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2 **Helium trapping in apatite damage: insights from (U-Th-Sm)/He**  
3 **dating of different granitoid lithologies**

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7 Alice Recanati<sup>1\*</sup>, Cécile Gautheron<sup>1</sup>, Jocelyn Barbarand<sup>1</sup>, Yves Missenard<sup>1</sup>,  
8 Rosella Pinna-Jamme<sup>1</sup>, Laurent Tassan-Got<sup>2</sup>, Andy Carter<sup>3</sup>, Eric Douville<sup>4</sup>,  
9 Louise Bordier<sup>4</sup>, Maurice Pagel<sup>1</sup>, Kerry Gallagher<sup>5</sup>

10  
11  
12 <sup>1</sup> GEOPS, Univ Paris Sud, CNRS, Université Paris-Saclay, Rue du Belvédère, Bât. 504,  
13 Orsay, F-91405, France

14 <sup>2</sup> Institut de Physique Nucleaire, Université Paris Sud, CNRS/IN2P3, 91405 Orsay, France,

15 <sup>3</sup> Dept. of Earth and Planetary Sciences, Birkbeck, University of London, UK

16 <sup>4</sup> Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL), CEA- CNRS-  
17 UVSQ, Université Paris-saclay, F-91191 Gif sur Yvette, France

18 <sup>5</sup> Géosciences Rennes, Université Rennes 1, Rennes, F-35042, France

19  
20 \* corresponding author

21 alice.recanati@u-psud.fr

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24 9 Figures

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35 **Abstract.** Apatite (U-Th-Sm)/He (AHe) thermochronometry is widely used to constrain  
36 thermal histories and rates of tectonic, exhumation, and erosion processes. However, data  
37 interpretation is often challenging, especially when the thermal history includes extended  
38 residence time in the He partial retention zone (HePRZ), with highly dispersed dates revealing  
39 the complexity of diffusion processes in natural systems. This study investigates chemical and  
40 physical factors that may have impacted He diffusion in apatite over long timescales in a  
41 context of protracted residence in the HePRZ. Nine samples from the Ploumanac'h pluton and  
42 North Tregor (Armorican Massif, France) were collected in granitoids, differing in  
43 petrography and chemistry. This area was chosen because these samples underwent a similar  
44 thermal history since ~300 Ma. We report new (U-Th-Sm)/He dates, along with apatite  
45 fission-track (AFT) data, as well as lithological and chemical characterization. The results  
46 show dispersed (U-Th-Sm)/He dates, ranging from  $87\pm 7$  to  $291\pm 23$  Ma, whereas central AFT  
47 dates vary from  $142\pm 6$  to  $199\pm 9$  Ma. Current predictive models for He diffusion and fission-  
48 track annealing in apatite could not reproduce the two datasets together. However, this  
49 apparent discrepancy gives insight into the parameters influencing He diffusion at geological  
50 timescales. The data confirm that radiation damage enhances He trapping, as the AHe dates  
51 are positively correlated to effective uranium (eU) concentration. The He age dispersion for  
52 constant eU content cannot be explained just by variations in grain size or chemical  
53 composition. To explore the potential influence of recoil damage trapping behavior and  
54 annealing kinetics on AHe dates, we tested a new diffusion model from Gerin et al. (2017).  
55 Given the expected model of the thermal history provided by AFT inversion, we investigated  
56 the influence of the trapping energy on AHe dates. The AHe date variations can be explained  
57 only if the trapping energy evolves from one crystal to another, increasing with the amount of  
58 damage. For a given trapping energy, minor variations in the recoil-damage annealing rate  
59 can consistently explain most of the remaining dispersion of the AHe dates.

## 60 1. Introduction

61 Apatite (U-Th-Sm)/He (AHe) thermochronometry is widely used to determine the  
62 thermal histories of mountain ranges and sedimentary basins, as apatite crystals retain  
63 radiogenic helium at low temperature (<150°C) (e.g. House et al., 1998, Ehlers and Farley  
64 2003, Stock et al., 2006; Reiners and Brandon, 2005, Valla et al., 2011; Herman et al., 2013).

65 (U-Th-Sm)/He thermochronometry is based on the accumulation of radiogenic  $^4\text{He}$  in  
66 apatite crystals, generated by  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  alpha decay chains, and to a lesser extent  
67 by  $^{147}\text{Sm}$  alpha decay. Interpretation of a set of AHe dates is not straightforward though,  
68 especially when the cooling history is complex or long (e.g. Green et al., 2006; Green and  
69 Duddy, 2006; Shuster et al., 2006; Lepretre et al., 2015) as this often produces high levels of  
70 intra and intersample dispersion. The extent to which such dispersion reflects complex He  
71 behavior during diffusion in apatite has yet to be fully explained.

72 Our knowledge of He diffusion in apatite has improved over the last decade due to  
73 numerous experiments and atomistic models (e.g. Farley 2000; Shuster et al., 2006; Cherniak  
74 et al., 2009; Bengston et al., 2012; Djimbi et al., 2015). In natural apatite, damage is produced  
75 during U-Th-Sm decay (alpha and recoil damage) and natural fission of  $^{238}\text{U}$  and may  
76 undergo annealing (self repair) at elevated temperatures (Chaumont et al., 2002; Shuster and  
77 Farley, 2009). The level of preserved damage produced by alpha decay in an apatite can  
78 influence helium retention (reduced diffusion) due to the trapping of He atoms in the damaged  
79 areas which act as holes within the crystal structure (Shuster et al., 2006; Shuster and Farley,  
80 2009; Gautheron et al., 2009; Gerin et al., 2017). Damage density depends on U-Th-Sm  
81 contents and on the damage-annealing rate, which varies with crystal chemistry and thermal  
82 history (Chaumont et al., 2002, Shuster and Farley, 2009; Gautheron et al., 2013, Fox et al.,  
83 2014).

84 Currently, two main models take into account the effect of damage trapping and

85 annealing on He retention in apatite (Flowers et al., 2009; Gautheron et al., 2009). Both  
86 models imply that the diffusion coefficient decreases with increasing damage fraction (or  
87 effective track density). Damage annealing is known to make the apatite lattice more  
88 diffusive for He atoms (Shuster and Farley, 2009), and has been suggested to be sensitive to  
89 apatite chemical composition as is fission track annealing (Gautheron et al., 2013). In the  
90 absence of specific damage annealing studies, alpha recoil damage and fission tracks are  
91 generally assumed to behave similarly. However, recent simulations from Fox and Shuster  
92 (2014) indicate that alpha damage may anneal slower than fission tracks.

93         Recently, a new radiation damage diffusion model was published by Gerin et al.  
94 (2017), and was implemented in QTQt for the purpose of our study. In this model, the closure  
95 temperature in undamaged apatite is assumed to be 30-40°C (Djimbi et al., 2015), and  
96 diffusion processes kinetics decrease with the alpha damage content as a function of damage  
97 retentivity. This last parameter is controlled by the trapping energy that adds to the activation  
98 energy, with a linear He trapping behavior.

99         Further work is required to better understand the long-term controls on helium  
100 retentivity in apatite, such as He damage trapping efficiency, the damage annealing rates, and  
101 the influence of microvoids in apatite (Zeitler et al., 2017). To this end, it is desirable to study  
102 the natural variability in helium dates (as in Green et al., 2006; Gautheron et al., 2009, 2013).  
103 Careful selection of apatite crystals is paramount for such work, as implantation, broken  
104 grains with mineral inclusions can also lead to significant scatter in AHe dates distribution  
105 (Vermeesh et al., 2007; Spiegel et al., 2009; Gautheron et al., 2012; Beucher et al., 2013;  
106 Brown et al., 2013; Murray et al., 2014; Janowski et al., 2017).

107         This study focuses on data from samples in the North Armorican Massif (western  
108 France), and particularly within the Ploumanac'h pluton and North Tregor massif. This region  
109 represents a single geological and tectonic domain and the samples experienced a common

110 thermal history since Carboniferous time. As the massif includes a variety of lithologies, the  
111 samples were selected specifically to assess whether petrography and apatite chemistry can  
112 account for the dispersion in the AHe datasets. The present work combines low temperature  
113 thermochronology, including apatite (U-Th-Sm)/He and fission-track analysis, and sample  
114 petrography/chemistry from the hand specimen to the mineral scale.

115         The aim of this study is to better understand the behavior of helium in apatite, as well  
116 as to test the current models for alpha damage accumulation and annealing. We first  
117 investigate the AHe date dispersion as a function of different physical and chemical  
118 parameters. Then, we try to model our dataset using the Flowers et al. (2009) and the  
119 Gautheron et al. (2009) models. We investigated the role of the damage retentivity and  
120 damage annealing kinetics to reproduce our dataset. To this aim, we used the new Gerin et al.  
121 (2017) model, as it is based on the most recent physical representation from Djimbi et al.  
122 (2015) and has a linear trapping behavior. We tweaked the trapping energy and damage  
123 annealing characteristics in order to predict our dataset. This approach give new insights into  
124 radiation damage and into the role of apatite chemistry on helium retentivity.

## 125 **2. Geological setting**

126         The Armorican Massif is located in northwestern France, bounded by the English  
127 Channel to the north, the Atlantic margin to the south, and the Paris Basin to the east (Fig.  
128 1A). It is composed of Proterozoic and Paleozoic rocks that experienced the Cadomian and  
129 Variscan orogenic phases, respectively at 620-540 Ma and 416-299 Ma (Peucat, 1986; Pin  
130 and Peucat, 1986; Chantraine et al., 2001; Ballèvre et al., 2009). Large scale thrusting  
131 occurred along two NW-SE shear zones, dividing the massif into four main domains: the  
132 North, Central, and South Armorican zones, and the Leon domain to the north-west (Ballèvre  
133 et al., 2009). The Tregor unit lies in the North Armorican zone, and is delimited to the  
134 northwest by the Pink Granite Coast. The exposure along this coast reveals the Variscan

135 Ploumanac'h pluton ( $303 \pm 15$  Ma, whole-rock Rb/Sr age, Vidal et al., 1981;  $301 \pm 1.7$  to  
136  $309 \pm 2.5$  Ma, zircon U-Pb, Ballouard et al., 2015), emplaced within a Cadomian magmatic  
137 complex: the North Tregor batholith ( $\sim 615$  Ma), itself intruded within a  $\sim 2$  Ga old Icartian  
138 host gneiss (U/Pb zircon ages; Auvray et al., 1980, Graviou, 1984), as illustrated in Fig. 1B.

139 This magmatic complex of the Ploumanach pluton is composed of three concentric  
140 bands (Barrière, 1977a; b): (i) the innermost two-mica granite and leucogranite (Fig. 1C,  
141 purple bands), (ii) a fine-grained granite (Fig 1C, orange band), and (iii) a coarse-grained pink  
142 granite (Fig. 1C, red band). The outermost pink granite grades from monzo- to syenogranite  
143 (Barrière 1977a; 1981). The outer two bands of magmatic rocks are co-genetic, formed during  
144 the initial magmatic injection (Barriere et al., 1977a). They were derived from a sub-alkaline  
145 magma, whereas the innermost white granites originate from the later cooling of a high-  
146 alumina magma, or from several non-cogenetic magma pulses (Albarède et al., 1980). The  
147 three concentric bands were emplaced at  $\sim 8$  km depth (i.e.  $\sim 2$  kbars, Barrière et al., 1977b),  
148 and now crop out due to significant erosion.

149 The absence of post-Variscan sedimentary deposits close to the studied area makes  
150 paleogeographic studies difficult. However, at a regional scale, the post-Variscan history of  
151 the Armorican Massif includes several geologically constrained phases of burial and erosion.  
152 During the Permian and Triassic the massif was exposed and the climate was arid, which  
153 favored fluvial and eolian sediment transport northward into depocenters (Owen, 1976;  
154 Ballèvre et al., 2012). These continental sediments are preserved in the northern seas  
155 (Western Approaches trough, English Channel) and onshore along the eastern margin of the  
156 Armorican Massif, nearby the Paris Basin. A marine transgression occurred during the  
157 Jurassic, and drowned most of the Armorican Massif. Evidence for this event is preserved in  
158 outcrops in the Normandy hills (Bessin et al., 2014), but also in offshore deposits in the  
159 Northern Approches Trough and adjacent basins (Menpes, 1997; Ziegler, 1987). A major

160 unconformity between Late Triassic and Jurassic deposits marks the initiation of this event,  
161 and is revealed by seismic data and well logs (Ruffell, 1995). Uplift in the early to mid-  
162 Cretaceous led to a second erosion phase (Owen 1976; Guillocheau et al., 2003), related to the  
163 rifting of the Bay of Biscay. It was possibly followed by a Upper Cretaceous marine  
164 transgression that is recorded by flints and chalk remnants (Hillis, 1991), followed by  
165 Pliocene sedimentation.

### 166 **3. Sampling and methods**

#### 167 **3.1. Sample collection and preparation**

168 We collected nine samples to represent the natural chemical and lithological variation  
169 on the North Armorican massif, mostly on the Ploumanach pluton (Fig. 1B and C). Eight of  
170 the samples were selected out of a single intrusive body over a small geographic area  
171 (sampling interval: 1-2 km, area: 35 km<sup>2</sup>). Sample elevation is constant and low, as the pluton  
172 is close to sea level, and the pluton is not cut by any major fault. Therefore, the sampling  
173 procedure was designed so the sample thermal history should be similar for all of the samples.  
174 Any significant dispersion in AHe dates should thus be due to other causes (experimental,  
175 compositional, or other unknown other parameters).

176 The rocks (La Clarté and Traouieros granites) are porphyritic, with cm-size crystals.  
177 Five of the samples belong to the outermost band, i.e. the coarse-grained pink granites of  
178 “Traouieros” and “La Clarté” (samples PL1 to 4, and PL8, Fig. 1C, red band). Sample PL6 is  
179 similar to the “Traouieros” granites, but is part of an intrusion within the adjacent fine-grained  
180 band (Fig. 1C, orange band). Those granites include biotite and hornblende. The center of the  
181 pluton (Fig. 1C, purple bands) was also sampled, represented by sample PL7 (innermost  
182 leucogranite) and PL11 (two-mica granite). Both samples include biotite and muscovite. A  
183 single granodiorite specimen was also collected from the North Tregor batholith (PL10),  
184 located ~30km east of the Ploumanach pluton. Sample locations and petrography are reported

185 in Fig. 1B and C, and listed in Table 1.

186 Thin sections were prepared for each rock sample and apatite grains were separated  
187 using standard crushing, sieving, density and magnetic methods, and hand-picked using a  
188 binocular microscope. Apatite crystal selection for AHe and AFT analyses as well as apatite  
189 dissolution was performed at the GEOPS low temperature thermochronology laboratory  
190 (University of Paris Sud, France). U, Th, Sm, and major, trace and rare earth element (REE)  
191 composition was determined on the He dated apatite crystals at the LSCE (Gif sur Yvette,  
192 France). Major, trace, and rare earth element contents were determined on AFT dated grains  
193 and mounted apatite crystals, using electron microprobe and LA-ICPMS measurements  
194 performed respectively at at ISTERre (Grenoble, France) and UCL (London, UK).

