
259 cathodoluminescence (CL) using a Reliotron CL system in Géosciences Rennes (5 kV
260 acceleration voltage and 600 μ A beam current) and a Cambridge Image Technology (CITL)
261 Mark 5 CL system at University of Mons, Belgium (15 kV acceleration voltage and 500 μ A
262 beam current). The CL system at UMons is also equipped with a CITL COS8200 fiber-based
263 optical spectrometer with a 350-1100 nm spectral range and 4 nm resolution. Spectral
264 calibration was performed with a standard Hg-Ar lamp. The spectra were not corrected for total
265 system response. Backscattered electron (BSE) images and chemical maps were performed
266 using the Cameca SX-100 electron probe micro analyze (EPMA) at IFREMER (Plouzané,
267 France). Analyses were performed using a 15 keV accelerating voltage and a beam diameter of
268 15 μ m. Beam currents of 10 nA and 20 nA were used for spot analyses and elemental mapping,
269 respectively. Standards were: apatite (F $K\alpha$, TAP crystal, counting time of 30s; P $K\alpha$, LPET,

270 60s; Ca K α , PET, 30s), albite (Si K α , TAP, 30s; Na K α , TAP, 30s), strontianite (Sr L α , TAP,
271 30s), pyromorphite (Cl K α , LPET, 60s), Si-Al-Ca glass with 4w.% La (La L α , LPET, 30s),
272 barium sulfate (S K α , PET, 30s), Si-Al-Ca glass with 4w.% Ce (Ce L α , PET, 60s), andradite
273 (Fe K α , LLIF, 60s), rhodonite (Mn K α , LLIF, 60s), gallium arsenide (As L α , TAP, 60s).

274 U-Pb geochronology of apatite was conducted by in-situ laser ablation inductively coupled
275 plasma mass spectrometry (LA-ICP-MS) at Géosciences Rennes using a ESI NWR193UC
276 excimer laser coupled to a quadripole Agilent 7700x ICP-MS equipped with a dual pumping
277 system to enhance sensitivity. The methodology used to perform the analyses can be found in
278 Pochon et al. (2016) and in Electronic Material ESM1. Ages, calculated using the ISOPLOT
279 software (Ludwig 2001), are provided with their 2 σ uncertainties. All the isotopic ratios as well
280 as the corresponding U and Pb contents in ppm are provided in ESM 2.

281 *Uranium oxide U-Pb dating*

282 Petrography and imaging of selected polished thin sections and mounts of uranium oxide
283 samples were carried out at the GeoRessources Laboratory (Nancy, France) and the Centre de
284 Recherches Pétrographiques et Géochimiques (CRPG, Nancy, France). U-Pb dating was carried
285 out at the CRPG by secondary ion mass spectrometry (SIMS). The U oxide samples were first
286 examined using reflected light microscopy. We then selected chemically homogeneous area
287 with high radiogenic lead content suitable for SIMS analyses based on BSE images obtained
288 using a JEOL J7600F, a HITACHI S-4800 (GeoRessources) or a JEOL 6510 (CRPG) scanning
289 electron microscope and major element analyses obtained using a CAMECA SX100 EPMA
290 (GeoRessources). U-Pb isotope analyses were performed using a CAMECA IMS 1270 SIMS.
291 The complete methodology is described in ESM 3. Due to the common Pb-rich character of the
292 U oxides ($50 < {}^{206}\text{Pb}/{}^{204}\text{Pb} < 11000$), a common lead correction based on the measured ${}^{204}\text{Pb}$
293 content and the Pb isotopic composition calculated using the model of Stacey and Kramers
294 (1975) at the estimated age of the U oxide was applied to the analyses. All the isotopic ratios

295 are provided in ESM 4 and ages, calculated using the ISOPLOT software (Ludwig 2001), are
296 provided with their 2σ uncertainties.

297 **Results**

298 *Uranium oxide petrography*

299 Uranium oxide U-Pb dating was performed on 6 mounts or thin sections belonging to the
300 AREVA collection from the Kerlech (Lignol), Rosglas and Qu errien (Kerjean) deposits as well
301 as three deposits in the region of Guern (Fig. 3: Quistiave, Kerroch and a sample referenced as
302 “undifferentiated-Guern” in the AREVA collection).

303 In the Guern region, the mineralization is described as brecciated quartz veins, following
304 N120 – 130° oriented faults, which mostly occur in tectonized contacts between the porphyritic
305 and equigranular leucogranites close to micashists enclaves and/or small stocks of quartz
306 monzodiorites (Marcoux 1982; Cuney 2006) (Fig. 3). The Quistiave deposit consists of two
307 veins orientated WNW-ESE and dipping SW, occurring about 80 m apart. The veins are more
308 or less parallel to alternating bands of a porphyritic biotite-rich – muscovite granite and an
309 equigranular muscovite-rich and biotite-poor leucogranite, and minor pegmatite veins. The
310 uranium mineralization occurs as discontinuous lenses along these structures and has been
311 mined to a depth of 95 m. The wall zone of the vein is made by a ~1 cm thick quartz comb
312 where U oxide nodules (up to 50 cm in diameter) have grown. The center part was, then, filled
313 by brecciated quartz, chalcopyrite, galena, sphalerite, marcasite, covellite and bismuthinite
314 (Cuney 2006). The analyzed sample corresponds to a nodule of pseudo-spherulitic to spherulitic
315 U oxide (Ur1) brecciated by microfractures mainly filled with quartz, chalcopyrite, galena,
316 sphalerite and a product of alteration of Ur1 (Alt Ur1; Fig. 4a). In the sample from the Kerroch
317 deposit, the mineralization occurs as clusters of millimeter sized spherulitic U oxides
318 disseminated in a leucogranitic granitic country rock and crosscut by ~10 μm wide veins filled

319 with quartz and sulfides (Fig. 4b). In the last sample from the Guern region (undifferentiated-
320 Guern, Fig. 4c), 0.05 to 2 mm wide veins filled by quartz and spherulitic to pseudo-spherulitic
321 U oxide crosscut the leucogranitic country rock.

322 In the Kerlech (Lignol) deposit, 50 cm to 1 m wide mineralized quartz veins oriented N-S
323 to N°170 crosscut the equigranular leucogranite from the contact with the sedimentary rocks
324 (Marcoux 1982; Cuney 2006) (Fig. 3). Vein filling began with a quartz comb followed by fine
325 grained quartz bearing U oxide and chalcopyrite. The last infilling event is represented by
326 barren quartz (Cuney 2006). In the studied sample, a centimeter large cluster of U oxide
327 spherules (Ur1) up to 500 µm in length occurs in fine grained (< 1 mm) quartz together with Fe
328 oxides. The first generation of spherulitic U oxide (Ur1) is brecciated by a second generation
329 of U oxide (Ur2) accompanied by quartz, galena and locally bismuthinite (Fig. 4d).

330 Rosglas and Quérrien (Kerjean) are both classified as episyenite type deposits and are found
331 in the equigranular leucogranite facies (Fig. 3). At Rosglas the episyenite forms a nearly
332 cylindrical subvertical column at the intersection of two faults. In the Rosglas sample, the
333 mineralization occurs as millimeter large clusters of U oxide spherules, with a diameter of 10
334 to 100 µm, disseminated in episyenitized leucogranitic country rocks (Fig.4e). In the Quérrien
335 (Kerjean) sample, the mineralization occurs as brecciated millimeter-wide pseudo-spherulitic
336 U oxides veinlets or clusters disseminated in episyenitized leucogranitic country rocks. In the
337 BSE images, U oxides, crosscut by numerous millimeter-wide fractures, are characterized by
338 the presence of light grey zones interpreted as unaltered (Ur1) and dark grey zones interpreted
339 as altered U oxide (Alt Ur 1; Fig. 4f).

340 *Uranium oxide U-Pb dating*

341 Chemically homogeneous areas that had undergone little or no post-crystallization
342 alteration were selected for U-Pb SIMS dating (Fig. 4). Yet, most analyses plot in a discordant

343 position in the Wetherill concordia diagram (Wc) (Fig. 5), suggesting radiogenic Pb losses
344 which could be the result of hydrothermal alteration (Fig. 4).

345 For the Quistiave (Guern) deposit (Fig. 5a), the 8 analyses plot in a discordant position in
346 the Wc diagram and define a poorly constrained upper intercept of 290 ± 69 Ma (MSWD = 4.1)
347 and a lower intercept of 7 ± 130 Ma. If the discordia is anchored at 0 Ma, assuming a recent Pb
348 loss, an upper intercept of 286 ± 10 Ma (MSWD = 3.5) is obtained. For the Kerroch (Guern)
349 deposit (Fig. 5b), the 30 analyses are discordant to concordant and display an important
350 scattering in the Wc diagram. A poorly constrained upper intercept of 307 ± 86 Ma (MSWD =
351 1.3) and a lower intercept of 89 ± 66 Ma are obtained. For the last sample from the Guern region
352 deposits (Guern – undifferentiated; Fig. 5c), the 15 discordant analyses, affected by Pb loss,
353 define a relatively well-constrained upper intercept of 268 ± 8 Ma (MSWD = 1.2) and a lower
354 intercept of -15 ± 24 Ma in the Wc diagram. Assuming a recent Pb loss, an upper intercept of
355 273 ± 3 Ma (MSWD = 1.2) can be calculated. For the Kerlech (Lignol) deposit (Fig. 5d), the
356 13 analyses plot in a discordant position and display an important scattering in the Wc diagram.
357 The data define a poorly constrained upper intercept of 258 ± 89 Ma (MSWD = 1.3) and a lower
358 intercept of -18 ± 210 Ma. If the discordia is anchored at 0 Ma in the Wc diagram, an upper
359 intercept of 267 ± 11 Ma (MSWD = 1.2) is obtained. Regarding the sample from the Rosglas
360 deposit (Fig. 5e), the 12 discordant analyses, affected by Pb loss, define a relatively well-
361 defined upper intercept of 303 ± 32 Ma (MSWD = 0.5) and a lower intercept of 33 ± 130 Ma.
362 A comparable upper intercept of 296 ± 4 Ma (MSWD = 0.5) is obtained by anchoring the
363 discordia at 0 Ma. Finally, for the Qu errien (Kerjean) deposit, the 14 analyses plot in concordant
364 to discordant positions in the Wc diagram reflecting variable degree of Pb loss (Fig. 5f). The
365 data define a well-constrained upper intercept of 219 ± 5 Ma (MSWD = 0.9) with a lower
366 intercept of -8 ± 69 Ma. By anchoring the discordia at 0 Ma, an identical upper intercept of 219
367 ± 5 Ma (MSWD = 0.8) is obtained.

368 *Whole-rock geochemistry and U-Th distribution*

369 Some major element compositions of whole-rock samples from the Pontivy-Rostrenen
370 complex are reported in the Q-P diagram (Fig 6a). Leucogranite, monzogranite and quartz
371 monzodiorite plot mostly in the field defined for granite-adamellite, ademellite and quartz
372 monzodiorite, respectively, whereas the episyenite sample from the Prat Mérien deposit plots
373 in the granite field and the two episyenite samples from the Poulprio deposit plot out of the field
374 defined for magmatic rocks. The episyenites, which result from leucogranite metasomatism
375 (Alabosi 1984), display evidence of an important dequartzification combined with potassic
376 alteration for Poulprio and a slight dequartzification for Prat Mérien (Fig. 6a). Loss on ignition
377 (LOI) between ~4 and 8 wt.% in the episyenite samples reflects the presence of carbonates and
378 clay minerals (montmorillonite; Alabosi 1984), whereas LOI are below 2 wt.% for the unaltered
379 leucogranite. The episyenite sample from Part Mérien is also enriched in P₂O₅ (1.19 wt.%;
380 Table 1) compared to other episyenites (P₂O₅ < 0.7 wt.%; Table 1) and unaltered leucogranite
381 samples (P₂O₅ < 0.5 wt.%; Ballouard et al. 2017b). Moreover, all episyenite samples display
382 higher As content relative to leucogranite with values from 20 to 95 ppm (Table 1) in
383 episyenites whereas the As content in leucogranite is generally below 11 ppm (Ballouard et al.
384 2017b).

385 In the Th vs. U diagram (Fig. 6b), monzogranite and quartz monzodiorite samples are
386 characterized by high Th/U values mostly above 3, low U contents from ~3 to 9 ppm and a
387 poorly-defined correlation between Th and U. In contrast, the Th/U ratios and U contents are
388 highly variable in the leucogranite and range from ~0.1 to 5 and ~3 to 27 ppm, respectively.
389 Among leucogranite, the lowest Th/U ratios (< 1) and highest U contents (> 15 ppm) are
390 displayed by the equigranular leucogranite and the late equigranular leucogranite whereas Th/U
391 ratios above 1 and U contents below or equal to 15 ppm are found in the porphyritic
392 leucogranite. U correlates negatively with Th for the late equigranular leucogranite samples

393 whereas no clear correlation appears for the porphyritic and equigranular leucogranite. In the
394 episyenite, the Th/U ratios range from 0.9 to 0.01 with a U content from 17 to 48 ppm for
395 Poulprio and of 113 ppm for Prat Mérien. The Prat Mérien episyenite is characterized by an
396 extremely low Th content of 0.6 ppm.

397 *Apatite petro-geochemistry*

398 Apatite is a common accessory mineral in all the magmatic rocks from the Pontivy-
399 Rostrenen complex. In this study, chemical analyses (Table 2) were performed on separated
400 apatite grains from one porphyritic leucogranite (PONT-1: 13 grains), two equigranular
401 leucogranites (PONT-10: 13 grains, PONT-26: 14 grains), the late equigranular leucogranite
402 (PONT-20: 13 grains), one episyenite (MS-81-66-PM: 10 grains), one monzogranite (PONT-
403 22: 20 grains) and two quartz-monzodiorites (PONT-7:14 grains, PONT-23: 15 grains). In all
404 these samples, the F content in the apatite crystals is always above 0.75 apfu indicating that
405 they are fluoroapatite (Table 2).

406 In all leucogranite and episyenite samples, apatite grains appear as squat prisms up to 500
407 μm in length. In leucogranite, the crystals display generally yellow-green colors in
408 cathodoluminescence (CL) with irregular patchy zoning (Fig. 7a) not visible in the BSE images.
409 In these CL images, the dark yellow-green color characteristic of the grain cores generally
410 evolves toward light yellow-green or even locally blue colors for the rims. CL emission spectra
411 reveal mainly Mn^{2+} and minor REE^{3+} activated luminescence in both yellow-green zones (Fig.
412 8a). The change from dark yellow-green to light yellow-green color is associated with a
413 decrease in the Fe and Mn contents in the chemical maps (Fig. 7a) and in the Mn vs. Fe diagram
414 (Fig. 9), suggesting that the decrease of the CL intensity is due to quenching of Mn-activated
415 CL by Fe or self-quenching effect by Mn (Kempe and Götze 2002). In samples PONT-10 and
416 26, the cores are characterized by Mn and Fe contents from 0.04 to 0.17 and 0.02 to 0.07 apfu,
417 respectively, whereas in the rims, the Mn and Fe contents range from < 0.01 to 0.11 and < 0.01

418 to 0.06 apfu, respectively (Fig. 9). In the episyenite samples, apatite crystals commonly display
419 irregular zoning with yellow-green, blue, blue-grey or red-pink colors in CL images (Fig 7b).
420 Blue-grey CL zones are commonly observed as overgrowth or as patches and veins in yellow-
421 green and blue zones whereas red-pink zones are observed as late overgrowth for the other
422 zones (Fig. 7b). CL spectra reveal mainly Mn^{2+} with minor Eu^{2+} and REE^{3+} activated
423 luminescence in yellow-green zones whereas the CL activation by both Eu^{2+} and REE^{3+}
424 increases in the other zones (Fig. 8b). As seen on the spectra, the differences in CL color in
425 these apatite grains result from subtle changes in the emission caused by three main activators:
426 Eu^{2+} (blue), Mn^{2+} (yellow-green) and Sm^{3+} (red). Interestingly, the CL spectra in blue, blue-
427 grey and red-pink zones commonly show activation by Eu^{3+} (Fig. 8b). Crystals or zones with a
428 yellow-green color in the CL images are generally characterized by elevated Mn and Fe
429 contents from 0.08 to 0.12 and 0.03 to 0.04 apfu, respectively, whereas blue (and blue-grey)
430 CL zones display Fe and Mn contents < 0.01 apfu (Fig. 9). In contrast, the REE content as
431 indicated by La and Ce concentration does not significantly change (Table 2), suggesting that
432 the increased REE activation in blue and blue-grey CL zones relative to yellow-green CL zones
433 is not due to an increase in REE concentration but rather to a decrease in Mn^{2+} . Zones with a
434 red-pink color in CL images are brighter in the BSE images (Fig. 8b) and are characterized by
435 Fe and Mn below detection limit, an elevated average As content of 0.28 apfu (commonly $<$
436 0.01 apfu in other grains) and an elevated average OH content of 0.21 apfu (generally around
437 0.1 in other crystals; Table 2). There is a perfect match between dark red CL and As-rich bands
438 (Fig. 7b). Incorporation of As in apatite structure likely explains the decrease of CL intensity
439 since As has been reported as a CL inhibitor (Perseil et al. 2000; Kempe and Götze 2002). The
440 combined increase in both As and OH contents marks the evolution toward the johnbaumite
441 end-member $[Ca_5(AsO_4)_3(OH)]$.

