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- 1 Alteration of magmatic monazite in granitoids from the Ryoke belt (SW Japan):
- 2 processes and consequences
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20 ABSTRACT

21 The alteration of magmatic monazite and its consequences for monazite geochronology are 22 explored in granitoids from the western part of the Rvoke belt (Iwakuni-Yanai area, SW 23 Japan). Biotite-granite samples were collected in two plutons emplaced slightly before the 24 main tectono-metamorphic event: one, a massive granite (Shimokuhara) adjoins schistose 25 rocks affected by greenschist facies metamorphism; and the second, a gneissose granite 26 (Namera) adjoins migmatitic gneiss that experienced upper-amphibolite facies conditions. 27 Despite contrasting textures, the granite samples have similar mineral modes and 28 compositions. Monazite in the massive granite is dominated by primary domains with limited 29 secondary recrystallization along cracks and veinlets. It is variably replaced by 30 allanite+apatite±xenotime±Th-U-rich phases. The outermost rims of primary domains yield a 31 weighted average 206 Pb/ 238 U date of 102 ± 2 Ma while the Th–U phases show Th–U–Pb dates 32 of 58 ± 5 and $15-14 \pm 2-3$ Ma. Monazite in the gneissose granite preserves sector- or 33 oscillatory-zoned primary domains cross-cut by secondary domains enriched in Ca, Y, U, P and containing numerous inclusions. The secondary domains preserve concordant ²⁰⁶Pb/²³⁸U 34 35 dates spreading from 102 ± 3 to 91 ± 2 Ma while primary domain analyses are commonly 36 discordant and range from 116 to 101 Ma.

37 Monazite alteration textures in the two granites chiefly reflect differences in their post-38 magmatic histories. In the massive granite, monazite replacement occurred via a nearly 39 stoichiometrically-balanced reaction reflecting interaction with an aqueous fluid enriched in 40 Ca+Al+Si±F during hydrothermal alteration of the granitic assemblage, likely below 500 °C. 41 In the gneissose granite a small amount of anatectic melt, probably derived from the 42 neighboring metasedimentary rocks, was responsible for a pseudomorphic recrystallization of 43 monazite by dissolution-reprecipitation above 600 °C. Regardless of whether monazite 44 underwent replacement or recrystallization, primary monazite domains preserve the age of

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45 magmatic crystallization for both plutons (102 ± 2 Ma and 106 ± 5 Ma). Conversely, the age 46 of monazite alteration is not easily resolved. Monazite replacement in the massive granite 47 might be constrained using the Th-U-rich alteration products; with due caution and despite probable radiogenic Pb loss, the oldest date of 58 ± 5 Ma could be ascribed to chloritization 48 during final exhumation of the granite. The spread in apparently concordant ²⁰⁶Pb/²³⁸U dates 49 50 for secondary domains in the gneissose granite is attributed to incomplete isotopic resetting 51 during dissolution-reprecipitation, and the youngest date of 91 ± 2 Ma is considered as the age 52 of monazite recrystallization during a suprasolidus metamorphic event. These results reveal a 53 diachronous, ca. 10 Ma-long HT history and an overall duration of about 15 Ma for the 54 metamorphic evolution of the western part of the Ryoke belt. 55

56 Keywords: Monazite replacement, monazite recrystallization, dissolution-reprecipitation,

57 partial melting, LA-ICP-MS geochronology, Ryoke plutono-metamorphic belt

58 INTRODUCTION

59 Monazite, a lanthanide- and actinide-bearing phosphate, is a valuable petrological and 60 geochronological tool. In metamorphic rocks, it can be used to decipher the age and 61 Pressure–Temperature (P-T) conditions of distinct orogenic events (e.g. Williams et al. 1999; 62 Foster et al. 2004; Engi 2017) while in magmatic rocks it is useful for dating pluton 63 emplacement (e.g. Harrison et al. 1995; Be Mezeme et al. 2006), especially in cases where 64 zircon dates appear unreliable (Kusiak et al. 2004; Piechocka et al. 2017). However, monazite 65 is also known to be a versatile mineral (e.g. Catlos, 2013), so its petrochronological potential 66 must be carefully assessed.

67 In granitoids from the Ryoke belt (SW Japan), age dating of different samples from 68 the same pluton revealed a discrepancy between magmatic zircon and monazite dates. There are cases of ²⁰⁶Pb/²³⁸U zircon dates being older than Th–U–Pb monazite dates (Skrzypek et al. 69 70 2016), and vice versa (Takatsuka et al. 2018a, b). When monazite dates are younger than 71 zircon's, it is commonly suspected that monazite reacted during post-magmatic processes, as 72 the mineral is prone to fluid-induced compositional modifications and/or age resetting (e.g. 73 Kelly et al. 2012; Seydoux-Guillaume et al. 2012; Villa and Williams 2013; Taylor et al. 74 2014). Thus, our objective was to identify monazite alteration textures and document possible 75 age discrepancies when zircon and monazite from the same sample are dated with the same 76 method.

The monazite-bearing granitoid samples described by Skrzypek et al. (2016) for zircon dating (Iwakuni-Yanai area, western part of the Ryoke belt) represent a suitable material, as they contain monazite with common alteration textures and different age populations. In this contribution we document in detail the post-magmatic monazite alteration textures and evaluate their impact on monazite's geochronological record. We focus on two granite samples that have (1) crystallization ages slightly older (~5 Ma) than the main (regional)

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83 tectono-metamorphic event recorded in the neighboring metasedimentary rocks, (2) similar 84 whole-rock compositions, and (3) contrasting monazite textures. The massive Shimokuhara 85 granite lies next to schistose rocks where regional conditions did not exceed ~2.5 kbar/550 °C (Ikeda 2004), whereas the gneissose Namera granite adjoins migmatitic gneissose rocks where 86 regional metamorphism reached ~3 kbar/675 °C (Ikeda 2004; Fig. 1). As such, these two 87 88 granites provide an opportunity to explore the response of magmatic monazite to post-89 emplacement metamorphism of varying grade. First, we report the mineralogy, texture and 90 mineral mode of biotite-granite samples from both plutons to assess their degree of similarity. 91 We then document their monazite textures, and date monazite in situ by laser ablation 92 inductively-coupled mass spectrometry (LA-ICP-MS). The results are used to identify the 93 processes involved in the alteration of magmatic monazite and the consequences for age 94 dating of magmatic/metamorphic events.

95 BACKGROUND: ALTERATION OF MAGMATIC MONAZITE

96 The alteration of magmatic monazite follows two mechanisms: *replacement* by other phases 97 and/or recrystallization of new monazite. The most commonly observed monazite 98 replacement texture in (meta-)igneous rocks involves the peripheral growth of allanite, apatite 99 and minor Th–U-bearing phases following a nearly balanced reaction (e.g. Murata et al. 1957; 100 Broska and Siman 1998). Interestingly, monazite replacement by allanite and/or apatite is 101 reported in granitoids that underwent no metamorphism (Broska et al. 2005), greenschist 102 facies hydrothermal alteration (~340 °C; Ward et al. 1992; Poitrasson et al. 2000), and lower-(5-6 kbar/550-600°C; Finger et al. 1998) to upper-amphibolite facies metamorphism (4-8 103 104 kbar/685-825°C; Broska et al. 2005). An extreme example of monazite replacement by 105 Aln-Ap at the magmatic stage is attributed to an increase in the Ca content of the melt (Dini 106 et al. 2004). Other cases of monazite replacement at low temperature (290-350 °C) involve

the growth of pyrite + brockite (Poitrasson et al. 2000) or thorite + fluorapatite + synchysite
(Didier et al. 2013) assemblages.

109 Monazite recrystallization corresponds to the formation of new domains with a 110 composition that differs from the initial magmatic grain. In this case a balanced reaction is 111 difficult to infer and it is only possible to document the relative element enrichment/depletion 112 with respect to primary monazite. Recrystallization may result in monazite evolving towards a 113 cheralitic [Ca_{0.5}Th_{0.5}PO₄], huttonitic [ThSiO₄], Heavy REE (HREE)-richer or U-richer 114 composition (Jefferies 1985; Ward et al. 1992; Poitrasson et al. 1996; 2000; Townsend et al. 115 2000; Didier et al. 2013). Again, monazite recrystallization is observed across a wide range of 116 P-T conditions; it is ascribed to relatively low-T chloritization or greisenization (Jefferies 117 1985; Ward et al. 1992; Poitrasson et al. 1996; 2000; Didier et al. 2013) and high-T, late- to 118 post-magmatic fluid-mineral interaction (Hawkins and Bowring 1997). There is no evident 119 link between P-T conditions and the composition of newly-formed monazite, but the extent 120 of recrystallization seems to increase with metamorphic grade. Townsend et al. (2000) 121 observed that monazite recrystallization occurs along micron-scale veins in low-grade samples (~1.5 kbar/< 350-400 °C), whereas larger patchy domains are formed at higher grade 122 123 (~3-4 kbar/400-700 °C).