### 195 **3.2. Apatite fission track (AFT) analysis**

196 Apatite grains were mounted in epoxy, polished, and etched at 5M HNO<sub>3</sub> for 20 seconds  
197 at 20±1 °C. AFT ages were obtained using the external detector method, following the zeta  
198 procedure (Hurford and Green, 1982). Analyst J. Barbarand's zeta value was 359±8 for the  
199 CN5 dosimeter glass. Apatite mounts were covered by muscovite external detectors, and  
200 irradiated at the Garching facility (München, Germany) with a nominal fluence of 5×10<sup>15</sup>  
201 neutrons/cm<sup>2</sup>. Detectors were subsequently etched for 20 minutes in 40% HF at 20±1°C. AFT  
202 ages are reported as the central age at ±1 σ (Galbraith and Laslett, 1996). Mean etch pit size  
203 (Dpar) was also measured. Tracks were counted and measured at x1250 magnification, using  
204 an optical microscope and a digitizing tablet (Laslett et al., 1994). Results are presented in  
205 Table 2.

### 206 **3.3. Apatite (U-Th-Sm)/He dating**

207 Apatite grains were carefully selected for AHe dating based on size, morphology, and  
208 on the absence of any optically detectable inclusion. Six to fifteen replicates per sample were  
209 analyzed. The analytical procedure is similar to that described by Gautheron et al. (2013)

210 Crystal dimensions were evaluated under a binocular microscope, and equivalent sphere radii  
211 (Rs) were calculated to represent the He diffusion domain (e.g. Gautheron and Tassan-Got,  
212 2010). Ejection factors ( $F_T$ ) were determined using Monte Carlo simulation (Ketcham et al.,  
213 2011; Gautheron et al., 2012). Individual grains were examined twice in order to check for  
214 any unrecognised He-rich inclusions.

215 U, Th and Sm concentrations were measured by isotopic dilution using a quadrupole  
216 ICP-QMS series<sup>II</sup> CCT Thermo-Electron at the LSCE (Gif/Yvette, France). Apatite grains  
217 were dissolved in a HNO<sub>3</sub> solution spiked with <sup>235</sup>U, <sup>230</sup>Th, and <sup>149</sup>Sm. Analyses were  
218 calibrated using internal and external age standards, including Durango apatite and Limberg  
219 Tuff (Kraml et al., 2006; McDowell et al., 2005). Results of the Durango apatite are presented  
220 in supplementary information (Table S1). The one-sigma error on each AHe age amounts to  
221 8%, reflecting the analytical error and the uncertainty on the ejection factor  $F_T$  correction. All  
222 AHe data are reported in Table 3, including the effective uranium concentration, ie.  
223  $eU=U+0.24\times Th$ . Sm content was not measured in the first batch of grains (in italics in Table  
224 3). For this dataset, the Sm content indicated in the table is the mean of the more recent  
225 analyses on the same sample. It was used for the (U-Th-Sm)/He date calculation. This  
226 approximation affects the AHe dates for the six grains with low eU (<20ppm), indicated by an  
227 asterisk in Table 3. However, the contribution of Sm on AHe dates is less than the analytical  
228 resolution for eU content higher than ~10 ppm (i.e. 8%). Additionally, three crystals showed  
229 anomalously high Th/U ratios compared to the other grains from the same sample, and will  
230 not be taken into consideration in this study, as the AHe dates may have been influenced by  
231 undetected U-Th rich inclusions such as thorite or uraninite, or incomplete apatite dissolution.  
232 When the AHe dates are used in order to compare to AFT data or to geological features, then  
233 it is appropriate to correct the AHe dates from the ejection factor  $F_T$  (Gautheron et al., 2012).  
234 This factor is computed from the crystal dimensions, and account for the fact that the helium

235 atoms located beneath the grain boundarie can be lost. Therefore, the correction aims at  
236 increasing the AHe in reason of this helium leak. Small crystals thus lose a larger proportion  
237 of helium during ejection than large grains and that's why the FT ejection factor depend on  
238 grain geometry. Note that in QTQt software, the entry parameter is the raw age, that we left in  
239 Figures 6, 7, and 8.

#### 240 **3.4. Major, minor and trace elements analyses**

241 Apatite chemical composition was determined on some of the dated (U-Th-Sm)/He  
242 crystals, on AFT grain mounts and on some additional apatite fractions. The cation content of  
243 several individual (U-Th-Sm)/He dated apatite grains was quantified using the LSCE's  
244 quadrupole ICP-MS series<sup>II</sup> CCT Thermo-Electron, including P, Ca, Mn, Sr, Ba, and REE.  
245 The measurements were performed on the same solutions previously used for U, Th, Sm  
246 analyses. Calibration was performed with internal and external standards that were analyzed  
247 several times during the experiment, similar to Cros et al. (2014). Reference material NIST  
248 1640a and BCR (containing REE; Kent et al., 2007) were used for data validation. Relative  
249 standard deviation (RSD) on the NIST standard was <3%, except for P (15%). For REE, the  
250 RSD was remarkably low on BCR (0.5 to 2 %). Data treatment includes a correction  
251 regarding the reference values for NIST 1640, as well as a linear correction of the drift during  
252 the analytical sequences. Selected results can be found in Table 3, and all data can be found in  
253 supplementary Table (S2).

254 Fifty apatite grains per sample were selected and mounted in resin for Electron Probe  
255 Micro-Analysis (EPMA). The mean composition (in equivalent oxide weight percent) and  
256 standard deviations for each sample are reported in Table 4. Analyses were carried out at  
257 ISTERre (Grenoble University, France), with a JEOL-JXA-8230 microprobe. Measurements  
258 included anions (F, Cl) and cations (Ca, P, S, Si, metals). Three repeated Durango  
259 measurements were made for standardization after 40 apatite grain analyses. Errors on

260 standards were relatively low for major elements: from 0.1% for CaO to 1.7 % for F <10% for  
261 all elements except Fe and Y (22 and 35%). Minor corrections were applied to account for the  
262 small drift in Ca and P. Counting errors on X-ray intensities were negligible for Ca, P, F, Si,  
263 Ce (<10%). Due to larger errors, Cl, Na, and La results are interpreted qualitatively.

264 Finally, the major, minor and trace element compositions of the AFT and AHe dated  
265 grains were also investigated. The AFT dated grains were analyzed using EPMA.  
266 Measurements were undertaken at ISTERre using a similar protocol to that described above,  
267 except that Pr was not analyzed. Additionally, three samples were analyzed using LA-ICPMS.  
268 Laser ablation was coupled to an ICPMS system (Agilent 7700) at UCL, London (UK) in  
269 order to determine the elementary concentrations in Mn, Sr, Ba, Nb, and rare earth element.  
270 Calibrations were based on external standards NIST 610 and 612 (Jochum et al., 2011).

## 271 **4. Results**

### 272 **4.1 AFT data**

273 AFT dating was performed for all samples except PL3 and PL8. The granite and  
274 granodiorite samples have similar AFT ages, with a central age ranging from  $142\pm 6$  to  $199\pm 9$   
275 Ma (Fig. 1C). All samples passed the chi-square test at the 5% level, indicating that the range  
276 of single grain ages are consistent with a single population. Granitoid sample PL4 has a  
277 slightly older AFT age:  $199\pm 9$  (Table 2, Fig. 1C). Note that: (i) this sample has a similar Cl  
278 content than other samples, except PL10 that has slightly more Cl, (ii) PL4 is richer in Y and  
279 otherwise has a similar composition to the other samples (as well as a similar Dpar value).

280 Track length distributions are similar for all samples. Mean track length ranges  
281 between  $12.5\pm 1.3$  and  $13.0\pm 1.2$   $\mu\text{m}$  (Fig. 1C, Table 2), with low standard deviation ( $<1.4$   
282  $\mu\text{m}$ ). Individual lengths vary between 8 and 16  $\mu\text{m}$ , and the distribution of lengths is  
283 unimodal. The mean fission track-etch pit diameter (Dpar) ranges from  $1.3\pm 0.1$  to  $1.5\pm 0.1$  for  
284 all samples (Table 2). These values are quite low compared to those of Carlson et al. (1999),

285 yet they were cross-calibrated following a comparison with etching experiments in London,  
286 yielding a value of  $D_{par}$  of 1.5  $\mu\text{m}$  which is comparable to the values given by the theory of  
287 Carlson et al. (1999). Therefore, our  $D_{par}$  values are cross-calibrated and comparable to those  
288 given in Barbarand et al. (2003).

#### 289 **4.2 (U-Th-Sm)/He dating**

290 Alpha ejection corrected (U-Th-Sm)/He dates for the nine samples range between  
291  $87\pm 7$  and  $291\pm 23$  Ma. The effective uranium concentration (eU) is extremely variable from  
292 one grain to another, ranging from 1 to 557 ppm and the Th/U ratios also fluctuate, between  
293 0.01 and 7.5. Apatite grain size, expressed as the equivalent spherical radius, varies between  
294 34 and 105  $\mu\text{m}$ , with a mean value of 59  $\mu\text{m}$ .

295 AHe age dispersion correlates with eU (Fig. 2A): most AHe dates lie on a hyperbolic  
296 curve increasing with effective uranium (eU). Below the eU=60-80 ppm threshold, AHe rise  
297 with eU from  $87\pm 7$  Ma (eU<2 ppm) to >200 Ma. The granodiorite sample (PL10) has the  
298 youngest AHe dates and the lowest eU (<20 ppm). Compared to this end-member, the  
299 hornblende-bearing granite samples PL 2, PL 3, and PL 6 have older AHe dates ( $162\pm 13$  to  
300  $250\pm 20$  Ma; mean of  $202\pm 22$  Ma) and higher eU (44 to 326 ppm).

301 Some variations in (U-Th-Sm)/He dates appear to be independent of eU, as observed  
302 on Fig. 2A. For instance, for eU $\approx$ 80 ppm (75-85 ppm), AHe are scattered between  $178\pm 14$   
303 and  $261\pm 21$  Ma. This dispersion is observed even among different grains from the same  
304 sample. For instance, apatite crystals from sample PL2 range between  $176\pm 14$  and  $234\pm 19$   
305 Ma.

#### 306 **4.3 Apatite geochemistry**

307 The chemical composition of apatite crystals analyzed via EPMA is presented in Table  
308 4 and Fig. 3. Results show an almost constant major element chemistry for all apatites:  
309 CaO= $54.2\pm 0.5$ wt%, P<sub>2</sub>O<sub>5</sub>= $41.1\pm 0.7$  wt% and F= $3.7\pm 0.1$  wt%. In more detail, crystal

310 composition in CaO ranges from 52 to 56 wt%, with an approximately Gaussian distribution  
311 (Fig. 3A). P<sub>2</sub>O<sub>5</sub> content lies between 39 and 44 wt%. Most crystals have F=3.4 to 3.9 wt%  
312 (Fig. 3B), except for sample PL11 which is F-rich (3.6 to 4.8 wt%). Note that the theoretical  
313 maximum F composition is 3.77 wt% assuming no Cl or OH and only Ca and P cations  
314 (Kecham, personal communication). This composition is very close to that of Durango  
315 apatite, as reported by Barbarand et al. (2003): CaO=54.7 wt%, P<sub>2</sub>O<sub>5</sub>=41.2 wt%, and F=3.5  
316 wt%.

317 Apatite crystals from the leucogranite (PL11) also have slightly different minor  
318 element chemistry, which results in a bimodal distribution of Fe, Mn, and Na (Fig. 3C and D).  
319 They contain higher Fe (0.4±0.1 wt%), Mn (0.5±0.1 wt%), and Na (0.12±0.02 wt%) contents  
320 while being LREE-poor (e.g. Ce=0.1±0.03 wt%, Fig. 3E). Variations in sample lithology are  
321 accompanied by differences in apatite geochemistry: minerals from the different petrographic  
322 facies presented above (section 2) can be distinguished by their chemical composition (Fig.  
323 3). Crystals from the granodiorite sample PL10 are characterized by high Cl contents (0.2±0.1  
324 wt%, Fig. 3F) and low F (3-3.8 wt%, Fig. 3B). They also have lower REE (e.g. Ce=0.3±0.04  
325 wt%, Fig. 3E) than monzo and syenogranites. The apatite crystals in granitoid (PL4) are Na-  
326 rich (0.1 ±0.03 wt%, Fig. 3C) and Y-rich (0.9±0.3 wt%, Fig. 3H). Conversely, LREE contents  
327 are relatively low (e.g. La=0.1 wt%, Fig. 3G).

328 In addition to these measurements, we combined AHe dating and chemical analysis of  
329 sixty apatite crystals in order to enable direct comparison between AHe date and chemical  
330 composition. Results are included in Table 3. In general, there are no obvious chemical  
331 differences between the specimens. However, the pink granitoid PL4 is richer in REE than  
332 other samples with four crystals having ΣREE >3% (cf. Table 3, where ΣREE is the sum of  
333 all analyzed REE). PL4 also has relatively low Sr contents (85-215 ppm). On the other hand,  
334 the granodiorite sample PL10 is Sr-rich (382-816 ppm) and ΣREE-poor (5730-10604 ppm).

335 Complete chemical contents can be found in supplementary information (Table S1, Figure  
336 S1).

## 337 **5. Discussion**

### 338 **5.1. Variations in AHe dates**

339 AHe dates vary between  $85\pm 7$  and  $291\pm 23$  Ma within the studied samples and seems  
340 to be controlled at first-order by the eU content (Fig. 2A). Additionally, the AHe dates can be  
341 plotted against the estimated alpha dose (Fig. 2B), which is the amount of alpha particles that  
342 were produced in apatite since the beginning of accumulation. This alpha dose was calculated  
343 from radionuclide concentrations, given the quantity of alpha particles produced by each  
344 radionuclides and the radioactive decay equation. We assumed that damage has accumulated  
345 for at least 250 Myrs, as the pluton likely was at too high temperature to accumulate damage  
346 before this date given the thermal history. 250 Myrs is also the oldest individual track dates,  
347 which indicate that the pluton was cold enough to retain tracks at this time.

348 On both figures, the AHe dates are broadly positively correlated with eU (for eU < 60-  
349 80 ppm) and alpha dose (below  $\sim 2-3 \times 10^{16}$  alpha/g). This confirms that crystal damage  
350 strongly controls He trapping in apatite. The present dataset highlights for the first time that  
351 the relationship between He retention and damage accumulation is more complex than  
352 assumed in current models (Flowers et al., 2009 and Gautheron et al., 2009).