442 In the monzogranite sample, apatite appears as squat or elongated prisms up to 500 μm in
443 length homogeneous in BSE images and displaying a generally homogeneous yellow-green CL
444 color (Fig. 7c). CL spectra reveal a major Mn^{2+} with a moderate REE^{3+} activated luminescence
445 (Fig. 8c). Rarely, apatite grains display a zonation on Si chemical map with a slight decrease of
446 the Si contents from core (~ 0.02 apfu) to rim (< 0.01 apfu) (Fig. 7c). The Mn (0.02 – 0.03 apfu)
447 and Fe (0.01 – 0.03 apfu) contents are generally lower than those found in the apatite grains
448 from the leucogranite (Fig. 9). Regarding the quartz monzodiorite samples, apatite crystals
449 appear as squat or elongated prisms up to 200 μm in length. These apatite crystals appear as
450 homogeneous in BSE images and display a predominant purple CL color with purple-green
451 rims (Fig. 7d). CL emission spectra reveal a major REE^{3+} and Eu^{2+} with minor Mn^{2+} activated
452 luminescence. Here again, slight changes in spectra have an impact on the resulting CL color,
453 with the more greenish CL being induced by a decrease in Sm^{3+} (red) relative to Dy^{3+} (green)
454 Accordingly, EPMA analyses show a slight decrease of the LREE content ($[\text{La} + \text{Ce}] \sim 0.01$
455 apfu to below detection limits) from core to rim and these crystals are characterized by low Fe
456 and Mn contents below 0.01 apfu (Fig. 9). Note that here the emission peak at 415 nm (Fig. 8d)
457 could be related to structural defects associated with REE substituting for Ca^{2+} (Marshall 1980)
458 as Eu^{2+} emission occurs at about 450 nm (e.g. Blanc et al. 2000) as it is the case in episyenite
459 apatite (Fig. 8b).

460 *Apatite U-Pb dating*

461 Apatite U-Pb analyses were performed for all the samples presented in the last section with
462 the exception of the quartz monzodiorite PONT-23. The results are reported in Tera-
463 Wasserburg diagrams (Fig. 10).

464 In leucogranite, the analyses are discordant with $^{207}\text{Pb}/^{206}\text{Pb}$ ratios ranging from 0.148 to
465 0.537. Due to the small size of the rims observed in the CL images, analyses were almost
466 exclusively performed on grains cores. For the porphyritic leucogranite sample PONT-1 (Fig.

467 10a), 23 analyses on 19 different grains define a poorly defined lower intercept of 285.4 ± 8.5
468 Ma (MSWD = 6.8). Using the common Pb composition calculated at 285 Ma (Stacey and
469 Kramers 1975), the analyses yield two populations for the ^{207}Pb corrected ages; 307.2 ± 7.1 Ma
470 (MSWD = 2.2, n = 9) and 286.2 ± 3.8 Ma (MSWD = 1.6, n = 14), respectively. For the
471 equigranular leucogranite sample PONT-10 (18 analyses from 16 grains; Fig. 10b), the
472 unforced discordia yields a lower intercept of 270.4 ± 6.7 Ma (MSWD = 2.5). Once again, two
473 populations are obtained for the corrected ^{207}Pb ages, calculated using the common Pb
474 composition at 270 Ma, and yield mean values of 294.9 ± 7.4 Ma (MSWD = 0.12) and $279.9 \pm$
475 2.9 Ma (MSWD = 0.42), respectively. For the equigranular leucogranite PONT-26 (21 analyses
476 from 15 grains; Fig. 10c), we obtain a lower intercept of 272.8 ± 2.9 Ma (MSWD = 1.2). Two
477 populations of ^{207}Pb corrected ages are obtained and yield mean values of 299.9 ± 4.3 Ma
478 (MSWD = 0.26) and 279.6 ± 2.1 Ma (MSWD = 0.59), respectively. Then, for the late
479 equigranular leucogranite sample (PONT-20; 24 analyses from 18 grains; Fig. 10d), a poorly
480 constrained lower intercept at 278.0 ± 11.0 Ma (MSWD = 8.7) is obtained and analyses yield
481 two populations of ^{207}Pb corrected ages with a mean value of 297.1 ± 3.0 Ma (MSWD = 1.6)
482 and 280.9 ± 2.1 Ma (MSWD = 0.63), respectively. There is no clear correlation between the
483 apparent ^{207}Pb corrected ages obtained for the apatite grains from leucogranite and their relative
484 Mg and/or Fe contents.

485 For the episyenite sample (MS-81-66-PM; Fig. 10e), the discordant analyses display highly
486 variable common Pb contents with $^{207}\text{Pb}/^{206}\text{Pb}$ values ranging from 0.122 to 0.861. No analyses
487 were performed on the blue-grey or pink-red CL zones because of their small sizes. In this
488 sample, 9 analyses out of 6 grains presenting yellow-green or blue colors and characterized by
489 $^{207}\text{Pb}/^{206}\text{Pb}$ values below 0.300, display a well-defined discordia and yield a lower intercept age
490 of 289.0 ± 10.0 Ma (MSWD = 0.54). This age is identical within error to the mean ^{207}Pb

491 corrected age of 286.5 ± 3.8 Ma (MSWD = 0.31). The other data characterized by a higher
492 common Pb content are more scattered and were therefore not used.

493 Regarding the monzogranite sample (PONT-22; Fig. 10f), the 23 analyses from 22 grains
494 are discordant with $^{207}\text{Pb}/^{206}\text{Pb}$ values ranging from 0.268 to 0.729. The analyses yield a well-
495 defined lower intercept of 317.8 ± 4.9 Ma (MSWD = 1.2) identical within error to a mean ^{207}Pb
496 corrected age of 320.6 ± 3.1 Ma (MSWD = 0.76).

497 Finally, for the quartz-monzodiorite sample (PONT-7; Fig. 10g), the 17 analyses from 13
498 grains are discordant with $^{207}\text{Pb}/^{206}\text{Pb}$ values from 0.316 to 0.628. The data align on a discordia
499 which yields a well-defined lower intercept of 298 ± 13 Ma (MSWD = 1.17) identical within
500 error to a mean ^{207}Pb corrected age of 310.4 ± 7.5 Ma (MSWD = 0.31).

501 **Discussion**

502 *U behavior in the Pontivy-Rostrenen complex*

503 In contrast to the quartz monzodiorite and monzogranite samples, characterized by low U
504 contents (< 9 ppm) and elevated Th/U values mostly above 3, the leucogranite is characterized
505 by both highly variable U contents (~3 to 27 ppm) and Th/U ratios (~0.1 to 5) (Fig. 6b). The
506 Th/U is an indicator of the nature of the U bearing minerals in granitoids and the high Th/U
507 ratios (> 2) measured in some samples suggest that most of their U is hosted in refractory
508 mineral phases such as zircon, titanite or allanite for quartz monzodiorites and zircon or
509 monazite for leucogranite and monzogranite (Cuney 2014). On the other hand, low Th/U values
510 (< 1) and U contents of tens of ppm in peraluminous leucogranitic melts favor the crystallization
511 of magmatic uranium oxides at the expense of monazite and up to 80-90 % of the available U
512 can be hosted in U oxide (Friedrich et al. 1987; Peiffert et al. 1994, 1996; Förtser 1999; Förster
513 et al. 2008; Cuney 2014). In leucogranitic magmas, extraction of accessory minerals
514 incorporating limited amounts of U, such as monazite and zircon, during fractional

515 crystallization (Ballouard et al. 2017b) likely induced an increase of the U contents in the melt
516 and a decrease of the Th/U values. Such behavior, well-illustrated by the negative correlation
517 between Th and U in the late equigranular leucogranite (Fig. 6b), probably triggered the
518 crystallization of uranium oxides in the most evolved leucogranitic melts. In contrast, there is
519 no correlation between Th and U for the porphyritic and equigranular leucogranites (Fig. 6b).
520 We propose that the very variable Th/U values displayed by these facies can be attributed to a
521 combination between magmatic evolution (uranium oxides crystallization), hydrothermal
522 alteration and/or surface weathering (uranium oxides leaching), low Th uranium oxides being
523 particularly prone to dissolution by infiltrating oxidized fluids (Förster 1999, Förster et al.
524 2008). In the episyenite sample, the high U contents suggest hydrothermal U enrichment while
525 the extremely low Th content of the Prat Mérien sample suggest either high degree of magmatic
526 fractionation or monazite leaching, the latter being documented in some episyenites (López-
527 Moro et al. 2013).

528 *Age of the uranium mineralization*

529 U-Pb analyses of uranium oxides provide evidence for a complex behavior of the U-Pb
530 system depending on the samples. A first group composed of the Guern-undifferentiated,
531 Rosglas and Quérien (Kerjean) samples yield relatively well-constrained, unforced upper
532 intercepts of 268 ± 8 Ma, 303 ± 32 Ma and 219 ± 5 Ma, respectively whereas the Quistiave
533 (Guern), Kerroch (Guern) and Kerlech (Lignol) samples, belonging to the second group, yield
534 poorly constrained unforced upper intercepts of 290 ± 69 Ma, 307 ± 86 Ma and 258 ± 89 Ma
535 respectively (Fig. 5). In Figure 11, the analyses from the two groups are reported in a $^{206}\text{Pb}/^{207}\text{Pb}$
536 versus $^{235}\text{U}/^{207}\text{Pb}$ concordia diagram that is useful to discriminate between Pb loss and Rn
537 leakage processes in uranium oxides (Ludwig and Simmons 1992). In this diagram, samples
538 from the first group only display evidence of Pb loss whereas samples from the second group
539 display evidence for both Pb loss and Rn leakage. It is not possible to constrain precisely the

540 timing of the Rn leakage in these samples but this process seems to have occurred during the
541 Mesozoic (Fig. 11b). In contrast, unforced lower intercept ages close to 0 suggest recent Pb loss
542 for all samples excepted Kerroch (Guern: Fig. 5). Therefore, we suggest that the poorly
543 constrained upper intercept U-Pb ages obtained for samples from the second group are the result
544 of a complex combination of Pb loss and Rn leakage. Despite that the Wc diagrams suggest an
545 Upper Carboniferous to Lower Permian age for the crystallization of these samples (Fig. 5), we
546 argue in the followings that only the samples from the first group (Fig. 11a) can be used to
547 constrain precisely the timing of the U mineralizing events in the Pontivy-Rostrenen district.

548 In the Guern region, the Guern-undifferentiated sample yields an unforced upper intercept
549 age of 268 ± 8 Ma which is identical within error to the upper intercept of 273 ± 3 Ma obtained
550 if the discordia is anchored at 0 Ma (Fig. 5c). Therefore, we suggest that this deposit formed
551 273 ± 3 Ma ago. Analyses on uranium oxides from the Rosglas deposit yield an unforced
552 intercept of 303 ± 32 Ma identical within error to the forced upper intercept age of 296 ± 4 Ma
553 that we interpret as the age of formation (Fig. 5e). Then, the U oxides from the Qu errien
554 (Kerjean) deposit yield two identical well-constrained upper intercepts of 219 ± 5 Ma (Fig. 5f),
555 interpreted as the age of their crystallization. To sum up, the hydrothermal U deposits from the
556 Pontivy-Rostrenen complex mostly formed during the Lower Permian, from ca. 300 to 270 Ma,
557 but some U deposits formation or U remobilization also occurred during the Triassic around
558 220 Ma as illustrated in the Qu errien (Kerjean) deposit.

559 ***Apatite as a proxy to date emplacement and/or alteration ages?***

560 *Apatite dating in rocks unaffected by fluids*

561 In the quartz monzodiorite and the monzogranite, apatite grains are unzoned or display
562 discrete regular zonation on CL images (Fig. 7c and d), which suggest that these crystals have
563 kept their magmatic signature and were not affected by significant hydrothermal processes. U-

564 Pb dating of apatite grains from the quartz monzodiorite yield a lower intercept of 298.0 ± 13.0
565 Ma identical within error to a mean ^{207}Pb corrected age of 310.4 ± 7.5 Ma (Fig. 10g). The mean
566 ^{207}Pb corrected age is identical within error to the zircon U-Pb age of 315.2 ± 2.9 Ma obtained
567 for this facies (Ballouard et al. 2017b) (Table 3), and can be interpreted as reflecting the
568 emplacement for this quartz-monzodiorite intrusion. In the monzogranite, apatite grains provide
569 comparable intercept and mean ^{207}Pb corrected ages of 317.8 ± 4.9 Ma and 320.6 ± 3.1 Ma (Fig.
570 10f), respectively. Both ages are identical within error to the zircon U-Pb age of 315.5 ± 2.0
571 obtained on this sample (Ballouard et al. 2017b) (Table 3) and can be interpreted as reflecting
572 the emplacement age of the intrusion.

573 *Apatite dating in rocks affected by fluids*

574 In contrast to the monzogranite and quartz monzodiorite apatite crystals, the apatite grains
575 from leucogranite and episyenite display petro-geochemical evidence for pervasive
576 hydrothermal alteration. Indeed, these apatite grains show patchy irregular zoning in the CL
577 images and in the Fe and Mn chemical maps, likely reflecting fluid interactions processes
578 (Fig.7a). The decrease in the Mn and Fe contents, generally observed from the core to the rims
579 (Fig. 7a and 9) likely reflects the transition toward a more oxidized environment during
580 hydrothermal events. Indeed, Mn and Fe are more compatible in apatite in reduced conditions
581 as Mn^{2+} and Fe^{2+} substitute easily to Ca^{2+} (Miles et al. 2014). In addition to the irregular patchy
582 zoning reflecting Fe and Mn mobility, apatite grains from the episyenite also display features
583 likely hydrothermal in origin. The formation of new apatite is in agreement with the elevated
584 whole-rock P content (1.2 wt.%) measured for this sample (MS-81-66; Table 1). Late apatite
585 overgrowths, having a red-pink color in CL images, show enrichment in As and OH
586 characteristic of a substitution toward the johnbaumite end-member (Fig. 7b). The observed
587 OH enrichment suggests that these zones crystallized in a H_2O -rich environment whereas the
588 increase of the As content reflects the high oxygen fugacity of the hydrothermal fluids as As^{5+}

589 will substitute more easily for P^{5+} than As^{3+} . Moreover, apatite zones interpreted as
590 hydrothermal in origin (blue, blue-grey or red-pink CL zones in episyenite sample MS-81-66;
591 Fig 7b) show a slight but significant Eu^{3+} activation (Fig. 8b) that could also be interpreted as
592 related to the oxidized character of the mineralizing fluid (Roeder et al. 1987; Mitchell et al.
593 1997). Roder et al. (1987) argued that no conclusive interpretation regarding the redox state of
594 the fluids can be drawn from the observation of Eu^{3+} activation in the CL of apatite because
595 apatite is expected to concentrate preferentially Eu^{3+} over Eu^{2+} . However, in our episyenite
596 apatite, the involvement of oxidized fluids is supported by the occurrence of As^{5+} and the
597 observed Eu^{3+} activation could be regarded as an additional evidence.

598 In the episyenite sample MS-81-66 (Fig. 10e), apatite grains yield a lower intercept of 289.0
599 ± 10 Ma and a mean ^{207}Pb corrected of 286.5 ± 3.8 Ma identical within error. Considering the
600 alteration evidence depicted above, we thus argue that dating of these apatite grains provides
601 the age of the episyenitization (i.e. the metasomatism of the leucogranite) at 286.5 ± 3.8 Ma.

602 In each of the leucogranite samples, the data obtained by apatite U-Pb dating reveal a
603 complex behavior with regard to their U-Pb system. First of all, the lower intercept obtained
604 for leucogranite are characterized by rather high MSWD values (between 2.5 and 8.7) with the
605 exception of equigranular leucogranite PONT-26 (MSWD=1.2). This probably means that the
606 scattering of the data can be attributed to geological event(s) rather than to an analytical
607 problem. This is further amplified by the fact that lower intercept and ^{207}Pb corrected ages are
608 systematically different and, when available, are always younger than the emplacement ages
609 (Ballouard et al. 2017b; see Table 3). As outlined earlier, all the apatite grains from leucogranite
610 show evidence for some interaction with fluids. At a first order, this means that the U-Pb system
611 in these grains has been affected by late fluid circulations. In all cases, the mean ^{207}Pb corrected
612 ages define two different populations for each sample: a first one returning ages in the range
613 294.9 ± 7.4 Ma to 307.2 ± 7.1 Ma, and a second one with ages ranging from 279.6 ± 2.1 Ma to

614 286.2 ± 3.8 Ma (Fig. 10a-d). In Figure 12, the calculated U contents in the apatite grains are
615 reported as a function of the corresponding ²⁰⁷Pb corrected ages. Regarding the apatite grains
616 from the monzogranite ([U] ~7 – 90 ppm), quartz-monzodiorite ([U] ~12 – 56 ppm), late
617 equigranular leucogranite ([U] ~48 – 184 ppm) and episyenite ([U] ~95 – 263 ppm) samples,
618 the U content is relatively constant and there is no correlation with the ²⁰⁷Pb corrected ages. In
619 contrast, the U contents in the apatite from the equigranular (and the porphyritic leucogranite)
620 increase as the ²⁰⁷Pb corrected ages get younger. This inverse correlation between the apatite
621 grain apparent ages and their U contents likely shows that the fluids which interacted with these
622 apatite grains were U-rich.