124 GEOLOGICAL OUTLINE

The Ryoke belt is an association of Cretaceous plutonic and metamorphic rocks located in SW Japan (e.g. Nakajima 1994). The Iwakuni-Yanai area (W part of the Ryoke belt) exposes Late Jurassic accretionary-wedge sediments metamorphosed along a low-*P*/high-*T* gradient and intruded by abundant granitoids (Fig. 1; e.g. Brown 1998). From north to south, the weaklymetamorphosed sediments transition to a low-grade schist zone and then a high-grade gneiss zone (Fig. 1; Higashimoto et al. 1983). The tectono-metamorphic history involves an initial contact metamorphic event followed by the growth of syn-tectonic parageneses that are used

to define the E-W trending, so-called regional metamorphic zones (Fig. 1; Ikeda 1998;

133 Okudaira et al. 2001; Skrzypek et al. 2016).

134 For metamorphic rocks, U-Pb zircon dating in the highest-grade Grt-Crd zone 135 brackets the timing of suprasolidus conditions between 103 and 97 Ma (Nakajima et al. 2013; 136 Skrzypek et al. 2016). Metamorphic monazite in the Kfs-Crd and Grt-Crd zones shows 137 features that indicate growth at mainly prograde to peak temperature conditions (600-800 138 °C), but U-Pb monazite dates ranging from ca. 100 down to 85 Ma are obtained, so that the 139 persistence of medium-T conditions (> 200-300 °C) until 85 Ma was proposed (Skrzypek et 140 al. 2018). U-Pb zircon ages for granitoids, in agreement with structural and petrological 141 observations, indicate pluton intrusion before (105-100 Ma), during (~100 Ma) and after (~96 142 Ma) the regional metamorphic peak (Fig. 1; Herzig et al. 1998; Nakajima et al. 2013; 143 Skrzypek et al. 2016). The present study focuses on the granitoids (Shimokuhara and Namera) 144 emplaced in the sediments before the metamorphic peak, and therefore affected by subsequent 145 regional metamorphism.

146 The Shimokuhara granite abuts schistose rocks of the Bt and Ms-Crd metamorphic 147 zones (Fig. 1). It is a massive, biotite-bearing granite that commonly shows biotite 148 chloritization, and locally contains K-feldspar phenocrysts, garnet, tourmaline and allanite as 149 primary phases (Murakami 1970; Higashimoto et al. 1983; Moutte and Iiyama 1984). The 150 contact with metasedimentary rocks gently dips to the east, and a metamorphic aureole 151 defined by andalusite porphyroblasts extends up to ~ 1 km east of the exposed contact 152 (Skrzypek et al. 2016). The pluton is thought to have intruded at a maximum depth of 2–3 km 153 (Murakami 1970). Peak P-T estimates for regional metamorphism in the neighboring 154 schistose rocks range from 1 kbar/450 °C to 2.5 kbar/550 °C (Ikeda 2004). For a biotite-155 granite sample, U-Pb dating of sector- and oscillatory-zoned zircon by LA-ICP-MS revealed 206 Pb/ 238 U dates ranging from 108 to 102 Ma, with 105 ± 3 Ma being taken as the age of 156

granite intrusion (Skrzypek et al. 2016). Monazite dating of two granite samples by the chemical isochron method (CHIME) yielded 87.3 ± 1.6 and 86.6 ± 2.1 Ma, and these results were also regarded as the age of granite intrusion (Suzuki et al. 1996). K–Ar dating on biotite from a two-mica granodiorite sample gave 87 Ma (data from Kawano and Ueda 1966, recalculated by Higashimoto et al. 1983).

162 The Namera granite lies next to gneissose rocks of the Kfs-Crd zone (Fig. 1). It is 163 mainly a muscovite-biotite-bearing granite with a moderate gneissosity defined by aligned 164 biotite, but it locally shows a granodioritic or leucocratic facies. Accessory garnet and fibrous 165 sillimanite are reported, with pinitized cordierite observed in the leucocratic facies (Nureki 166 1974; Higashimoto et al. 1983). The contact with metasedimentary rocks gently dips towards 167 the east; isolated granite outcrops within the metasedimentary succession (Kfs-Crd zone; not 168 shown in Fig. 1) suggest that the Namera granite extends eastwards and lies a few meters 169 below the metasediments. Peak P-T conditions of regional metamorphism in the neighboring 170 gneissic rocks are estimated at 2-3 kbar/590-675 °C (Ikeda 2004). For a biotite-granite sample, U-Pb dating of oscillatory-zoned zircon by LA-ICP-MS vielded ²⁰⁶Pb/²³⁸U dates 171 172 between 107 and 100 Ma, with 105 ± 2 Ma being regarded as the age of granite intrusion 173 (Skrzypek et al. 2016). Monazite CHIME dates for three granite samples cluster at $91-89.9 \pm$ 174 3.2 Ma and are also ascribed to granite intrusion (Suzuki et al. 1996). A K-Ar biotite date of 175 86.6 ± 4.3 Ma was obtained from a hornblende-biotite granite sample (Higashimoto et al. 176 1983).

177 ANALYTICAL METHODS

Two samples were collected for each granite and renamed for simplicity. The main samples for the Shimokuhara granite (EY99A, renamed SHK) and Namera granite (EY117A, renamed NAM) are the same as those described by Skrzypek et al. (2016) for U–Pb zircon dating. They are used for both petrological descriptions and monazite geochronology. These samples

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located close to the pluton margins were selected to explore the effects of post-emplacement metamorphism whose P-T conditions are recorded in the adjacent metamorphic rocks. The additional samples (EY152A, renamed SHK2; EY140A, renamed NAM2) were collected slightly farther from the pluton margin (100–300 m) and are used for comparing monazite textures and compositions. Sample locations are indicated on Figure 1.

187 Mineral compositions were analyzed at Kyoto University and at the University of Graz. In order to use a low beam current (500 pA), chlorite was measured on a scanning 188 189 electron microscope (Hitachi S3500H, Kyoto) equipped with an energy-dispersive system 190 (EDS) at 20 kV. All other minerals were measured with a JEOL JXA-8105 (Kyoto) and a 191 JEOL JXA-8530F Plus (Graz) electron microprobe (EMP) at 15 kV and with a 3µm beam 192 diameter. Analyses of all silicates except Aln were done at 10 nA and corrected with the ZAF 193 method, while those of REE±Th±U-bearing minerals were done at 150 nA and corrected 194 following the Bence and Albee (1968) method with the updated α -factor table of Kato (2005). 195 The analytical procedure for REE±Th±U-bearing minerals followed published guidelines (e.g. 196 Scherrer et al. 2000; Pyle et al. 2005; Suzuki and Kato 2008) and is available as Supplemental 197 Material together with the full set of monazite analyses. The REE measured for most 198 monazite analyses are La, Ce, Pr, Nd, Sm and Gd, but several analyses with the additional 199 HREE Tb to Yb were performed for Mnz-Xtm thermometry. Monazite [A4(TO4)4] is described according to A-site (Ca²⁺, REE³⁺, Y³⁺, Th⁴⁺, U⁴⁺, Pb²⁺) and T-site (P⁵⁺, Si⁴⁺) 200 201 cations. Its composition is expressed according to the end-members X_{mnz} [($\Sigma_{REE}+Y$)/A], X_{cher} 202 [2Ca/A] and X_{hut} [(Th+U+Pb-Ca)/A], where A represents the sum of A-site cations. For 203 Mnz–Xtm thermometry, $X_{(HREE,Y)}$ in monazite is defined as $[(\Sigma_{HREE}+Y)/A]$, with Σ_{HREE} 204 including elements from Gd to Yb. For EMP age dating of the Th-U phases, the accuracy was checked with several analyses of "ekanite" from Okkampitiya, Sri Lanka (207Pb/206Pb 205

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age=562.1 \pm 0.8 Ma; Nasdala et al. 2017) that yielded a weighted average Th–U–Pb age of 564 \pm 6 Ma (n=5; MSWD=0.15).

Several calibrations are available for Mnz–Xtm thermomety. While the calibration of Seydoux-Guillaume et al. (2002) considers the effect of Th, it is derived for a relatively high temperature range (800–1100 °C). We prefer to use the calibrations of Heinrich et al. (1997) and Pyle et al. (2001) as they rely on natural samples for a lower temperature range (400–700 °C). These two calibrations differ, probably because they are based on samples from different metamorphic gradients, and we use both to obtain the range of likely temperature conditions associated with the growth of Mnz–Xtm pairs in direct contact.

215 Electron microprobe element mapping of thin sections (~ 2 x 3 cm areas) was performed at 15 kV, 60 nA, 10 µm beam diameter, 15 µm step size and 4 ms dwell time for 216 217 the CaK α , AlK α , PK α , FeK α , NaK α lines and the BSE signal. An in-house Matlab \odot routine 218 was used to identify the different minerals based on image analysis of the combined maps, 219 and to calculate modal abundances (Table 1). The granite mesonorm (Table 1) was calculated 220 following the procedure of Mielke and Winkler (1979) implemented in GCDkit (Janoušek et 221 al. 2006) using whole-rock analyses of Skrzypek et al. (2016) and with all Fe expressed as 222 FeO so as to exclude normative magnetite, since both granites belong to the ilmenite series of 223 Ishihara (1977).