353 Additionally, for a given range of eU, AHe dates appear to be strongly scattered  
354 (standard deviation of  $\pm 20\%$  at  $\pm 50$  Ma). Another control mechanism is required to explain  
355 this scatter. Other factors than eU can be considered to influence He retention: (i) apatite  
356 crystal size, which controls the diffusion domain size (Reiners and Farley, 2001), (ii) grain  
357 morphology (Brown et al., 2013; Beucher et al., 2013), (iii) alpha implantation from rich U-  
358 Th neighbor minerals (Spiegel et al., 2009; Murray et al., 2014; Janowski et al., 2017), (iv) U

359 and Th zonation (Farley et al., 2011); Ault and Flowers 2012), and (v) apatite chemistry, more  
360 especially Cl content, which is often considered to control the damage annealing parameter by  
361 analogy with AFT studies (Ketcham et al., 2007; Gautheron et al., 2013), or (vi) apatite eU  
362 zonation, which might contribute to this dispersion.

363 No obvious relationship between AHe dates and apatite crystal size was found: we  
364 failed to evidence any correlation between AHe dates and crystal equivalent radius size (Fig.  
365 4A), as the correlation coefficient is 0.04. Additionally, we examined whether a correlation  
366 exist within five restricted eU bins. The answer is negative: crystal size does not contribute to  
367 AHe data scatter. Results are presented in supplementary information. Thus crystal size does  
368 not directly influence He retention here. Note that apatites from the leucogranite (PL11), the  
369 two-mica granite (PL7), and the granitoid (PL4) are older than most apatites from monzo- and  
370 syenogranites (Fig. 2). Therefore, the variation in rock lithology seems to affect He date  
371 dispersion. Additionally there is dispersion in the apatite AHe dates from the same rock  
372 sample, e.g. PL2. Yet, at the  $2\sigma$  uncertainty, most of these ages would be concordant.  
373 General influence of He implantation on AHe date dispersion is to rule out because thin  
374 section examination revealed no U-Th rich cluster around apatite. Thus, we rule out He  
375 implantation as a process that would have dispersed systematically the AHe dates within a  
376 sample .

377 We kept a record of 5 grain morphologies, as indicated in table 4B, following the  
378 number of pyramidal termination (0, 1, 2), and the number of broken faces (0, 1, or 2). There  
379 is no correlation between individual crystal AHe age and crystal length (Fig. 4B), and we  
380 observe no systematic trends with grain morphology. This absence of correlation is indicated  
381 by the fact that the correlation coefficient is nul, revealing that the crystal size and length do  
382 not influence significantly the AHe dates. We also showed that this correlation is absent for  
383 restricted eU bins. Thus, the crystal size does not control AHe dates. Additionally, we exclude

384 the hypothesis of helium implantation formulated here as explaining the whole AHe date  
385 dispersion. Indeed we examined sample thin sections which evidenced that apatite crystals are  
386 not close to U-Th rich mineral host and are not clustered. eU zonation could also induce a  
387 dispersion in AHe date, but not significant as no detectable zonation was noticed during AFT  
388 analysis.

## 389 **5.2. AHe dates of highly damaged samples**

390 The apatite crystals have eU ranging from 0 to 600 ppm. The high values of eU are  
391 quite rare, as usually it does not exceed 200 or 300 ppm. Additionally, the alpha doses range  
392 from  $4 \times 10^{15}$  to  $5 \times 10^{17}$   $\alpha/g$ , and thus exceed the usual range. For instance, Flowers et al. (2009)  
393 have investigated helium closure temperatures for apatites until  $10^{17}$   $\alpha/g$ . Therefore, our set of  
394 apatite is interesting and rare in its chemical composition and damage content. Studying  
395 helium retention in those apatites, especially the most damaged apatite, is thus quite new and  
396 interesting.

397 Only a dozen of crystals have eU > 150 ppm and alpha doses  $> 6 \times 10^{16}$   $\alpha/g$  so it is  
398 difficult to reach general conclusions about helium retention in highly damaged apatites. Yet,  
399 it is interesting to note that the mostly damaged apatites – from sample PL4 (557 ppm eU, and  
400  $5 \times 10^{17}$   $\alpha/g$ ) have low AHe dates ( $\sim 152$  Ma). This value is lower than the plateau age of 200-  
401 250 Ma reached by the apatites after the threshold of eU of 60 ppm (see the AHe vs eU plot in  
402 Figure 2A). Additionally, the ten crystals with the highest alpha doses ( $> 6 \times 10^{16}$   $\alpha/g$ ) have  
403 AHe dates decreasing with the alpha dose (Figure 2B). This negative correlation is confirmed  
404 by a relatively high correlation coefficient ( $\sim 0.5$ ). A figure is provided in the supplementary  
405 information for additional details (Figure S2), focused on these ten highly damaged apatites.  
406 Yet, this correlation coefficient is increased by the presence of the highest-alpha dose apatite.

407 Therefore, it is difficult to conclude because of the too small number of highly  
408 damaged apatites. But our dataset suggests that below a threshold of  $2\text{-}3\times 10^{16}$   $\alpha/\text{g}$ , helium  
409 retention increase with alpha doses while for higher values than  $6\times 10^{16}$   $\alpha/\text{g}$  it decreases  
410 (Figure 2B). These preliminary observations should be confirmed by studying the AHe ages  
411 in more highly damaged apatites in different geological contexts. Yet, to our knowledge it is  
412 the first time that such a decrease in helium retention at high alpha doses is observed in  
413 apatite.

414 Note that in zircon, such phenomenon was already reported by Guenthner et al. (2013)  
415 and Ketcham et al. (2013). They proposed that beyond a certain alpha dose, alpha-recoil  
416 damage interconnect and He effectively percolates through zircon. Poor He retentivity in  
417 highly damaged zones is an expected characteristic of percolation. Therefore, if future works  
418 confirmed that it also occurs in apatite, then the increase in damage dose would cause  
419 damage to cluster at low alpha doses and then to percolate after a threshold dose.

### 420 **5.3. AHe dates and apatite geochemistry**

421 The geochemistry of apatite crystals can theoretically explain part of the AHe date  
422 dispersion via a change in He diffusivity (Wolf et al., 1998; Miro et al., 2006; Djimbi et al.,  
423 2015). However, no obvious relationship between individual grain age and chemistry was  
424 found. The only element whose concentration correlates with AHe dates is Sr (Fig. 5A). This  
425 inverse correlation is significant at the 5% level ( $n=35$ , correlation coefficient =  $-0.37$ ). The  
426 two end-members are samples PL4 and PL10. Sample PL4 has high AHe dates ( $150\pm 15$  to  
427  $280\pm 28$  Ma) and low Sr contents ( $<211$  ppm), and sample PL10 has low AHe dates ( $<171\pm 17$   
428 Ma) and high Sr contents ( $>382$  ppm). However, the correlation between AHe dates and Sr  
429 contents cannot be properly interpreted since Sr content is also inversely correlated with U  
430 content and eU (Fig. 5C), which is the main factor controlling helium retention rate. The

431 relationship between AHe dates and Sr is therefore most easily explained by the fact that both  
432 are correlated with U content (eU).

433 Similarly, ions that substitute for calcium in apatite may affect fission track annealing  
434 kinetics (Carlson et al., 1999; Barbarand et al., 2001; 2003; Ketcham et al., 2007). However,  
435 no clear correlation between date and chemistry was found for any other element. The  
436 correlation coefficients – which detect the presence of a correlation between two variables -  
437 often are null, as for Ba and La (Fig. 5B and D). This implies that major element substitution  
438 chemistry and AHe ages are independent. Therefore, we suggest that either there is no  
439 chemical control on AHe dates or the measured elemental concentrations do not vary enough  
440 to impact significantly the measured AHe dates.

441 Additionally, we were able to calculate the fission track annealing parameter  $rmro$   
442 from the entire chemical composition of the samples, estimated from EPMA measurements  
443 (from Ketcham et al., 2015). This parameter combines all chemical contents. Results are  
444 presented in supplementary information (Table S3). It reveals that only sample PL11 and PL4  
445 have a slightly different value of  $rmro$  (respectively 0.80 and 0.81), compared to other  
446 samples that have a higher value of  $rmro$  (0.82-0.83). This is in agreement with the  
447 lithological difference that exists between both samples (a two-mica granite and a granitoid)  
448 compared to other samples that all are granites and granodiorite. It main explain some part of  
449 AHe dispersion (  $<50$  Ma= 20%, as is illustrated in Figure 2A, the AHe dates of PL11 are  
450 slightly higher than those of pink granite samples). Nevertheless, there is an intrinsic  
451 dispersion in AHe dates within the pink granites (red dots), that cannot be explained by a  
452 variation in the chemical composition nor in the  $rmr_o$  parameter (which is 0.82).

## 453 **5.4 (U-Th-Sm)/He dates simulation: insights into He behaviour in damaged apatite**

### 454 **5.4.1. Inconsistency between AFT and AHe dates**

455           The (U-Th-Sm)/He dates are mostly older than AFT dates, except for sample PL10.  
456   We ran a series of inverse simulations of AHe and AFT date datasets together, using QTQt  
457   software (Gallagher et al., 2009; Gallagher, 2012). Inversion of the datasets was first  
458   performed without any constraints on the possible thermal history, given the formation age  
459   (~300 Ma). A second simulation was then carried out with one-single constraint at the time of  
460   rock formation: 300 Ma ago the pluton was at “higher temperature” relatively for the low  
461   temperature thermochronometers systems (>200°C). A third simulation was performed,  
462   incorporating constraints based on the geological history described in section 2: (i) a Permo-  
463   Triassic exhumation phase to surface temperature, as the massif is known from independent  
464   geological to have been exposed emerged during this period, (ii) Cretaceous and  
465   Maastrichtian cooling at surface temperature, as the massif was uplifted during that time.

466           We tested both the Flowers et al. (2009) and the Gautheron et al. (2009) models. Both  
467   models suggest a reheating event in earliest Jurassic time. Based on the presence of Triassic  
468   detrital rocks ~180 km northwest of our study area (i.e Cotentin), and marine sediments in  
469   Normandy, Ballèvre et al. (2012) suggests that a denudation event occurred during the  
470   Triassic, followed by Jurassic reheating, as illustrated Figure 6A. This Triassic peneplanation  
471   event is classically described regionally in the Armorican Massif (Guillocheau et al., 2003),  
472   the Central Massif (Barbarand et al., 2001), and even North Africa (Gentil, 1912; Michard et  
473   al 2008). Either with or without geological constraints, none of these He diffusion models  
474   reproduce both the AHe and AFT datasets. More specifically, the old AHe dates cannot be  
475   explained given the AFT dates. The models underestimate the AHe dates, and overestimate  
476   the AFT dates. Several examples of these inverse model results are provided in the  
477   supplementary section (Fig. S3 and S4).

478           We thus decided to construct a geological plausible thermal history by inverting only  
479   the AFT dataset, using the geological constraints (Black boxes in Fig. 6A). In doing this, we

480 assume the fission track inversion technique is reliable, as it has been improved since 30  
481 years, although innovations in fission track analysis can still be carried out. We use the most  
482 recent track annealing model implemented in QTQt (Ketcham et al., 2011), which we assume  
483 is enough constrained for the purpose of our study. The aim of our paper is not to discuss the  
484 regional thermal history nor the fission track annealing kinetics, but rather to use a reasonable  
485 T-t path in order to improve the (U-Th-Sm)/He techniques.

486 Four geological constraints were used: (i) the pluton was emplaced at high pressure  
487 and temperature 300 Ma ago (box 1, Fig. 6A; Barrière et al., 1977a), (ii) the rock was at near  
488 surface temperature during the Triassic (box 2), and the Maastrichtian, (iii) the pluton has  
489 been at surface temperature for less than 50 Ma (box 4). The present day temperature was also  
490 set at  $10\pm 10^{\circ}\text{C}$ . Box 3 results from a thermochronometric constraint: as the samples were not  
491 entirely annealed, the rock did not exceed  $110^{\circ}\text{C}$ . QTQt enabled the fission track diameter  
492  $D_{\text{par}}$  to vary. AFT annealing parameter was calculated for each grain from these values.

493 Figure 6A present the result of the inversion of all samples analysed for fission-tracks  
494 (ages and lengths). It is a set of possible T-t paths, with different probabilities given the  
495 dataset. The most probable T-t paths are represented in red, while the less probable are in blue  
496 (Fig. 6A). We decided to select the “expected model” (black line in Fig. 6A) as our preferred  
497 thermal history, as this history represents a weighted model (Gallagher 2012, 2015), which is  
498 the most probable given the AFT dataset. For information, the “maximum likelihood” model  
499 was very similar to the “expected” model. In the supplementary information, we added a  
500 comparison of the predicted ages and the observations for different (T-t) paths (Figure S5).  
501 The less probable paths (in blue in Figure 6A) do not reproduce the AFT ages. Only the  
502 expected model fits several of the AFT dates. Figure 6B represent the predicted AFT ages  
503 against the observations for the expected model (green squares). The expected model fits the  
504 highest dates around 180-200 Ma, but slightly overestimates the lowest range of AFT dates

505 (140-150 Ma).

506 The numerical inversion of the samples with lowest dates (e.g.: PL1 only) was also  
507 performed with AFT ages only, and with ages and lengths. Both simulations provide the same  
508 thermal history, which are presented in supplementary information (Figure S6). It evidences  
509 that in order to reproduce the lowest dates, the Jurassic heating has to be 110°C if we inverse  
510 only PL1. Figure 6B presents the predicted fission track ages for this thermal history (yellow  
511 square). The younger AFT ages (140-180 Ma) are reproduced, but the highest AFT date (200  
512 Ma) is slightly underestimated.

513 Nevertheless, as all samples were collected over a small (km<sup>2</sup> scale) area, which is not  
514 crosscut by any major fault, and at the same elevation, all of the samples underwent the same  
515 thermal history (and so the same Jurassic heating). For instance, sample PL1 and PL2 are  
516 separated from only 200 m and have undergone a similar geological and thermal history, but  
517 have AFT ages differing by 45 Ma. No particular chemical feature appears to distinguish PL1  
518 and PL2, and their resulting annealing parameter calculated from Ketcham et al. (2015) is  
519 similar (0.82). The difference in AFT ages thus likely evidences that other parameters than the  
520 thermal history and the apatite chemical composition influence the fission track annealing  
521 kinetics. As it is not taken into account yet in fission track models, we will not be able to  
522 reproduce both the highest and the lowest AFT ages with a single thermal history. We  
523 decided to reproduce the highest dates around from Siddall 1993; 207±9 Ma for PL-14-1 from  
524 Ballouard et al. 2015). Therefore, in the following, the expected model presented in Figure 6A  
525 will be used.