623 In order to determine if we can extract meaningful ages from this dataset, we decided, for
624 each sample, to keep only the population returning the youngest ²⁰⁷Pb-corrected ages (Fig. 13).
625 The resulting unforced lower intercept ages are 285.9 ± 7.4 Ma (MSWD=2.1; PONT-1:
626 porphyritic leucogranite), 266 ± 11 Ma (MSWD=2.7; PONT-10: equigranular leucogranite),
627 272.8 ± 6.4 Ma (MSWD=0.39; PONT-26: equigranular leucogranite) and 278 ± 15
628 (MSWD=0.84; PONT-20: late equigranular leucogranite). Individually, all these ages are
629 comparable with their respective ²⁰⁷Pb-corrected ages. We therefore conclude that the youngest
630 mean ²⁰⁷Pb-corrected ages represent, for each sample, the best estimate of the time when the
631 fluids interacted with the rocks. The other, older, ages probably represent partially reset apatite
632 grains and are therefore meaningless.

633 As a conclusion, apatite U-Pb dating of episyenite and leucogranite provide evidence for
634 fluid infiltration from ca. 290 Ma (episyenitization) to 275 Ma (late alteration of leucogranite).
635 This period is contemporaneous with the Lower Permian ages obtained on most of the uranium
636 deposits in the Pontivy-Rostrenen district (Fig. 5). This, together with the fact that the apatite
637 grains affected by the fluids are richer in uranium than those that have been partly affected, or

638 not-affected, suggest that these hydrothermal fluids were the same than those at the origin of
639 the formation of U deposits (Fig. 12).

640 *Metallogenic model and regional implication*

641 In the Pontivy-Rostrenen complex, the main U mineralization is hosted in N170° oriented
642 quartz veins (Kerlech – Lignol, Bonote) or as brecciated quartz veins (Guern region) and
643 episyenite bodies (Prat Mérien, Poulprio) where the mineralized zone follows N120-130° and
644 N130-160° oriented brittle lineaments, respectively. As illustrated in Figure 14 and proposed
645 by Marcoux (1982), the N170° oriented mineralized quartz veins can be interpreted as tension
646 gashes accommodating late dextral movement along the SASZ, whereas the formation of other
647 deposits could be related to second order faulting also associated with deformation along the
648 SASZ. Muscovite Ar-Ar and zircon or monazite U-Th-Pb dating on syntectonic leucogranite
649 and mylonites from the SASZ (Tartèse et al. 2011b; 2012; Gapais et al. 2015) bracketed the
650 ductile deformation along the SASZ between ca. 315 and 300 Ma. According to uranium oxide
651 and apatite U-Pb dating on U deposits and leucogranite from the Pontivy-Rostrenen complex,
652 respectively, brittle deformation which allowed the mineralizing fluids to circulate was active
653 from ca. 300 to 270 Ma.

654 On a regional scale, U deposition in the Pontivy-Rostrenen complex was contemporaneous
655 with U mineralizing events in the Mortagne (from ca. 290 to 260 Ma; Cathelineau et al. 1990)
656 (Fig 2) and Guérande (from ca. 300 to 275 Ma; Ballouard et al. 2017a; Figs. 2 and 14) districts.
657 In the Guérande district, the main perigranitic vein mineralization (Pen Ar Ran) occurs in a
658 graben structure localized above the apical zone of the Guérande leucogranite. The formation
659 of the graben and U mineralization relates to a brittle-ductile deformation which followed the
660 emplacement of the Guérande intrusion in an extensional setting at ca. 310 Ma (Ballouard et al.
661 2015, 2017a) (Fig. 14). From 310 to 300 Ma, the SASZ acted as a transfer zone between the
662 South Armorican Massif, a thickened domain in extension, and the Central Armorican Massif,

663 a thin domain under pervasive dextral wrenching (Gumiaux et al. 2004a; Gapais et al. 2015).
664 According to uranium oxide U-Pb dating, this tectonic configuration was likely active until the
665 middle Permian under mostly brittle conditions. U deposition in the Armorican Massif is
666 contemporaneous with the main U mineralizing phase in the whole EVB (300 – 260 Ma;
667 Cathelineau et al. 1990; Tischendorf and Förster 1994; Romer et al. 2010; Ballouard et al.
668 2017a) and follow the main period of formation of W ± Sn granite-related deposits in the Massif
669 Central (Fig. 1) that mostly occurred from ca. 335 to 300 Ma (Harlaux et al. 2017).

670 As outlined earlier, the apatite grains from leucogranite of the Pontivy-Rostrenen complex
671 and their episyenites show evidence for an interaction with oxidized hydrothermal fluids from
672 ca. 290 to 275 Ma, contemporaneous with the formation of the U deposits. A surface-derived
673 origin is favored as numerous indications for the infiltration of meteoric-derived fluids at depth,
674 from oxygen isotopes and fluid inclusions studies, exist in rocks associated with the SASZ:
675 quartz veins (Lemarchand et al. 2012), leucogranite (Tartèse and Boulvais 2010) and mylonite
676 (Tartèse et al. 2012). For example, a mylonite from the Guillomarch quarry on the southern-
677 edge of the Pontivy-Rostrenen complex (Fig. 3) displays whole-rock $\delta^{18}\text{O}$ values as low as 1.7
678 ‰, which indicates interaction with a low- $\delta^{18}\text{O}$ fluid derived from the surface (Tartèse et al.
679 2012). Surface-derived fluids, likely oxidized, are thus able to transport an important quantity
680 of U in solution (Dubessy et al. 1987). During the late Carboniferous and Early Permian, the
681 SASZ and the detachments likely acted as major channels for surface-derived oxidized fluids
682 which have the capacity to dissolve magmatic uranium oxides in fertile intrusions such as the
683 Guérande (Ballouard et al. 2017a), Questembert (Tartèse et al. 2013) and the Pontivy-Rostrenen
684 leucogranites (Fig. 14).

685 In the Guérande district (Figs. 2 and 14), fluid inclusion analyses of a quartz from a quartz-
686 uranium oxide vein of the Pen Ar Ran deposit, indicate a low salinity mineralizing fluid (1-6
687 wt.% eq. NaCl) with trapping temperatures in the range 250 – 350 °C (Ballouard et al. 2017a).

688 The trapping temperatures and the salinities of fluid inclusions in the Guérande district are
689 overall comparable with those obtained in U deposits from the Mortagne district (Fig. 2) and
690 the northwestern part of the Massif Central (Fig. 1), with salinity and temperature generally in
691 the range 0 – 7 wt.% eq. NaCl and 150-250 °C, respectively (Cathelineau 1982; Cathelineau et
692 al. 1990; Lespinasse and Cathelineau 1990; Cuney and Kyser 2008). The low salinity measured
693 in the fluid inclusions from these deposits are in agreement with the contribution of meteoric-
694 derived fluids although the elevated trapping temperatures and the variation of salinity values
695 suggest mixing processes with metamorphic- or basin-derived fluids, as it was already proposed
696 for the FMC (Turpin et al. 1990) and the Bohemian Massif (Křibek et al. 2008; Dolníček et al.
697 2013).

698 In the Guérande district, the uranium precipitated at the contact with reducing lithologies,
699 such as black shales (Cathelineau et al. 1981; Ballouard et al. 2017a). In the Pontivy-Rostrenen
700 district, most deposits occurred to the north of the Pontivy intrusion close to the contact with
701 sedimentary country rocks or micaschists enclaves, which likely played a role in the U
702 precipitation processes. In parallel, regional-scale strike slip faults can act as channels for lower
703 crustal reduced metamorphic fluids, in addition to surface derived fluids, such as those
704 documented in regional quartz veins along the SASZ (Lemarchand et al. 2012). These fluids
705 can in turn be involved in U precipitation (Fig. 14). Permian basins are not preserved in the
706 Armorican Massif, with the exception of its northeastern part where shales and red sandstones
707 were deposited in fluvial or lacustrine environments (Ballèvre et al. 2013). However, in the
708 Massif Central, intra-continental basins locally filled by bituminous shales, were proposed as
709 the source of reduced fluids that caused U to precipitate (Turpin et al. 1990; Maignac and
710 Cuney 1999). These Permian basins, that formed during a major episode of extension affecting
711 all western Europe (Kroner and Romer 2013), have a dominant half-graben structural style with

712 a common transtensional character (McCann et al. 1990) attesting for the role of detachments
713 and strike-slip faults in the control of the sedimentation, as illustrated in Figure 14.

714 Around 300 Ma, convective fluid infiltration in the Armorican Massif was enhanced by heat
715 from a regional late crustal magmatism event as shown by the emplacement of the late
716 equigranular leucogranite in the Pontivy-Rostrenen district (304.7 ± 2.7 Ma; Ballouard et al.
717 2017b) and leucogranitic dykes in the Guérande area (302.5 ± 2.0 Ma; Ballouard et al. 2015,
718 2017a). The Permian period in the EVB is marked by an abnormal heat flux in the crust and the
719 mantle, as illustrated, for example, by important granitic magmatism in Cornwall (Chen et al.
720 1993), in Iberia (Gutiérrez-Alonso et al. 2011) and in Central Europe (Finger, 1997; Romer et
721 al. 2007). This heat flux combined with the high heat producing character of the granites
722 enriched in radioactive elements (Vigneresse et al. 1989) likely sustained an elevated
723 geothermal gradient in the upper crust, able to enhance convective fluid flow (Scaillet et al.
724 1996). In the Guérande and Pontivy district, apatite fission tracks dating suggests that
725 leucogranite was at a temperature above 120°C , or at a depth above 4 km for a geothermal
726 gradient of $30^{\circ}\text{C}/\text{km}$, during the formation of the deposits (Fig. 14) (Ballouard, 2016; Ballouard
727 et al. 2017a).

728 Finally, a last U mineralizing or remobilization event occurred at ca. 220 Ma in the Pontivy-
729 Rostrenen district (Rosglas deposit; Figs. 3 and 5d). This mineralizing event is sub-
730 contemporaneous with the emplacement of microgabbro dikes in the western part of the
731 Armorican Massif between 210 and 195 Ma which marks the first step of Atlantic rifting
732 (Caroff et al. 1995; Ballèvre et al. 2013). This tectonic event likely caused fluid flow responsible
733 for late, discrete, U mobilization. Triassic and Lower Jurassic U mineralizing or mobilization
734 events are also recorded in the Mortagne district (ca. 200 Ma) and the Massif Central (ca. 210
735 – 170 Ma), and have been attributed to the tectonic movements at the origin of the opening of
736 the Tethys (Cathelineau et al. 1990; Cathelineau et al. 2012).

737 **Conclusion**

738 In the Late Carboniferous Pontivy-Rostrenen composite intrusion, intragranitic
739 hydrothermal U mineralization is associated with the emplacement of peraluminous
740 leucogranite. Mineralization is hosted in quartz veins associated with brittle structures related
741 to strike-slip deformation along the SASZ. Our study of the U deposits and their magmatic
742 country-rocks leads to the following conclusions:

743 (1) In the peraluminous monzogranite and metaluminous quartz monzodiorite samples, low
744 U contents (< 9 ppm) and elevated Th/U values (> 3) suggest that most of their U is
745 hosted in refractory minerals such as zircon and monazite for the former and zircon,
746 titanite and allanite for the latter. For the peraluminous leucogranite, the highly variable
747 U contents (~3 – 27 ppm) and Th/U ratios (~0.1 to 5) suggest that, in some samples,
748 crystallization of magmatic uranium oxide followed by uranium oxide leaching during
749 subsequent hydrothermal alteration and weathering occurred.

750 (2) Apatite is a powerful tool both for dating and tracing fluids in the system. Apatite grains
751 from the monzogranite and quartz monzodiorite samples are unzoned or display regular
752 zonation in CL images suggesting that these crystals kept their magmatic signature.
753 Apatite U-Pb dating of these samples yield ages around 315 Ma which can be interpreted
754 as emplacement or cooling ages. Apatite grains from leucogranite or episyenites
755 samples display irregular zoning in CL (or BSE) images attributed to the mobility of Fe
756 and Mn or As during an oxidized hydrothermal event involving surface-derived fluids.
757 Apatite U-Pb dating of episyenite and leucogranite samples yield ages from ca. 290 to
758 275 Ma, interpreted to date the oxidized hydrothermal event. In the leucogranite facies
759 associated with U deposits, the younger apatite grains are enriched in U compared to
760 older ones, suggesting that these oxidized fluids were involved in the formation of U
761 deposits.

762 (3) U-Pb dating of uranium oxide from the U deposits revealed a main Permian U
763 mineralizing phase from 300 to 270 Ma synchronous with the oxidized hydrothermal
764 event recorded by apatite grains from leucogranite. A late U mineralization or
765 remobilization event also occurred during the Triassic at ca. 220 Ma.

766 On a regional scale, U deposition from 300 to 270 Ma in the Pontivy-Rostrenen complex is
767 contemporaneous with the main U mineralizing phase in the Armorican Massif and the
768 European Variscan belt. During this period, late brittle dextral deformation along the SASZ was
769 synchronous with discrete extension in the South Armorican Domain suggesting a continuum
770 of the ductile deformation which occurred in the region during Late Carboniferous from ca. 315
771 to 300 Ma. Detachment zones and regional scale strike-slip faults acted as major channels for
772 oxidized surface-derived fluids which were in turn able to dissolve magmatic uranium oxide
773 from fertile peraluminous leucogranite and then form hydrothermal U deposits due to the
774 interaction with reducing lithologies and/or crustal basin derived fluids.

775 **Acknowledgment**

776 This study was supported by 2012-2013 NEEDS-CNRS and 2015-CESSUR-INSU (CNRS)
777 research grants attributed to M. Poujol. We want to thank AREVA (in particular D. Virlogeux,
778 J.M. Vergeau and P.C. Guiollard) for providing uranium oxide samples, historical reports and
779 data and for fruitful discussions. We are grateful to Y. Lepagnot (Geosciences, Rennes) for
780 crushing the samples. Many thanks to J. Langlade (IFREMER, Brest), O. Rouer, S. Matthieu
781 and L. Salsi (SCMEM - Géoressources, Nancy) for their technical supports during EPMA and
782 SEM analyses. Thank you to N. Bouden (CRPG, Nancy) for the help during SIMS analyses.
783 J.M., M.C. and M.C. would also like to thank AREVA for the permission to publish the present
784 manuscript. This manuscript benefited from the comments of R. Shail and an anonymous
785 reviewer. We want to thank R.L. Romer and G. Beaudoin for editorial handling.

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- 1075

1076 **Captions**

1077 Figure 1: Schematic representation of the west European Variscan belt representing the main terranes
1078 (Ballèvre et al. 2009, 2013, 2016) and showing the relationship between Variscan granitoids and
1079 hydrothermal U deposits (after Cuney and Kyser 2008). The granite typology is based on the 1:1,000,000
1080 geological maps of France (Chantaine et al. 2003), Portugal and Spain (Civis Llovera, 2015) and the 1:
1081 500,000 geological maps of Czechoslovakia (Fusán et al. 1967) and Czech Republic (Cháb et al. 2007).
1082 NASZ: north Armorican shear zone; NBSASZ: northern branch of the south Armorican shear zone;
1083 SBSASZ: southern branch of the south Armorican shear zone. NEF: Nort-sur-Erdre fault. Mineral
1084 abbreviations according to Whitney and Evans (2010).

1085

1086 Figure 2: (a) Schematic structural map of the Armorican Massif. (b) General geological map of the
1087 Armorican Massif identifying the different type of Carboniferous granites, according to Capdevila
1088 (2010), and the locations of the principal U deposits. The geological map is modified from Chantraine
1089 et al. (2003), Gapais et al. (2015) and Ballouard et al. (2017b). NASZ: north Armorican shear zone.
1090 SBSASZ: southern branch of the south Armorican shear zone. NBSASZ: northern branch of the south
1091 Armorican shear zone. Fe-K granites: ferro-potassic granites. Mg-K granites: magnesio-potassic
1092 granites. Mineral abbreviations according to Whitney and Evans (2010).

1093

1094 Figure 3: Geological map of the Pontivy-Rostrenen magmatic complex showing the different magmatic
1095 units and locations of U deposits. Sample locations from this and previous studies are shown on the
1096 map. The map is redrawn from Euzen (1993), Ballouard et al. (2017b) and from the 1: 50,000 BRGM
1097 geological maps of Pontivy (Dadet et al. 1988), Rostrenen (Bos et al. 1997), Plouay (Bechennec et al.
1098 2006) and Bubry (Bechennec and Thiéblemont 2009). SASZ: south Armorican shear zone. The types of
1099 uranium deposits and their main orientations are from Marcoux (1982) and Cuney (2006). Qst:
1100 Quistiave; Krh: Kerroch; PM: Prat Mérien; PP: Poulprio; Sul: Sulliado; Qsn: Quistinic; Krl: Kerlech
1101 (Lignol); Bnt: Bonote; Rsg: Rosglas; Qrn: Quérien (Kerjean); Krs: Kerségalec. Guill.: Guilligomarch.