Monazite dating by LA-ICP-MS was performed at Gakushuin University, Tokyo, on separated grains mounted in epoxy, using a NWR-213 Nd:YAG laser with a 10 μ m spot diameter. The ablated material was carried to an Agilent 8800 single-collector tandem quadrupole ICP-MS for monitoring ²⁰⁴(Pb+Hg), ²⁰⁶Pb, ²⁰⁷Pb, ²⁰⁸Pb, ²³²Th¹⁶O and ²³⁸U¹⁶O. Fifteen analyses of unknowns were bracketed by five measurements of 44069 monazite (Aleinikoff et al. 2006) and three of NIST SRM 610 glass (Pearce et al. 1997) for normalization. Namaqualand monazite (1034.4 ± 5.8 Ma concordia age by LA-sector field-

231 ICP-MS; Goncalves et al. 2016) was used as a consistency standard and concordant data 232 yielded the following weighted average ages (n=3 concordant analyses out of 5 sequences): 206 Pb/ 238 U age=1061 ± 39 Ma (MSWD=1.40), 207 Pb/ 235 U age=1072 ± 47 Ma (MSWD=3.20), 233 206 Pb/ 207 Pb age=1092 ± 91 (MSWD=5.20). Error calculations used the standard error (SE, 2 σ 234 235 level) in order to take into account the large number of primary reference material analyses. 236 Considering the typical uncertainty on Pb/U ratios, monazite analyses with a concordance ratio [U–U cc(%)=(²⁰⁶Pb/²³⁸U date)/(²⁰⁷Pb/²³⁵U date)*100] between 97 and 103% are referred 237 238 to as concordant. The results and full analytical procedure are available as Supplemental 239 Material. Data were plotted with Isoplot 3.76 (Ludwig 2004).

240 SAMPLE DESCRIPTION

241 Samples SHK & SHK2

242 The main Shimokuhara granite sample (SHK) was collected at the eastern margin of the 243 pluton, a few meters away from the contact with schistose rocks from the Bt zone (Fig. 1). 244 There, the Shimokuhara pluton exhibits a leucocratic, biotite-bearing facies with some K-245 feldspar phenocrysts (~2 cm in length), plagioclase and quartz aggregates with a noticeable 246 rounded shape (Fig. 2a). Centimeter-scale xenoliths of the neighboring metasediments and 247 small garnet (< 1 mm in diameter) are also observed in the granite. Sample SHK is massive 248 and contains Qtz, Kfs, Pl, Bt with accessory Grt, Ap, Zrn, Mnz, Xtm and secondary Ms, Chl, 249 Ep, Aln, Ttn.

In thin section, large (up to 1 cm in diameter) and rounded aggregates of quartz with undulatory extinction dominate (Fig. 2a). K-feldspar phenocrysts ($Or_{87-93}Ab_{7-13}$) contain albite rods and locally show the microcline twin. The core of euhedral and concentrically zoned plagioclase ($An_{18-47}Ab_{52-81}Or_1$) is replaced by fine-grained brownish material, probably damourite (Fig. 2a, c). Biotite flakes (X_{Fe} =0.68–0.71; F=0.27–0.68 wt%) are interstitial and

variably replaced by chlorite (brunsvigite, X_{Fe} =0.67–0.70), K-feldspar, muscovite (F=0.27–0.40 wt%) and titanite (Fig. 2c). Garnet (up to 400 µm in diameter) is included in Kfeldspar phenocrysts and has virtually no inclusion. No mineral preferred orientation is visible under the optical microscope (Fig. 2a).

259 Sample SHK2 was collected 300 m from the mapped contact with metasedimentary 260 rocks. It is a massive granite with Qtz, Kfs, Pl, Bt and accessory Chl, Ilm, Rt, Mnz, Xtm, Zrn,

Aln, Ap. Biotite chloritization appears to be less pervasive than in sample SHK.

262 Samples NAM & NAM2

263 The main Namera granite sample (NAM) was collected less than a hundred meters away from 264 the contact with migmatitic gneissose rocks from the Kfs-Crd zone (Fig. 1). There, the 265 Namera pluton shows a leucocratic, fine- to coarse-grained biotite-bearing facies and locally 266 contains muscovite, garnet, greenish pinitized cordierite and sillimanite. Several outcrops in 267 the eastern part of the pluton show a weak to moderate gneissosity defined by biotite 268 alignment, but an undeformed facies resembling the Shimokuhara granite is also observed 269 (Fig. 1). The granite is locally cut by cm-thick quartz veins, garnet-bearing aplitic veins and Qtz-Kfs-Ms pegmatites, the latter being nearly parallel to the gneissosity. The outcrop of 270 271 sample NAM shows cm-thick, garnet-bearing leucocratic veins mostly parallel and locally 272 oblique to the gneissosity. A NE-SW striking fault is mapped in the vicinity of the sampling 273 locality, but no signs of it are visible on the outcrop. Sample NAM has a moderate gneissosity 274 (Fig. 2b) and contains Qtz, Kfs, Pl, Bt with accessory (pinitized) Crd, Grt, Ap, Zrn, Mnz, Xtm 275 and secondary Ms, Chl, Hem.

276 K-feldspar phenocrysts ($Or_{80-90}Ab_{10-20}$) exhibit a straight to wavy outline, are locally 277 perthitic and contain albite stringlets. Plagioclase ($An_{05-39}Ab_{60-94}Or_{01-02}$) is found as large 278 crystals (up to 0.5 cm in length), small polygonal grains (~100 µm in diameter) or in fine-279 grained intergrowths with secondary muscovite. Plagioclase locally shows an irregular outline

280 and its core is replaced by skeletal muscovite and minor biotite (Fig. 2b). Quartz shows 281 undulatory extinction and forms slightly elongated ribbons parallel to the gneissose fabric 282 (Fig. 2b). Biotite (X_{Fe}=0.59-0.63; F=0.34-0.56 wt%) appears as equant flakes or elongate 283 laths that define the gneissosity, and is slightly replaced by chlorite (brunsvigite, $X_{Fe}=0.63$) 284 (Fig. 2b). Garnet (up to 200 µm in diameter) is included in plagioclase. Pools of fine-grained 285 plagioclase and Ms-Bt intergrowths surround minute relicts of pinitized cordierite (200 µm in 286 length). The textural position of both garnet and former cordierite suggests they are of 287 magmatic origin. Secondary muscovite (F=0.17-0.29 wt%) is preferentially developed inside 288 K-feldspar where it forms a network of Ms-Qtz veins (Fig. 2d). A few myrmekites are 289 observed at Kfs-Pl contacts.

Sample NAM2 is a gneissose granite from an outcrop where no leucocratic veins or
pegmatites are observed. It contains Qtz, Kfs, Pl, Bt and accessory Grt, pinitized Crd, Ms,
Chl, Mnz, Xtm, Zrn, Ap. The gneissose fabric is slightly stronger than in sample NAM.

293 Comparison of mineral modes

294 The whole-rock data published by Skrzypek et al. (2016) show that, despite a different appearance, samples SHK and NAM have a similar major and trace element composition. For 295 296 further comparison, mineral modal abundances were calculated using whole-rock analyses 297 and EMP mapping of thin sections (Table 1). Granite mesonorm calculations indicate a 298 similar amount of Kfs, Qtz, Pl and Bt in SHK and NAM (Table 1). Element mapping of \sim 3.5 299 x 2 cm thin sections fails to accurately determine the modal amount of phenocrystic phases 300 (Kfs, Qtz, Pl), but can be used for the smaller phyllosilicates. It reveals that the modal amount 301 of Bt in NAM is almost equal to that of Bt plus Chl in SHK (Table 1). Since textural 302 observations indicate that chlorite forms at the expense of biotite, it can be deduced that the 303 original amount of biotite in both samples was identical, as suggested by the mesonorm 304 calculations.

305 PETROLOGY OF MONAZITE AND ASSOCIATED PHASES

306 Sample SHK

307 Monazite forms euhedral to skeletal grains (\sim 50 to 250 µm in length) with generally few 308 inclusions (Fig. 3a). It is located inside biotite or more commonly at the boundary between 309 biotite and quartz/plagioclase. Monazite grains that were separated from the hand specimen 310 are mostly euhedral, and monazite observed in thin section tends to be more subhedral to 311 anhedral. Large euhedral monazite has micro-cracks along which allanite, hematite and 312 xenotime are observed (Fig. 3b, f), and is surrounded by apatite and a fringe of zoned allanite 313 that becomes REE-poorer towards the rim (Fig. 3b, c). Skeletal monazite is surrounded by 314 large allanite (~100 μ m), small Th–U phases (< 30 μ m in diameter) and minor xenotime (Fig. 315 3c). Elongate grains of oscillatory-zoned allanite are found at chlorite rims and are locally 316 separated from chlorite by a titanite film (Fig. 3d). In general, monazite occurs together with 317 allanite in the vicinity of chloritized biotite. Xenotime has two modes of occurrence; lone 318 matrix grains form euhedral overgrowths around zircon, whereas tiny anhedral xenotime (< 319 30 µm in length) occurs next to or in contact with altered monazite (Fig. 3b, c, f).

320 Primary and secondary monazite domains are distinguished in both in situ and 321 separated grains. Primary domains have sector- or oscillatory-zoned textures that occupy the 322 majority of most monazite grains (Fig. 3e). The primary domains may be subdivided into (1) 323 core domains with a juxtaposition of BSE-bright and BSE-dark sectors, and (2) thin ($\leq 20 \, \mu m$ 324 in width) and BSE-dark outermost rim domains (Fig. 3e). The primary core and outermost rim 325 domains are differentiated by a sharp contrast on BSE images (Fig. 3e). Secondary domains 326 correspond to altered parts of the primary monazite zoning. They are generally associated 327 with the presence of cracks that cross-cut the primary zoning; on BSE images they appear as 328 darker zones up to $\sim 20 \ \mu\text{m}$ in width, or as brighter and thin (< 5 μ m) veinlets (Fig. 3f).