526 The expected thermal history model was used for a series of forward simulations of  
527 the dataset. These simulations reproduced the oldest AFT data correctly according to HeFTy  
528 (Ketcham, 2005) and QTQt (Gallagher 2012) simulations. However, our forward simulations  
529 cannot explain the AHe data given the chosen Tt path. Given the Flowers et al. (2009) and the

530 Gautheron et al. (2009) models, the predicted AHe versus eU relationship is not consistent  
531 with the observations: both models underestimate the AHe dates (see figure 6C and 6D, green  
532 squares).

533 We also tested the sensitivity of the forward simulations for variation in the maximum  
534 Jurassic temperature. Figure 6B represent the AFT dates and Figure 6C and 6D are the AHe  
535 vs eU relationship predicted for a Jurassic burial temperature of 80°C (red triangles)  
536 compared to the expected heating of 130 °C (green squares). The results show that even  
537 though AFT dates are strongly dependent on the heating temperature (see Figure 6B), the  
538 AHe dates are not. The simulations performed with a higher Jurassic burial temperature are  
539 slightly lower, but this does not affect significantly the AHe vs eU relationship (Figure 6C  
540 and 6D).

541 If we inverse the AHe dataset without AFT constraints, the expected Jurassic burial  
542 temperature is 50°C using both the Flowers et al. (2009) model and the Gautheron et al.  
543 (2009) model (see Figure S7 and S8 in supplementary data. Nevertheless, if we reduce the  
544 Jurassic burial temperature from 130 to 80°C, the helium retention slightly increase but not  
545 enough to reproduce the modeled AHe/eU curve (Figure 6C and 6D). Additionally, with a  
546 50°C burial temperature, the simulated AFT ages would be significantly older than measured  
547 (205-220 Ma vs 140-200 Ma). With a 40°C reheating, as suggested by the Gautheron et al.  
548 (2009) model, the AFT ages would even range from 230 to 250 Ma (forward simulations).  
549 Therefore, whatever the thermal history is selected, the current models underpredict helium  
550 retention and cannot reproduce the observations. Even excluding the fission track lengths, we  
551 cannot reproduce both AFT and AHe dates given a single thermal history.

552 To summarise, neither of the current damage accumulation and annealing models for  
553 apatite can reconcile both thermochronological datasets. Inversion of AFT and AHe dates  
554 failed to reproduce AFT and AHe data together. The old AHe dates compared to AFT dates

555 indicate that: (i) the AHe system records earlier parts of the thermal history than AFT, and (ii)  
556 He retention in apatite is high, revealing that damage annealing is probably slow or that  
557 damage retention is high. Thus, the parameters used in the current models to describe helium  
558 retention (e.g. activation energy) and damage annealing kinetics do not manage to predict  
559 helium retention.

#### 560 **5.4.2. Reconciling AFT and AHe datasets: a new model**

561 Failure to produce thermal history models that fit both the AFT and AHe data suggest  
562 that either the AFT models are wrong or, as already mentioned in section 5.1, He behaviour in  
563 apatite is more complex than described by previous models (Gautheron et al., 2009; Flowers  
564 et al., 2009). To investigate He trapping in apatite, and its evolution with the damage dose, the  
565 recent He diffusion model from Gerin et al. (2017) was implemented in the QTQt software.  
566 This recent model has the advantage that the helium retention efficiency can be easily  
567 modified by the user, through a parameter called the trapping energy  $\Delta Ea$ .

568 In their contribution, Gerin et al. (2017) propose a relationship between radiation  
569 damage accumulation and the diffusion properties, where the diffusion coefficient  $\tilde{D}$   
570 decreases with the damage fraction  $f(x)$  following the equation:

$$571 \quad \tilde{D}(x,t) = \frac{D(T)}{\left[ 1 + f(x) \exp\left(\frac{\Delta E_a}{RT}\right) h \right]} \quad (1),$$

572 where  $D(T)$  is the diffusion coefficient in undamaged apatite;  $\Delta Ea$  is the additional  
573 energy that is required for He, once it has entered into a damage site, to diffuse back into the  
574 lattice (see Shuster et al., 2006; Gautheron et al., 2009); and  $h$  is the percentage of annealing  
575 ranging from 0 to 1. In the Gerin et al. (2017) model, initial He diffusion parameters  
576 (diffusion coefficient  $D_0$  and activation energy  $E_a$ ) are adopted from multi scale quantum  
577 based Density Function Theory (DFT) calculations (Djimbi et al., 2015). Such calculations  
578 yield a He closure temperature of  $\approx 30-40$  °C for undamaged apatite.

579 Here, we used the same approach but the alpha damage annealing parameter is  
580 hereafter termed  $rmr_0'$ , to distinguish it from the  $rmr_0$  parameter used to describe AFT. This  
581 new parametrization of  $rmr_0'$  may help to reconcile AFT and AHe data (Fox and Shuster,  
582 2014), which was not possible in previous models that considered that track and recoil  
583 damage anneal at the same rate (i.e.  $rmr_0=rmr_0'$ ). Following Ketcham et al. (2007), the value  
584 of  $rmr_0$  can be calculated either from the  $D_{par}$  measurements, or from the Cl content, and  
585 varies between 0.6 and 0.83 in most natural apatites, even though in rare crystals it goes down  
586 to 0. In the case of the Ploumanac'h rocks, the  $rmr_0$  value calculated from  $D_{par}$  values is 0.85  
587 (Ketcham et al. 2007), while that calculated from the Cl PFU contents, issued from EMP  
588 analyses is 0.83, except for sample PL10 (0.82). More recently, Ketcham et al. (2015) propose  
589 that  $rmr_0$  is multi-compositional. We calculated the values of  $rmr_0$  using this recent study:  
590 0.82-0.83 for all samples except PL11 (0.80), PL4 (0.81). Results can be found in  
591 supplementary informations. Unfortunately, we did not analyze sample PL7, that likely has a  
592 different composition due to its lithological specificity.

### 593 **5.4.3. Investigation of damage trapping energy ( $\Delta E_a$ )**

594 We first estimate the range of trapping energy  $\Delta E_a$  that is required to reconcile the  
595 AHe with the thermal history obtained from AFT data and assumed geological constraints.  
596 The  $\Delta E_a$  value depends on damage topology, i.e damage shape, size, and connectivity. In the  
597 case of simple vacancies, Gerin et al. (2017) estimated that  $\Delta E_a$  is  $\sim 30 \pm 5$  kJ/mol, based on  
598 DFT calculations. Shuster and Farley (2009) also predict experimental values in the same  
599 order of magnitude ( $\sim 25$  kJ/mol). In addition, Gerin et al. (2017) suggest that damage  
600 clustering increases  $\Delta E_a$  to higher than 50 kJ/mol.

601 Using our preferred thermal history (Fig. 6A), we ran several forward simulations in  
602 which we vary the damage trapping energy  $\Delta E_a$  from 30 to 90 kJ/mol. Results are

603 summarised in Fig. 7, where the AHe vs eU relationship was modeled for different values of  
604  $\Delta Ea$ . With  $\Delta Ea=30$  kJ/mol (red triangles) or 40 kJ/mol (yellow losangles), the predicted AHe  
605 dates are significantly lower than observed values (Fig 7A) so these trapping energies are not  
606 sufficient to retain enough helium. However, for the trapping energy  $\Delta Ea$  of 70 kJ/mol (green  
607 squares), the model dates better fit the mean observed AHe dates (blue circles).

608 Note that for the lowest eU values (eU<20 ppm), the trapping energy of 70 kJ/mol  
609 overestimate the AHe dates, while for higher eU values it correctly reproduce the  
610 observations. Thus, the whole data dispersion cannot be reproduced if all of the crystals have  
611 the same  $\Delta Ea$  value. Therefore, we decided to vary the  $\Delta Ea$  value for each grain from samples  
612 PL10 and PL4, i.e. samples with the lowest and the highest eU. The results are presented in  
613 Fig. 7B and 7C. For each crystal, we determined the  $\Delta Ea$  value required so that the predicted  
614 AHe agrees reasonably well with the measured AHe date so that in a prediction vs.  
615 observation diagram, the data points lie around the 1:1 straight line (Figure 7B and 7C).  $\Delta Ea$   
616 values are represented on a color scale from pink to red, and are indicated on the plots. Please  
617 remind that these simulations were performed using one single Tt path, the “expected model”  
618 that fits AFT dates. At this stage, the  $rmr_0'$  parameter was chosen equal to  $rmr_0$  (=0.81 and  
619 0.82), calculated from the chemical composition measurements (using Ketcham et al., 2015).  
620 We will come back to the role of a variable  $rmr_0'$  later.

621 For sample PL10, the simulation showed that the required  $\Delta Ea$  is relatively high (46<  
622  $\Delta Ea$  <87 kJ/mol), given the assumption made and the chosen thermal history. Additionally,  
623  $\Delta Ea$  is the main parameter to increase the predicted AHe date, and thus it increases with the  
624 measured AHe date (Fig. 7C, purple to red triangles). As AHe positively correlates with eU in  
625 PL10,  $\Delta Ea$  also increases with damage density (Fig.8). Damage clustering (recombination, or  
626 gathering) may explain this implied increase in trapping energy.

627 This can be inferred from our understanding of the diffusion process at atomic scale:  
628 helium migration between sites is restricted due to its repulsion by neighbour atoms. To  
629 overcome this effect, helium atoms have to provide an additional energy. The energy that  
630 needs to be furnished in order to fit an atom (or one mole) of helium from outside of the  
631 crystal into an insertion site is called the “insertion energy” which amounts to 0.66 eV  
632 (Djimbi et al., 2015) in apatite, i.e. 64 kJ/mol. Extended damage, like cavities, are zones  
633 where the neighbor atoms are distant. Therefore, the repulsion effect drops, and the energy  
634 barrier height equals the insertion energy. On the other hand, small damage sites are  
635 neighbored by repelling atoms, which favors helium jump to another site. The energy barrier  
636 is thus smaller than for larger damage sites.  $\Delta Ea$  quantifies the increase in the required energy  
637 barrier induced by a defect. This rise in energy is caused by the increase in the distance  
638 between helium and neighbour atoms that reduce the repulsion effects. Depending on the size  
639 of the defect site, the value of  $\Delta Ea$  ranges from 0 kJ/mol (no trapping effect in small-size  
640 defect site with high repulsion effects) to 64 kJ/mol (large cavities, with efficient trapping).  
641 This range of values is also documented in the DFT calculation in the Gerin et al. (2017)  
642 study, where the values of  $\Delta Ea$  (20-50 kJ/mol) are beyond the insertion energy. Note that if  
643 the interaction between helium and neighbour atoms was attractive at moderate distance, the  
644  $\Delta Ea$  values could theoretically be higher than the insertion energy.

#### 645 **5.4.4. Investigation of alpha recoil damage annealing behaviour**

646 In this section, we used a similar approach, but kept the  $\Delta Ea$  value constant, and  
647 investigated the  $rmr_0'$  values required to reproduce AHe dates. Fig. 9A and 9B present the  
648 results of two simulations, performed with  $rmr_0'$  value of 0.7 and 0.9 (red triangles and green  
649 squares respectively), all other parameters being constant ( $\Delta Ea=40$  or  $70$  kJ/mol). For AFT  
650 analysis,  $rmr_0$  is 0.8-0.83, but we explore here any other variation of  $rmr_0'$ . The modeled AHe  
651 are negatively correlated to  $rmr_0'$  (red triangles are above green squares). This is consistent, as

652 alpha-recoil annealing enhances He diffusion (Gautheron et al., 2009; Shuster and Farley,  
653 2009).

654 As is revealed by Figure 9A, except for low eU values, the AHe vs eU relationship is  
655 better reproduced with  $\text{rnr}_o'=0.7$  (red triangles) than with  $\text{rnr}_o'=0.9$  (green squares). This  
656 reveals that alpha-recoil damage likely anneal slowly – maybe more slowly than fission  
657 tracks-, as suggested by Fox and Shuster (2014). Note that a high trapping energy is required,  
658 as otherwise the observed AHe vs eU relationship would not be reproduced (Figure 9B).  
659 Therefore, the conclusions of section 5.4.3 still stand.

## 660 **5.5. Implications regarding He trapping law**

661 The trapping energy ( $\Delta Ea$ ) quantifies the damage topology as it increases with the  
662 damage site size and when defects cluster as proposed by Gerin et al. (2017). As illustrated in  
663 Fig. 8, the present study suggests that – given the assumption made and the chosen expected  
664 Tt path – a positive correlation between  $\Delta Ea$  and eU can explain the data at low eU (in the  
665 sample PL10). The conclusions from all the parameter tweaking ( $\Delta Ea$  and  $\text{rnr}_o'$ ) is that in  
666 order to reproduce both the AHe and the AFT dataset the trapping energy has to be high  
667 enough (>46 kJ/mol) and to vary between crystals. This is the main difference between the  
668 Gerin et al. (2017) model and other radiation models: in QTQt, other radiation models such as  
669 the Flowers et al. (2009) or Gautheron et al. (2009) model consider that the activation energy  
670 is constant for all crystals. Within the Gerin et al. (2017) model, the trapping energy can be  
671 varied between grains, and that's why we can reproduce our dataset.

672 Our study suggests that this trapping energy increases with the alpha dose below a  
673 threshold. It may reveal that: (i) in low damaged apatites, defect clustering occurs with dose  
674 accumulation, while (ii) after a damage dose threshold is reached, the damage site size  
675 declines or defect declustering occurs. Defect clustering in low damaged apatites likely leads  
676 to an increase in He retentivity, due to the creation of microvoids in apatite (Zeitler et al.,

677 2017). The geometry of damaged zones may be drastically modified in highly damaged  
678 apatites, as already mentioned earlier. If this were also a significant process for high eU  
679 apatite, the damage accumulation physics of Gerin et al. (2017) would not be applicable, and  
680 another model should be elaborated specifically for highly damaged samples.

## 681 **6. Conclusion**

682 Our study investigated variations in (U-Th-Sm)/He date from samples with differing  
683 chemistry and petrography, which have undergone a protracted low temperature thermal  
684 history. Fission track analysis and geological constraints show that the samples were first  
685 exhumed after the Variscan orogeny, and then were buried until 110°C during the Jurassic,  
686 before being exhumed once more. Our study investigated some parameters that have the  
687 potential to influence He diffusion in apatite.

688 Results show dispersed (U-Th-Sm)/He dates, ranging from  $80\pm 8$  to  $291\pm 29$  Ma, which  
689 are mostly older than the central AFT dates ( $142\pm 6$  to  $199\pm 9$  Ma). In detail, the (U-Th-  
690 Sm)/He dates increase with the effective uranium content (for eU < 60-80 ppm) and effective  
691 recoil track density ( $< 2-3\times 10^{16}$  alpha/g). This confirms that crystal damage strongly controls  
692 He trapping in apatite. For higher alpha doses, ten crystals preliminarily suggest a decrease in  
693 helium retention above a threshold in damage content; but this should be investigated further  
694 by future works. Apatite grain chemistry was also investigated to test its potential impact on  
695 AHe dates. Our data suggest that either there is no chemical control on AHe dates or our  
696 measured elemental concentrations do not vary enough to impact significantly on the  
697 measured AHe dates.