1102

1103 Figure 4: Backscattered electron images for uranium oxides (Ur) from uranium deposits of the Pontivy-
1104 Rostrenen district. (a) Pseudo-spherulitic U oxide crosscut by a fracture filled with quartz, alteration
1105 product of U oxide (Alt Ur1) and galena (Quistiave deposit). (b) Spherulitic U oxide nodule from the
1106 Kerroch deposit. The nodule that occur inside a granitic country-rock is crosscut by several veinlets
1107 filled with sulfides. (c) Vein of pseudo-spherulitic U oxides inside a granitic country rock
1108 (Undifferentiated-Guern). (d) U oxide in a quartz vein from the Kerlech deposit. The first generation of
1109 spherulitic U oxide (Ur1) was breccified by quartz and a second generation of U oxide (Ur2). (e) Cluster
1110 of U oxide spherules occurring inside a granitic country rock (Rosglas deposit). (f) U oxides from the
1111 Quérien (Kerjean) deposit. Dark gray zones (alt Ur1) corresponds to the alteration of the first generation
1112 of U oxide. Red dashed ellipses represent the location of SIMS U-Pb analyses with the corresponding
1113 $^{207}\text{Pb}/^{206}\text{Pb}$ age. The white bar represents 100 μm .

1114

1115 Figure 5: Wetherill concordia diagrams displaying the analyses made on uranium oxides from the (a)
1116 Quistiave, (b) Kerroch, (c) undifferentiated-Guern, (d) Kerlech, (e) Rosglas, (f) Quérien uranium
1117 deposits. Ellipses and age errors are reported at 2σ .

1118

1119 Figure 6: Geochemical diagrams showing the whole-rock compositions of samples from the Pontivy-
1120 Rostrenen magmatic complex. (a) Q-P diagram (after Debon and Le Fort 1988) where the dashed fields
1121 delimit the location of common igneous rock: gr = granite, ad = adamellite (monzogranite), gd =
1122 granodiorite, to = tonalite, sq = quartz syenite, mzq = quartz monzonite, mzdq = quartz monzodiorite, s
1123 = syenite, mz = monzonite, mzgo = monzogabbro and go = gabbro. Q-P parameters are expressed in
1124 molar proportion multiplied by 1000. The grey arrows represent the compositional evolution of
1125 leucogranite during episyenitization. (b) U versus Th diagram where the pink arrow represents the
1126 theoretical evolution of a peraluminous leucogranitic melt during fractional crystallization and the red
1127 and green arrows represent the evolution of a sample during uranium oxide (UO_2) and monazite (Mnz)

1128 leaching, respectively. The sample compositions are from Ballouard et al. (2017b), Cotten (1975),
1129 Alabosi (1984), Euzen (1993), Bechennec et al. (2006, 2009) and Tartèse et al. (2012).

1130

1131 Figure 7: Selected images of apatite grains from magmatic and metasomatic rocks of the Pontivy-
1132 Rostrenen complex. (a) Cathodoluminescence (CL) images and chemical maps of Fe and Mn for apatite
1133 from leucogranite. (b) CL, backscattered electron (BSE) images and chemical map of As for apatite
1134 from an episyenite. (c) CL image and chemical map of Si for apatite from a monzogranite. (d) CL and
1135 BSE images for apatite from a quartz-monzodiorite. Numbers in yellow represent the associated ^{207}Pb
1136 corrected age whereas dashed circles and associated numbers represent the location of CL emission
1137 measurements (Fig. 8). The white bar represents 100 μm .

1138

1139 Figure 8: Cathodoluminescence emission spectra of representative apatite grains from (a) leucogranite,
1140 (b) episyenite, (c) monzogranite and (d) quartz monzodiorite of complex. The locations of analyses are
1141 indicated in Figure 7.

1142

1143 Figure 9: Fe versus Mn diagram displaying the EPMA analyses made on apatite grains from the
1144 granitoids of the complex. APFU = atoms per formula unit.

1145

1146 Figure 10: Tera-Wasserbug concordia diagrams with the corresponding ^{207}Pb corrected ages for LA-
1147 ICP-MS analyses made on apatite grains from (a) porphyritic leucogranite, (b-c) equigranular
1148 leucogranite, (d) late equigranular leucogranite, (e) episyenite, (f) monzogranite and (g) quartz
1149 monzodiorite of the complex. ^{207}Pb corrected ages are calculated using the common Pb composition at
1150 the lower intercept age (Stacey and Kramers 1975). Dashed ellipses in (e) represent analyses not used
1151 for ages calculations. Ellipses and errors on ages are reported at 2σ . The main period of U deposit
1152 formation in the complex is reported for comparison (green double arrow).

1153

1154 Figure 11: $^{235}\text{U}/^{207}\text{Pb}$ versus $^{206}\text{Pb}/^{207}\text{Pb}$ concordia diagram displaying the uranium oxide analyses from
1155 the uranium deposits of the complex and reporting the theoretical trajectories of uranium daughter loss
1156 (Ludwig and Simmons 1992).

1157

1158 Figure 12: Diagram reporting the U content of apatite from the granitoids of the complex as a function
1159 of the corresponding ^{207}Pb corrected age obtained by LA-ICP-MS.

1160

1161 Figure 13: Tera-Wasserbug concordia diagrams reporting LA-ICP-MS apatite analyses characterized by
1162 young ^{207}Pb corrected ages for the (a) porphyritic leucogranite, (b-c) equigranular leucogranite and (d)
1163 Late equigranular leucogranite (Fig. 10). The main period of U deposits formation in the complex is
1164 reported for comparison (grey double arrow). Ellipses and age errors are reported at 2σ .

1165

1166 Figure 14: Schematic bloc diagram summarizing the geodynamic context of U mineralization formation
1167 in the Armorican Variscan Belt from 300 to 270 Ma. A schematic cross section representing the
1168 topography at the end of the Variscan orogeny is represented in the background. NASZ: north Armorican
1169 shear zone; NBSASZ: northern branch of the south Armorican shear zone; SBSASZ: southern branch
1170 of the south Armorican shear zone; Grn: Guern; PP: Poulprio; PM: Prat Mérien; Krl: Kerlech (Lignol);
1171 Bnt: Bonote; Mn: Métairie-Neuve; Pnr: Pen Ar Ran. See text for details.

1172

1173 Table 1: Whole-rock major and trace elements composition of episyenite samples.

1174

1175 Table 2: Average chemical composition of apatite.

1176

1177 Table 3: Comparison between the different U-Pb ages obtained on zircon (Ballouard et al. 2017b) and
1178 apatite grains from samples of the Pontivy-Rostrenen complex.

Figure 1

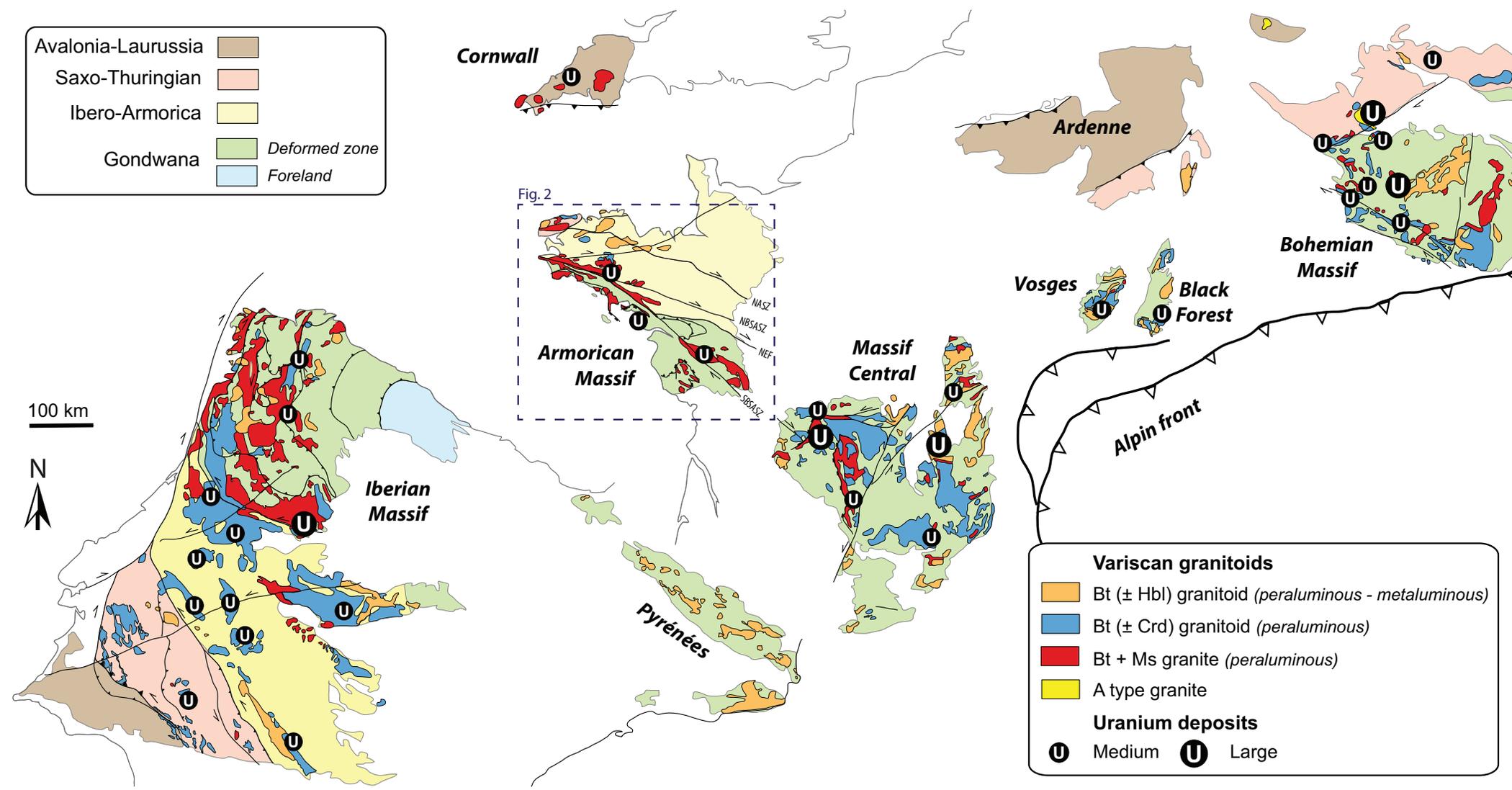


Figure 2

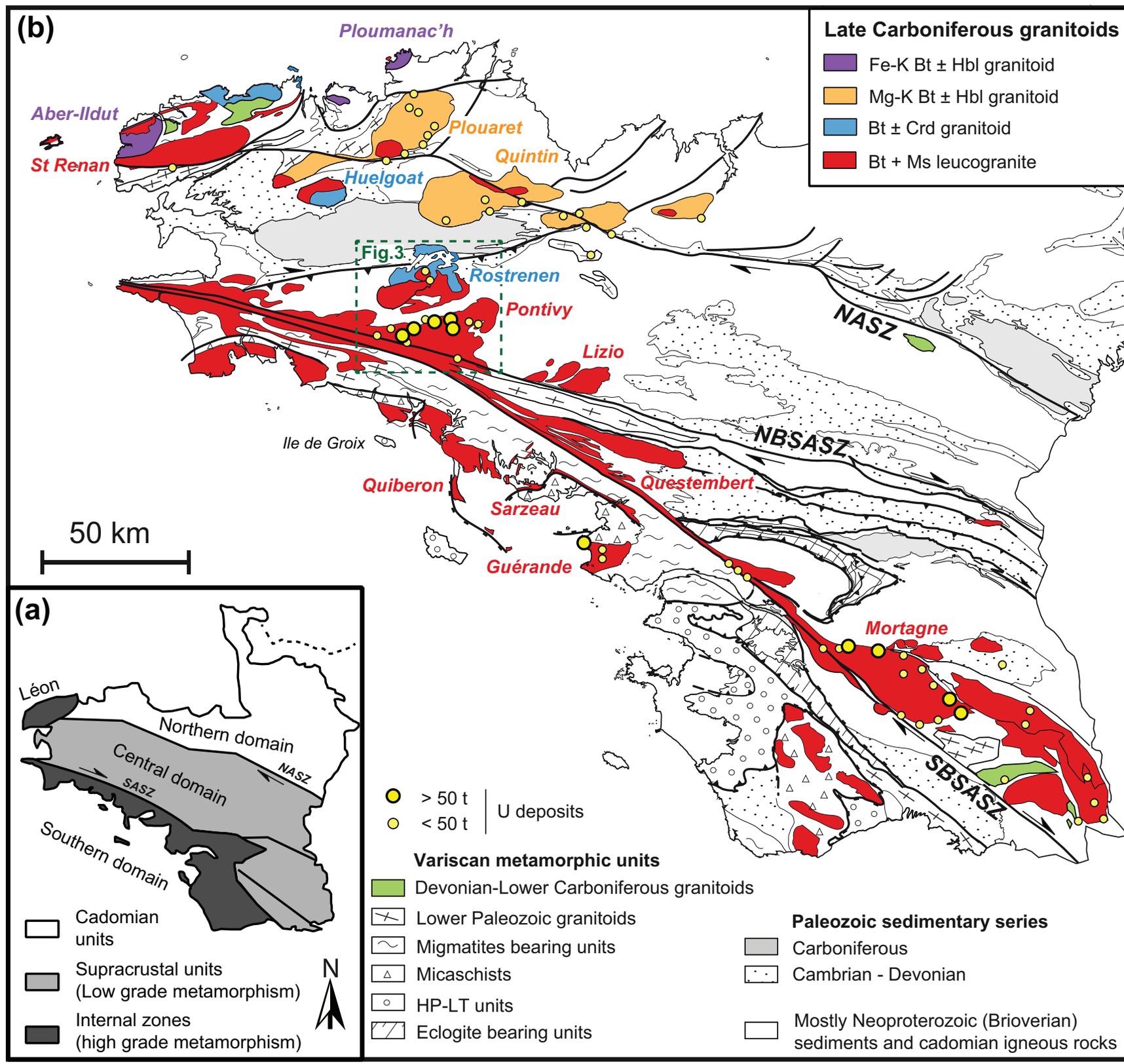
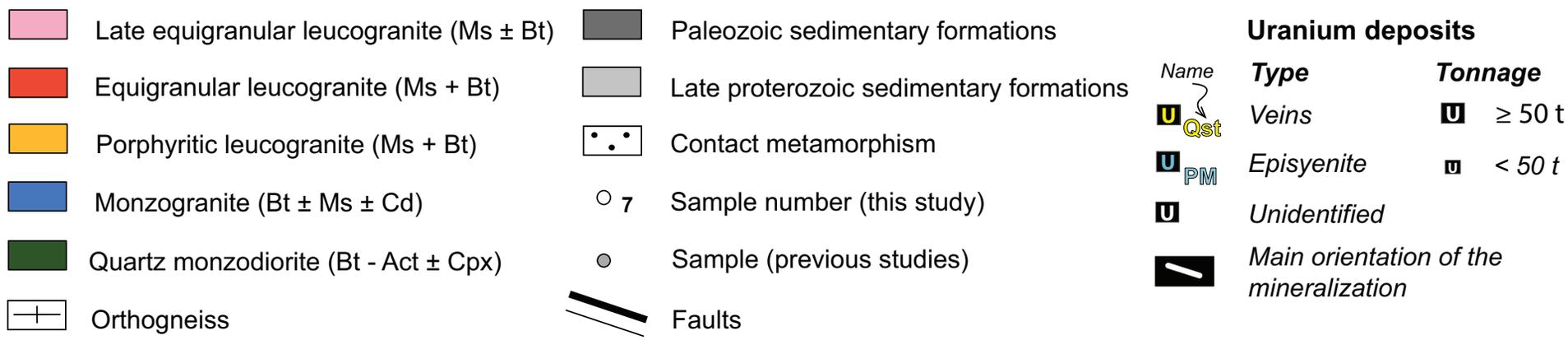
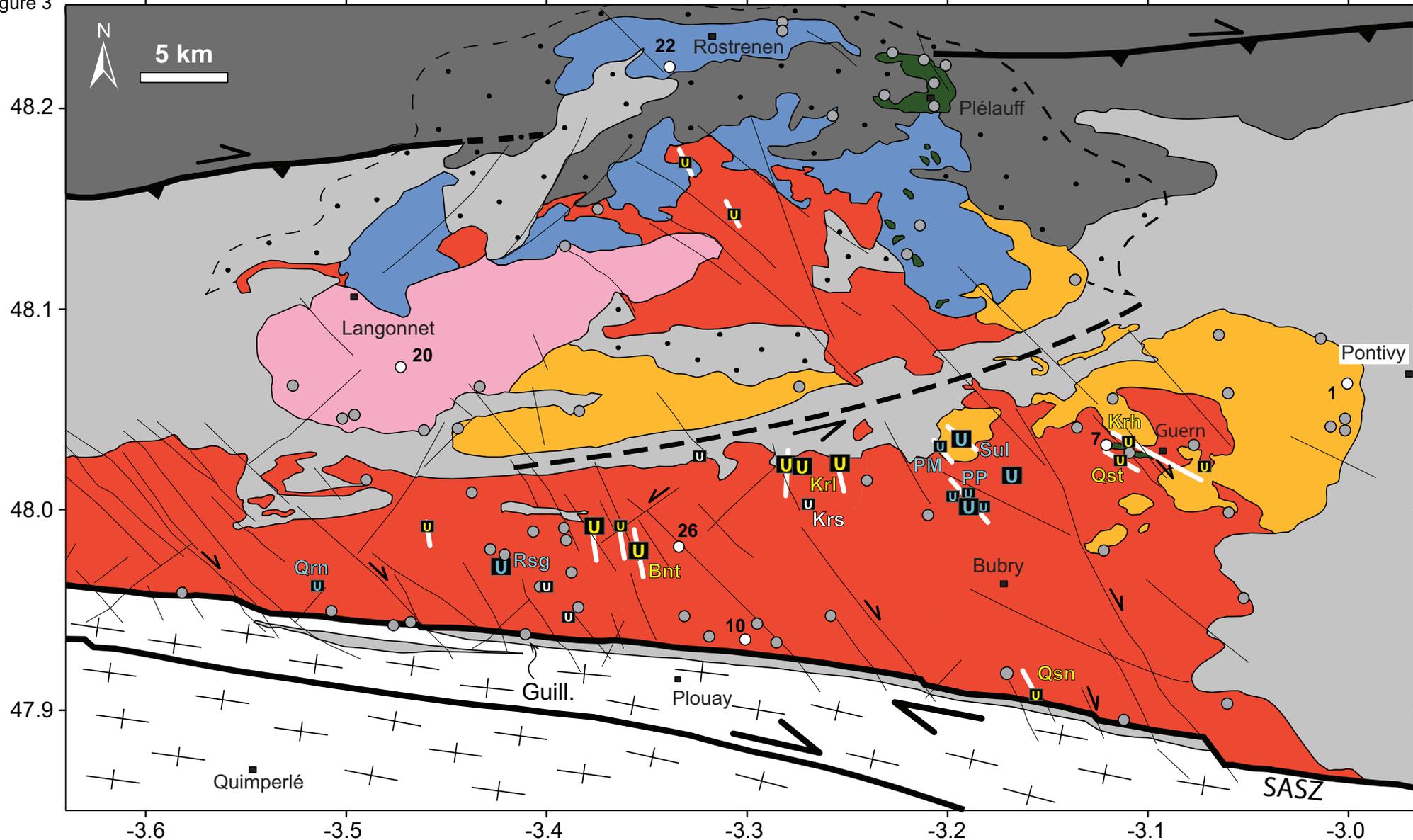
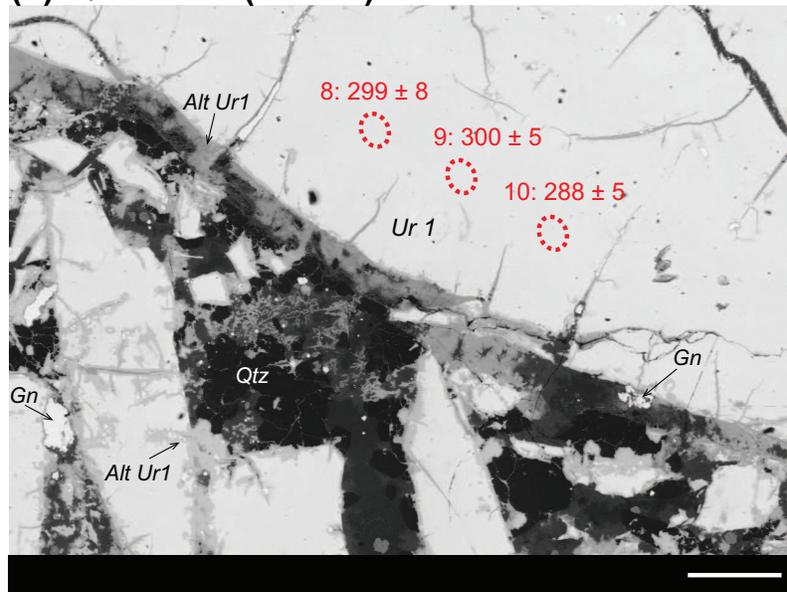