Monazite-(Ce) compositions are dominated by a huttonite-type substitution [REE³⁺ + 329 $P^{5+} \leftrightarrow (Th,U)^{4+} + Si^{4+}$ (Fig. 4a). The analyses define a trend with an evolution towards the 330 331 monazite end-member from the primary core (X_{hut} =0.02-0.13; X_{cher} =0.05-0.10; X_{mnz} =0.78-0.92; 48 analyses on 36 grains) to the outermost rim (X_{hut} =0.01-0.03; 332 X_{cher}=0.05-0.07; X_{mnz}=0.91-0.93; 9 analyses on 9 grains) domains (Fig. 4a, Table 2). This 333 334 evolution is due to Si + U + Th decrease and P + Light REE (LREE) increase while Y and 335 HREE remain constant. Analyses of the secondary domains follow the huttonitic trend 336 defined by primary domain analyses, with some excess in $U \pm Th \pm Pb$ being responsible for a 337 slight deviation (Fig. 4a). Secondary domain analyses define two subgroups based on Y and U 338 contents; one shows markedly low Y ($Y_2O_3=0.55-1.03$ wt%) and U ($UO_2 < 0.15$ wt%), whereas the other has Y ($Y_2O_3=1.65-3.27$ wt%) and U ($UO_2=0.32-1.22$ wt%) contents in the 339 340 range of primary domain analyses (Fig. 4b). With respect to primary domains, the Y-poor 341 subgroup of secondary domains is depleted in Y, U, Th, Ca and enriched in both LREE and 342 HREE.

343 Allanite analyses yield low oxide sums (94-97 wt%) that are ascribed to 344 metamictization, since Th-richer spots tend to have lower totals. The composition is mainly that of allanite-(Ce) with recalculated Fe³⁺/Fe_{tot} ratios of 0.04-0.32 (normalization to 25 345 negative charges) pointing to some feriallanite component, and Altot (1.99-2.27 a.p.f.u.) 346 indicating some proportion of the Ep-Czo component. The analyses of BSE-bright cores show 347 Σ_{REE} between 0.71 and 0.80 a.p.f.u., negligible actinide contents (< 0.02 a.p.f.u.) and about 348 0.08–0.14 wt% fluorine. One spot from a darker rim overgrowth has a Σ_{REE} of 0.54 a.p.f.u., 349 350 actinide abundances below detection limits and about 0.05 wt% fluorine.

Lone matrix xenotime (observed in thin section) is enriched in U and Er, Tm, Yb (i.e. HREE with an ionic radius close to that of U), whereas tiny xenotime occurring in monazite alteration textures (separated monazite grains) is enriched in Gd, Tb, Dy (i.e. HREE with an

ionic radius close to that of Th) and slightly in Ce and Th. When in direct contact with monazite, xenotime is always adjacent to the BSE-dark, Y-poor secondary domains (Fig. 3f). Six analyses of Y-poor secondary monazite domains in contact with xenotime show a nearly constant $X_{(HREE,Y)}$ (0.060 ± 0.006) and yield temperature estimates of 471 and 562 °C with the calibration of Pyle et al. (2001) and Heinrich et al. (1997), respectively.

359 The tiny Th–U phases (< 30 μ m in diameter) are not easily quantified but valid 360 analyses (not influenced by the surrounding minerals) yield a constant oxide sum of about 90 361 wt% (Table 2). They correspond to Th-rich (ThO₂=51.67-62.77 wt%) silicates with significant amounts of U (UO₂=2.14-12.82 wt%), P (P₂O₅=0.71-9.21 wt%) and Y 362 363 $(Y_2O_3=0.84-3.40 \text{ wt\%})$. Their formula approaches (Th, U, REE)(Si, P)O₄ with the missing 10 364 wt% being ascribed to free H_2O (Table 2); they can be considered as a monoclinic (huttonite; 365 note that monazite is also monoclinic) or tetragonal (thorite) polymorph of (hydrous) ThSiO₄ (Fig. 4c). Electron microprobe age dating of the Th–U phases was attempted due to their high 366 367 actinide content. The cores of euhedral grains (~30 µm in diameter) included in a K-feldspar 368 phenocryst yield Th–U–Pb dates of 369 ± 6 and 262 ± 7 Ma, whereas the rims show much 369 younger dates of $23-21 \pm 3$ Ma. These inclusions are, however, not related to monazite and 370 their ages are not discussed further. The other, generally smaller (< 20 μ m) Th–U phases 371 occur in the matrix and are always associated with allanite around anhedral monazite; one 372 grain preserves a Th–U–Pb date of 58 ± 5 , three grains yield $15-14 \pm 2-3$ Ma and four other 373 grains have PbO below the detection limit.

374 Sample SHK2

375 Large (100 μm in diameter) and euhedral monazite grains surrounded by a thin fringe of 376 Aln–Ap with local xenotime and Th–U phases are common, but skeletal aggregates 377 associated with more abundant apatite are also observed (see Supplemental Material for BSE 378 images). Monazite mostly preserves a sector zoning pattern that is affected by micro-cracks

and locally riddled with tiny holes. One euhedral and zoned allanite (~ 500 μ m in length) is entirely included in a plagioclase grain within a Kfs phenocryst and is regarded as primary in origin. Other allanite occurrences correspond to thin (~ 50 μ m in diameter) laths next to monazite or along the cleavages of chloritized biotite. Large matrix xenotime (~ 100 μ m in diameter) shows oscillatory zoning cross-cut by BSE-dark zones. Overall, the textures are similar to those observed in SHK, with more occurrences of large euhedral monazite that might be correlated with the lesser degree of chloritization.

386 Monazite domains similar to those distinguished in SHK are identified (Fig. 4a, b). 387 The primary domains show a dominant huttonitic substitution and an increase in the monazite 388 component from the core (X_{hut} =0.04-0.15; X_{cher} =0.03-0.05; X_{mnz} =0.80-0.92) to the outermost 389 rim ($X_{hut}=0.04-0.07$; $X_{cher}=0.03-0.04$; $X_{mnz}=0.89-0.94$). A few BSE-dark secondary domains 390 could be analyzed and their composition mostly falls in a subgroup with markedly low Y 391 $(Y_2O_3 < 0.77 \text{ wt\%})$ and U (UO₂ < 0.05 wt%) contents. Only one valid analysis of a Th–U 392 phase could be obtained. Like in SHK, it is a Th-rich silicate (Fig. 4c) with a somewhat higher 393 amount of REE ($\Sigma REE_2O_3=15.30$ wt%) and PbO below the detection limit.

394 Sample NAM

Monazite occurs as subhedral, rounded or anhedral grains (~20 to 250 μ m in length) with a variable amount of inclusions (Fig. 5a). Monazite is found inside biotite, plagioclase and at the boundary between major minerals. Tiny zircon in contact with monazite is common, and only one relatively large (~100 μ m in diameter) and oscillatory-zoned xenotime is found in textural equilibrium with monazite in the matrix. Other xenotime occurrences are scarce, minute inclusions in monazite. Micro-cracks are rare and are not associated with extensive alteration of the original zoning (Fig. 5c, f).

402 Two contrasting domains are observed in both *in situ* and separated monazite. All 403 grains exhibit a primary sector- or oscillatory-zoned part that is randomly cross-cut and

404 replaced by a secondary BSE-dark and inclusion-rich domain (Fig. 5c, e, f). The proportion of 405 primary to secondary domains varies; only a few grains fully preserve their primary zoning 406 (Fig. 5d), most grains exhibit a juxtaposition of both domains, and some grains are dominated 407 by secondary domains in which only patches of primary domains remain (Fig. 5c, e, f). In 408 general, the secondary domains are preferentially developed in monazite core (Fig. 5e) but 409 examples of replacement occurring at the rim are also found (Fig. 5f).

410 Primary domains are nearly inclusion-free, whereas secondary domains are riddled 411 with inclusions that range from a few microns up to \sim 50 µm in diameter (Fig. 5a, e, f). The 412 inclusions occur as single grains or more commonly form multi-phase aggregates with a 413 rounded or tubular shape (Fig. 5g, h). A survey of 906 inclusions in 84 grains reveals the 414 presence of, in order of decreasing abundance: Zrn, Ms, Kfs, Qtz, Hem, Bt, Po, Pl and scarce 415 Chl, Ap, Xtm and Ant (Fig. 6; Supplemental Material). Among these inclusions, Zrn, Ms, Kfs, Qtz, Bt, Po, Pl, Ap and Xtm are thought to have formed during the magmatic or metamorphic 416 417 stage while Hem, Chl and Ant are considered as later alteration products. The most common 418 assemblages are Ms-Qtz and Qtz-Kfs-Pl±Bt. A significant number of Si-rich inclusions is 419 also observed (Fig. 5g, h); they appear as orange to brownish blebs under the optical 420 microscope, and their EDS spectrum shows a prominent Si peak with additional Al, K, Na, Ca 421 or Fe peaks (Fig. 6a). They are easily damaged during Raman analysis and their spectrum 422 does not reveal any discernible vibration band outside that of the monazite host (Fig. 6b).

Primary and secondary domains are clearly distinguished based on their compositions (Fig. 4, Table 2). Analyses from primary domains follow a dominant huttonitic substitution (X_{hut} =0.01-0.07; X_{cher} =0.06-0.14; X_{mnz} =0.80-0.94; 41 analyses on 29 grains), whereas those from secondary domains cluster on a narrow cheralite-monazite exchange trend (X_{hut} =0-0.02; X_{cher} =0.10-0.15; X_{mnz} =0.84-0.89; 29 analyses on 23 grains; Fig. 4a). With respect to primary domains, secondary domains are depleted in Si, LREE and enriched in Ca, Y, U, P, HREE

429 while Th remains constant. The strongest contrast is seen in the Y and U contents (Fig. 4b); 430 they are on average low in primary domains ($Y_2O_3=0.91$ wt%; $UO_2=0.19$ wt%) and 431 significantly higher in secondary ones ($Y_2O_3=2.30$ wt%; $UO_2=1.43$ wt%).