698 Current He radiation damage models fail to reproduce the present dataset adequately  
699 and they cannot reconcile the apparent discrepancy between AFT and AHe dates. To  
700 determine whether this discrepancy can be resolved we implemented the recent He diffusion  
701 model by Gerin et al. (2017) and an alpha damage annealing law similar to the Ketcham et al.

702 (2007)'s model for fission tracks. We tested the sensitivity of the model to variations in alpha  
703 recoil damage trapping energy ( $\Delta Ea$ ) and annealing kinetics. Thus, we determined the  $\Delta Ea$   
704 and  $rmr_0'$  parameters required to reproduce the data.  $Rmro'$  characterizes annealing kinetics  
705 of alpha damage. (U-Th-Sm)/He and AFT dates can be adequately modeled together using (i)  
706 enhanced trapping, or (ii) reduced annealing (or a combination of both). The first mechanism  
707 requires high damage trapping energy values ( $\Delta Ea$  of 40-80 kJ/mol), which exceeds estimates  
708 for a single vacancy from DFT calculations ( $\sim 30$  kJ/mol). This may be understandable if the  
709 accumulated individual damage sites coalesce to form single connecting vacancies, thereby  
710 creating extended damaged domains.

711 For high alpha doses, if the decrease of AHe dates is confirmed by future works, then  
712 we suggest a similar behavior than previously described in zircon by Ketcham et al. (2013),  
713 involving topological modification or damage interconnection. Variations in annealing  
714 kinetics induce some dispersion in AHe dates, but cannot explain the observed AHe scattering  
715 without any change in damage trapping energy. The second key point revealed by the samples  
716 from Tregor is that alpha recoil probably anneals slower than fission tracks. Future models  
717 should take this into consideration and investigate damage annealing mechanisms.

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## **Table captions**

**Table 1:** Sample lithology and location.

**Table 2:** AFT data.

**Table 3:** AHe age data and chemical composition for select elements.

**Table 4:** Apatite mean composition (wt%).

## Figure captions

**Figure 1: Geological setting and sample location.** (A) Regional map of northwestern France, and location of the Armorican massif. Pink areas are outcropping Paleozoic basement massifs. (B) Geological map of the “pink granite coast” (Brittany, France), and location of the Ploumanac’h pluton. (C) Lithological map of the Ploumanac’h intrusion, and fission track analysis results (this study). Sample locations, crystallization ages, central AFT ages, and mean fission track lengths are shown.

**Figure 2: Individual AHe dates as function of (A) effective U concentration and (B) alpha dose.** The colors indicate sample location and lithology: PL10 (cyan squares), PL11 (dark purple diamonds), PL7 (light purple triangles), PL4 (yellow circles), PL6 (dark red circles), and external pink granites (red circles). Alpha dose was calculated from U, Th, and Sm contents over 250 Ma. In (B) only data where U, Th, and Sm were analyzed are shown.

**Figure 3: Histograms of apatite chemical composition.** Measurements were made on 339 single crystals, using EPMA. The dots represent the raw data, with the same color code than in Figure 3. Purple dots represent sample PL11. Units are oxide weights percent.

**Figure 4: AHe dates as a function of (A) equivalent grain size and (B) crystal length.** Pyr.: pyramidal termination. Bf.: Broken face. L: crystal length. Equivalent sphere radius size ( $R_s$ ) was calculated based on measured individual grain dimensions.

**Figure 5: Individual AHe dates as function of chemistry.** Relationship between AHe date and Sr (A), Ba (B), and La concentrations (D). Relationship between eU and Sr content (C). Error bars represent maximum uncertainties, i.e. 8% for AHe, 15% for Sr and La and eU, and 10 % for Ba. The color coding is the same as for Fig. 1, 3, and 4.

**Figure 6: Thermal history reconstruction and forward simulations.** (A) Thermal history reconstruction resulting from inverse simulation of the AFT dataset, along with geological constraints, using QTQt (Gallagher, 2012). (B) Forward simulations of AFT dataset using this (T-t) path (green squares) or a 80°C Jurassic reheating (red squares) using QTQt. The yellow square is issued from the inversion of PL1 only. (C) Forward simulation performed using the Flowers et al. (2009) model, with two different burial temperature, carried out with QTQt. (D) Forward simulations using the Gautheron et al. (2009) model, carried out using QTQt for two different burial temperatures. The error bar on raw AHe ages is 5%.

**Figure 7: Evolution of the AHe dates as a function of the trapping energy.** Forward simulations of AHe dates using Gerin et al. (2017), with constant damage annealing rate ( $\text{rmro}'=\text{rmro}$ ). (A) Forward simulation results of the AHe vs eU relationship obtained for  $\Delta E_a=30$  kJ/mol (red triangles), 40 kJ/mol (yellow losangles), and  $\Delta E_a=70$  kJ/mol (green squares), compared to observations (blue dots). (C) and (D):  $\Delta E_a$  calibration obtained for sample PL10 and PL4. Colors and numbers indicate  $\Delta E_a$  values (see color code). Squares represent AFT data and triangles represent AHe dates.

**Figure 8: Evolution of the inferred trapping energy  $\Delta E_a$  with the effective uranium (eU).** Results for PL4 and PL10, which required trapping energy range from 46 to 120 kJ/mol.

**Figure 9: Damage annealing simulation.** Forward simulations of AHe dates were performed using Gerin et al. (2017), using the thermal history obtained from geological and AFT constraints. Alpha damage annealing rate is quantified by a variable  $\text{rnr}_0'$ . (A) AHe vs eU relationship obtained for  $\text{rnr}_0'=0.7$  (red triangles) and 0.9 (green squares) with  $\Delta Ea=70$  kJ/mol and (B)  $\Delta Ea=40$  kJ/mol.

Figure 1

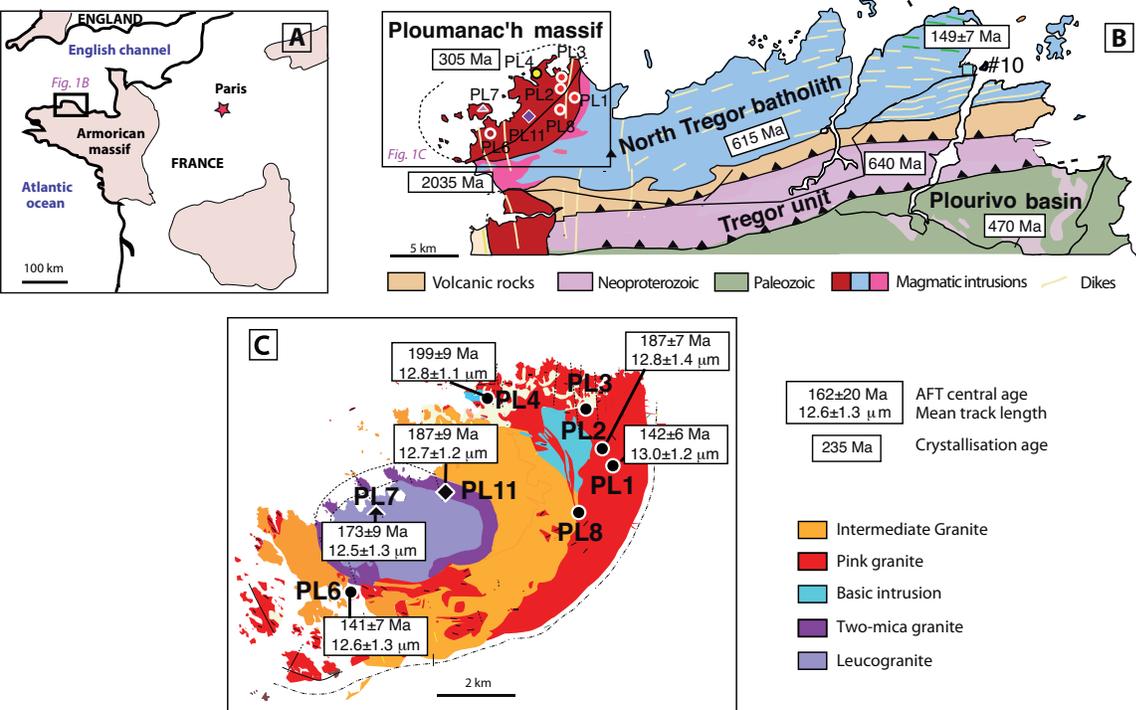


Figure 2

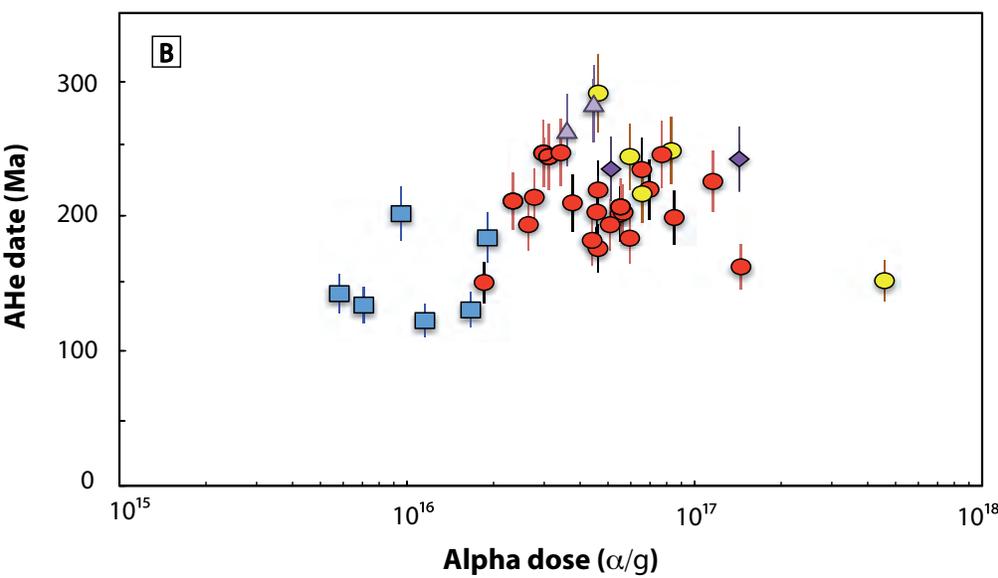
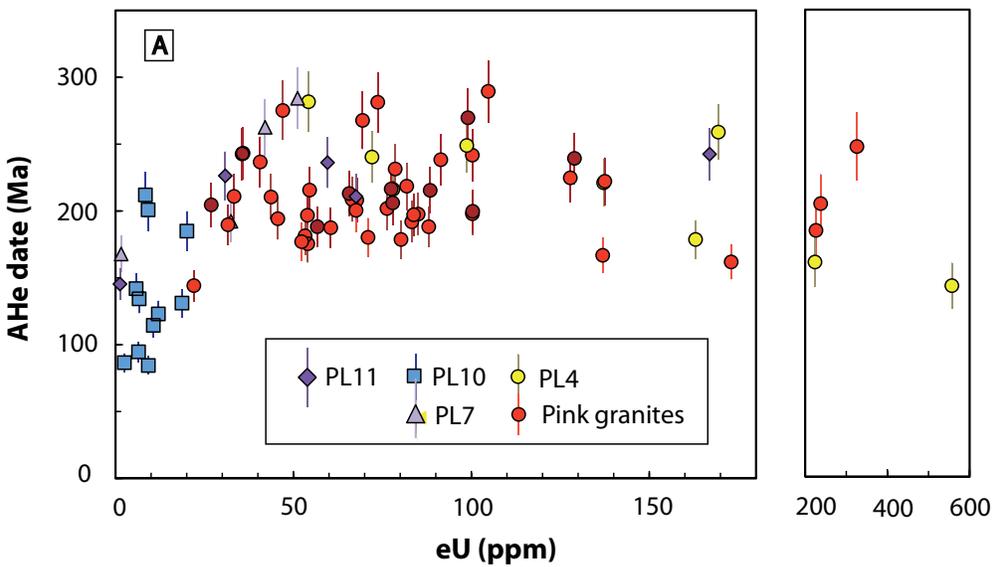