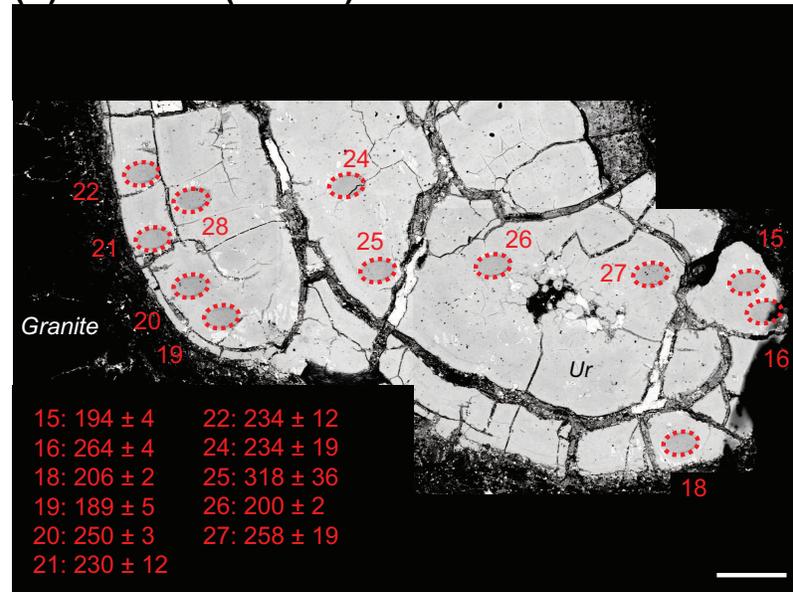
Figure 3



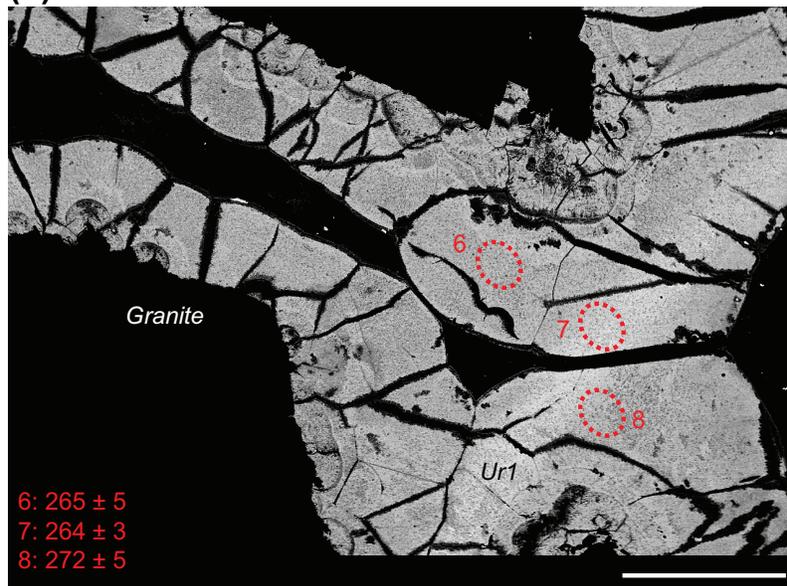
(a) Quistiave (Guern)



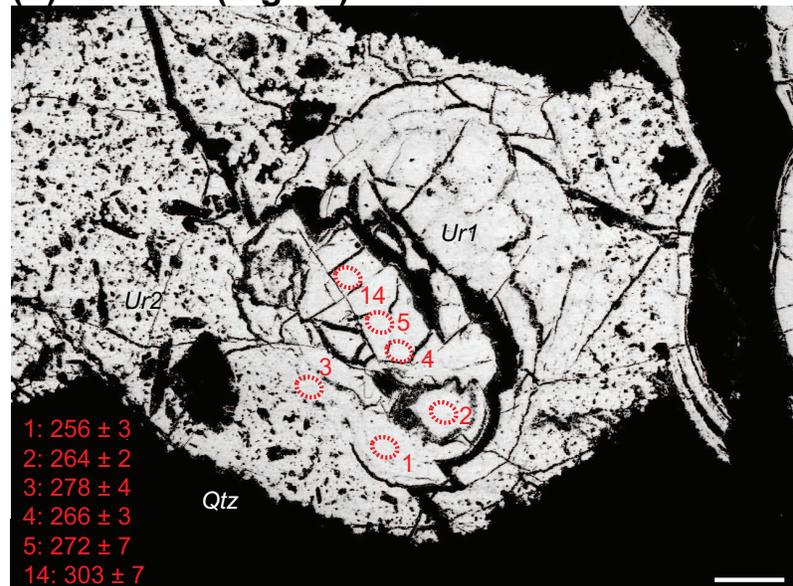
(b) Kerroch (Guern)



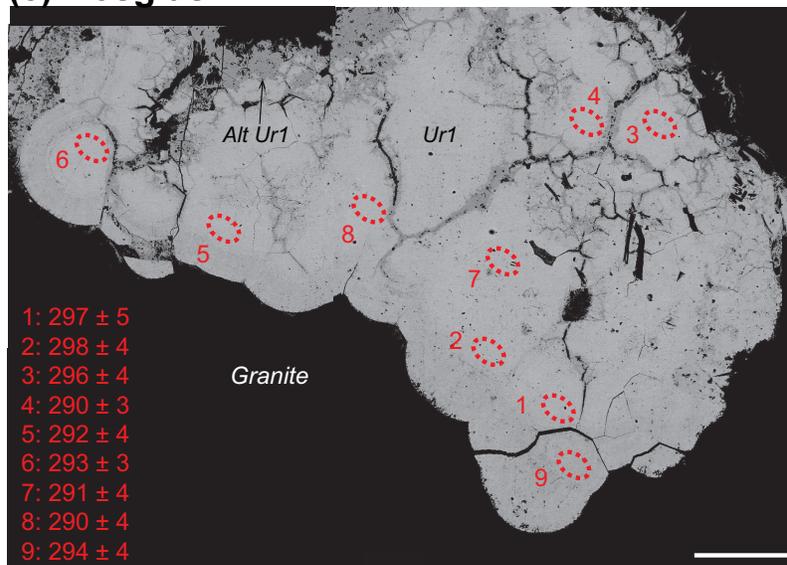
(c) Undifferentiated-Guern



(d) Kerlech (Lignol)



(e) Rosglas



(f) Qu errien (Kerjean)