Compared to the large matrix xenotime grain, minute xenotime included in monazite is enriched in La, Ce and depleted in U, Er, Tm, Yb. Xenotime inclusions in monazite, like other minerals, always occur inside the secondary domains. Five analyses of secondary monazite domains in contact with included xenotime show a slightly varying $X_{(HREE,Y)}$ (0.089 \pm 0.018) and yield temperature estimates of 584 and 664 °C with the calibration of Pyle et al. (2001) and Heinrich et al. (1997), respectively.

438 Sample NAM2

439 Monazite is found inside Bt, Kfs or between major minerals; it mostly has a euhedral and 440 slightly rounded shape, but skeletal grains are locally present (see Supplemental Material for 441 BSE images). Monazite shows oscillatory or sector zoning cross-cut by BSE-dark zones with 442 tiny holes or inclusions. Only a few inclusions in monazite could be identified in this thin 443 section; these are Zrn, Po, Ms, Xtm, Chl, Hem, Bt, Qtz and K-Al-bearing Si-rich phases. Tiny 444 $(\leq 30 \,\mu\text{m} \text{ in length})$ grains of skeletal allanite are locally found in between the major minerals 445 or along biotite cleavages. Euhedral to anhedral matrix xenotime is common and shows sector 446 or oscillatory zoning cross-cut by BSE-dark zones next to chloritized biotite. Monazite 447 textures are globally similar to those observed in sample NAM, but one difference is the 448 limited occurrence of tiny allanite in NAM2.

Monazite analyses reveal the same contrast between primary and secondary domains as observed in NAM (Fig. 4a, b). Primary domain analyses follow a huttonitic exchange $(X_{hut}=0.02-0.08; X_{cher}=0.02-0.06; X_{mnz}=0.86-0.97)$ and have relatively low Y and U contents $(Y_2O_3=0.42-1.90 \text{ wt}\%; UO_2=0.04-0.38 \text{ wt}\%)$. Conversely, secondary domain analyses

453 indicate a cheralitic exchange (X_{hut} =0-0.04; X_{cher} =0.01-0.11; X_{mnz} =0.85-0.99) and are

454 commonly enriched in Y and U ($Y_2O_3=1.49-3.03$ wt%; UO₂=0.15-3.26 wt%).

455 MONAZITE GEOCHRONOLOGY

456 Sample SHK

457 Twenty-six analyses were performed on fifteen grains, with two spots (16.4/16.6) located on 458 cracks being rejected due to severe discordance. The remaining 24 analyses are located on 459 primary core (15 spots, U/Th \leq 0.03) and outermost rim (9 spots, U/Th > 0.03) domains, and 13 are concordant. Concordant core analyses show 206 Pb/ 238 U dates spreading from 116 ± 3 to 460 98 ± 3 Ma (106 Ma on average; Fig. 7a, b) with no clear maximum in the probability density 461 distribution. The three concordant spots from the outermost rim parts yield a weighted 462 average ${}^{206}\text{Pb}/{}^{238}\text{U}$ date of 102 ± 2 Ma (MSWD=1.13, n=3), which is similar to the lower 463 464 intercept date tentatively calculated with all outermost rim analyses in an inverse Concordia 465 diagram $(102 \pm 2 \text{ Ma}, \text{MSWD}=2.2, n=9)$.

466 Sample NAM

467 Twenty-six analyses were performed on nineteen grains, with two severely discordant spots 468 (19.8/19.13) being discarded as they are affected by the presence of an inclusion or located at 469 a grain edge. The remaining 24 analyses are clearly differentiated based on their U/Th ratio; 470 they belong to primary (11 spots, U/Th \leq 0.01) and secondary (11 spots, U/Th \geq 0.10) domains, 471 two spots (20.14/20.15) with an intermediate U/Th ratio (0.02-0.03) being considered as 472 analyses of mixed domains. Primary domain analyses show a significant degree of discordance and their ${}^{206}\text{Pb}/{}^{238}\text{U}$ dates spread between 116 ± 6 and 101 ± 6 Ma, with only one 473 concordant spot (19.15) at 106 ± 5 Ma (Fig. 7d). The latter result agrees with the weighted 474 average ${}^{206}Pb/{}^{238}U$ date of 109 ± 2 (MSWD=2.00, n=11) calculated with all primary domain 475 476 analyses. Secondary domain analyses have a better concordance; 10 spots out of 11 are

477 concordant and their ${}^{206}\text{Pb}/{}^{238}\text{U}$ dates spread from 102 ± 3 to 91 ± 2 Ma (Fig. 7d). No clear 478 maximum arises from this spread, and no correlation between ${}^{206}\text{Pb}/{}^{238}\text{U}$ date and U/Th ratio 479 is apparent. For example, the two spots with the highest U/Th ratios yield the oldest (19.3: 480 102 ± 2 Ma, U/Th=0.21) and youngest (19.11: 91 ± 2 Ma, U/Th=0.21) ${}^{206}\text{Pb}/{}^{238}\text{U}$ dates of the 481 distribution (Fig. 7c).

482 MONAZITE ALTERATION PROCESSES

The SHK and NAM granite samples have, despite contrasting textures (Fig. 2a, b), nearly similar mineral modes (Table 1), whole-rock compositions, U–Pb zircon ages (Skrzypek et al. 2016) and U–Pb monazite ages for primary domains (Fig. 7b, d). These similarities are used to demonstrate that the different monazite alteration textures are not due to initial compositional differences, but to the respective P-T and chemical conditions of the metamorphic overprintings that affected the granites.

489 Monazite replacement at low to medium temperature (Shimokuhara granite)

490 Monazite in SHK and SHK2 mostly consists of primary domains for which sector or 491 oscillatory zoning and compositional variations along a single exchange trend suggest a 492 magmatic origin (Figs. 3e and 4a). However, numerous observations indicate alteration of 493 primary monazite. The change from a euhedral to a skeletal shape points to a variable degree 494 of monazite dissolution. Allanite, apatite, xenotime and Th-U phases - i.e. secondary 495 products that partly use elements originally contained in monazite – develop directly adjacent 496 to altered monazite, along its margins or cracks (Fig. 3b, c, f). Euhedral allanite is also found 497 farther from monazite along chlorite cleavages (Fig. 3d), indicating some REE mobility since 498 monazite is the main, primary LREE-carrier in the rock. Monazite replacement by Aln-Ap is 499 dominant, but recrystallization is additionally observed in the secondary, porous domains that 500 developed in cracks or at the rim of some grains (Fig. 3b, f).

501 The formation of hydrous phases such as allanite and chlorite next to altered monazite, 502 and indications of (limited) element transport strongly suggest that monazite replacement 503 involved the action of an aqueous fluid. The secondary domains also bear similarities with the 504 textures reported for monazite alteration experiments in the presence of fluids (e.g. 505 Hetherington et al. 2010). In several of these experiments the composition of secondary 506 monazite shows LREE enrichment and Y, Ca, Th, U depletion (Harlov et al. 2011; Williams 507 et al. 2011; Grand'homme et al. 2018), as observed for the Y-poor subgroup of secondary 508 domains (Fig. 4b). The effects of aqueous fluid on monazite dissolution were investigated for 509 a wide range of P-T conditions and fluid compositions (e.g. Oelkers and Poitrasson 2002; 510 Schmidt et al. 2007; Hetherington et al. 2010; Budzyń et al. 2011; 2015; 2017; Harlov et al. 511 2011; Williams et al. 2011; Grand'homme et al. 2018; Trail 2018). The main outcome of 512 experimental studies is that alkali-bearing fluids (NaOH, KOH) more efficiently lead to dissolution and recrystallization, while observations of natural samples insist on the role of 513 514 halogens in enhancing monazite dissolution (e.g. Förster 2001; Didier et al. 2013).

515 In addition to H_2O , a source of Ca ($\pm Al$, Si) has to be identified to explain the growth 516 of Aln-Ap after monazite (Fig. 3b, c). In a similar case of magmatic monazite replacement in 517 S-type granitoids, Broska et al. (2005) proposed plagioclase (for Ca) and biotite (for Al, Si, F) 518 as likely sources. In sample SHK biotite and the Ca-rich core of large plagioclase are 519 extensively altered (Fig. 2a, c), and biotite chloritization could have released some F 520 $(\sim 0.27-0.68 \text{ wt\%})$ that was subsequently incorporated in allanite $(\sim 0.08-0.14 \text{ wt\%})$. To 521 explain monazite replacement by Aln-Ap in granitoids, Broska and Siman (1998) proposed a 522 reaction that involves anorthite breakdown but not biotite chloritization. Following the 523 minerals observed in monazite alteration textures, we use the mineral compositions measured 524 in SHK to present a stoichiometrically-balanced reaction that tries to conserve most elements 525 (except HREE-Y, Th-U and F; see Supplemental material for the calculation procedure). The

526 main uncertainty in our approach lies in the elements that potentially arrived and escaped with 527 the fluid (+Ca, Si; -F, Na, K). The proposed reaction is: 1 Mnz + 5.462 Pl + 4.041 Bt + 3.828528 $Qtz + 3.078 H_2O + 1.148 Ca^{2+} \rightarrow 1.095 Aln + 0.316 Ap + 1.317 Chl + 1 Ttn + 3.268 Kfs + 1$ Ms + 2.780 Ab (+ 0.013 HREE-Y + 0.089 Th-U + 0.300 F). The slight excess of HREE-Y 529 530 and Th–U can be accounted for by the occurrence of tiny xenotime and Th–U phases in the 531 monazite alteration textures (Fig. 3b, c, f), while the high proportions of Kfs and Ab 532 compared to those observed in the chloritization textures suggest that some K and Na are 533 instead stored in damourite or were lost in the fluid together with excess F. The major 534 alteration process can therefore be explained by a nearly balanced replacement reaction that 535 leaves magmatic monazite as a metastable relict.