Figure 3

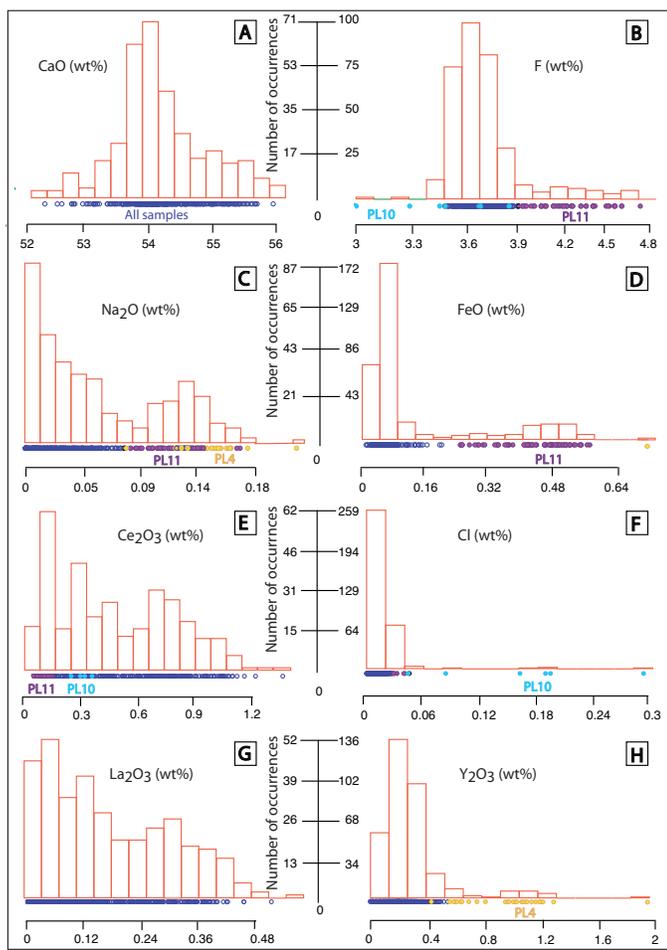


Figure 4

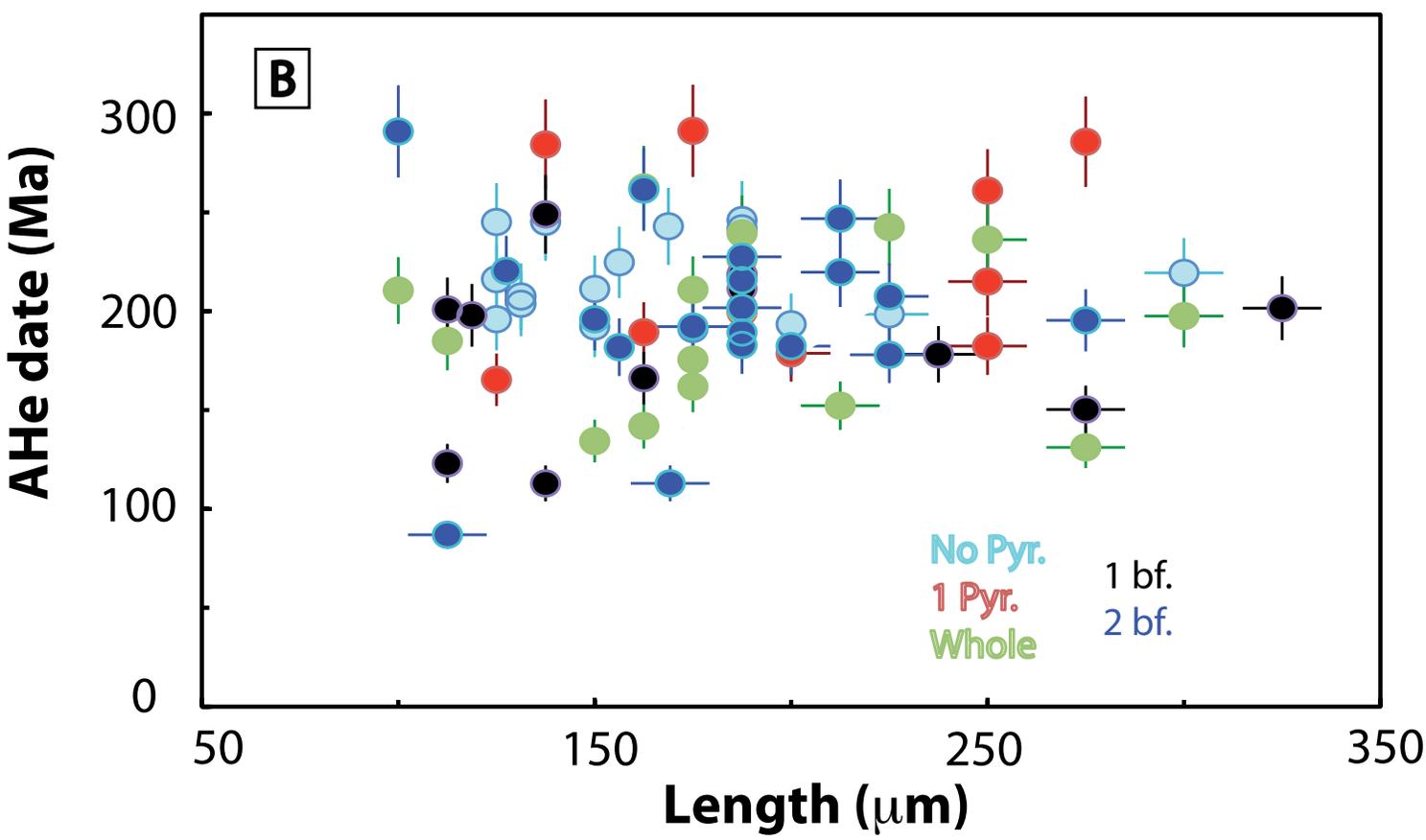
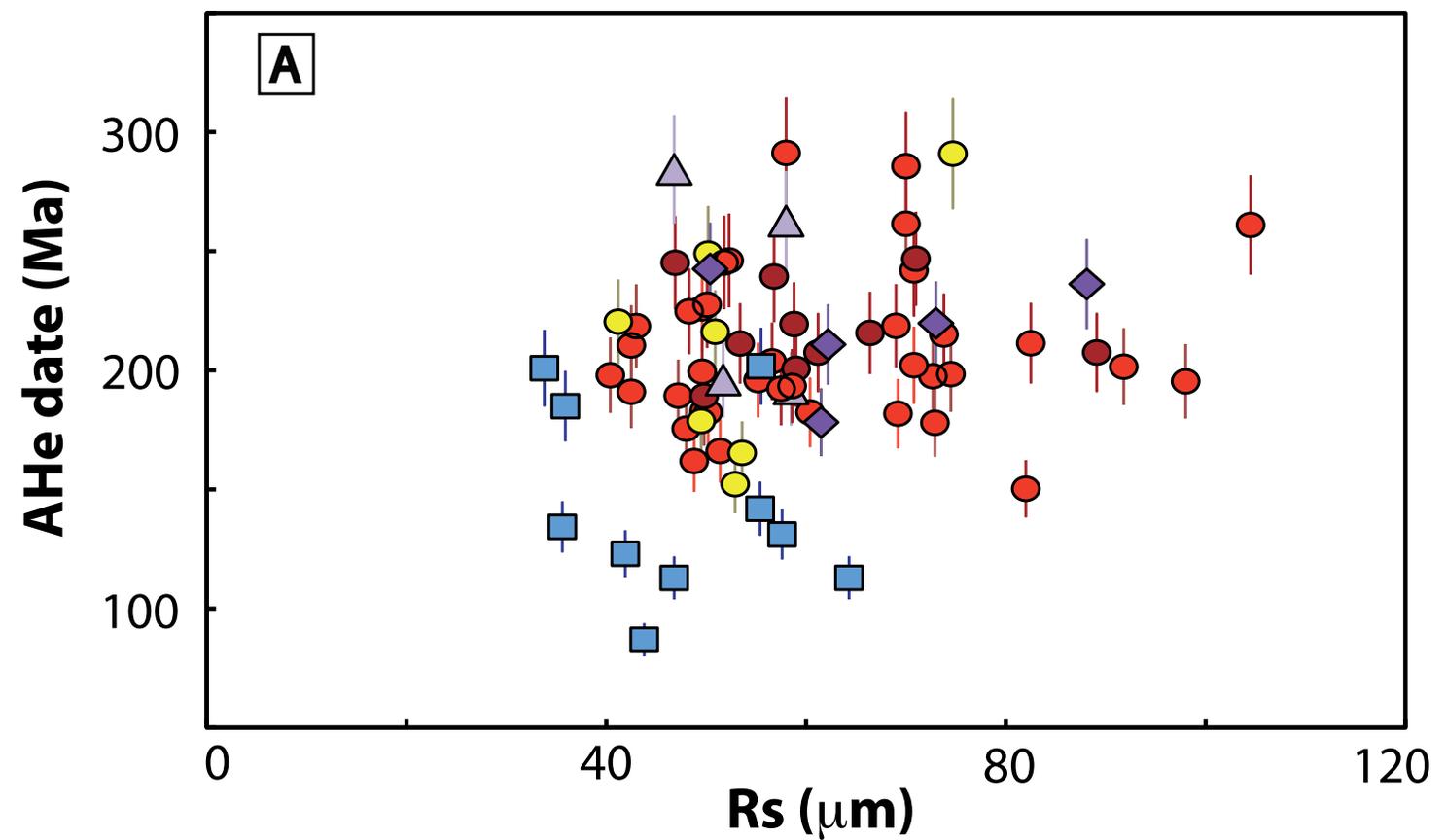


Figure 5

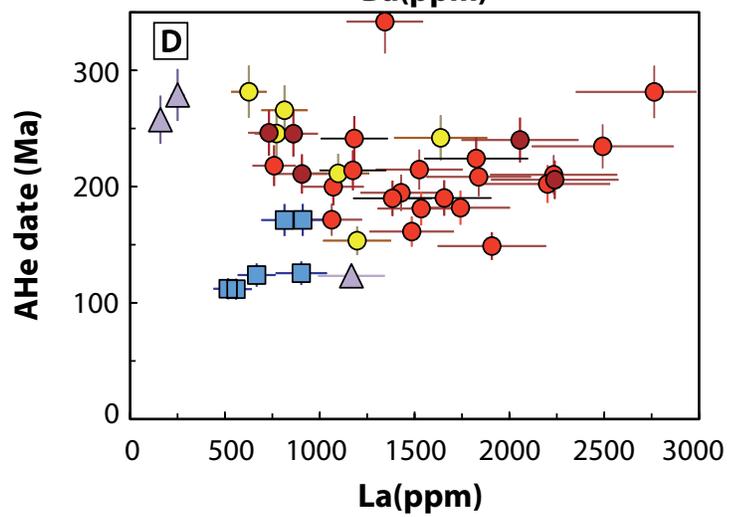
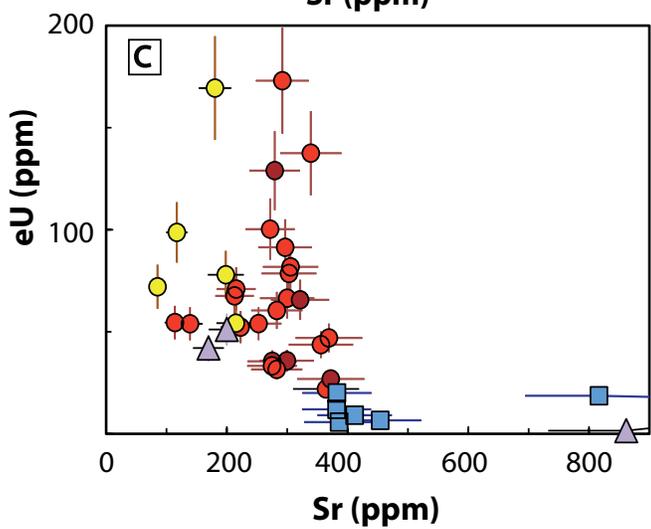
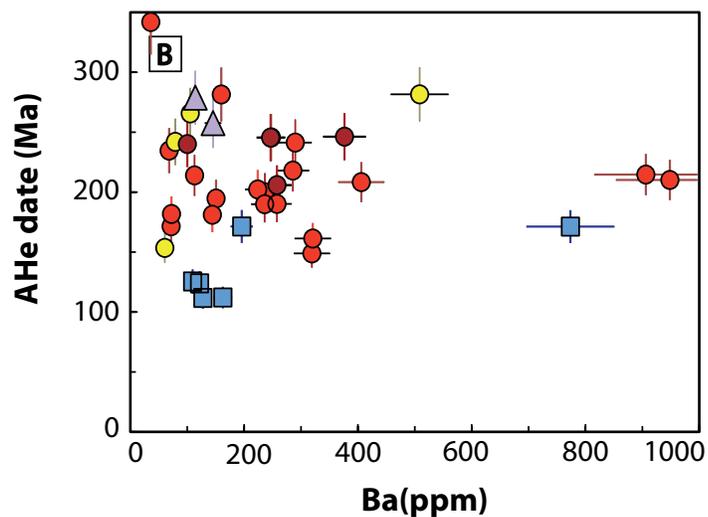
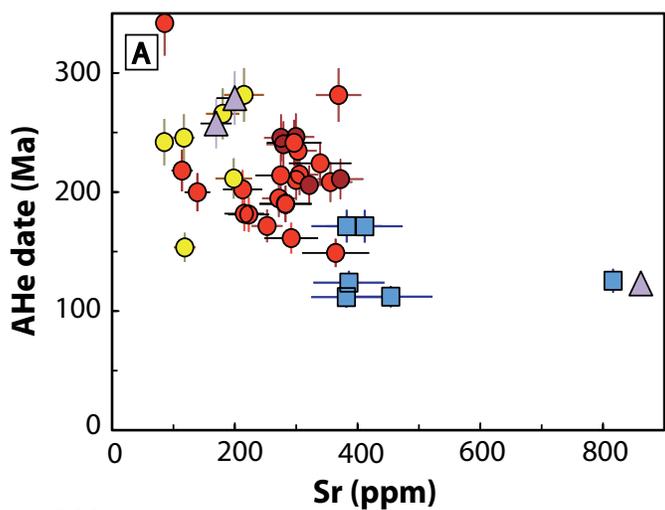