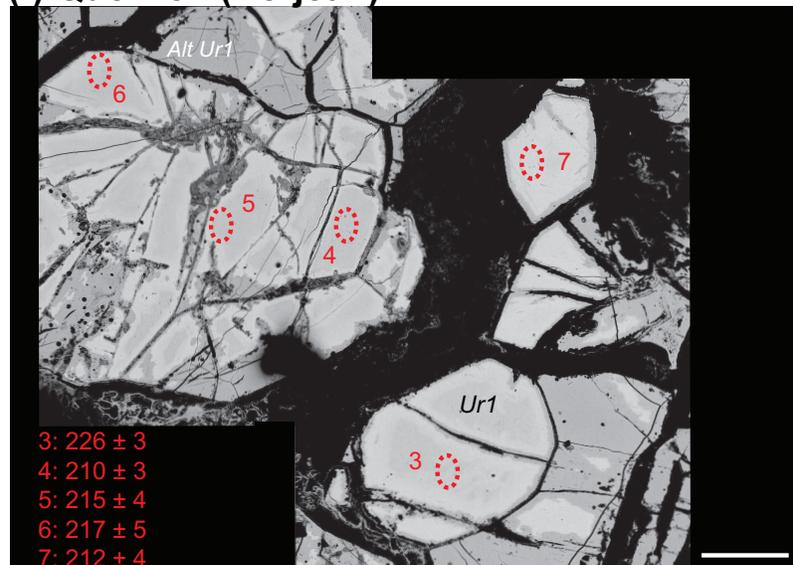


Figure 5

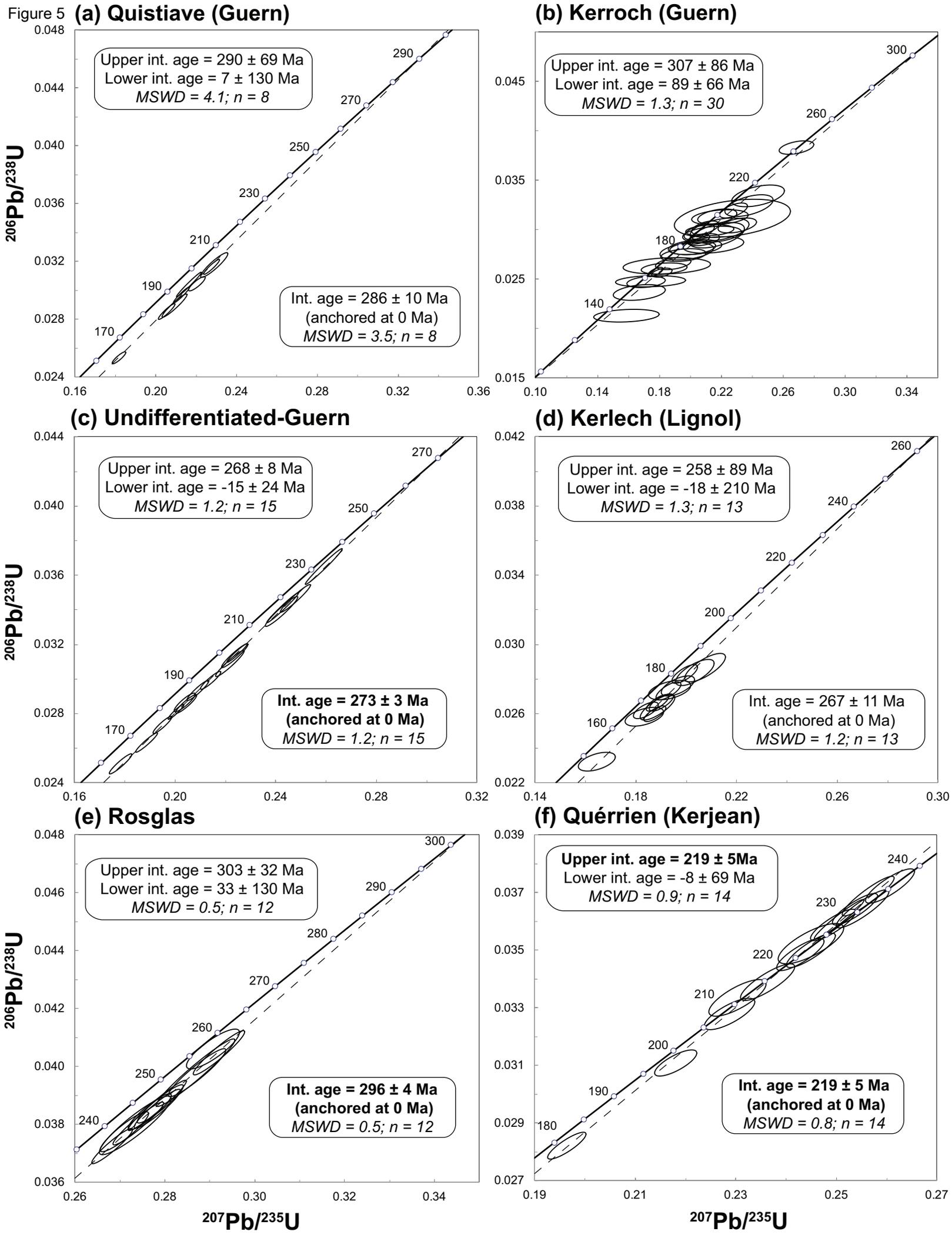


Figure 6

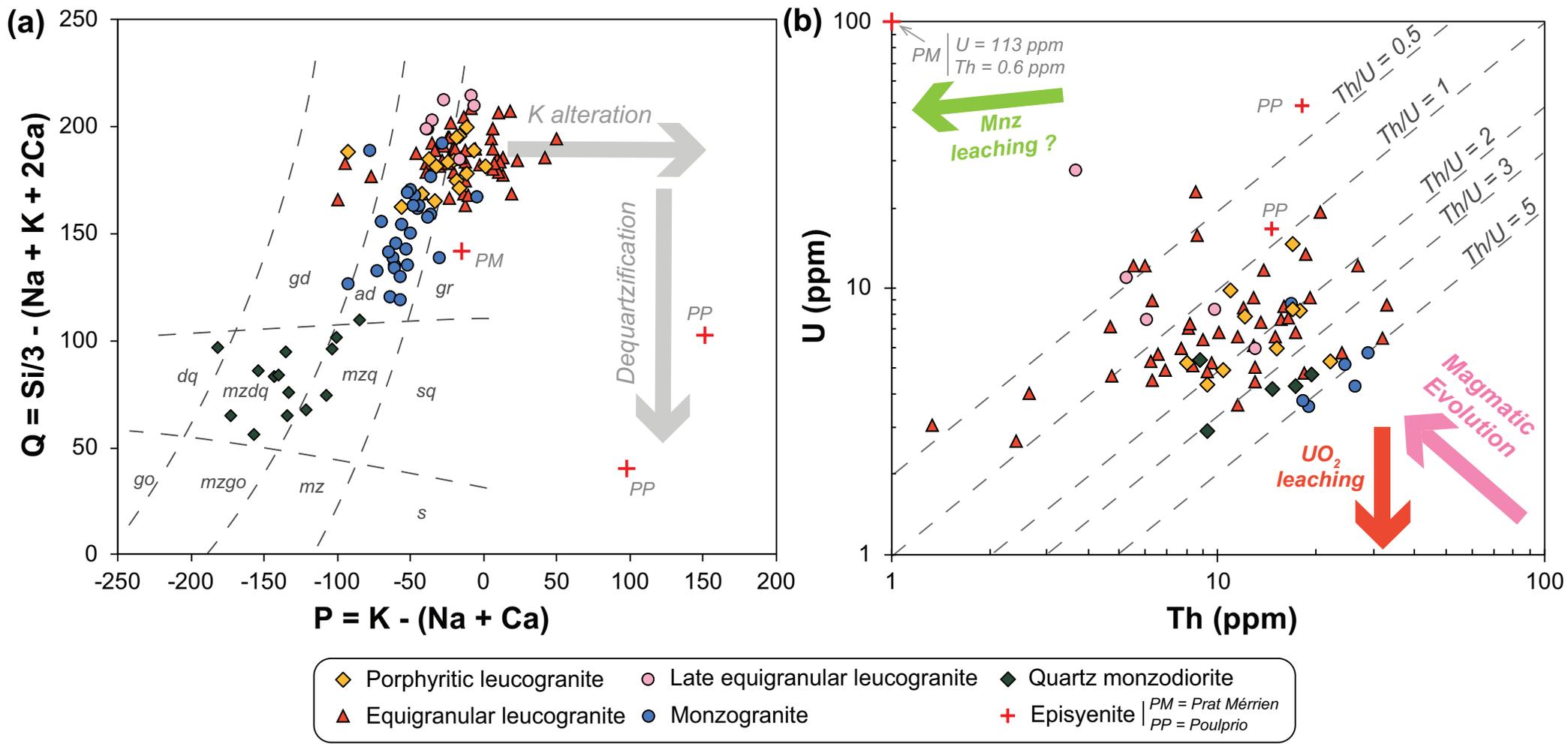


Figure 8

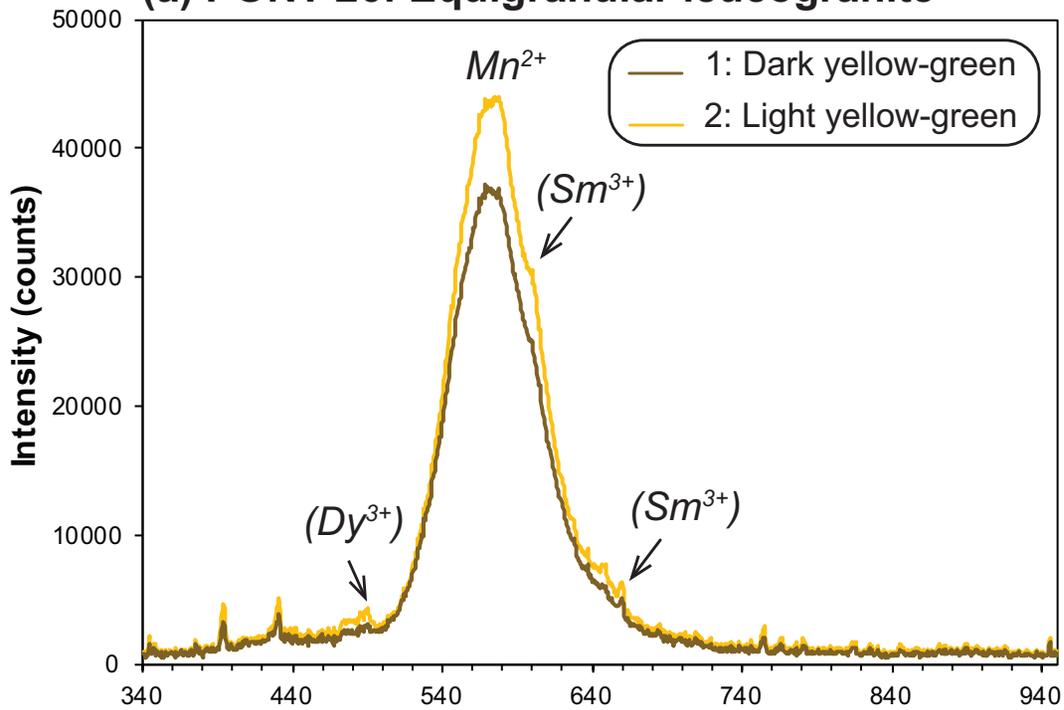
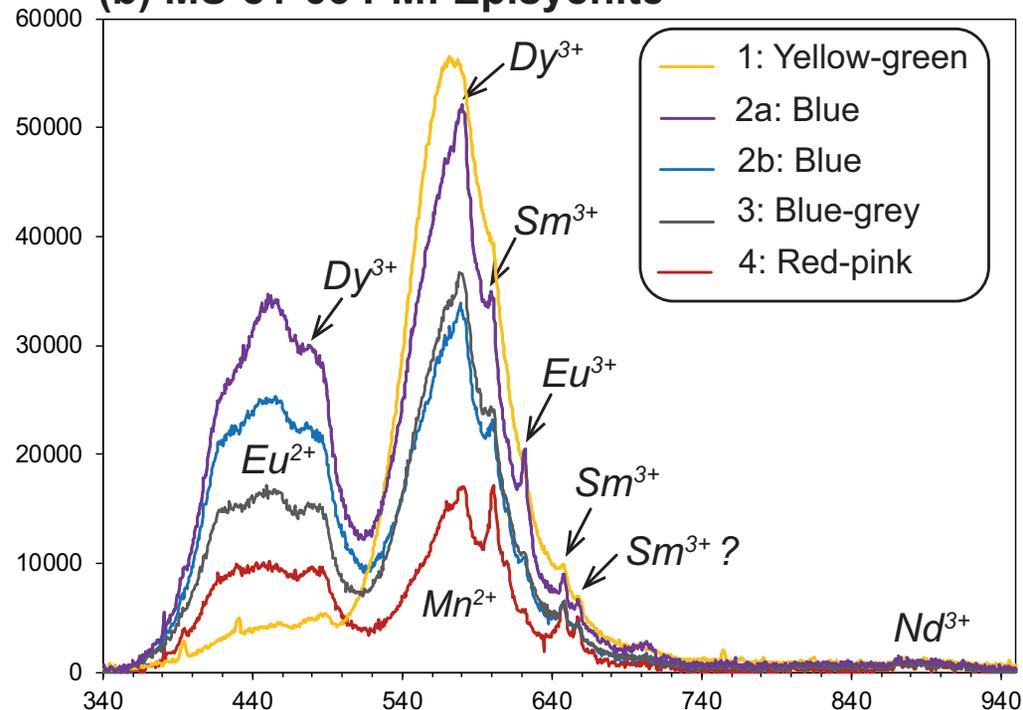
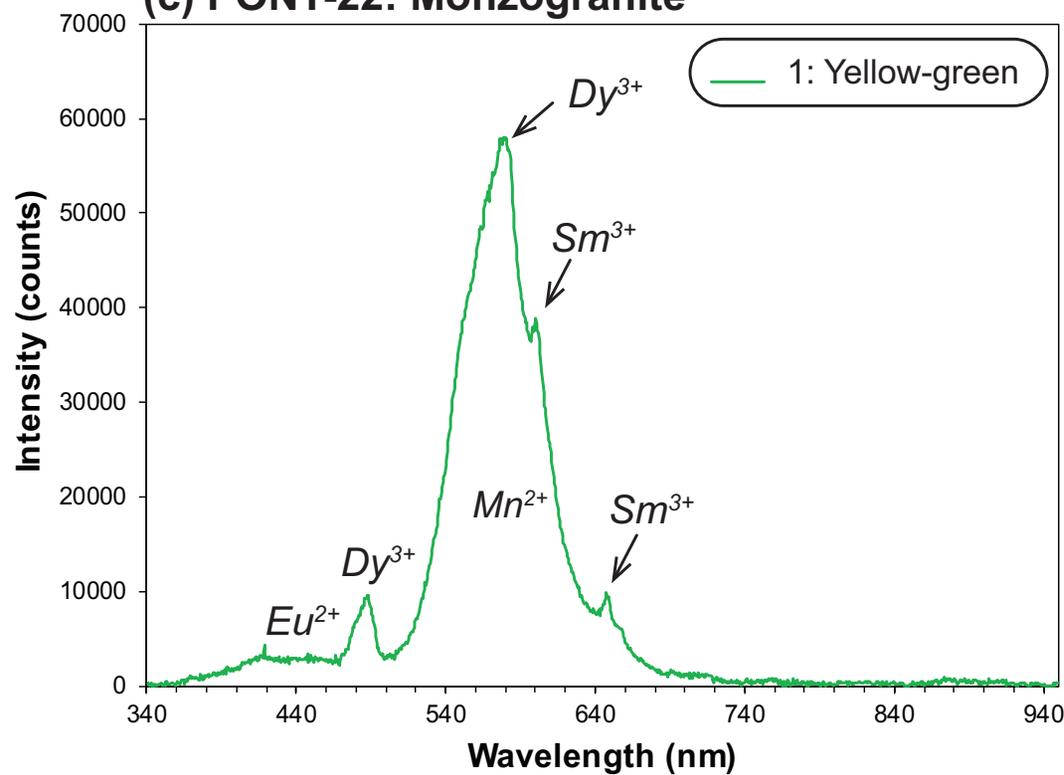
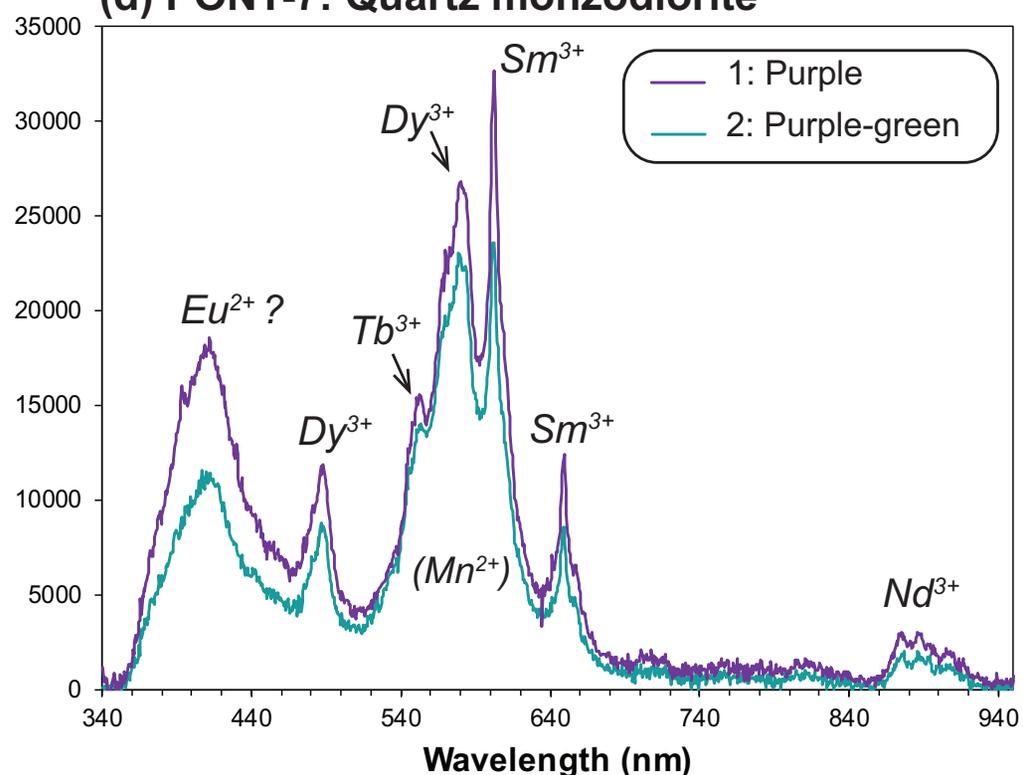
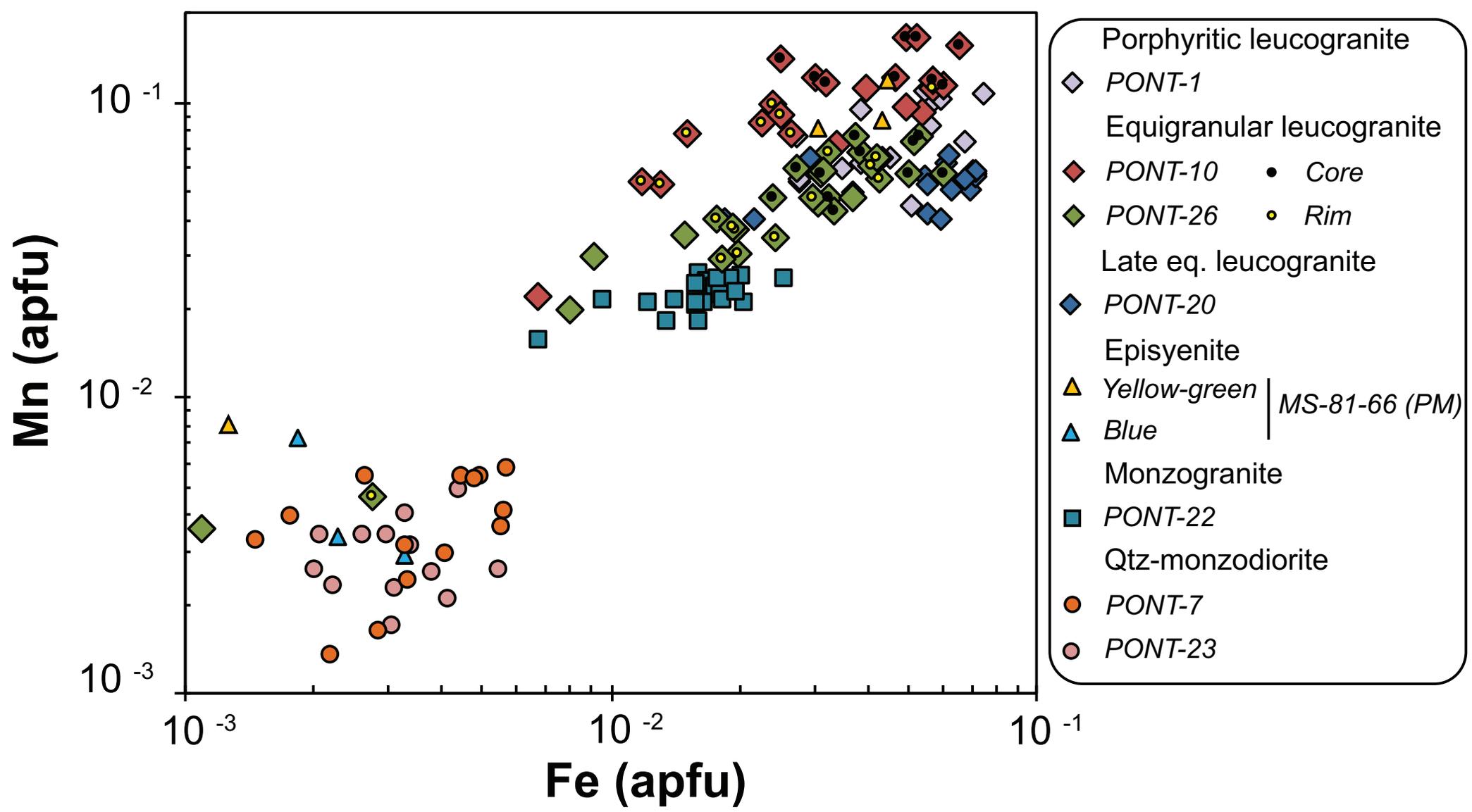
(a) PONT-26: Equigranular leucogranite**(b) MS-81-66 PM: Episyenite****(c) PONT-22: Monzogranite****(d) PONT-7: Quartz monzodiorite**