536 The formation of the observed replacement textures requires an increase in Ca activity 537 and H_2O influx. At the magmatic stage, the first condition can be met and lead to monazite replacement by Aln-Ap (Dini et al. 2004), but contemporaneous chlorite growth is unlikely. 538 539 Conversely, both conditions are satisfied by hydrothermal alteration for which plagioclase 540 resorption and biotite chloritization are commonly reported (e.g. Alderton et al. 1980; 541 Yuguchi et al. 2015). The results of Mnz-Xtm thermometry suggest that the rare secondary 542 domains in SHK formed at 471-562 °C. This range agrees with temperature estimates for the regional overprint in the neighboring schist zone (450-550 °C; Ikeda 2004), but the upper 543 544 temperature limit conflicts with the widespread occurrence of chlorite. This could suggest that 545 limited monazite recrystallization actually occurred at lower-amphibolite facies conditions (> 546 500 °C), whereas later monazite replacement was due to a more pervasive hydrothermal 547 alteration of the granite at greenschist facies conditions (< 500 °C). Similar monazite 548 replacement textures were reported in rocks that experienced peak metamorphic temperatures 549 of 340 °C (Poitrasson et al. 2000), 550-600 °C (Finger et al. 1998) and up to 685-825 °C 550 (Broska et al. 2005). Although the P-T conditions at which monazite replacement occurs are

likely controlled by the whole-rock Ca and Al contents (e.g. Spear, 2010), we propose that the textures can hint at a low- or high-grade origin. The presence of chlorite, fractured monazite and allanite growing farther from monazite would indicate the dominance of pervasive fluid circulation at greenschist facies conditions, whereas the formation of Aln–Ap coronas should reflect the increasing role of diffusion at higher metamorphic grade (Broska et al. 2005; Finger et al. 1998; 2016; Ondrejka et al. 2012).

557 Monazite recrystallization at high temperature (Namera granite)

558 Monazite is the dominant LREE-rich phase observed in samples NAM and NAM2. It is 559 composed of sector- and oscillatory-zoned primary domains whose composition lies on the same huttonitic trend as that defined by primary domain analyses from sample SHK (Figs. 4a 560 561 and 5d). This supports the magmatic origin of monazite in both samples. A significant part of monazite is also occupied by secondary domains enriched in Ca, Y, U and P (Fig. 4). The 562 563 formation of secondary domains generally preserves the shape of the original magmatic grain, 564 and it is necessary to clarify whether this pseudomorphic recrystallization occurred at the 565 solid state or under the action of a fluid phase.

566 Primary and secondary domains are separated by a sharp boundary on BSE images, 567 and their composition is clearly distinct (Figs. 4 and 5c, e, f). This stands against 568 intracrystalline diffusion, which should result in more gradual changes in terms of texture (see 569 the diffusion zone imaged by Gardés et al. 2006) and composition (e.g. Cherniak et al. 2004). 570 In addition, BSE images show that recrystallization does not necessarily start from the rim 571 and proceed inwards, but that a preferential recrystallization of the monazite core is dominant 572 (Figs. 5e and 7c). The latter observation suggests that a mobile medium was able to make its 573 way up to the more unstable core, possibly through precursor cracks. Thus, the action of a 574 fluid phase is inferred to explain the selective dissolution of primary monazite. Dissolution 575 was selective; it is striking that the primary domains have a narrow compositional range,

although a larger spread might be expected by analogy with the composition of primary monazite in sample SHK, which has the same bulk composition as sample NAM (Fig. 4a). The primary domains that are presently observed must have been the most robust parts during dissolution, whereas we suppose that the originally Th–U-richer monazite core was more reactive, more porous to allow fluid circulation, hence preferentially dissolved. The different solubilities of the initial domains could have been due to a higher degree of disequilibrium with respect to the fluid, and/or to denser radiation damage in the supposedly Th–U-rich core.

583 Dissolution of the primary domains had to be followed by precipitation of secondary 584 monazite at exactly the same place in order to explain the pseudomorph texture (Fig. 5f). Both 585 characteristics are accounted for by the dissolution-reprecipitation process detailed by Putnis (2002). A key aspect of this process is that a fluid-mineral interface has to be maintained so 586 587 that recrystallization can proceed (Putnis and Putnis 2007). In samples NAM and NAM2, recrystallization corresponds to the transformation of primary huttonitic monazite into 588 589 secondary cheralitic monazite (Fig. 4a). According to the unit cell parameters of natural 590 monazite-(Ce) (Ni et al. 1995), huttonite (Taylor and Ewing 1978) and Ca_{0.5}Th_{0.5}PO₄ (Terra et 591 al. 2008), this transformation should result in a unit cell volume decrease of at least $\sim 0.14\%$. 592 Although limited, the volumetric decrease indicates that a porosity could be maintained and 593 progressively filled by inclusions, the latter being distinctively abundant in the secondary 594 domains (Fig. 5a, e, f).

The major factor controlling the spatial and temporal extent of the dissolutionreprecipitation process is the fluid composition (Putnis 2009). In the case of NAM and NAM2 samples, it was a fluid able to initially dissolve monazite and to reprecipitate not only new monazite but also the whole range of inclusions that are found in the secondary domains (Zrn, Ms, Kfs, Qtz, Bt, Po, Pl, Ap, Xtm). Similar inclusions are also reported in zircon from the same sample (Qtz, Pl, Kfs, Ms, Bt, Hem, Ap, Xtm; Skrzypek et al. 2016). Because of these

601 constraints, the fluid medium is thought to be anatectic melt. Dissolution, transport and 602 reprecipitation of zircon, apatite or xenotime in the presence of alkaline fluids has been 603 reported (e.g. Tomaschek et al. 2003; Rubatto et al. 2008; Harlov 2011; Broom-Fendley et al. 604 2016), but at low pressure the solubility of both monazite and zircon is higher in granitic melt 605 than in aqueous fluid (e.g. Montel 1986; Watson 1979; Ayers and Watson 1991). In addition, 606 the rounded or tubular shape (Fig. 5g, h) and the nature of the most abundant multi-phase 607 inclusions in recrystallized monazite (Si-rich (melt?), Qtz-Kfs-Pl, Qtz-Ms assemblages) 608 strongly suggest that a hydrous granitic melt crystallized in the pores. A similar texture with 609 numerous and tiny multi-phase inclusions is reported for metamorphic monazite that grew in 610 the presence of leucogranitic melt (Bea and Montero, 1999).

611 The origin of the anatectic liquid that is thought to have favored monazite 612 recrystallization is debatable. Although small amounts of partial melt are difficult to 613 recognize, the Ms-Qtz network texture observed in thin section (Fig. 2d) might be evidence 614 of internal or external melt percolation. On the one hand, regional metamorphism in the 615 neighboring Kfs-Crd zone peaked at 2-3 kbar/590-675 °C (Ikeda 2004) and locally reached 616 the range of H₂O-present melting reactions for granitic compositions (e.g. Weinberg and 617 Hasalová 2015). The Namera granite might therefore have experienced a small degree of in 618 situ wet melting, which produced a liquid that could dissolve monazite, zircon and xenotime. 619 The latter mineral, which is scarcely observed in the matrix and relatively U-rich, represents a 620 likely source for U and Y enrichment in recrystallized monazite. On the other hand, 621 metasedimentary rocks in the Kfs-Crd zone show leucocratic layers that are ascribed to 622 partial melting (Brown 1998; Kawakami and Ikeda, 2003), and this external melt could 623 equally have infiltrated the Namera granite. The presence of undeformed Grt-bearing 624 leucocratic veins and Qtz-Kfs-Ms pegmatite on NAM outcrop supports this view, but a 625 similar observation is unfortunately not made on NAM2 outcrop. Melt provided by the 626 metasedimentary rocks would also have the advantage of being in greater disequilibrium with 627 the granitic assemblage, thereby enhancing monazite dissolution. It is therefore proposed that 628 monazite recrystallization occurred at 584–664 °C and was facilitated by the infiltration of 629 melt possibly derived from the neighboring metasedimentary rocks.

630 CONSEQUENCES FOR MONAZITE GEOCHRONOLOGY

631 Preservation of magmatic age

632 In monazite from sample SHK, the primary core and outermost rim domains show no difference in ²⁰⁶Pb/²³⁸U date, and concordant rim analyses yield a weighted average ²⁰⁶Pb/²³⁸U 633 634 date of 102 ± 2 Ma (Fig. 7a, b). In sample NAM, the only concordant analysis of primary 635 monazite domain yields 106 ± 5 Ma (Fig. 7d). Both results are within error of the U-Pb 636 zircon ages of 105 ± 3 Ma and 105 ± 2 Ma derived from the same SHK and NAM samples, 637 respectively (Skrzypek et al. 2016). These age similarities, together with well-defined sector 638 and oscillatory zoning patterns, support a magmatic origin for both zircon and monazite, and 639 indicate that the age of magmatic crystallization is preserved in the primary monazite 640 domains. Previous works similarly reported that monazite may partly retain its magmatic age in meta-igneous rocks metamorphosed up to \sim 700 °C, despite subsequent alteration (Finger et 641 642 al. 1998, 2016; Townsend et al. 2000). This view appears fully valid for sample SHK in which 643 the greenschist facies regional overprint did not lead to total monazite replacement, which 644 allows the magmatic age to be measured with a 10 µm laser spot. The magmatic age is less 645 well preserved in monazite from sample NAM, but is still accessible with the analytical 646 approach used in this study. The main observation is that the primary domain analyses are commonly discordant (Fig. 7d). Given that such a discordance is not observed in SHK, it is 647 648 suspected that the upper-amphibolite facies metamorphic overprint was responsible for some 649 disturbance of the primary isotopic system in NAM. The BSE images of primary domains,

however, do not hint at a perturbation of the original, magmatic sector zoning (Figs 5 and 7c).