Figure 6

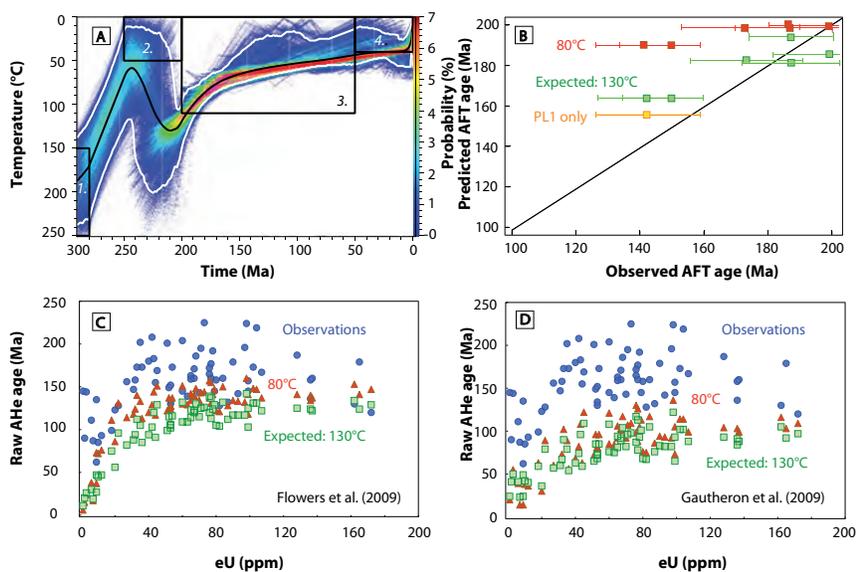


Figure 7

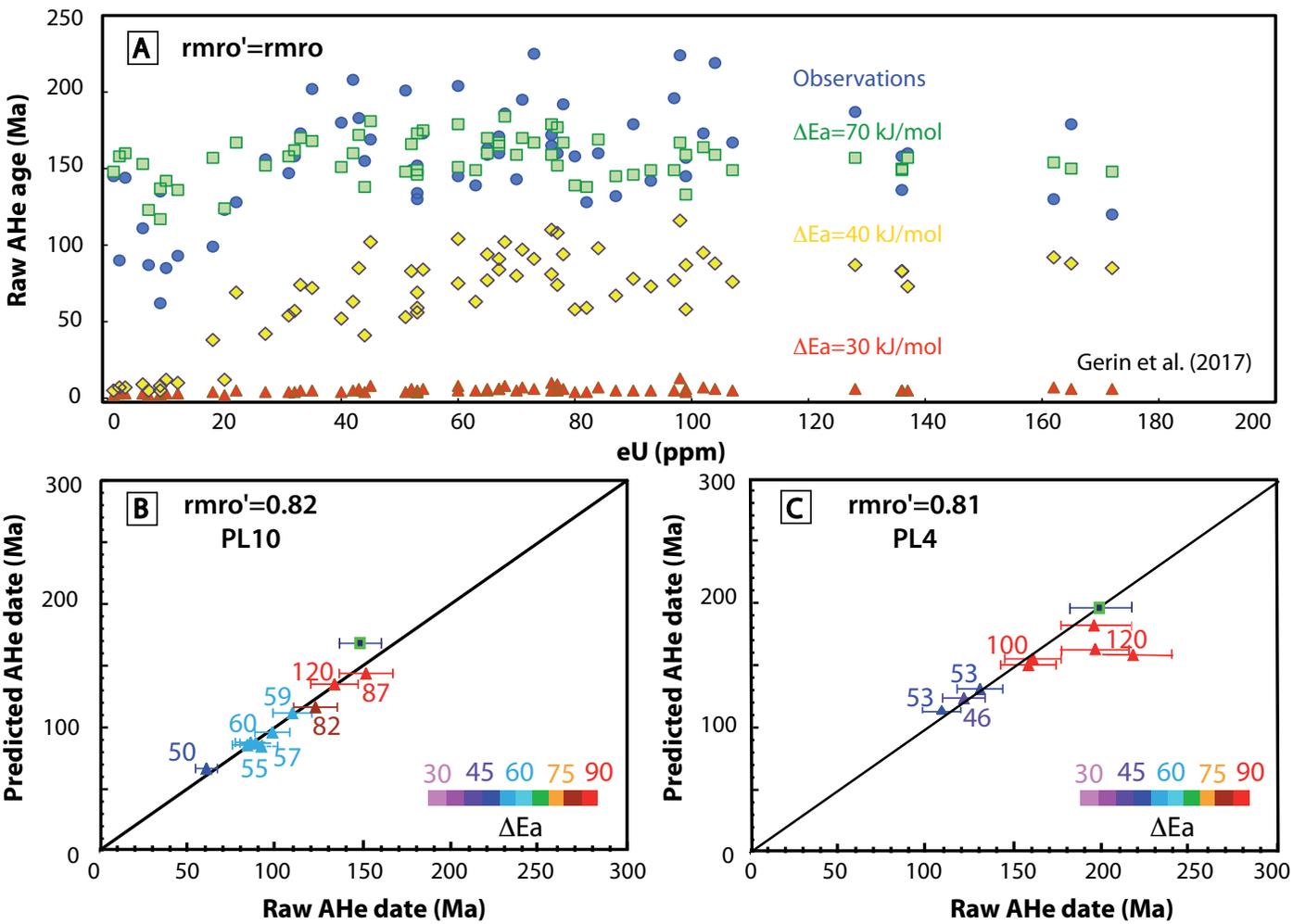


Figure 8

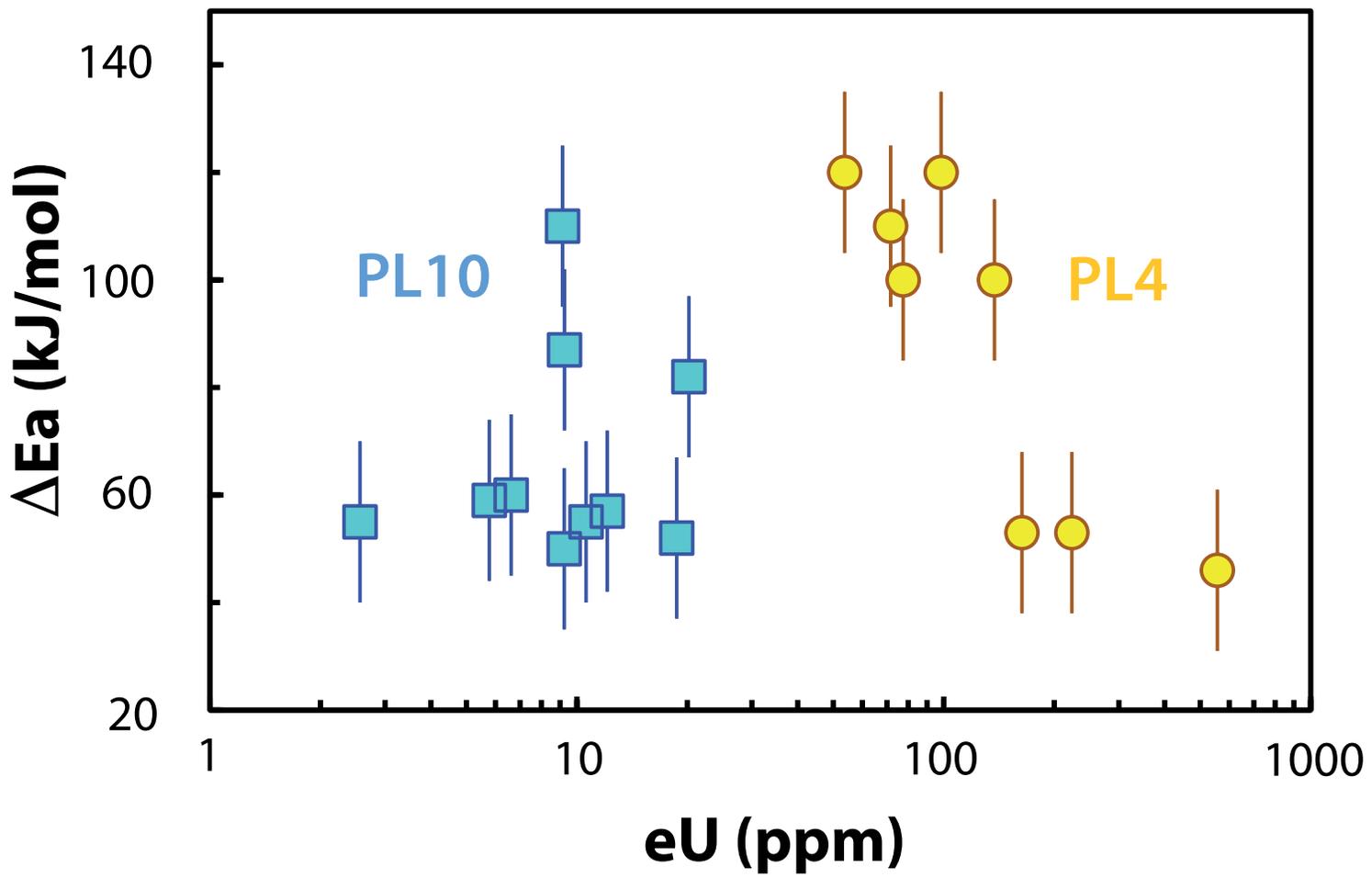
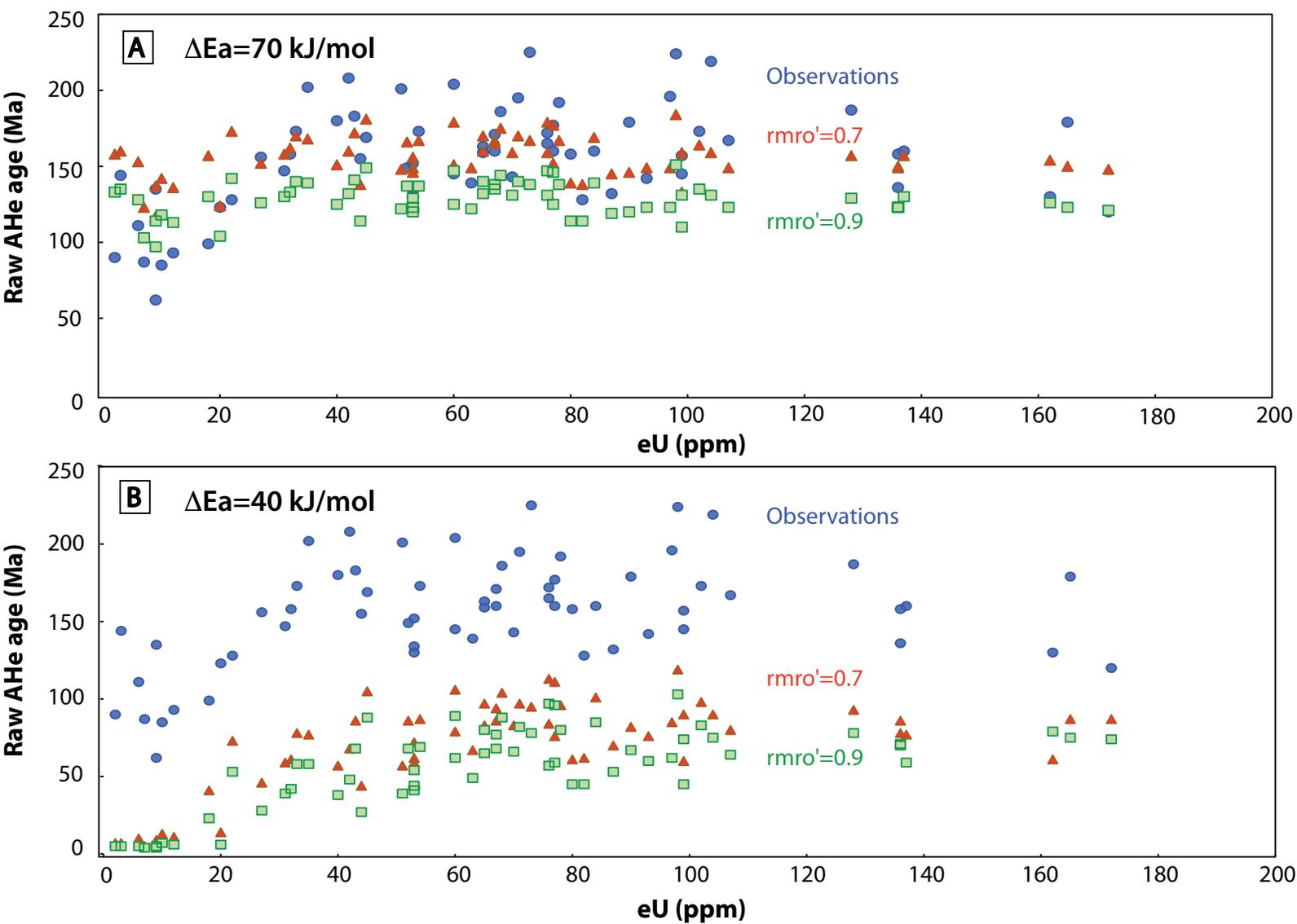


Figure 9



**Table 1:** Sample petrology and location.

<i>Samples</i>	<i>Petrography</i>	<i>Altitude (m)</i>	<i>Coordinates</i>
<b>Ploumanac'h extern layer</b>			
PL 1	Coarse-grained syenogranite	34	N48°48'54,4-W003°28'37,3
PL 2	Coarse-grained syenogranite	16	N48°49'11,6-W003°29'32,6
PL 3	Coarse-grained monzogranite	0	N48°49'47,9-W003°29'38,6
PL 4	Medium-grained granitoid	0	N48°49'52,7-W003°31'22,4
PL 6	Coarse-grained monzogranite	21	N48°47'09,7-W003°34'32,0
PL 8	Coarse-grained monzogranite	50	N48°48'19,9-W003°29'32,7
<b>Ploumanac'h inter layer</b>			
PL 7	Mono-mica leucogranite	28	N48°48'19,2-W003°34'18,8
PL 11	Two mica granite	3	N48°48'34,4-W003°32'50,1
<b>Trégor</b>			
PL 10	Granodiorite	0	N48°51'10,8-W003°12'39,3

**Table 2:** AFT data.

Sample name	No. of grains	No. of tracks	$\rho_s \times 10^6$ (tr/cm <sup>2</sup> )	N <sub>s</sub>	$\rho_i \times 10^6$ (tr/cm <sup>2</sup> )	N <sub>i</sub>	$\rho_d \times 10^5$ (tr/cm <sup>2</sup> )	N <sub>d</sub>	P( $\chi^2$ ) (%)	AFT $\pm \sigma$ (Ma)	D (%)	MTL $\pm$ SD ( $\mu$ m)	Dpar $\pm$ SD ( $\mu$ m)
PL 1	20	100	3.234	1504	2.742	1275	7	7671	63	142 $\pm$ 6	0.9	13.0 $\pm$ 1.2	1.3 $\pm$ 0.1
PL 2	23	102	3.849	2321	2.463	1485	7	7671	60	187 $\pm$ 7	1.4	12.8 $\pm$ 1.4	1.5 $\pm$ 0.1
PL 4	24	100	2.912	1491	1.750	896	7	7671	49	199 $\pm$ 9	1.8	12.8 $\pm$ 1.1	1.4 $\pm$ 0.1
PL 6	20	100	2.728	1506	1.893	1045	5.5	6040	54	141 $\pm$ 7	2.5	12.6 $\pm$ 1.3	1.9 $\pm$ 0.1
PL 7	23	78	2.593	1006	1.794	696	7	7671	85	173 $\pm$ 9	0.0	12.5 $\pm$ 1.3	1.4 $\pm$ 0.1
PL 10	23	NA*	3.764	1566	3.024	1258	7	7671	46	149 $\pm$ 7	2.8	NA	1.5 $\pm$ 0.5
PL 11	20	95	3.145	1557	2.008	994	7	7671	68	187 $\pm$ 9	1.2	12.7 $\pm$ 1.2	1.4 $\pm$ 0.1

\* NA=analysed

No. of grains and No. of tracks mean number of counted grains and number of measured tracks lengths