Figure 9



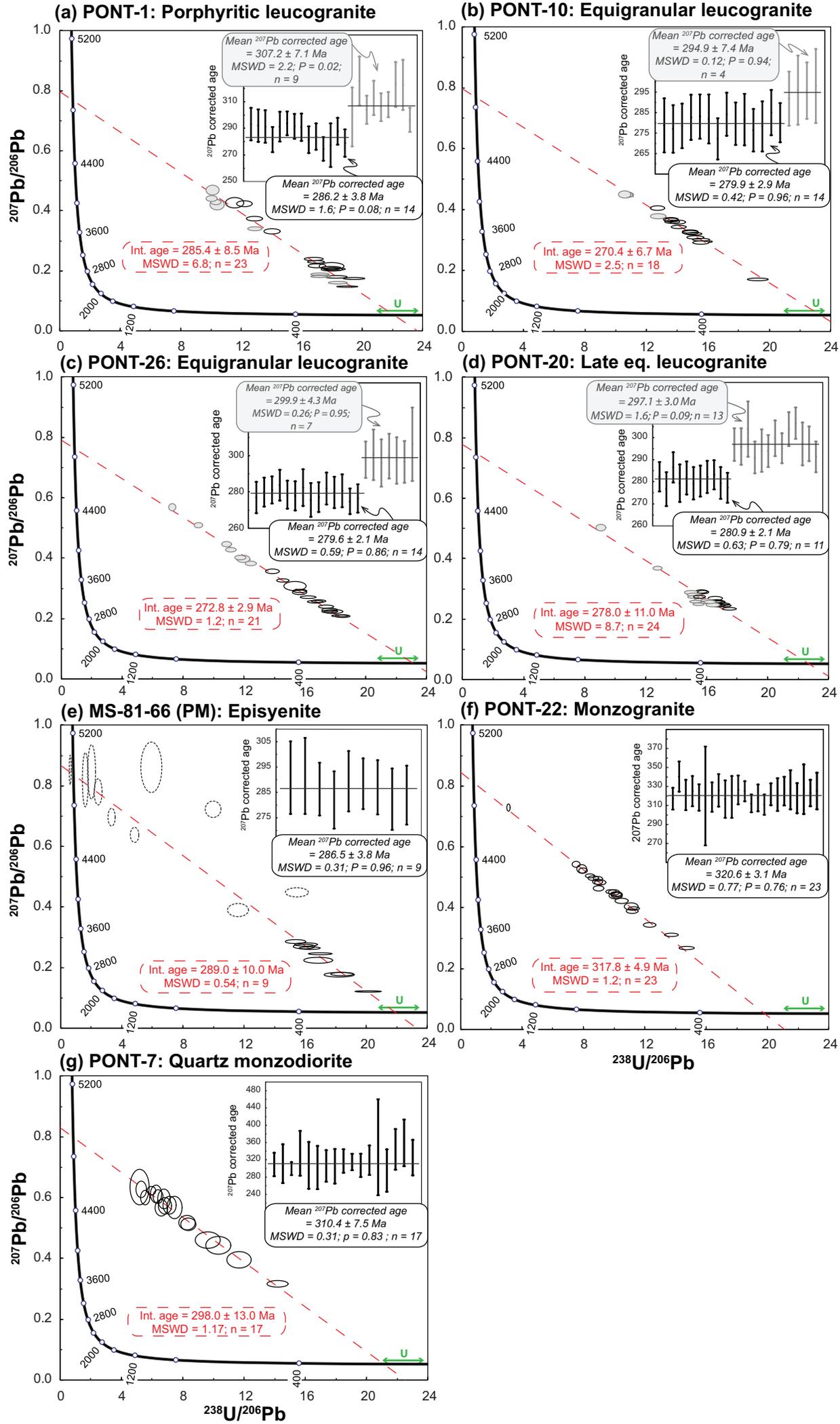


Figure 11

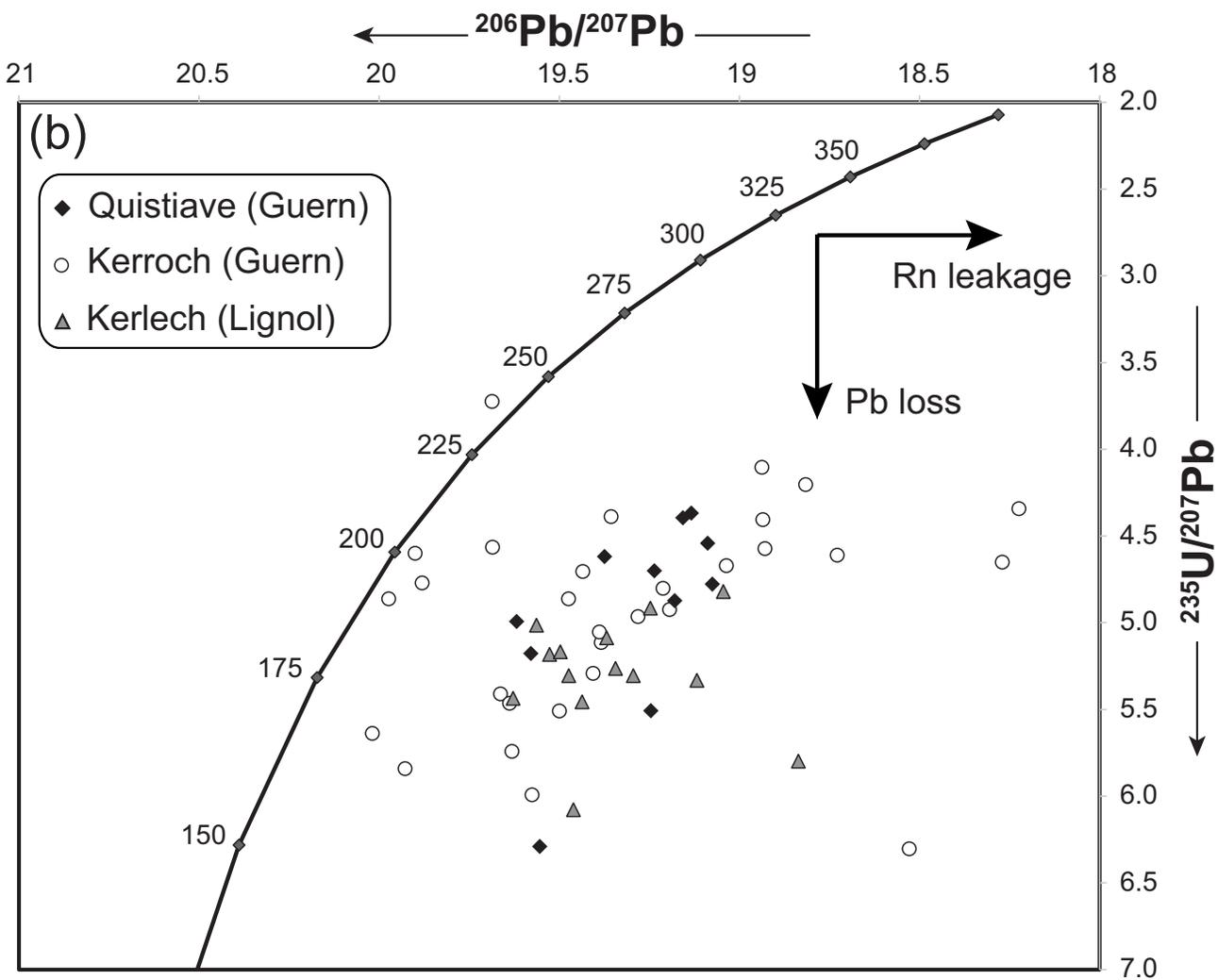
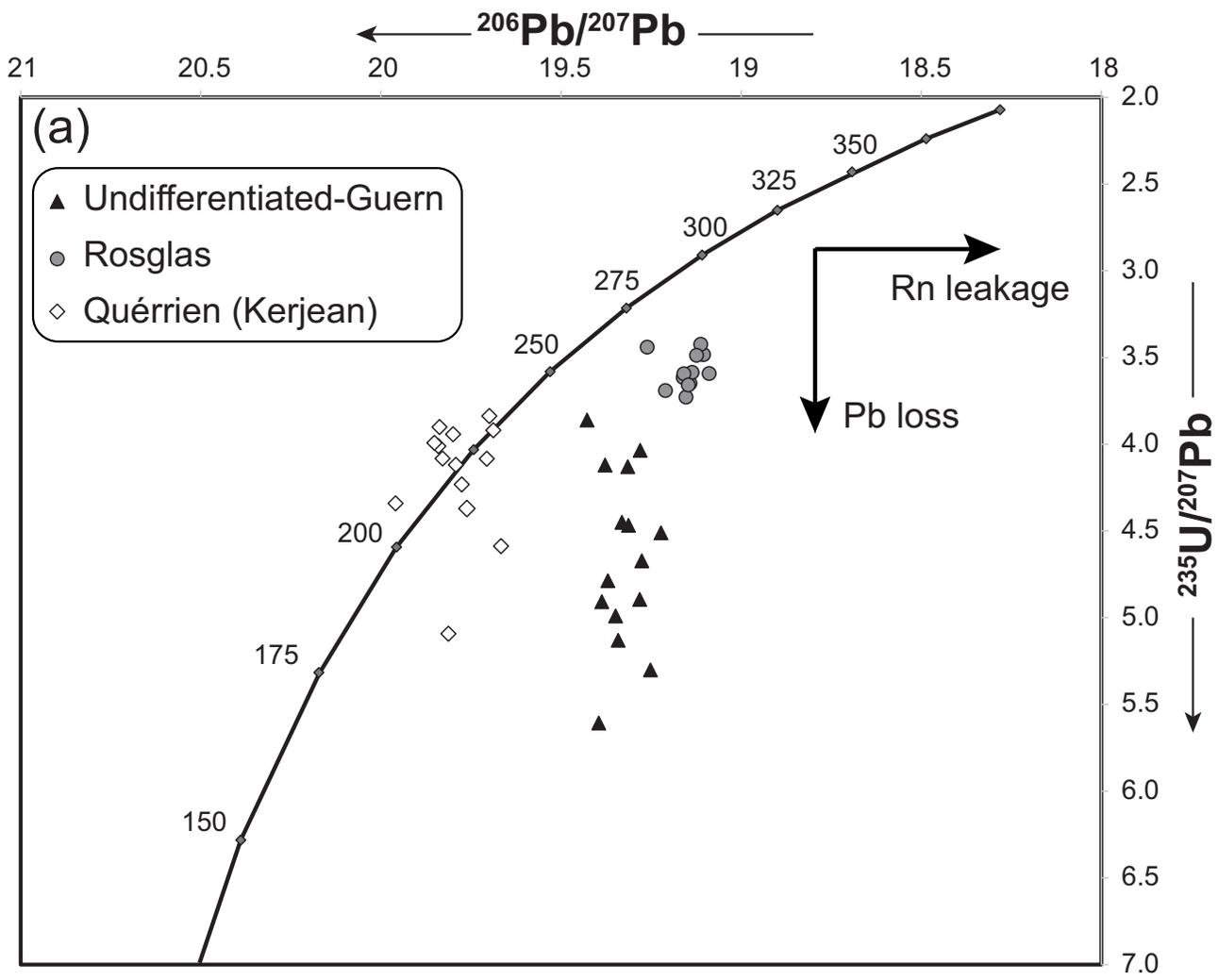
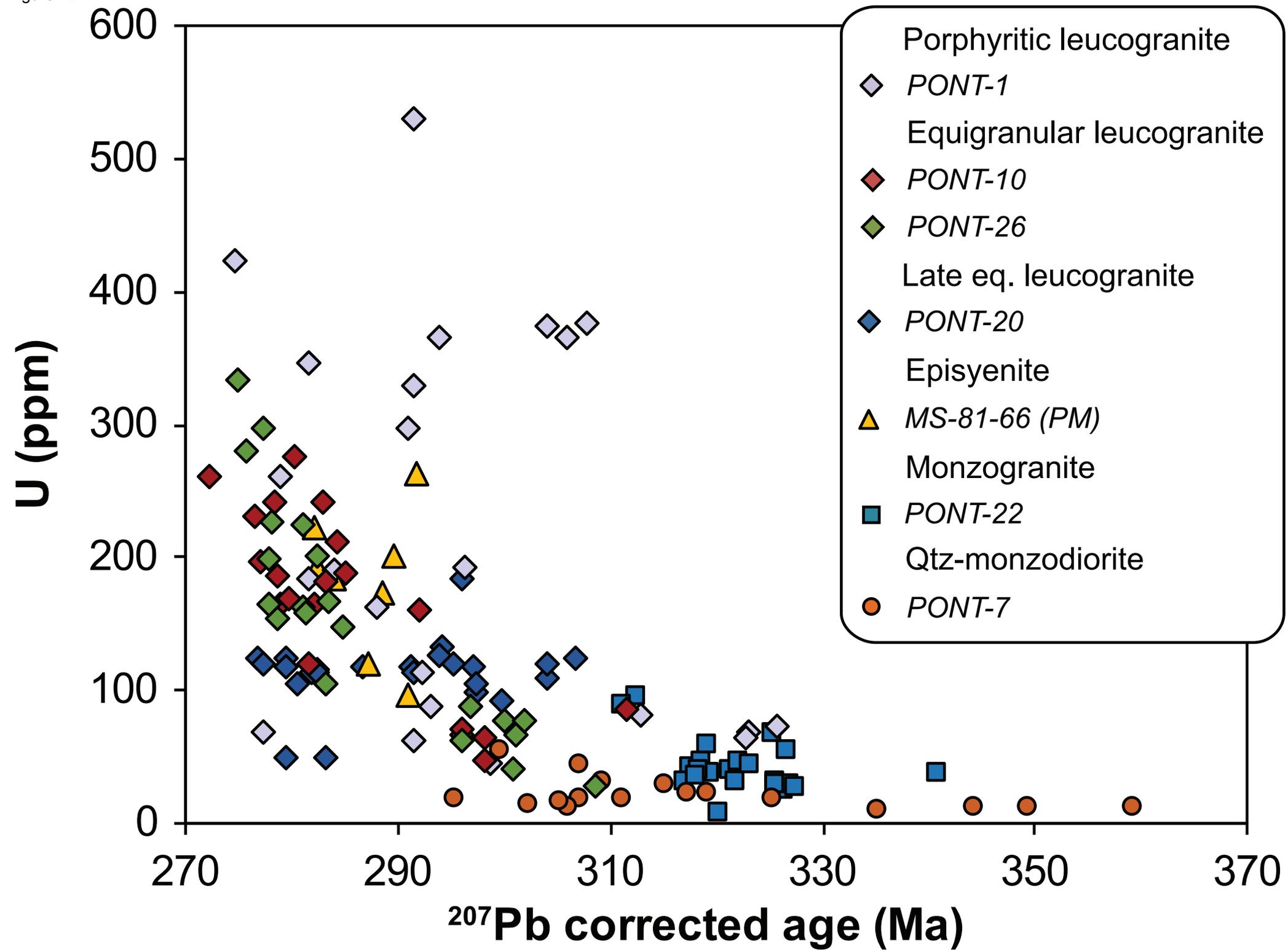