651 Identification of alteration age

652 Age dating of the secondary monazite domains in SHK was not attempted due to their small 653 size. In trying to analyze similar narrow veinlets by ion probe, Townsend et al. (2000) could 654 obtain nothing but mixed ages. Moreover, two spots located on micro-cracks show that severe 655 discordance is to be expected for the secondary domains (spot 16.6 in Fig. 7a). A few analyses 656 of the Th–U phases found next to altered monazite yield one Th–U–Pb date of 58 ± 5 and 657 three of $15-14 \pm 2-3$ Ma, with four nearly Pb-free spots (Fig. 4d). Because the Th–U phases are tiny anhedral grains that are thought to host ~ 10 wt% H₂O, the probability of radiogenic 658 659 Pb loss is high and the Th–U–Pb dates should be treated with extreme caution. We can simply 660 remark that the oldest Th–U–Pb date (58 ± 5 Ma) agrees with the apatite fission track age of 661 57.4 ± 5 Ma that is reported for a granodiorite sample in the southern part of the Iwakuni-662 Yanai area and ascribed to final exhumation of the Ryoke belt (Okudaira et al. 2001). It is 663 therefore possible that monazite replacement associated with chloritization actually occurred 664 during the Paleogene, when granitic rocks were ultimately brought closer to the surface.

665 The extensive recrystallization of secondary monazite in NAM gives the opportunity to date the new domains in situ. However, the age of recrystallization is difficult to constrain 666 from the spread of concordant ${}^{206}\text{Pb}/{}^{238}\text{U}$ dates (102 ± 3 to 91 ± 2 Ma) that is obtained for 667 668 secondary domains (Fig. 7d). A first interpretation would be to consider all results as true 669 recrystallization ages, in which case a ca. 10 Ma duration could be ascribed to the whole 670 dissolution-reprecipitation process. A second interpretation is to consider that the results 671 reflect isotopic mixing between the primary and youngest secondary compositions. This view 672 is supported by several examples of Pb incorporation and incomplete isotopic resetting during 673 monazite recrystallization by dissolution-reprecipitation (Teufel and Heinrich 1997; SeydouxGuillaume et al. 2002, 2012; Grand'Homme et al. 2016). In such a case, the youngest $^{206}Pb/^{238}U$ date of 91 ± 2 Ma can be considered as the minimum age for monazite recrystallization.

677 Resolving the zircon-monazite age discrepancy and the age of high-T metamorphism

678 No discrepancy between U–Pb zircon (105 ± 3 Ma; Skrzypek et al. 2016) and monazite dates 679 $(102 \pm 2 \text{ Ma})$ is revealed for sample SHK, because monazite recrystallization remains limited. 680 The CHIME monazite dates of 87.3 ± 1.6 and 86.6 ± 2.1 Ma reported for the Shimokuhara 681 granite (Suzuki et al. 1996) were obtained from a sample collected at the northern edge of the 682 pluton, next to a fault zone (K. Suzuki, personal communication). The best way to explain 683 these younger CHIME results is to assume that monazite recrystallization was more 684 pronounced in that sample, so that the CHIME dates actually constrain monazite recrystallization during regional metamorphism in the Ms-Crd zone (~500-550 °C; Ikeda, 685 686 2004). Conversely, dominant monazite replacement in SHK should be associated with lower 687 temperature chloritization (< 500 °C), and the Th–U–Pb dates obtained from Th–U phases 688 next to replaced monazite suggest that this alteration stage might be significantly younger 689 (Paleogene ?) and unrelated to regional metamorphism.

690 For sample NAM, the U-Pb age of secondary monazite domains (102-91 Ma) can be 691 up to ~ 10 Ma younger than that of magmatic monazite (106 ± 5 Ma) and zircon (105 ± 2 Ma; 692 Skrzypek et al. 2016). The comparison between our monazite analyses and those of Suzuki et 693 al. (1996) clearly shows that the CHIME dates ($91-89.9 \pm 3.2$ Ma; Suzuki et al. 1996) were 694 calculated by grouping spots from primary and secondary domains. Because the secondary 695 domains are U-rich and thus have a lower analytical uncertainty, they presumably had more 696 weight in the calculation of the Th* vs. Pb isochron, which led to results of 91–90 Ma (Suzuki 697 et al. 1996). As confirmed by our combined petrological and geochronological data, the age of

ca. 91 Ma cannot be ascribed to magmatic crystallization but rather reflects monaziterecrystallization during a subsequent metamorphic overprint.

700 The present work proposes that the Kfs-Crd zone was still subjected to suprasolidus 701 conditions at \sim 91 Ma, i.e. about 10 Ma later than in the deeper Grt–Crd zone (U–Pb zircon 702 ages of 103–97 Ma; Nakajima et al. 2013; Skrzypek et al. 2018). It is another argument for 703 the diachronous origin of the metamorphic field gradient exposed in the Ryoke belt (Skrzypek 704 et al. 2016), and agrees with the persistence of medium-T conditions until 89 ± 5 Ma, as 705 suggested by U-Pb monazite ages obtained in a gneiss sample from the Kfs-Crd zone 706 (Skrzypek et al. 2018). The accumulation of petrochronological data reveals that in the Ryoke 707 belt – the type locality for low-P/high-T metamorphism – the age at which high-T conditions 708 ended seems to become younger with decreasing depth in the metamorphic section.

709 CONCLUSIONS AND IMPLICATIONS

We investigated two plutons for which the selected hand specimens appear as fresh, massive or deformed biotite-granites. Under the microscope, the major minerals are not significantly altered, except for plagioclase damouritization and variable biotite chloritization in the massive granite, and the occurrence of Qtz–Ms veinlets and pinitization in the deformed granite. However, the shape and/or composition of magmatic monazite is commonly altered in both granites. The versatility of this accessory mineral offers a unique chance to constrain the nature and timing of post-magmatic metasomatic events that are not easily detected otherwise.

717 Monazite alteration textures – clues to metasomatic conditions

The two plutons arguably have a similar composition, but differences in their post-magmatic histories led to contrasting monazite alteration processes. Depending on the response of the granitic assemblage to post-emplacement P-T and chemical conditions, the interaction between the metamorphosed major and accessory minerals and a fluid medium will result in monazite replacement or recrystallization. On the one hand, the hydration of a granitic

723 assemblage at low temperature (≤ 500 °C) will trigger the alteration of Ca-plagioclase and the 724 chloritization of biotite. This will enrich the externally-derived aqueous fluid in Ca+Si+Al±F 725 and favor monazite replacement by allanite and apatite. We introduce a new reaction that 726 accounts for this process and that is expected in most monazite-bearing granitoids affected by 727 greenschist facies hydrothermalism. On the other hand, a limited amount of hydrous melt, 728 probably of external origin and percolating through the granitic assemblage, can trigger 729 monazite recrystallization by dissolution-reprecipitation at upper-amphibolite facies 730 conditions (> 600 °C). The recrystallized monazite domains are expected to host inclusions, 731 and their new composition will be controlled by the dissolution of other phases in the 732 presence of melt, with for instance Y and U enrichment reflecting xenotime resorption.

733 Monazite alteration textures – a chance for dating the thermal history

734 Age dating of monazite alteration textures can yield information on both the magmatic and 735 subsequent alteration stages. Monazite replacement leaves primary domains that show no 736 detectable modification of the isotopic system and preserve the age of magmatic 737 crystallization. The minute Th-U-rich alteration products of monazite can additionally yield 738 precise constraints on the timing of low-T metasomatism. Partial monazite recrystallization 739 causes a disturbance of the isotopic system in primary domains and limits the access to the 740 magmatic crystallization age. It also produces secondary domains that are large enough to be 741 analyzed, but in which an incomplete resetting of the primary isotopic signature complicates 742 the interpretation of the alteration age. The recrystallized domains nevertheless provide a 743 minimum age for the end of high-T conditions. Regardless of the process involved, monazite 744 alteration textures offer the possibility to date precise steps of a high- to low-T, post-magmatic 745 thermal evolution.

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1026 FIGURE CAPTIONS

FIGURE 1. Geological map of the Ryoke belt in the Iwakuni-Yanai area (SW Japan), with sampling localities. Inset shows the area within the Inner Zone of SW Japan (MTL=Median Tectonic Line). Granitoids are classified according to their emplacement timing relative to the main, regional tectono-metamorphic event (D1). Lithologies are after Sato (1933), Higashimoto et al. (1983) and the Geological Survey of Japan (2012). Metamorphic zone boundaries are after Ikeda (1993). Schist/gneiss boundary modified after Higashimoto et al. (1983). Mineral abbreviations follow IUGS recommendations after Kretz (1983).