**Table 3:** AHe age data and selected chemical contents.

<i>Name</i>	<i>Geometry</i>	<i>L</i> ( $\mu\text{m}$ )	<i>H</i> ( $\mu\text{m}$ )	<i>W</i> ( $\mu\text{m}$ )	<i>Rs</i> ( $\mu\text{m}$ )	<i>F<sub>T</sub></i>	<i>Masse</i> ( $\mu\text{g}$ )	<i><sup>4</sup>He</i> ( $\times 10^5$ ) ( <i>ncc/g</i> )	<i>U</i> ( <i>ppm</i> )	<i>Th</i> ( <i>ppm</i> )	<i>Sm</i> ( <i>ppm</i> )	<i>eU</i> ( <i>ppm</i> )	<i>Th/U</i>	<i>Age</i> <i>raw</i> ( <i>Ma</i> )	<i>Age c</i> ( <i>Ma</i> ) *	<i>Mn</i> ( <i>ppm</i> )	<i>Sr</i> ( <i>ppm</i> )	<i>Ba</i> ( <i>ppm</i> )	$\Sigma$ <i>REE</i> ( <i>ppm</i> )
<i>PL1F</i>	<i>1 py</i>	175	175	113	58	0.75	4.7	28.21	66	162	352	105	2.5	219	291±23	-	-	-	-
<i>PL1D</i>	<i>1 py</i>	250	238	238	105	0.86	22.5	19.17	46	99	254	69	2.2	224	261±21	-	-	-	-
<i>PL1G</i>	<i>2bf</i>	188	175	163	83	0.88	12.5	15.53	39	122	236	68	3.2	186	211±17	-	-	-	-
PL1terB	No py	188	125	119	69	0.79	8.9	11.55	33	91	129	55	2.8	173	219±18	702	114	286	9025
PL1bisA	2bf	275	125	150	82	0.85	16.5	3.43	11	46	58	22	4.2	128	150±12	449	364	319	12307
PL1QuA	No py	131	113	100	57	0.75	4.7	9.96	25	120	204	54	4.8	152	204±16	752	139	237	11428
PL 2																			
PL2-B	1+1	100	100	88	43	0.74	3.2	8.32	22	92	183	44	4.3	155	210±17	449	356	406	11791
PL2-C	1+1	175	88	88	48	0.74	2.6	8.51	28	108	235	54	3.9	130	176±14	353	253	72	7145
PL2-D	2bf	188	75	75	43	0.72	2.4	15.69	43	161	306	82	3.7	158	219±18	739	306	906	9872
PL2-E	2bf	119	81	69	40	0.73	1.6	17.61	53	196	354	100	3.7	145	198±16	412	272	151	9900
<i>PL2A</i>	<i>No py</i>	175	75	75	43	0.67	2.2	12.93	43	168	270	83	3.9	127	191±15	-	-	-	-
<i>PL2B</i>	<i>1bf</i>	225	138	138	73	0.82	9.7	14.23	39	172	270	80	4.4	145	178±14	-	-	-	-
<i>PL2C</i>	<i>1+1</i>	300	150	125	73	0.81	10.2	16.61	42	179	311	85	4.3	160	198±16	-	-	-	-
<i>PL2D</i>	<i>1bf</i>	188	100	88	50	0.73	3.5	8.78	28	107	301	53	3.9	134	183±15	-	-	-	-
<i>PL2E</i>	<i>1py</i>	163	88	88	47	0.70	2.4	14.09	46	175	270	88	3.8	132	189±15	-	-	-	-
<i>PL2F</i>	<i>1bf</i>	275	200	188	98	0.87	22.8	9.38	24	89	285	46	3.7	169	195±16	-	-	-	-
<i>PL2G</i>	<i>2bf</i>	325	200	163	92	0.85	21.8	16.04	38	160	321	76	4.2	172	202±16	-	-	-	-
PL2bisB	No py	225	138	119	75	0.81	11.8	13.66	34	134	97	66	3.9	163	202±16	452	303	68	16070
PL2bisC	1bf	163	138	125	70	0.82	8.9	18.41	40	161	129	79	4.0	192	234±19	855	86	36	12731
PL3																			
PL3-B	1+1	175	125	88	49	0.74	3.0	25.30	87	360	353	173	4.2	120	162±13	701	292	321	11534
<i>PL3A</i>	<i>1+1</i>	188	100	88	50	0.73	3.0	65.69	152	726	1223	326	4.8	165	226±18	-	-	-	-
<i>PL3B</i>	<i>2bf</i>	163	88	88	51	0.82	3.4	22.75	64	305	306	137	4.8	136	166±13	-	-	-	-
<i>PL3C</i>	<i>1bf</i>	150	100	100	55	0.78	3.9	44.15	106	548	633	238	5.2	153	196±16	-	-	-	-
<i>PL3D</i>	<i>1pyr</i>	250	125	113	50	0.72	4.2	35.80	109	489	513	226	4.5	130	182±15	-	-	-	-
PL3bisA	No py	188	144	113	71	0.79	9.7	13.18	29	162	118	68	5.6	160	202±16	479	213	224	16625
PL3bisB	1bf	200	119	88	60	0.78	6.7	12.33	35	150	162	71	4.3	143	182±16	496	215	72	13215
PL3bisC	1bf	156	138	125	69	0.82	8.6	9.49	27	106	125	52	3.9	149	182±15	529	223	144	12074
PL4																			
PL4-E	2py	213	113	100	59	0.79	3.9	81.36	240	1320	291	557	5.5	120	152±12	719	118	61	13837
PL4-H	np	125	113	100	53	0.74	3.4	15.08	35	177	285	78	5.0	160	216±17	884	198-	3217-	38140-
PL4-I	2bf	138	100	100	51	0.79	3.1	23.53	41	239	334	99	5.8	196	249±20	817	117	248	39516
<i>PL4B</i>	<i>1bf</i>	128	125	100	50	0.72	4.2	26.35	76	256	480	137	3.4	158	220±18	-	-	-	-
<i>PL4D</i>	<i>1py</i>	125	88	88	41	0.66	1.5	29.34	107	485	400	224	4.5	108	165±13	-	-	-	-

<i>PL4E</i>	<i>1py</i>	200	125	113	54	0.73	3.9	25.76	81	343	487	163	4.3	130	179±14				
PL4bisC	No py	169	163	125	75	0.80	11.0	17.01	33	161	114	72	4.8	195	243±19	893	85	79	16307
PL6																			
PL6-C	1+1	188	113	94	57	0.78	4.0	29.43	65	268	245	129	4.1	187	239±19	601	279	100	14173
<i>PL6A</i>	<i>1bf</i>	188	100	88	50	0.73	3.5	10.65	33	127	167	57	3.9	139	189±15	-	-	-	-
<i>PL6B</i>	<i>No py</i>	300	100	100	59	0.75	6.8	15.46	43	142	150	77	3.3	165	219±18	-	-	-	-
<i>PL6D</i>	<i>1bf</i>	188	150	125	66	0.80	7.4	21.68	50	221	174	88	4.4	173	216±17	-	-	-	-
<i>PL6E</i>	<i>1bf</i>	225	200	175	89	0.85	16.9	16.81	40	158	150	78	4.0	177	207±17	-	-	-	-
<i>PL6F</i>	<i>1+1</i>	188	113	113	59	0.78	4.4	18.99	52	199	150	100	3.8	157	201±16	-	-	-	-
PL6bisA	1bf	213	125	119	71	0.82	10.1	8.77	22	56	81	36	2.6	202	247±20	443	275	247	5388
PL6bisB	No py	125	94	75	47	0.71	2.8	7.72	23	56	128	36	2.5	173	245±20	436	299	376	4554
PL6bisC	No py	131	125	113	61	0.77	5.9	12.66	32	141	162	66	4.5	159	207±18	1017	321	258	15340
PL6bisD	No py	150	100	88	53	0.74	4.2	5.20	17	43	106	27	2.6	156	211±17	481	372	2817	5878
PL7																			
PL7-AA	1+1	163	125	100	58	0.79	3.9	10.68	39	11	155	42	0.3	208	263±21	2243	170	145	2047
PL7-BB	1py	138	94	88	47	0.71	2.1	12.66	48	13	241	51	0.3	201	284±23	3112	200	114	2912
<i>PL7A</i>	<i>No py</i>	125	100	100	52	0.74	3.1	0.29	1.3	0.6	139	2	0.5	145	196±16	-	-	-	-
<i>PL7B</i>	<i>1bf</i>	175	125	88	59	0.82	4.9	6.27	32.3	1.2	151	32	0.0	158	192±15	-	-	-	-
PL8																			
<i>PL8C</i>	<i>1bf</i>	188	113	88	50	0.73	3.7	22.05	56	218	212	128	3.9	167	228±18	-	-	-	-
<i>PL8D</i>	<i>1pyr</i>	275	125	125	70	0.79	8.4	20.36	38	150	250	74	4.0	225	286±23	-	-	-	-
<i>PL8E</i>	<i>1pyr</i>	188	100	88	50	0.71	3.0	16.09	51	177	277	84	3.5	142	199±16	-	-	-	-
PL8bisA	No py	200	88	88	59	0.76	10.4	5.60	16	64	89	32	3.9	147	193±15	590	283	258	12756
PL8bisB	1pyr	250	150	150	74	0.81	19.7	7.00	19	59	57	33	3.1	173	215±17	588	275	113	8439
PL8bisD	No py	188	100	75	52	0.73	4.5	8.84	22	76	152	41	3.4	180	246±20	-	-	-	-
PL8bisE	No py	138	113	100	52	0.73	3.3	19.79	48	180	262	91	3.7	179	245±20	492	297	290	9097
PL8bisC	No py	156	106	88	48	0.71	3.0	26.82	70	283	331	138	4.1	160	225±18	579	339	2822	13704
PL8sterA	No py	150	125	88	58	0.75	5.3	10.62	31	122	176	60	3.9	145	192±15	6278	283	236	13040
PL 10																			
PL10-A	1+1	113	69	69	36	0.67	1.0	3.09	12.8	30.3	306	20	2.4	123	185±15	959	382	196	5909
PL10-B	2bf	113	100	88	42	0.76	1.7	1.39	7.3	19.6	203	12	2.7	93	123±10	645	382	128	3430
PL10-D	2bf	113	63	63	34	0.67	1.0	1.54	4.7	18.5	250	9	3.9	135	201±16	1709	412	774	5233
PL10-F	1+1	150	63	63	36	0.65	1.2	0.72	4.1	10.5	199	7	2.6	87	134±11	681	454	163	3093
PL10-I	2py	275	100	94	58	0.76	4.7	2.27	10.5	34.2	157	19	3.3	99	131±10	692	816	109	5163
PL10-L	1+1	163	113	100	55	0.78	3.5	0.80	3.9	7.8	129	6	2.0	111	142±11	740	386	122	4053
<i>PL10A</i>	<i>1bf</i>	169	138	125	64	0.80	6.4	0.29	1.1	6.1	104	2	5.6	90	113±9	-	-	-	-
<i>PL10B</i>	<i>2bf</i>	138	100	88	47	0.75	2.6	1.10	5.0	23.3	100	11	4.6	85	113±9	-	-	-	-
<i>PL10G</i>	<i>1bf</i>	113	88	88	44	0.71	2.0	0.70	4.1	21.4	100	9	5.3	62	87±7	-	-	-	-
PL 11																			
PL11-C	1+1	225	88	88	50	0.74	3.5	36.36	100	278	707	167	2.8	179	243±19	-	-	-	-

PL11E	1+1	250	181	175	88	0.87	14.0	15.06	57.7	7.8	185	60	0.1	204	236±19	-	-	-	-
<i>PL11A</i>	<i>2bf</i>	238	188	113	62	<i>0.81</i>	8.3	<i>0.49</i>	<i>1.9</i>	2.5	239	1	1.3	144	178±14	-	-	-	-
<i>PL11B</i>	<i>1bf</i>	213	200	138	73	<i>0.83</i>	10.9	9.98	42.5	5.2	207	31	0.1	183	220±18	-	-	-	-
PL11-A	1+1	175	125	125	62	0.81	4.9	14.17	67.1	1.9	150	68	0.0	171	211±17	-	-	-	-

\* are alpha ejection corrected age.

Italic samples and results refer to data were a mean value of Sm content as been used in the AHe age calculation.

ΣREE is the sum of analyzed rare earth elements: La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Ho, Er, Tm, Yb and Lu.

**Table 4:** Apatite mean composition (wt%).

<i>Sample name</i>	<i>P<sub>2</sub>O<sub>5</sub></i>	<i>CaO</i>	<i>F</i>	<i>Cl</i>	<i>SO<sub>3</sub></i>	<i>SiO<sub>2</sub></i>	<i>FeO</i>	<i>MgO</i>	<i>MnO</i>	<i>Na<sub>2</sub>O</i>	<i>Y<sub>2</sub>O<sub>3</sub></i>	<i>La<sub>2</sub>O<sub>3</sub></i>	<i>Ce<sub>2</sub>O<sub>3</sub></i>	<i>Sm<sub>2</sub>O<sub>3</sub></i>	<i>SrO</i>	<i>Pr<sub>2</sub>O<sub>3</sub></i>	<i>Nd<sub>2</sub>O<sub>3</sub></i>	<i>(OH)</i>	<i>Total</i>
PL 1	40.59	54.16	3.75	0.01	0.02	0.56	0.06	0.01	0.09	0.04	0.39	0.14	0.45	0.01	0.00	0.08	0.36	0.00	100.7
	±0.45	±0.36	±0.25	±0.01	±0.03	±0.15	±0.04	±0.01	±0.02	±0.03	±0.09	±0.07	±0.18	±0.01	±0.01	±0.05	±0.13	±0.00	
PL 2	40.90	54.09	3.63	0.02	0.01	0.64	0.06	0.01	0.04	0.03	0.18	0.29	0.77	0.01	0.01	0.10	0.43	0.00	101.2
	±0.51	±0.47	±0.11	±0.00	±0.01	±0.22	±0.02	±0.01	±0.02	±0.03	±0.06	±0.11	±0.25	±0.01	±0.02	±0.05	±0.14	±0.00	
PL 3	41.17	54.13	3.68	0.02	0.02	0.66	0.07	0.01	0.05	0.03	0.23	0.24	0.69	0.01	0.01	0.11	0.44	0.00	101.6
	±0.49	±0.50	±0.13	±0.01	±0.05	±0.24	±0.04	±0.01	±0.01	±0.02	±0.09	±0.10	±0.26	±0.01	±0.02	±0.06	±0.15	±0.00	
PL 4	40.52	53.31	3.81	0.01	0.04	0.67	0.09	0.00	0.10	0.13	0.93	0.11	0.41	0.01	0.00	0.07	0.40	0.04	100.7
	±0.68	±0.52	±0.16	±0.01	±0.14	±0.29	±0.13	±0.01	±0.02	±0.03	±0.32	±0.06	±0.19	±0.01	±0.01	±0.05	±0.13	±0.13	
PL 6	40.41	54.11	3.59	0.01	0.02	0.53	0.10	0.01	0.05	0.01	0.16	0.20	0.53	0.01	0.02	0.07	0.31	0.00	101.5
	±0.69	±0.60	±0.15	±0.01	±0.05	±0.29	±0.04	±0.01	±0.01	±0.01	±0.09	±0.10	±0.28	±0.02	±0.04	±0.05	±0.17	±0.00	
PL 8	42.51	54.90	3.66	0.02	0.01	0.52	0.06	0.01	0.06	0.01	0.18	0.16	0.48	0.01	0.03	0.07	0.31	0.00	103.0
	±0.68	±0.51	±0.17	±0.00	±0.01	±0.28	±0.02	±0.01	±0.02	±0.01	±0.10	±0.09	±0.25	±0.01	±0.04	±0.05	±0.16	±0.00	
PL 10	41.45	54.59	3.45	0.16	0.01	0.17	0.14	0.02	0.12	0.06	0.09	0.12	0.29	0.00	0.09	0.03	0.15	0.00	100.9
	±0.39	±0.07	±0.29	±0.09	±0.01	±0.04	±0.04	±0.01	±0.03	±0.02	±0.02	±0.02	±0.04	±0.00	±0.02	±0.05	±0.03	±0.00	
PL 11	41.42	54.06	3.90	0.01	0.01	0.01	0.42	0.07	0.52	0.12	0.25	0.04	0.11	0.00	0.10	0.02	0.08	0.00	101.1
	±0.37	±0.27	±0.26	±0.01	±0.01	±0.02	±0.12	±0.02	±0.10	±0.02	±0.05	±0.03	±0.03	±0.01	±0.02	±0.03	±0.03	±0.00	

Here the mean and standard deviation of individual grain composition are presented. EMP analyses were carried out on 50 grains per sample.

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