Figure 12



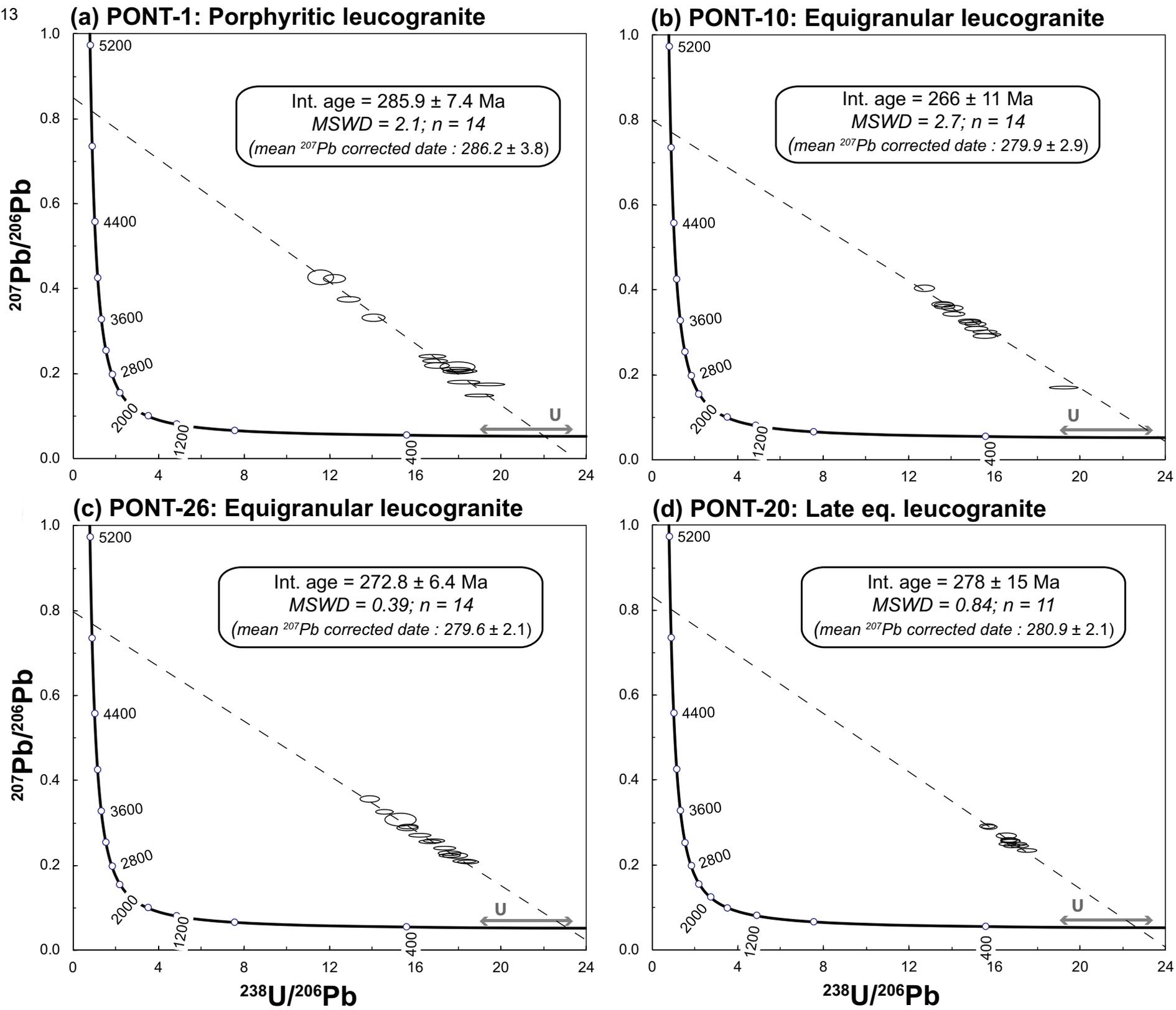


Figure 14

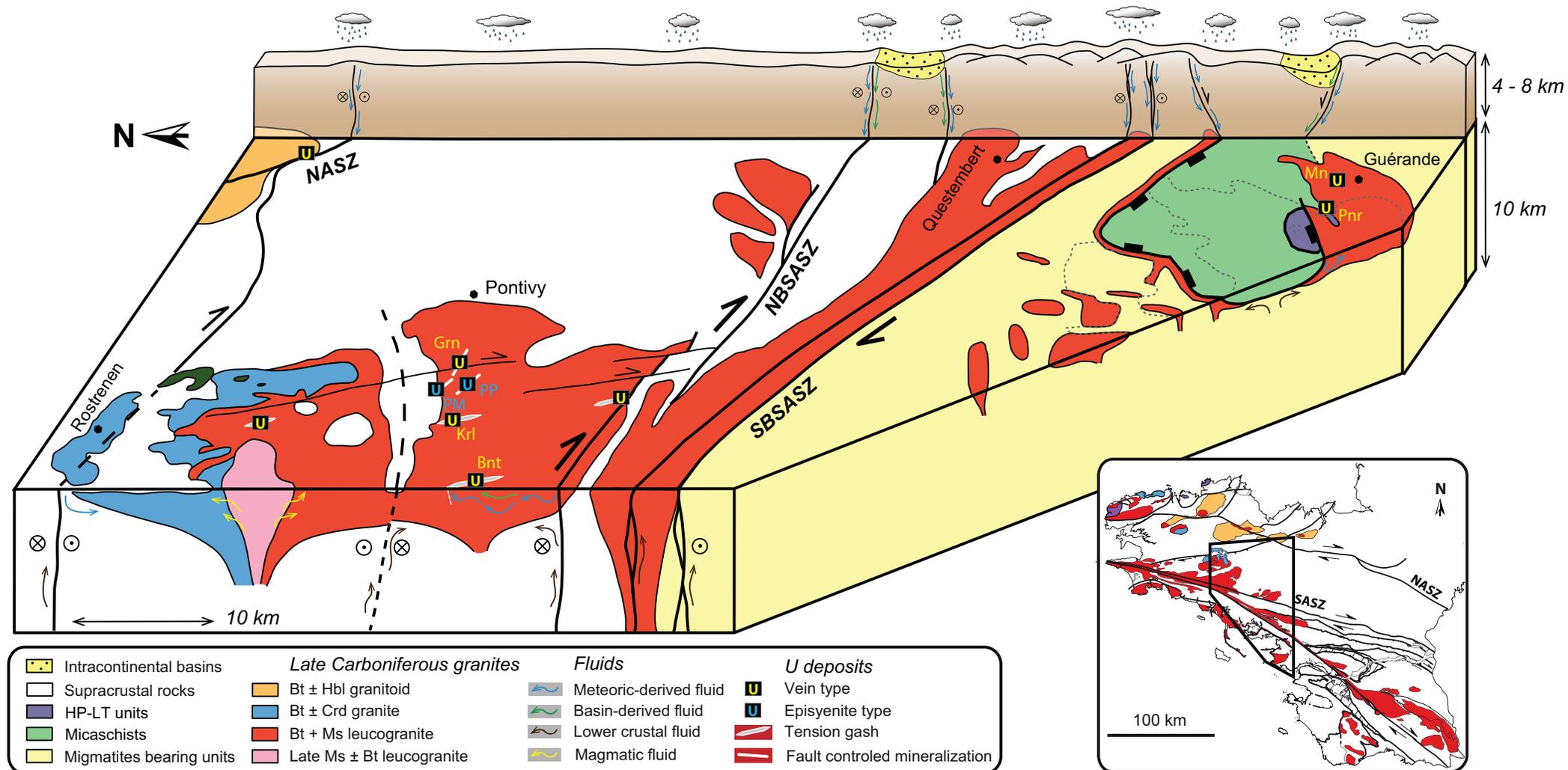


Table 1

Sample		MS-81-66	MS-81-32	MS-81-40
location		Prat-Mérien (PM)	Poulprio (PP)	
SiO ₂	wt.%	62.97	55.95	58.24
Al ₂ O ₃	wt.%	17.55	19.84	21.14
Fe ₂ O ₃	wt.%	4.40	3.01	1.64
MnO	wt.%	0.04	0.03	0.03
MgO	wt.%	1.00	1.85	1.18
CaO	wt.%	1.68	1.04	0.98
Na ₂ O	wt.%	2.67	0.39	2.42
K ₂ O	wt.%	4.77	8.59	9.08
TiO ₂	wt.%	0.13	0.43	0.39
P ₂ O ₅	wt.%	1.19	0.49	0.65
LOI	wt.%	4.13	8.23	4.08
Total	wt.%	100.52	99.85	99.82
Li	ppm	91	70	66
Cs	ppm	17.2	22.8	20.2
Rb	ppm	355	544	582
Sn	ppm	4.0	15.9	15.7
W	ppm	0.89	3.02	2.47
Ba	ppm	528	645	657
Sr	ppm	53.5	46.0	56.1
Be	ppm	8.1	34.4	8.2
U	ppm	113.20	48.42	16.53
Th	ppm	0.59	18.04	14.61
Nb	ppm	2.92	12.30	7.97
Ta	ppm	0.50	2.53	1.67
Zr	ppm	41.3	178.1	157.8
Hf	ppm	1.32	5.61	4.88
Bi	ppm	2.44	0.65	0.78
Cd	ppm	0.412	0.14	0.145
Co	ppm	7.05	1.48	3.25
Cr	ppm	14.72	9.441	8.305
Cu	ppm	25.55	bdl	7.167
Ga	ppm	24.3	30.7	32.4
Ge	ppm	1.18	1.37	1.20
In	ppm	bdl	0.523	0.139
Mo	ppm	bdl	bdl	bdl
Ni	ppm	15.54	bdl	bdl
Pb	ppm	52.1	30.5	25.4
Sc	ppm	3.36	3.71	2.94
Sb	ppm	2.403	2.229	5.07
V	ppm	22.0	22.5	16.0
Y	ppm	44.34	10.91	14.10
Zn	ppm	29.43	31.29	53.36
As	ppm	95.15	20.08	54.01
La	ppm	8.13	30.40	27.24
Ce	ppm	23.90	60.93	58.68
Pr	ppm	3.94	7.47	7.18
Nd	ppm	18.02	28.40	28.45
Sm	ppm	5.77	6.85	7.61
Eu	ppm	1.11	0.83	1.12
Gd	ppm	5.89	5.42	6.31
Tb	ppm	1.14	0.68	0.83
Dy	ppm	7.79	2.92	3.59
Ho	ppm	1.65	0.41	0.50
Er	ppm	4.45	0.84	1.00
Tm	ppm	0.66	0.11	0.13
Yb	ppm	4.35	0.68	0.77
Lu	ppm	0.61	0.10	0.11
A/NK		1.84	1.99	1.53
A/CNK		1.39	1.67	1.35

LOI: Loss on ignition; bdl: below detection limit

Table 2

Table 2: Average chemical composition of apatite.

Facies	Por. leucogranite		Equigranular leucogranite											
Sample Location Color CL	PONT-1		PONT-10						PONT-26					
	Yellow-green		Core Yellow-green		Rim Yellow-green		Unzoned Yellow-green		Core Yellow-green		Rim Yellow-green		Unzoned Yellow-green	
Analyses	n = 15	σ	n = 9	σ	n = 8	σ	n = 5	σ	n = 11	σ	n = 13	σ	n = 7	σ
CaO	52.42	0.71	51.97	0.44	53.17	0.55	53.16	0.91	53.21	0.39	54.19	0.73	53.95	0.45
SrO	bdl		bdl		bdl		bdl		bdl		bdl		bdl	
FeO	0.65	0.21	0.65	0.20	0.34	0.20	0.51	0.26	0.56	0.16	0.34	0.19	0.26	0.19
MnO	1.08	0.30	1.89	0.30	1.13	0.28	1.10	0.49	0.84	0.16	0.55	0.29	0.48	0.26
Na ₂ O	0.10	0.05	0.06	0.03	0.04	0.03	0.03	0.03	0.09	0.04	0.06	0.04	0.04	0.03
P ₂ O ₅	41.74	0.44	41.78	0.39	41.64	0.43	41.70	0.19	41.77	0.24	41.99	0.37	42.02	0.34
SiO ₂	0.01	0.01	0.02	0.01	0.01	0.01	0.02	0.02	0.01	0.02	0.02	0.02	0.03	0.02
SO ₂	0.01	0.02	0.02	0.02	0.01	0.01	0.02	0.02	0.01	0.01	0.01	0.02	0.02	0.02
As ₂ O ₃	bdl		0.01	0.01	0.01	0.01	0.01	0.02	bdl		0.01	0.01	0.01	0.01
Ce ₂ O ₃	0.07	0.05	0.09	0.05	0.07	0.04	0.05	0.04	0.06	0.04	0.03	0.03	0.06	0.05
La ₂ O ₃	0.03	0.03	0.02	0.02	0.01	0.01	0.02	0.04	0.01	0.02	0.02	0.02	0.02	0.02
Cl	0.012	0.007	0.003	0.003	0.008	0.006	0.008	0.003	0.004	0.003	0.005	0.005	0.002	0.003
F	3.316	0.101	3.311	0.107	3.318	0.066	3.235	0.123	3.286	0.093	3.342	0.095	3.385	0.108
Total	99.46	0.84	99.83	0.51	99.76	0.57	99.89	0.42	99.85	0.26	100.56	0.45	100.29	0.37
O=F	1.40	0.04	1.39	0.04	1.40	0.03	1.36	0.05	1.38	0.04	1.41	0.04	1.43	0.05
O=Cl	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Total*	98.06	0.81	98.43	0.49	98.36	0.58	98.52	0.41	98.47	0.25	99.15	0.44	98.86	0.36
Structural formula on the basis of a 12.5 oxygen equivalent														
Ca	4.80	0.05	4.75	0.04	4.86	0.05	4.85	0.08	4.86	0.03	4.91	0.06	4.90	0.03
Sr	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Fe	0.05	0.02	0.05	0.01	0.02	0.01	0.04	0.02	0.04	0.01	0.02	0.01	0.02	0.01
Mn	0.08	0.02	0.14	0.02	0.08	0.02	0.08	0.03	0.06	0.01	0.04	0.02	0.03	0.02
Na	0.02	0.01	0.01	0.00	0.01	0.00	0.00	0.00	0.02	0.01	0.01	0.01	0.01	0.00
P	3.02	0.02	3.02	0.01	3.01	0.01	3.01	0.01	3.01	0.01	3.01	0.02	3.01	0.01
Si	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
S	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
As	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ce	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
La	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
F	0.90	0.02	0.89	0.03	0.90	0.02	0.87	0.03	0.89	0.02	0.89	0.03	0.91	0.03
OH ^a	0.10	0.02	0.11	0.03	0.10	0.02	0.13	0.03	0.11	0.02	0.11	0.03	0.09	0.03

Facies	Late eq. leucogranite		Episyenite						Monzogranite		Quartz monzodiorite			
Sample Location	PONT-20		MS-81-66 (PM)						PONT-22		PONT-7		PONT-23	
Color CL	Yellow-green		Blue		Red-pink		Yellow-green		Yellow-green		Purple		Purple	
Analyses	n = 17	σ	n = 11	σ	n = 4	σ	n = 4	σ	n = 23	σ	n = 14	σ	n = 15	σ
CaO	52.41	0.67	54.47	0.30	52.42	0.67	53.03	1.36	53.21	0.63	53.62	0.51	53.87	0.41
SrO	bdl		bdl		0.08	0.01	bdl		bdl		bdl		0.01	0.01
FeO	0.77	0.24	0.01	0.02	0.05	0.03	0.42	0.28	0.23	0.05	0.05	0.02	0.04	0.02
MnO	0.75	0.12	0.03	0.03	bdl		1.04	0.66	0.31	0.04	0.05	0.02	0.04	0.02
Na ₂ O	0.12	0.02	0.01	0.01	0.02	0.02	0.08	0.06	0.12	0.02	0.02	0.01	0.02	0.02
P ₂ O ₅	42.61	0.29	42.79	0.38	37.15	0.43	42.64	0.37	42.04	0.50	41.63	0.40	41.59	0.59
SiO ₂	0.01	0.01	0.02	0.02	0.02	0.02	0.01	0.02	0.05	0.06	0.48	0.29	0.22	0.13
SO ₂	0.01	0.02	0.01	0.02	0.01	0.02	0.00	0.00	0.01	0.01	0.01	0.02	0.02	0.02
As ₂ O ₃	bdl		0.11	0.36	5.17	0.49	0.02	0.02	bdl		0.01	0.01	bdl	
Ce ₂ O ₃	0.09	0.05	0.03	0.04	0.08	0.04	0.04	0.03	0.03	0.03	0.24	0.15	0.19	0.12
La ₂ O ₃	0.03	0.02	0.01	0.01	0.04	0.04	0.03	0.03	0.01	0.01	0.06	0.05	0.07	0.06
Cl	0.011	0.004	0.005	0.005	0.007	0.010	0.078	0.054	0.030	0.009	0.055	0.014	0.090	0.030
F	3.323	0.107	3.377	0.136	2.784	0.114	3.305	0.126	3.395	0.097	3.332	0.094	3.430	0.090
Total	100.14	0.59	100.89	0.37	97.85	1.01	100.68	0.77	99.45	0.71	99.57	0.59	99.58	0.68
O=F	1.40	0.05	1.42	0.06	1.17	0.05	1.39	0.05	1.43	0.04	1.40	0.04	1.44	0.04
O=Cl	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.01	0.01	0.00	0.01	0.00	0.02	0.01
Total*	98.74	0.58	99.46	0.36	96.68	1.00	99.27	0.75	98.01	0.70	98.15	0.58	98.12	0.65
Structural formula on the basis of a 12.5 oxygen equivalent														
Ca	4.75	0.05	4.89	0.02	5.02	0.02	4.78	0.08	4.86	0.05	4.89	0.06	4.93	0.03
Sr	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Fe	0.05	0.02	0.00	0.00	0.00	0.00	0.03	0.02	0.02	0.00	0.00	0.00	0.00	0.00
Mn	0.05	0.01	0.00	0.00	0.00	0.00	0.07	0.05	0.02	0.00	0.00	0.00	0.00	0.00
Na	0.02	0.00	0.00	0.00	0.00	0.00	0.01	0.01	0.02	0.00	0.00	0.00	0.00	0.00
P	3.05	0.01	3.04	0.02	2.81	0.02	3.04	0.01	3.03	0.02	3.00	0.02	3.00	0.02
Si	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.04	0.02	0.02	0.01
S	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
As	0.00	0.00	0.01	0.02	0.28	0.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ce	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.01	0.00
La	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.01	0.00	0.00	0.01	0.00	0.01	0.00
F	0.89	0.03	0.90	0.03	0.79	0.03	0.88	0.03	0.91	0.03	0.90	0.03	0.93	0.02
OH ^a	0.11	0.03	0.10	0.03	0.21	0.03	0.11	0.03	0.08	0.03	0.09	0.03	0.06	0.02

Notes: oxide in wt.%, cationic contents in apfu. ^a: calculated OH cationic content. bdl: below detection limit; Por. Leucogranite: porphyritic leucogranite; Late eq. leucogranite: late equigranular leucogranite.

Table 3

Sample	Emplacement age (U-Pb zircon)	Unforced discordia dates	Age used for common Pb	²⁰⁷Pb corrected dates
Porphyritic leucogranite (PONT-1)	316.7 ± 2.5 Ma (MSWD = 1.2)	285.4 ± 8.5 Ma (MSWD=6.8)	285 Ma	307.2 ± 7.1 Ma 286.2 ± 3.8 Ma
Equigranular leucogranite (PONT-10)	310.3 ± 4.7 Ma (MSWD = 2.5)	270.4 ± 6.7 Ma (MSWD=2.5)	270 Ma	294.9 ± 7.4 Ma 279.9 ± 2.9 Ma
Equigranular leucogranite (PONT-26)		272.8 ± 2.9 Ma (MSWD = 1.2)	275 Ma	299.9 ± 4.3 Ma 279.6 ± 2.1 Ma
Late eq. leucogranite (PONT-20)	304.7 ± 2.7 Ma (MSWD = 0.57)	278 ± 11 Ma (MSWD = 8.7)	280 Ma	297.1 ± 3 Ma 280.9 ± 2.1 Ma
Episyenite (MS-81-66-PM)		289 ± 10 Ma (MSWD = 0.54)	290	286.5 ± 3.8 Ma
Monzogranite (PONT-22)	315.5 ± 2.0 Ma (MSWD = 1.5)	317.8 ± 4.9 Ma (MSWD = 1.2)	315	320.6 ± 3.1 Ma
Monzodiorite Quartzique (PONT-7)	315.2 ± 2.9 Ma (MSWD = 0.94)	298 ± 13 Ma (MSWD = 1.17)	315	310.4 ± 7.5 Ma

Note: Late eq. leucogranite: late equigranular leucogranite