FIGURE 2. Microscopic texture of SHK and NAM samples on (**a**, **b**) EMP scans of thin sections and (**c**, **d**) optical microscope photographs. (**a**) Sample SHK showing rounded Qtz aggregates, Kfs–Pl with straight margins and variably chloritized Bt. (**b**) Sample NAM showing elongated Qtz layers, aligned Bt and Kfs–Pl with irregular margins. The trace of the gneissosity is subparallel to the long axis of the thin section. (**c**) Occurrence of Mnz and Aln in relationship with variably chloritized Bt. SHK, right-hand side of the thin section, plane-

1040 polarized light. (d) Ms–Qtz network inside fragmented Kfs, and Mnz grains inside Bt. NAM,

1041 bottom left-hand side of the thin section, crossed polarizers.

1042 FIGURE 3. Monazite textures in sample SHK. (a) Optical microscope photographs of variably 1043 altered, separated monazite grains. White crosses indicate the location of laser pits. (b-f) BSE 1044 images of *in situ* textures and separated grains. (b) Euhedral monazite cross-cut by cracks and 1045 surrounded by Ap, Aln, Xtm and a Th–U phase next to chloritized Bt. (c) Anhedral monazite 1046 surrounded by zoned allanite, Th-U phase, Xtm and chloritized Bt. (d) Oscillatory zoned 1047 allanite protruding into the cleavages of chloritized Bt. Allanite is separated from chlorite by 1048 Al-rich titanite. (e) Virtually unaltered monazite with a sector-zoned core and a darker 1049 outermost rim. Limited alteration (darkening) is visible in the bottom right hand part. (f) 1050 Altered monazite with Ap, Aln, Xtm, Th–U phase, Chl and Bt present along micro-cracks. 1051 Recrystallization of secondary domains occurred around cracks (darkened parts) and veinlets 1052 (brightened parts). Contrast is adjusted to see the secondary minerals (left) and internal zoning 1053 (right). Same grain as on the right hand photograph in (a).

1054 FIGURE 4. Monazite composition in samples from the Shimokuhara (SHK, SHK2) and 1055 Namera (NAM, NAM2) granites. Data are from *in situ* and separated grains. (a) REE+Y+P 1056 vs. Th+U+Si diagram (cations for 16 oxygens) highlighting the dominant huttonic substitution 1057 for primary domain analyses in both samples, and the dominant cheralitic substitution for 1058 secondary domain analyses in NAM. (b) Y vs. U diagram (cation fraction on A site) showing 1059 Y-U depletion for some secondary domain analyses in SHK, and Y-U enrichment for 1060 secondary domain analyses in NAM. (c) Ternary diagram showing proportions of the 1061 monazite [REEPO₄], "cheralite" [Ca_{0.5}(Th,U)_{0.5}PO₄] and "huttonite" [(Th,U)SiO₄] end-1062 members for analyses of monazite (all samples) and Th-U phases (SHK & SHK2).

1063 **FIGURE 5.** Monazite textures in sample NAM. (a) Optical microscope photographs of 1064 variably altered, separated monazite grains. White crosses indicate the location of laser pits.

1065 (b-f) BSE images of *in situ* textures and separated grains. (b) Typical mode of occurrence of 1066 monazite inside matrix Bt, with (c) an internal structure showing relicts of a sector- or 1067 oscillatory-zoned primary domain cross-cut by a darker secondary domain. (d) Virtually 1068 unaltered monazite with a dominant oscillatory-zoned primary domain. A convoluted 1069 secondary domain is slightly developed in the bottom part. (e) Altered monazite with a dark 1070 and inclusion-rich secondary domain preferentially developed in the core (included minerals 1071 indicated). Isolated "islands" of primary monazite are left inside the secondary domain. (f) 1072 Altered monazite with a dark and inclusion-rich secondary domain developed at the grain 1073 margin (included minerals indicated). The right hand part preserves primary oscillatory 1074 zoning. (g-h) Close-up view of rounded and tubular, multi-phase inclusions in monazite.

FIGURE 6. Characterization of selected inclusions in secondary monazite domains from sample NAM. (**a**) Energy dispersion spectra with the most intense element peaks labelled. The analyzed inclusions are shown in Fig. 5g, h. (**b**) Raman spectra with the shift of the main vibration bands labelled. The analyzed inclusions are different from those in (**a**). All spectra are background-corrected and normalized to the maximum intensity of the most intense spectrum in each set (vertical intensity scale is arbitrary). The spectra of host monazite are shown for comparison.

FIGURE 7. Results of LA-ICP-MS monazite dating in (**a**,**b**) sample SHK and (**c**,**d**) sample NAM. (**a**, **c**) BSE images of analyzed monazite grains with representative results. Each spot location is labelled with spot name, 206 Pb/ 238 U date $\pm 2\sigma$ error (Ma) and concordance (%). Scale bar is always 20 µm. (**b**, **d**) Concordia diagrams showing concordant (cc=97–103%) and discordant (cc < 97% or cc > 103%) analyses for the different monazite domains. Graycoded symbols according to U/Th ratio are added in the centre of error ellipses for sample NAM.

 Table 1. Results of mineral mode calculations

	Or	Ab	An	ΡI	Qtz	Bt	Chl	Ms	Grt	Ар	Mnz	Ttn*	Hem	llm	Crn	Sum	Rest
Mesonorm																	
SHK	17.4	29.5	10.2	39.7	31.8	7.1	-	-	-	0.17	-	-	-	0.21	2.11	98.15	-0.26
NAM	15.3	30.7	8.8	39.6	33.9	6.9	-	-	-	0.24	-	-	-	0.19	2.31	98.22	-0.26
EMP mapping											ND						
SHK	24.7	-	-	30.2	32.7	3.6	4.1	-	0.07	0.05	0.01	0.36	-	-	-	95.71	4.29
NAM	19.1	-	-	38.6	31.2	7.1	-	1.4	0.01	0.05	0.01	-	low	-	-	97.48	2.52
Note: * Titanite mode includes allanite in the case of EMP mapping; <i>low</i> =<0.01; ND = no data.																	

Sample		SH	NAM			
Spot	99a-m41	99a-m34	99a-Mnz9	99a-Ac2	117a-m35	117a-Mnz17
Domain	primary core	primary rim	secondary	Th-U phase	primary	secondary
Wt% oxides						
SiO ₂	2.30	0.60	0.63	18.16	0.70	0.44
P_2O_5	27.31	29.70	29.39	0.78	29.63	29.29
CaO	1.08	0.83	0.11	1.26	0.86	1.30
ThO ₂	14.62	5.89	2.69	62.77	6.88	6.84
UO ₂	0.73	0.45	d.l.	5.64	0.15	0.45
Y_2O_3	2.53	3.22	0.74	1.10	0.65	1.46
La ₂ O ₃	11.35	13.31	12.80	0.195	16.90	11.50
Ce_2O_3	22.28	26.60	30.06	0.32	29.77	26.58
Pr ₂ O ₃	2.53	2.82	3.72	d.l.	2.92	3.14
Nd_2O_3	10.88	11.08	13.41	0.34	10.56	11.49
Sm ₂ O ₃	2.28	2.51	3.45	d.l.	1.27	3.19
Gd_2O_3	2.09	2.30	2.43	0.35	0.70	2.01
Tb_2O_3	n.a.	n.a.	0.19	n.a.	n.a.	0.19
Dy ₂ O ₃	n.a.	n.a.	0.57	n.a.	n.a.	0.61
Ho ₂ O ₃	n.a.	n.a.	d.l.	n.a.	n.a.	d.l.
Er ₂ O ₃	n.a.	n.a.	d.l.	n.a.	n.a.	0.14
Tm_2O_3	n.a.	n.a.	0.34	n.a.	n.a.	0.29
Yb ₂ O ₃	n.a.	n.a.	d.l.	n.a.	n.a.	d.l.
PbO	0.04	0.02	d.l.	0.198	d.l.	0.02
Total	99.98	99.34	100.53	91.12	100.97	98.92
Cations						
Si	0.366	0.094	0.099	0.988	0.109	0.069
P	3.681	3.932	3.912	0.036	3.908	3.934
Са	0.184	0.138	0.018	0.073	0.143	0.221
U	0.030	0.210	0.090	0.068	0.244	0.247
Ŷ	0.214	0.268	0.062	0.032	0.053	0.123
La	0.667	0.768	0.742	0.004	0.971	0.673
Ce	1.299	1.523	1.730	0.006	1.698	1.544
Pr	0.147	0.161	0.213	-	0.166	0.182
Sm	0.019	0.019	0.755	0.007	0.566	0.051
Gd	0.110	0.119	0.127	0.006	0.036	0.106
Tb	-	-	0.010	-	-	0.010
Dy	-	-	0.029	-	-	0.031
Ho	-	-	-	-	-	-
Er Tm	-	-	- 0.017	-	-	0.007
Yb	-	-	-	-	-	-
Pb	0.001	0.001	-	0.003	-	0.001
Total	7.968	7.985	7.995	2.000	7.990	8.003
Xmnz	0.811	0.908	0.971	0.059	0.901	0.879
Xcher	0.094	0.070	0.009	0.150	0.072	0.110
	0.095	0.022	0.020	0.792	0.027	0.011
T (°C) ^a	-	-	482	-	-	533
۲ (°C)	-	-	572	-	-	618

Table 2. Representative analyses of monazite domains and Th-U phase

Notes: n.a. = not analyzed; d.l. = below detection limit

Monazite: cations normalized to 32 charges; Th-U phase: normalization to 2 cations

^aCalibration after Pyle et al. (2001)

^bCalibration after Heinrich et al. (1997)



Figure 1

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Figure 3

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